

Study on magnon polariton in thin ferromagnetic film

Shugo YOSHII

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Shugo YOSHII

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Abstract of the dissertation

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Magnon-polariton, a strong coupling state of a quantized spin wave, magnon, and microwave photon has rapidly gained attention over the last decade. When the coupling strength is enhanced to exceed the relaxation time, the coupling state is “strong” coupling state, where the magnon and photon modes coherently exchange their energy with each other. A further, more strong coupling state, designated as "ultrastrong" coupling state, has also been demonstrated in magnon-polariton systems, wherein the rotating wave approximation is no longer applicable and counter-rotating terms emerge within the Hamiltonian coupling system. The counter-rotating term is necessary to realize the squeezing state and the entanglement of coupling system, which is the crucial excitation process in modern physics. While ultrastrong coupling has been achieved in magnon polaritons, that remains in few reports, and these coupling systems lack the freedom and simplicity to explore the fundamental physics in the ultrastrong coupling regime.

This dissertation demonstrates the ultrastrong coupling system of high- T_C superconducting resonator and thin ferromagnetic films, which has many tuning parameters and room for exploration of physics in ultrastrong coupling regime. To demonstrate this result, the author investigated the guiding principle to decrease the magnon and photon relaxation and finally achieved ultrastrong coupling system in thin ferromagnetic films. In addition, the author investigated the fundamental physics in ultrastrong coupling regime by using magnon polaritons.

In Chapter 1, the author briefly introduced magnonics, magnon polariton, the research background, and the purpose of this study.

This dissertation used several fundamental theories and phenomena of magnon and microwave photon to understand and investigate the physics of magnon-polariton in thin ferromagnetic films. In Chapter 2, the author introduced the theoretical essence to understand the physics in magnons and photons, and

Hamiltonian to describe the magnon polariton with counter-rotating term and A^2 term.

Wisdom for control of magnon damping in thin ferromagnetic films is crucial to design magnon polaritons with the desired relaxation rate of magnons. In Chapter 3, the author investigated the contribution to magnon damping in thin ferromagnetic films. Magnon damping in ferromagnetic materials is characterized by Gilbert damping constant, the phenomenologically introduced parameter. The intrinsic and extrinsic contributions to Gilbert damping in thin ferromagnetic films have been explored for a few decades. These days, magnons in ultrathin regime (< 2 nm-thick) has been attracting attention because of its tunability. Nevertheless, the phenomenon of magnon scattering at the surface has not been fully elucidated, despite the fact that it can be a dominant process, particularly in ultra-thin ferromagnetic films. Some theoretical models of typical magnon scattering at the ferromagnetic surface, namely two-magnon scattering, indicate that the surface magnetic properties, such as surface magnetic anisotropy, can exert a significant influence on two-magnon scattering. Nevertheless, there is a paucity of awareness regarding the experimental guiding principle of tunability of two-magnon scattering via control of surface magnetic anisotropy. The author conducted a comprehensive investigation into the influence of surface magnetic anisotropy on two-magnon scattering in a range of typical ferromagnetic materials, including Co and CoFe. Their findings revealed a fundamental principle for regulating magnon damping in thin ferromagnetic films.

The highly confined photon mode with a small volume enables to enhance the strong coupling strength between magnons and photons. Since the superconducting resonator on a chip has a high-quality factor, where photon mode is excited on the surface of the superconducting resonator, the superconducting resonator should be ideal photon media for strong coupling with magnon in thin ferromagnetic films. In Chapter 4, the superconducting resonator made of $\text{YBa}_2\text{Cu}_3\text{O}_7$ (YBCO) with a high critical temperature of about 90 K is established, and a high-quality factor of about 4000 is obtained. Expecting the stronger confinement of photon mode, the lumped element resonator is adapted. Optimizing the measurement setup and fabrication process is needed to achieve the superconducting resonator. Designing the sample box for installing the YBCO resonator enables the realization of the high-quality factor. While the external magnetic field and the stray magnetic field of ferromagnetic films could violate the superconductivity, the robustness of the YBCO resonator to temperature and magnetic field promises the ideal environment for coupling with ferromagnet. Microwave transmission measurement with different temperature and magnetic field provides the strong robustness of YBCO resonator.

Combining superconducting resonator and thin ferromagnetic films, the creation of magnon-polariton in thin ferromagnetic films is expected. In Chapter 5,

strong magnon-photon coupling in YBCO resonator system with a 30 nm-thick NiFe alloy (Py) bar is demonstrated, and ultrastrong coupling regime in multiple of Py bar with the same thickness is achieved by utilizing enhancement of coupling strength through the magnon bright mode. Py films with the thickness of 30 nm was fabricated on YBCO-LC resonator and magnetic field dependence of transmission spectra S_{12} was measured for visualizing the coupling state between magnon mode and photon mode. The anti-crossing of magnon and photon, signature of strong coupling state, was shown in the spectra for single, which coupling strength is 140 MHz. Increasing the number of Py bars with the condition that the magnon bright mode couples with a uniform photon mode enlarges the magnon mode volume coupled with photon mode. The coupling strength of YBCO resonator with multiple bars was 671 MHz, where the ratio of resonance frequency ω_r . Moreover, coupling strength g , $\frac{g}{\omega_r}$ is over 0.1, which is in ultrastrong regime. Bloch-Siegert Shift, resonance frequency shift due to counter rotating terms in coupling Hamiltonian, was observed in multiple structure while the shift is negligible in single Py structure. This observation is the critical evidence of the existence of the counter-rotating terms in magnon polaritons.

In Chapter 6, this dissertation was summarized.

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Chapter 1. Introduction

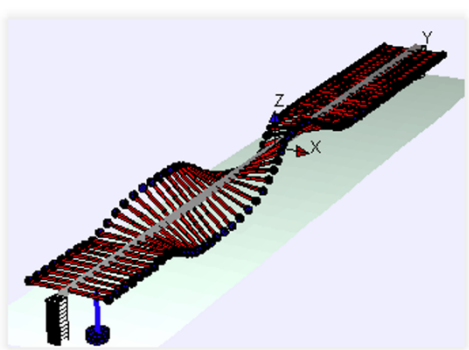
1.1 Abstract

From the end of the 19th century to the early 20th century, the work of Pierre Curie and Pierre Weiss established the Curie-Weiss law, which explains the temperature-dependent behavior of the collective magnetic moment in materials. It was recognized that strong magnetic materials, ferromagnets, have spontaneous magnetization below a critical temperature known as the Curie temperature. A few decades after the studies of Curie and Weiss, Lev Landau and Evgeny Mikhailovich Lifshitz theoretically predicted the existence of collective excitations of magnetization, known as ferromagnetic resonance. In 1946, ferromagnetic resonance was demonstrated by Griffiths's experiment. This is the starting point for studying collective magnetization dynamics in ferromagnetic materials, which have been studied as spin-wave. Recently, quantized spin-waves, or magnons, have been a fascinating research target for novel information technologies. In this chapter, the author briefly introduces magnonics and the research background of this dissertation.

1.2 Magnonics

Ferromagnetism is one of condensed matter physics' most fundamental and well-studied phenomena. It is the basis for many technological applications, from data storage devices to magnetic sensors. In a ferromagnetic material, atomic magnetic moments, which arise primarily from electron spin, tend to align parallel to each other due to the exchange interaction. This results in spontaneous magnetization even in the absence of an external magnetic field. The collective behavior of these spins gives rise to unique magnetic properties, such as hysteresis and domain formation, which are essential for the functionality of modern magnetic materials. In addition to their static magnetic properties, ferromagnetic materials are capable of generating dynamic excitations of collective magnetizations, known as spin waves, when subjected to external forces such as thermal and electromagnetic waves. These waves arise from the precessional motion of individual spins in a coordinated manner, propagating through the material like waves on a string. The quanta of these spin waves are called magnons, a type of quasiparticle that embodies the collective excitation of the spin system. Magnons are bosonic particles and can be described using quantum mechanics, with each magnon corresponding to a discrete energy state [1]. The quantization of spin waves and their collective properties place magnons at the forefront of modern condensed matter physics research.

(a) Mechanical wave on a string



(b) Spin wave

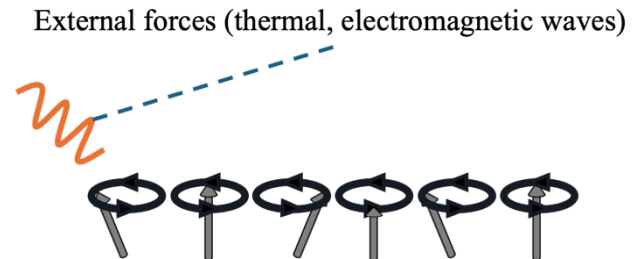


Figure 1-1. Schematics of (a) mechanical waves on a string and (b) collective magnetization dynamics, spin waves, excited by external forces such as thermal and electromagnetic waves. The image in (a) is quoted by [2].

The study of magnons and their potential applications has given rise to the rapidly developing field of magnonics [1,3–7]. Magnonics, much like its electronic counterpart, aims to control and utilize quasiparticles—magnons—within magnetic systems for information processing. The field holds promise as an alternative to traditional electronics, offering potentially lower power consumption and faster signal transmission by leveraging the unique properties of magnons. Magnons can propagate over relatively long distances without charge transport, eliminating Joule heating—a key limitation in conventional electronics [8–12]. This makes magnonics an attractive candidate for developing low-power, high-speed

information technologies, including memory and logic devices [13,9,14–16]. In magnonic systems, magnons can be excited and manipulated through various external stimuli, such as magnetic fields, electric currents, and thermal gradients. The frequency and wavelength of magnons can range from gigahertz to terahertz, making them suitable for a broad spectrum of communication and data processing applications. Significantly, magnons can also interact with other quanta, such as phonons (lattice vibrations) [17,18,18–23], photons (electromagnetic wave) [24–29] and magnons [30–33], opening the door to hybrid quantum technologies that could integrate magnonics, spintronics, phononics, and photonics with magnonics as depicted in Fig. 1.2.

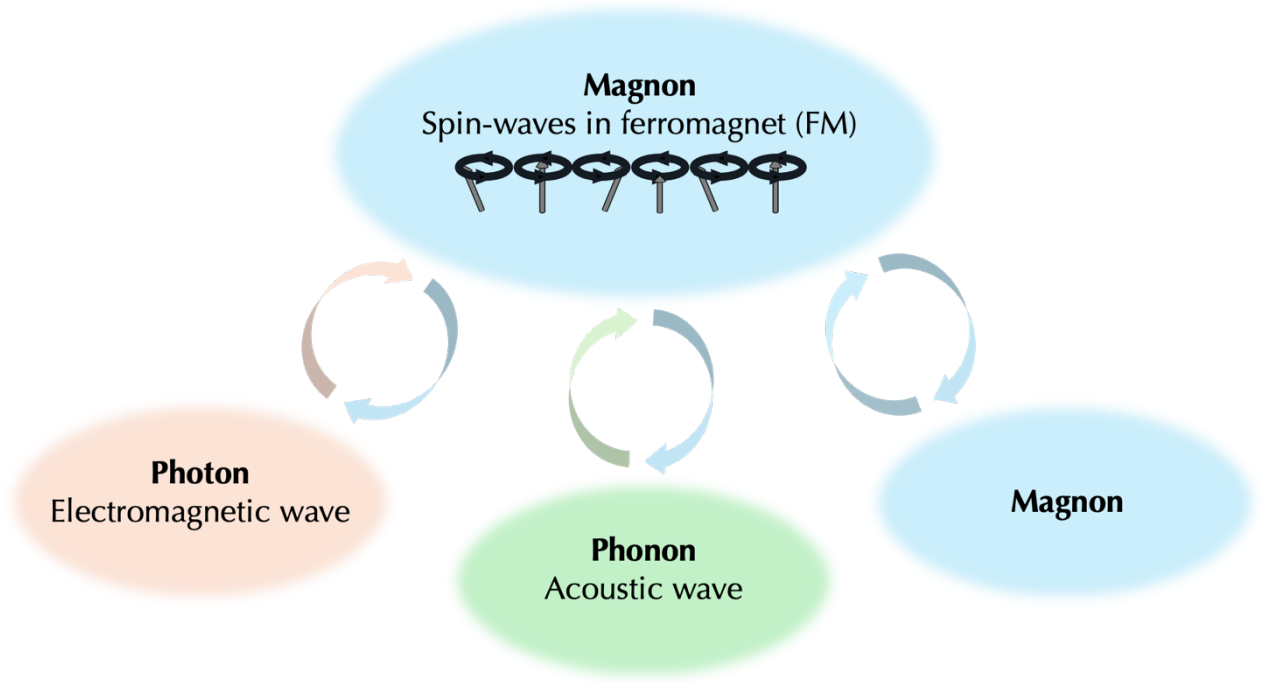


Figure 1-2. Concept of a quantum hybrid system composed of magnons, photons, and phonons.

Recent advances in material science and nanotechnology have enabled the fabrication of nanoscale magnetic structures, where magnon behavior can be precisely controlled. Thin films and patterned magnetic elements allow researchers to engineer magnon systems in on-chip devices, leading to the potential application of novel information processing on the microscale. These developments have introduced the concept of "magnonic devices," which could be analogous to electronic circuits but operate using magnons instead of electrons [34,35]. Recently, studies on reservoir computing with magnon systems in patterned thin ferromagnetic films have started to attract significant attention [36–38]. It has been demonstrated that strong coupling of magnons in on-chip ferromagnetic thin films on superconducting resonator with confined photon mode on the surface of superconducting resonator, which allows for strong coupling with small volume of magnons [28,29,39]. Such magnonic devices could revolutionize data processing and microwave communication, where

manipulating high-frequency spin waves is critical. While magnonics is still in its early stages compared to more mature fields like spintronics, its potential for impact is vast. Controlling magnon mode in thin ferromagnetic films is a significant challenge that researchers are actively addressing. Additionally, understanding magnon dissipation mechanisms, magnon-magnon interactions, and the influence of external factors like temperature and magnetic anisotropy on magnon dynamics in thin ferromagnetic films is critical for developing practical magnonic devices.

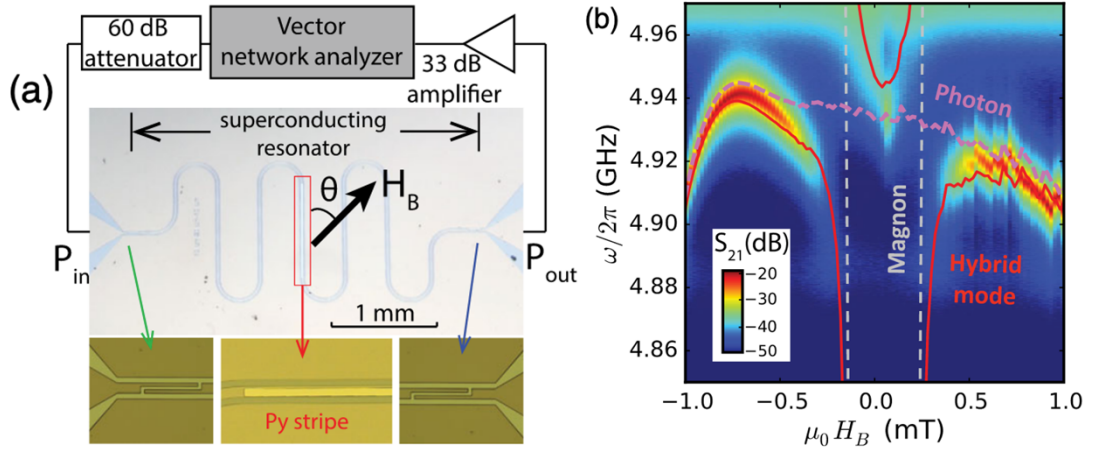


Figure 1-3. (a) Microscope image of superconducting resonator with 30 nm-thick Py strip and measurement setup for transmission spectra. (b) Transmission spectra S_{21} with external magnetic field dependence, where magnon and photon mode show polariton mode showing anti-crossing against each mode. These figures are quoted by the reference [40].

In summary, magnonics represents a novel approach to using quantum mechanical principles for practical applications. This field offers a promising pathway toward more efficient, scalable, and versatile information technologies by harnessing the dynamic magnon behavior in ferromagnetic materials.

1.3 Magnon polariton

Light-matter interaction is a fundamental concept that drives many technological advancements, from photonics to quantum information [41,42]. When light interacts with a material, it can couple with elementary excitations, creating hybrid quasiparticles called polaritons. Polaritons combine the properties of both photons and material excitations, allowing for the manipulation of light on scales smaller than its wavelength, which is crucial for next-generation information processing technologies. Among these, magnon polaritons—formed by coupling photons with magnons—have emerged as a promising platform for advancing magnetic and optical devices in magnonics [24,25,27–29]. The significance of magnon polaritons lies in their dual nature: they inherit the coherence and long-range propagation capabilities of photons and the magnetic properties of magnons. This coupling creates new opportunities for controlling both light and magnetic excitations in a single

platform, enabling applications in microwave signal processing, quantum information, and low-power devices. Figure 1-4 illustrates the differentiation of the coupling regime for magnon polaritons. Strong coupling regimes arise when magnons and photons coherently exchange energy, with their coupling strength surpassing relaxation. This can be realized by confining electromagnetic waves in resonant cavities, enhancing interaction and longer coherence time. The regime known as “ultrastrong coupling” is gaining attention, characterized by a coupling strength to resonance frequency ratio exceeding 0.1, at which point the rotating wave approximation no longer holds. The ultrastrong coupling regime is essential for entangling each quantum, forming a foundational technology for quantum information processing applications. When the ratio exceeds 1, counter-rotating terms in the Hamiltonian become predominant, leading to uncharted territory in quantum electrodynamics physics. The achievement of ultrastrong coupling between magnons and photons makes magnon polaritons highly attractive for integrating magnon-based and photon-based quantum technologies.

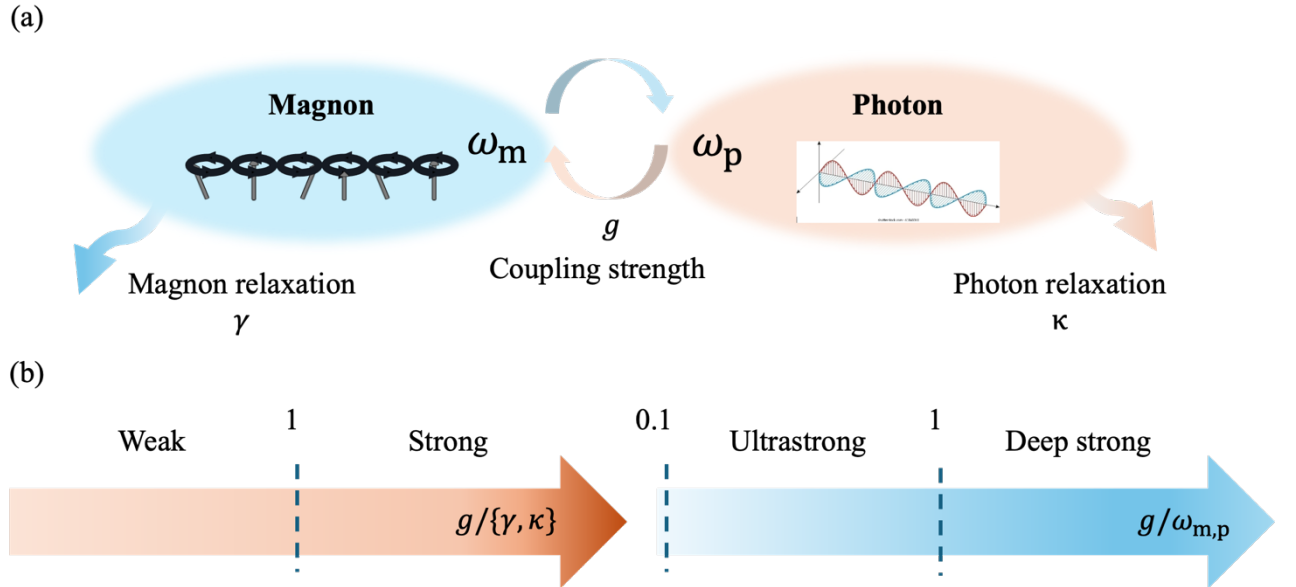


Figure 1-4. (a) Schematics of magnon-photon coupling with resonance frequency $\omega_{m,p}$ for magnon and photon mode, respectively, where g , γ , and κ are coupling strength, magnon relaxation, and photon relaxation. (b) Regimes of the coupling state. Red arrow shows the criteria of weak or strong coupling between magnons and photons, which is determined by the relation of coupling strength and their relaxation rates. Blue arrows show the criteria from ultrastrong coupling regime, which is determined by the ratio of coupling strength and the resonance frequency.

Thin-film ferromagnetic materials provide a promising platform for studying and utilizing magnon polaritons. The tunable properties of thin films, such as their thickness and magnetic anisotropy, allow researchers to precisely control the coupling strength, enabling scalable and adaptable magnon polariton devices. Moreover, the ability to stack adjacent

metals facilitates the integration of magnon phenomena, including nonreciprocity [43–49], nonlinearity [50–52], and magnon-current conversion [53–57]. Thin films are also compatible with modern integrated circuits, offering a pathway to practical, miniaturized devices that utilize magnon polariton effects for real-world applications. The development of magnon polaritons in thin-film ferromagnetic systems promises to bridge the gap between magnonics and photonics, offering new functionalities that neither magnons nor photons alone could achieve. These hybrid quasiparticles can be used to control the flow of information in ways that leverage both the low-energy dissipation of spin waves and the speed and coherence of electromagnetic waves. Such capabilities open up the possibility of developing efficient, high-speed technologies for signal processing, communication, and quantum computing.

In summary, magnon polaritons in thin-film ferromagnetic materials represent a cutting-edge frontier in fundamental research and technological development. Their unique properties offer a novel means of combining light and spin manipulation, positioning them at the forefront of future innovations in low-power, high-speed, and scalable information technologies.

1.4 Research purpose and outline of this dissertation

This dissertation aims to develop a robust platform for magnon-polaritons in thin ferromagnetic films through several key objectives: (i) outlining guiding principles for designing an optimal magnon system, (ii) developing a high quality-factor resonator to enable strong coupling with thin ferromagnetic films, and (iii) attaining the ultrastrong coupling regime for magnon-polaritons within these films.

First, the author explains the background physics, such as collective magnetization excitation, relaxation of magnons, quantization of magnons and photons, and Hamiltonian of magnon polaritons (Chapter 2). Investigating contributions to magnon damping in thin ferromagnetic films is a crucial study for optimizing the magnon systems. Although intrinsic damping and surficial damping from the spin pumping effect have been investigated, the author focused intently on the surficial magnon scattering resulting from surface magnetic anisotropy of ferromagnetic films, particularly significant in the ultrathin regime (around 2 nm). The guiding principle for suppressing magnon damping in thin ferromagnetic films is summarized in Chapter 3 of this dissertation. Chapter 4 details the design and fabrication of a superconducting resonator using $\text{YBa}_2\text{Cu}_3\text{O}_7$ (YBCO). The superconducting resonator plays a critical role in achieving strong coupling between magnons and photons. Its highly confined photons and high-quality factor are essential for observing coherent magnon-photon interactions, especially in the thin ferromagnetic films discussed earlier. Finally, Chapter 5 presents the realization of magnon polaritons in thin ferromagnetic films using the YBCO resonator. This chapter focuses on the experimental verification of magnon polariton formation and explores the implications for future magnonic and hybrid quantum technologies. In addition, the author demonstrated the realization of the ultrastrong coupling regime in magnon polaritons excited in thin ferromagnetic films. The potential of magnon

polaritons for coherent information transfer between magnetic and electromagnetic systems is thoroughly examined. In Chapter 6, the author summarizes this dissertation.

Chapter 2. Collective magnetization dynamics, and interaction between magnons and photons

2.1 Abstract

In this chapter, the author introduces the fundamental physics and theories for the measurement. The subjects include ferromagnetism, exchange interaction, dipole-dipole interaction, ferromagnetic resonance, the Gilbert damping constant, the quantization of magnons and photons, and the Hamiltonian of magnon polaritons.

2.2 Ferromagnetism and its fundamental feature of collective magnetization

Ferromagnetism is a fundamental magnetic phenomenon observed in certain materials, notably iron, cobalt, nickel, and some of their alloys. These materials exhibit a strong and permanent magnetic moment even in the absence of an external magnetic field, a property known as spontaneous magnetization. This behavior contrasts sharply with non-magnetic materials, where atomic magnetic moments are randomly oriented, resulting in no net magnetization. The unique properties of ferromagnetic materials arise from the collective alignment of atomic magnetic moments, governed by exchange interactions and dipole-dipole interactions. At the atomic level, electrons possess an intrinsic property called spin, which gives rise to a magnetic moment—essentially making each electron a tiny magnet. Additionally, the orbital motion of electrons around the nucleus contributes to the atom's overall magnetic moment. In most materials, these magnetic moments are randomly oriented due to thermal motion, leading to the cancellation of individual moments and resulting in no net macroscopic magnetization. As follows below, understanding the unique features of ferromagnetism is crucial for understanding magnon dynamics in ferromagnets.

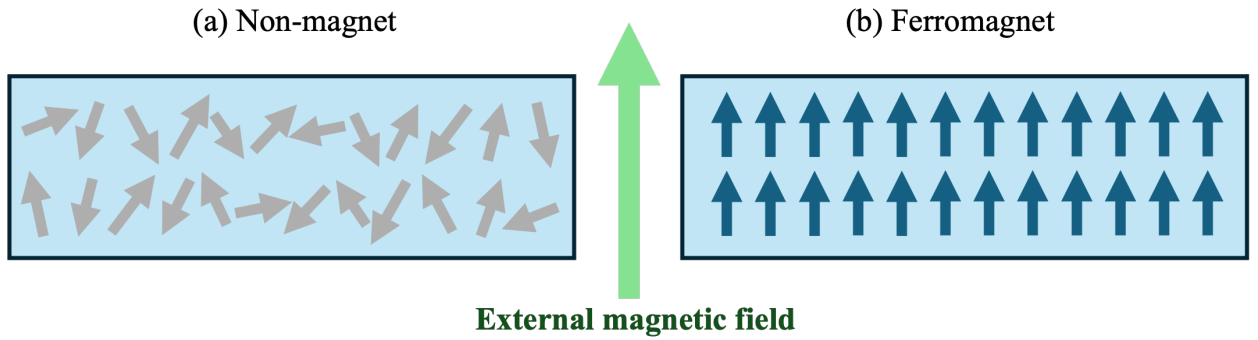


Figure 2-1. Schematic of stabilized magnetizations in (a) non-magnet and (b) ferromagnet when an external magnetic field is applied. The gray and blue arrows depict magnetizations in materials.

Exchange interaction

The exchange interaction is a quantum mechanical effect that plays a pivotal role in ferromagnetism. It arises from the Pauli exclusion principle, which states that no two electrons can simultaneously occupy the same quantum state. The Hamiltonian for the exchange interaction is expressed by the following equation:

$$\mathcal{H}_{\text{exchange}} = -2J \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+1}, \quad (1)$$

where J and $\mathbf{S}_i$ is the nearest neighbor exchange constant and operator of spin at site i that represents magnetic moment at site i , respectively. When electrons are close to each other, their spins tend to align in a parallel or antiparallel fashion if it lowers the system's energy depending on the sign of J . In ferromagnetic materials ($J > 0$), the exchange

interaction causes neighboring electron spins to align parallel over large regions, despite the presence of thermal agitation that would generally disrupt such alignment. This parallel alignment minimizes the energy of the system, making it a more stable configuration. The strength of the exchange interaction in ferromagnetic materials is significant, determining the dispersion relation of magnon mode with finite wave number k .

Dipole-Dipole interaction

The dipole-dipole interaction is a magnetic interaction between the magnetic moments of atoms or ions, arising from their intrinsic magnetic fields. Unlike the exchange interaction, which is short-range and quantum mechanical, the dipole-dipole interaction is a long-range, classical electromagnetic force. The Hamiltonian for the dipole-dipole interaction of spins in materials is expressed as follows:

$$\mathcal{H}_{\text{dipole}} = \frac{1}{2}(g\mu_B)^2 \sum_{i,j} \left[\frac{\mathbf{S}_i \cdot \mathbf{S}_j}{r_{ij}^3} - \frac{3(\mathbf{S}_i \cdot \mathbf{r}_{ij})(\mathbf{S}_j \cdot \mathbf{r}_{ij})}{r_{ij}^5} \right], \quad (2)$$

where g and μ_B are the g-factor of electrons and Bohr magneton, respectively. $\mathbf{r}_{ij}$ is the difference of spin sites $\mathbf{r}_{i,j}$ defined as $\mathbf{r}_i - \mathbf{r}_j$. It influences the orientation of magnetic domains relative to each other and tends to align magnetic moments to minimize the external magnetic field produced by the material. This interaction affects how collective magnetization is arranged within the material. The combination of exchange and dipole-dipole interactions results in complex collective magnetization dynamics crucial for understanding the macroscopic magnetic properties of ferromagnetic materials.

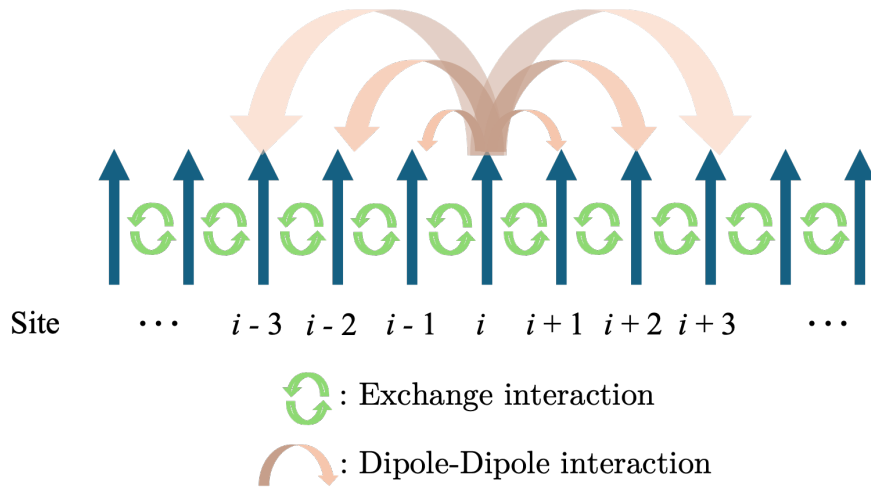


Figure 2-2. Schematic of the exchange interaction and dipole-dipole interaction in aligned magnetic moment. Here, the parallel magnetizations in a ferromagnet and the dipole field only from magnetization at site i are assumed. While exchange interaction is valid for neighboring magnetizations, dipole-dipole coupling works between long-distance magnetizations.

Contrast with Magnetic Materials

In non-magnetic materials, the magnetic moments of electrons are randomly oriented without strong interactions to align them (as in paramagnetic materials). When an external magnetic field is applied, these materials exhibit only a weak and temporary magnetization. The induced magnetization is typically several orders of magnitude smaller than that observed in ferromagnetic materials and disappears almost immediately after the external field is removed.

Collective Behavior of Magnetization

The key distinction of ferromagnetic materials lies in the collective behavior of their magnetic moments. The strong exchange interaction causes a cooperative alignment of spins over large numbers of atoms, leading to substantial magnetic domains with uniform magnetization. This collective phenomenon results in emergent properties do not present in non-magnetic materials. Due to the energy barriers associated with domain wall movement, the cooperative alignment is stable and can persist over time, even in the absence of an external magnetic field.

2.3 Magnetic anisotropy effect

In addition to exchange and dipole-dipole interactions, ferromagnetic materials often exhibit anisotropy—directional dependence of their magnetic properties. This directional dependence plays a crucial role in determining the behavior of magnetization, magnetization dynamics, and the overall magnetic characteristics of the material. Two primary types of magnetic anisotropy are (a) shape magnetic anisotropy and (b) magnetocrystalline anisotropy. Furthermore, the magnetic anisotropy created at the interface between the ferromagnetic layer and adjacent nonmagnetic layers, known as surface magnetic anisotropy, plays a vital role in the magnetization dynamics of thin ferromagnetic films. The material tends to magnetize along the easy directions to minimize the total magnetic energy, where the sum of magnetic anisotropy energy, including shape, crystalline, and surface magnetic anisotropy, determines its easy axis. Understanding these effects is essential for comprehending how ferromagnetic thin films respond to external field and how magnons behave in thin ferromagnetic films.

Shape magnetic anisotropy

Shape anisotropy arises from the demagnetizing fields generated due to the geometry of the ferromagnetic material. When a ferromagnetic object is magnetized, it creates its own magnetic field, which in turn affects the distribution of magnetization within the material. The demagnetizing field depends on the shape of the material. It can either enhance or oppose the magnetization in certain directions, leading to preferred directions of magnetization that minimize the system's energy.

Demagnetization fields are defined as internal magnetic fields that act to oppose the process of magnetization within the material in question. Its strength and orientation depend on the shape of the ferromagnet. For example, in a ferromagnetic film, the demagnetizing field is the weakest in the plane, making this direction energetically favorable for magnetization. Due to the demagnetizing fields, certain directions within the material become "easy" directions for magnetization, while others become "hard" directions.

The energy associated with shape anisotropy can be expressed using demagnetizing factors $N_{x,y,z}$, which are coefficients that depend on the material's geometry. The shape magnetic anisotropy energy E_{shape} for a uniformly magnetized ferromagnet is generally given by:

$$E_{\text{shape}} = \frac{1}{2}(N_x M_x^2 + N_y M_y^2 + N_z M_z^2), \quad (3)$$

where $M_i (i = x, y, z)$ denotes the component of magnetization in the material and $N_x + N_y + N_z = 4\pi$. In thin films, assuming that the film plane is x-y plane, $N_x = N_y = 0$ and N_z becomes 4π . The energy expression of shape magnetic anisotropy is vital for determining the frequency of collective magnetization excitation (ferromagnetic resonance).

Magneto crystalline anisotropy

Magneto crystalline anisotropy originates from the interaction between the magnetic moments of electrons and the crystal lattice of the material. This effect causes the energy of the system to depend on the direction of magnetization relative to the crystallographic axes, leading to preferred directions known as easy axes. The primary mechanism behind magnetocrystalline anisotropy is spin-orbit coupling. In ferromagnetic materials, the electron spins (magnetic moments) interact with their orbital motion, which is influenced by the electric fields of the surrounding ions in the crystal lattice. This coupling links the orientation of spins to specific directions in the lattice. Since the magnetocrystalline anisotropy emerges only when the ferromagnet is crystalized, amorphous and polycrystalline materials should not have this anisotropy. In this dissertation, magnetocrystalline anisotropy is less critical since the fabricated films are assumed to be amorphous and polycrystalline.

Surface magnetic anisotropy

Surface magnetic anisotropy is a crucial effect that arises due to the broken symmetry and altered electronic environment at the surface or interface of ferromagnetic materials. This phenomenon occurs regardless of whether the material is amorphous, polycrystalline, or single crystalline, as long as a surface or interface exists. The altered coordination of atoms at the surface leads to changes in spin-orbit coupling and exchange interactions, significantly affecting the material's magnetic properties, particularly in thin films. One of the most prominent effects of surface magnetic anisotropy is perpendicular magnetic anisotropy (PMA), where the magnetization tends to align perpendicularly to the plane of the film. PMA is particularly strong in ultrathin films or at interfaces with heavy

metals such as platinum or palladium, where spin-orbit coupling is enhanced. In these systems, surface contributions to anisotropy can dominate over bulk contributions, especially as the film thickness decreases.

The total anisotropy energy of a thin film can be expressed as the sum of the contributions from the bulk and surface:

$$E_{\text{total}} = K_{\text{bulk}}t + K_{\text{surface}}, \quad (4)$$

where t is the film thickness, K_{bulk} is the bulk anisotropy constant, and K_{surface} is the surface anisotropy constant. As the film becomes thinner, the surface magnetic anisotropy term plays a dominant role in determining the magnetic properties in thin ferromagnetic films.

Surface magnetic anisotropy is essential for understanding the magnetism of ferromagnetic thin films, whether being amorphous, polycrystalline, or crystalline. This effect is critical in designing magnetic properties in ultra-thin ferromagnetic film ($< \sim 2$ nm). The author investigated the effect of surface magnetic anisotropy on magnetization damping in Chapter 3, which gives informative guidelines for controlling magnon relaxation in thin ferromagnetic films.

2.4 Collective magnetization dynamics – magnons -

Understanding the dynamic behavior of magnetization in ferromagnetic materials is crucial for both fundamental physics and technological applications to magnonics. The time-dependent evolution of the magnetization vector within these materials is governed by the Landau-Lifshitz-Gilbert equation. This equation describes how magnetization responds to effective magnetic fields, including external fields and internal interactions. A key phenomenon arising from magnetization dynamics is ferromagnetic resonance, which corresponds to uniform precessional modes of the magnetization vector, often associated with magnons at zero wavenumber ($k = 0$). Extending beyond FMR, finite-wavenumber magnons represent collective excitations involving spatial variations in magnetization.

The Landau-Lifshitz-Gilbert equation provides a comprehensive framework for modeling the dynamics of magnetization $\mathbf{M}$ in ferromagnetic materials. It is given by:

$$\frac{d\mathbf{M}(t)}{dt} = -\gamma\mu_0\mathbf{H}_{\text{eff}} \times \mathbf{M}(t) + \frac{\alpha}{M_s}\mathbf{M}(t) \times \frac{d\mathbf{M}(t)}{dt}. \quad (5)$$

Here, $\gamma = g\mu_B/\hbar$ is the gyromagnetic ratio (here, g is the g-factor, μ_B is the Bohr magneton, and $\hbar$ is the Dirac constant), μ_0 is the vacuum permeability, $\mathbf{H}_{\text{eff}}$ is the effective magnetic field, and α is the Gilbert damping parameter, respectively. The first term on the right-hand side represents the precession of the magnetization around effective magnetic field $\mathbf{H}_{\text{eff}}$. This term reflects the torque exerted on the magnetic moments by the effective magnetic field including the external magnetic field, magnetic anisotropy field, exchange interaction, and demagnetizing fields, causing them to precess. The second term introduces phenomenological magnetization damping through the Gilbert damping

parameter α , accounting for the loss of energy from the precessional motion to the lattice and other degrees of freedom. This term ensures that the magnetization eventually aligns with the effective field over time. The LLG equation captures the essence of magnetization dynamics by balancing the precessional motion and the dissipative processes.

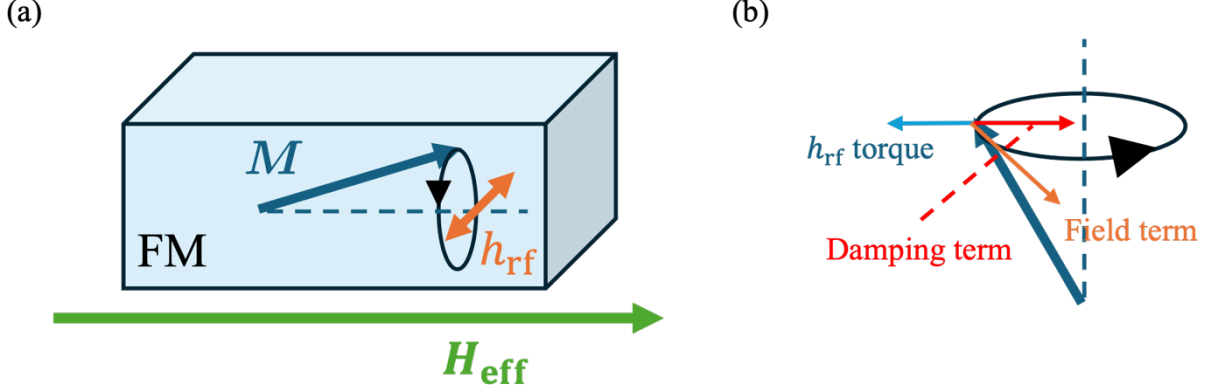


Figure 2-3. (a) Schematic of magnetization dynamics when the effective static magnetic field H_{eff} and alternating magnetic field with GHz frequency. According to Eq. (5), the magnetization precesses in anticlockwise. Fig. (b) shows external forces to the magnetization dynamics. In the FMR condition, torque equals the damping term, causing the magnetization to keep precessing.

Ferromagnetic resonance is a phenomenon where the magnetization precesses coherently around an effective magnetic field when subjected to an external oscillating magnetic field at a resonant frequency, where an alternating magnetic field generates anti-damping torque against the damping term (see Fig. 2-3). In FMR experiments, a uniform external field is applied, and the system is probed with microwave radiation. At the resonance condition, the energy from the microwave field is efficiently absorbed by the ferromagnet, leading to a peak in the absorption spectrum. The resonance frequency f_{FMR} for thin ferromagnetic films with in-plane external magnetic field is expressed by Kittel formula derived from Eq. (5) as follows [58]:

$$f_{\text{FMR}} = \frac{\gamma\mu_0}{2\pi} \sqrt{H(H + M_{\text{eff}})}. \quad (6)$$

Here, M_{eff} is the effective magnetization composed of demagnetization field and magnetic anisotropy field. FMR corresponds to the excitation of magnons with zero wavenumber ($k = 0$), meaning that the precession is uniform throughout the material. In this mode, all the magnetic moments precess in phase, resulting in a collective oscillation of the magnetization vector without spatial variation unlike $k \neq 0$ magnons, where the phase of the magnetization precession varies periodically in space (see Fig. 2-4). Measuring FMR delivers crucial insights for investigating magnon modes and relaxation.

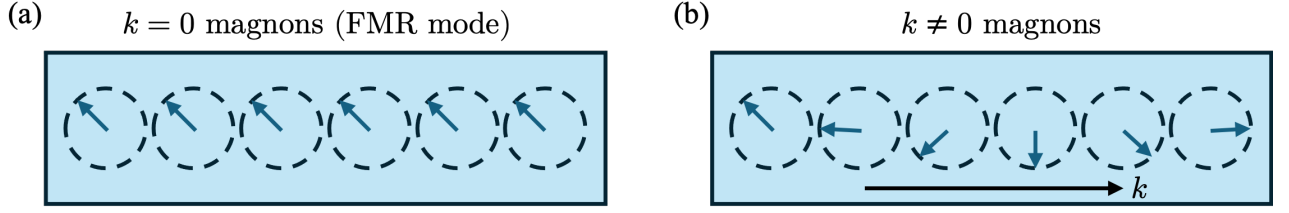


Figure 2-4. Schematics of spin-waves in thin ferromagnetic films with (a) $k = 0$ (FMR mode) and (b) $k \neq 0$.

In general, the direction of magnetization in thin ferromagnetic films with magnetic anisotropy does not necessarily coincide with the direction of the external magnetic field, which requires another expression of FMR mode when an external magnetic field is applied not along an easy axis. For the general expression of FMR in the condition that external magnetic field is applied to the ferromagnetic film as shown in Fig. 2-5, the resonance equation is formulated by Smit and Beljers [59] as follows:

$$\left(\frac{2\pi f_{\text{FMR}}}{\gamma}\right)^2 = \frac{1}{M_S^2 \sin \theta} \left[\frac{\partial^2 E_{\text{total}}}{\partial \theta^2} \frac{\partial^2 E_{\text{total}}}{\partial \phi^2} - \left(\frac{\partial^2 E_{\text{total}}}{\partial \theta \partial \phi} \right)^2 \right], \quad (7)$$

where E_{total} is the total of magnetic energies such as Zeeman energy, magnetic anisotropy energy and demagnetization energy, and M_S is the saturation magnetization. Assuming E_{total} according to the materials and the shape of the ferromagnet, we can derive the angular dependence of resonance frequency, which is useful for estimation of the magnetic parameters such as magnetic anisotropy. For instance, E_{total} for thin ferromagnetic films is defined with uniaxial magnetic anisotropy energy K_u as follows:

$$E_{\text{total}} = -HM_S \left[\sin \theta_H \sin \theta \cos(\phi - \phi_H) + \cos \theta_H \cos \theta \right] - 2\pi(M_S)^2 \sin^2 \theta + K_u \sin^2 \theta. \quad (8)$$

Here, θ_H and ϕ_H are the polar angle and azimuthal angle of the external magnetic field. Substituting Eq. (8) into Eq. (7) and considering the equilibrium condition, $\frac{\partial E_{\text{total}}}{\partial \theta} = \frac{\partial E_{\text{total}}}{\partial \phi} = 0$, we obtain the resonance equation as follows [60]:

$$\left(\frac{2\pi f_{\text{FMR}}}{\gamma}\right)^2 = [\mu_0 H_{\text{res}} \cos(\theta - \theta_H) - M_{\text{eff}} \cos 2\theta][\mu_0 H_{\text{res}} \cos(\theta - \theta_H) + M_{\text{eff}} \sin^2 \theta], \quad (9)$$

$$\mu_0 H_{\text{res}} \sin(\theta_H - \theta) + \frac{M_{\text{eff}}}{2} \sin 2\theta = 0. \quad (10)$$

Here, we assumed to rotate the external magnetic field in the X-Z plane and no magnetic anisotropy in plane ($\phi = \phi_H = 0$). Fitting of the experimental results of the angular dependence of FMR with these equations provides the estimation of M_{eff} , including

magnetic anisotropy of ferromagnetic thin films and the magnetization direction in ferromagnetic films when a certain magnetic field is applied.

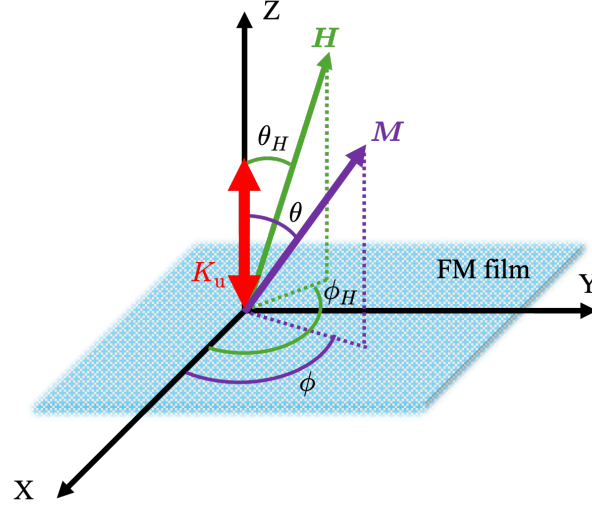


Figure 2-5. Schematic of magnetization and external magnetic field in thin ferromagnetic films with magnetic anisotropy, where K_u is uniaxial magnetic anisotropy defined along the perpendicular of ferromagnetic film. K_u emerged from crystallinity and surface magnetic anisotropy.

In summary, the study of FMR offers vital insights into the influence of magnetic anisotropy on magnon modes. This understanding is essential for engineering magnons in thin ferromagnetic films before investigating magnon-photon coupling states.

2.5 Magnon relaxation in thin ferromagnetic films

Another crucial parameter of magnons is the relaxation rate related to magnetization damping – the loss of energy from the precessional motion of magnetization in ferromagnetic materials – where its energy is dissipated into the lattice, mobile electrons, and the other degrees of freedom. In thin films, surface effects and spin-orbit coupling can enhance damping, influencing the lifetime and coherence of magnon modes. Materials with lower magnetization damping are desirable in applications where long-lived precessional motion is needed, such as in magnonic devices. Conversely, high-damping materials are useful in systems where rapid magnetization switching is required for magnetic memory devices. Understanding the physics of magnetization damping is crucial for analyzing the dynamics of magnetization and designing magnonic devices on demand.

FMR measurement is one of the primary ways also to measure magnetization damping in addition to resonance frequency. In an FMR experiment, the resonance condition is achieved when the frequency of the alternating magnetic field matches with the natural precession frequency of the magnetization. When measuring the absorption spectrum due to FMR, the half-width at half-maximum (HWHM) of the resonance peak, ΔH , provides a direct measure of the magnetization damping in the system. A broader linewidth indicates

greater energy loss during the precessional motion, while a narrower linewidth corresponds lower damping as shown in Fig. 2-6.

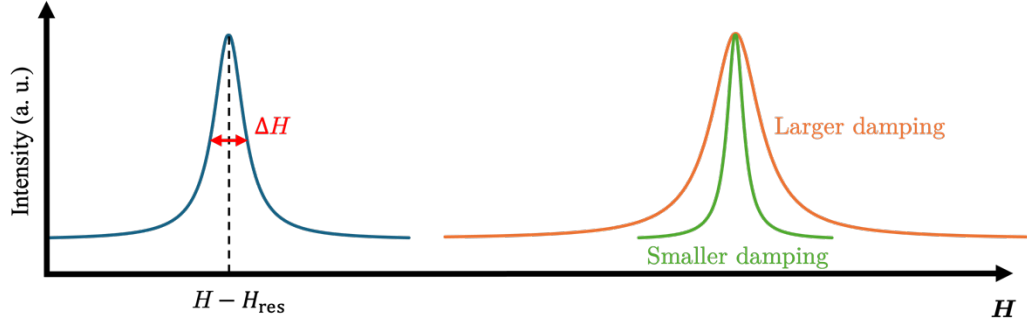


Figure 2-6. Absorption spectra in the FMR measurement. The blue curve depicts a typical FMR peak described by the Lorentzian function. Orange and green curves show the comparison of FMR peaks with larger (orange) and smaller (green) damping.

The HWHM, ΔH , in FMR is related to the Gilbert damping constant (α) via the following relationship,

$$\mu_0 \Delta H = \mu_0 \Delta H_0 + \frac{2\pi\alpha f_{\text{FMR}}}{\gamma}, \quad (11)$$

where ΔH_0 is inhomogeneous linewidth due to the variation in the magnetization precession that is generated by the inhomogeneity of ferromagnetic materials, such as inhomogeneity of composition and roughness of the film [61–63]. While the Gilbert damping constant is introduced phenomenologically in the LLG equation, as shown in Eq. (5), its contributions have been examined by numerous researchers. For instance, the key contributions in thin ferromagnetic films are decomposed with the FM thickness t_{FM} as follows:

$$\alpha = \alpha_{\text{int}} + \alpha_{\text{SP}} \frac{1}{t_{\text{FM}}} + \beta_{\text{TMS}} \frac{1}{t_{\text{FM}}^2}, \quad (12)$$

where α_{int} , α_{SP} and β_{TMS} are the thickness-independent intrinsic damping constant of the FM layer, the damping constant due to spin pumping, and the two-magnon scattering coefficient, respectively. Below is a detailed description of these contributions.

Intrinsic Gilbert damping (α_{int})

Intrinsic Gilbert damping is a fundamental characteristic of ferromagnetic materials, primarily resulting from magnon-phonon and magnon-electron scattering [64,65]. Magnon-phonon scattering occurs due to spin-orbit interaction and spin-flipping scattering, which allows the transfer of angular momentum from magnetization precession to the lattice or impurities, leading to energy loss. As this scattering is tied to lattice vibrations, its damping effect intensifies at elevated temperatures. Although phonon energy dissipation predominates

at high temperatures, magnon-electron scattering can surpass phonon scattering at lower temperatures. These phenomena are inherent to the material and generally do not depend on the film's thickness in bulk samples. For instance, $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (YIG), a common ferrimagnet, exhibits relatively low spin-orbit interactions, high crystallinity, and a reduction in magnon-electron scattering due to the lack of conduction electrons. This results in a Gilbert damping that is quite low, approximately 10^{-4} , even in 25 nm thick YIG films [66], although Gilbert damping of typical 3d transition ferromagnetic films shows $\sim 10^{-2}$. Recent studies have revealed that the ferromagnetic metallic alloy with a specific composition, $\text{Co}_{25}\text{Fe}_{75}$, has ultralow intrinsic damping $\sim 10^{-3}$, which is attributed to the significant suppression of the magnon-electron scattering by adjusting the composition of Co and Fe [67–69] as shown in Fig. 2-7 (a). Following the principle of suppression of intrinsic magnetic damping even in ferromagnetic metallic films, FeAl alloys show also ultralow Gilbert damping comparable with CoFe alloys [70] (see Fig. 2-7 (b)). Recent progress in understanding the origin of intrinsic Gilbert damping significantly expands the selection of ferromagnetic materials for low magnon damping.

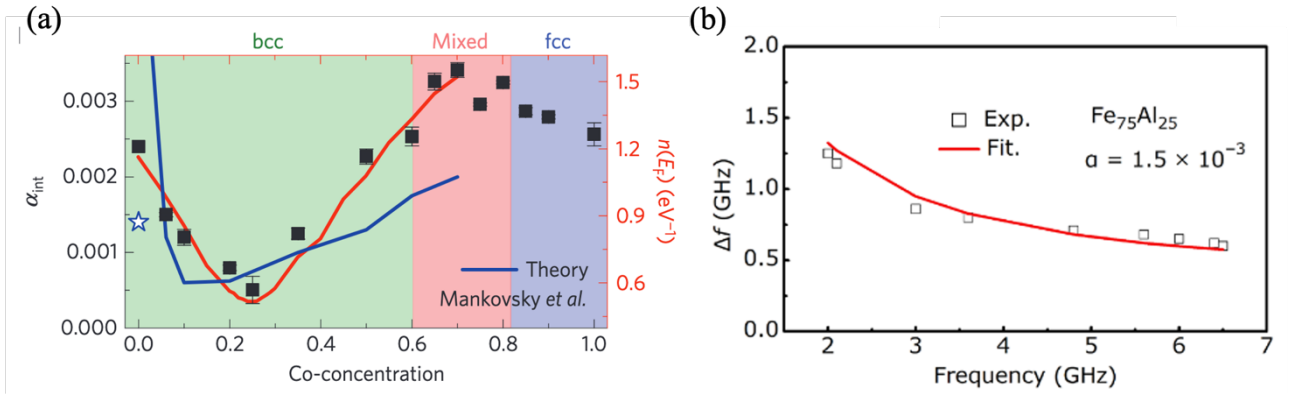


Figure 2-7. (a) Co concentration dependence of intrinsic Gilbert damping in CoFe alloys. In the Co concentration of 25%, CoFe alloys showed the lowest α_{int} , which trend corresponds with the calculated DOS at the Fermi energy. (b) Corresponding frequency width dependence of the frequency for 20 nm-thick $\text{Fe}_{75}\text{Al}_{25}$ films, which provides Gilbert damping parameter of 1.5×10^{-3} . These figures are quoted by [67] for Fig. (a) and [70] for Fig. (b).

Spin pumping (α_{SP})

Spin pumping occurs between a ferromagnetic material and an adjacent non-magnetic layer, typically a heavy metal like platinum and Tantalum [71–74]. In this mechanism, the precessing magnetization in the ferromagnet generates a spin current that is pumped into the nonmagnetic adjacent layer as shown in Fig. 2-8. This process leads to an additional source of angular momentum loss, increasing the effective damping constant. Studies on the damping constant due to spin pumping have been quite important in the context of spintronics since

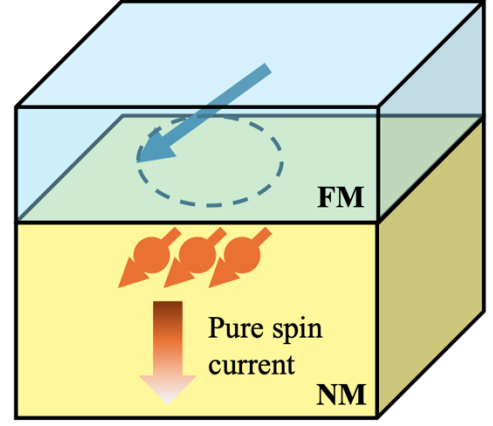


Figure 2-8. Schematic image of spin pumping. The pumped spin current from FM is flowing in nonmagnetic adjacent layer beside ferromagnetic layer.

spin pumping can be a source of pure spin current, which has been enthusiastically investigated for application to spintronics devices. Furthermore, the detection of generated pumped spin current by magnons enables observation of magnon-photon coupling state through the inverse spin Hall effect (ISHE), the spin-charge conversion in strong spin-orbit interaction materials such as platinum and Tantalum [53–56]. In addition, harnessing the detection of pumped spin current using ISHE allows for the tomography measurement of the magnon mode [57], which is potentially applicable to the detection of the squeezed state of magnon polaritons.

The spin pumping contribution is more significant in thin films because the interface plays a relatively larger role. As the thickness of the ferromagnetic layer decreases, more of the magnetization precession energy is transferred to the non-magnetic layer, leading to increased damping. Whereas there are intriguing aspects to spin pumping, it also adds to magnon damping in thin ferromagnetic films, and we need to reduce the unnecessary damping caused by spin pumping. Plenty of studies on spin pumping could provide an informative guiding principle for harnessing spin pumping without wasteful magnon damping.

Two-magnon scattering (β_{TMS})

In the past two decades, the ultrathin ferromagnetic films ($< \sim 2$ nm) have been attracting attention because of fascinating surface magnetic properties such as electronic gate tunability [75–78] and Dzyaloshinskii-Moriya interaction [47], which are expected to introduce novel functionalities into magnonics. While ultrathin ferromagnetic films are a fascinating platform for magnons, the enhancement of magnon relaxation in ultrathin ferromagnetic films has hampered its usage as the magnon medium. The notable damping mechanism in the ultrathin regime of ferromagnetic films is two-magnon scattering (TMS) because TMS term in Eq. (12) is proportional to the inverse square of t_{FM} , which is

extremely enhanced in ultrathin regime. It involves the scattering of uniform magnetization precession ($k = 0$ mode) into non-uniform spin wave modes due to the presence of exchange interaction and dipole interaction, which can be generally described by the energy transition between different wave numbers with the same energy, as shown in Fig. 2-9 (see section 2.6 for the dispersion relation of magnons). Some reports indicated that the two-magnon process in thin ferromagnetic films is valid in the presence of defects [79] and exchange coupling [80] of ferromagnetic films. Arias and Mills demonstrated the general expression of TMS, ΔH_{TMS} , in ultrathin ferromagnetic as follows:

$$\Delta H_{\text{TMS}} = \frac{8H_{\text{su}}^2 H_0^2 b^2 p}{\pi D (B_0 + H_{\text{su}} + H_0)^2} \times \sin^{-1} \left(\frac{H_0^{1/2}}{(B_0 + H_{\text{su}})^{1/2}} \right), \quad (13)$$

where D , b , and p is the exchange stiffness, the height of the defect, and the fraction of the surface covered by defects. Here, it is defined that $B_0 = H_0 + 4\pi M_S$ and $H_{\text{su}} = 2K_{\text{surface}}/t_{\text{FM}} M_S$. Although the ideal flat surface of ultrathin ferromagnetic film ($b = 0$) vanishes ΔH_{TMS} , it is quite difficult to eliminate defects at the surface of the ferromagnetic films deposited by some instrument such as sputtering system and electron beam deposition, which provides a finite ΔH_{TMS} . With a finite b , ΔH_{TMS} is approximately proportional to the surface magnetic anisotropy as follows:

$$\Delta H_{\text{TMS}} \sim \beta_{\text{TMS}} \frac{1}{t_{\text{FM}}^2} \propto H_{\text{su}}^2 = \left(\frac{2K_{\text{surface}}}{M_S} \right)^2 \frac{1}{t_{\text{FM}}^2}. \quad (14)$$

Therefore, the TMS coefficient β_{TMS} is theoretically predicted to be proportional to the square of the surface magnetic anisotropy field, $h_{\text{su}} = \frac{2K_{\text{surface}}}{M_S}$, where the manipulation of surface magnetic anisotropy in ultrathin ferromagnetic films is crucial for controlling TMS. Chapter 3 discusses the guiding principles for managing TMS.

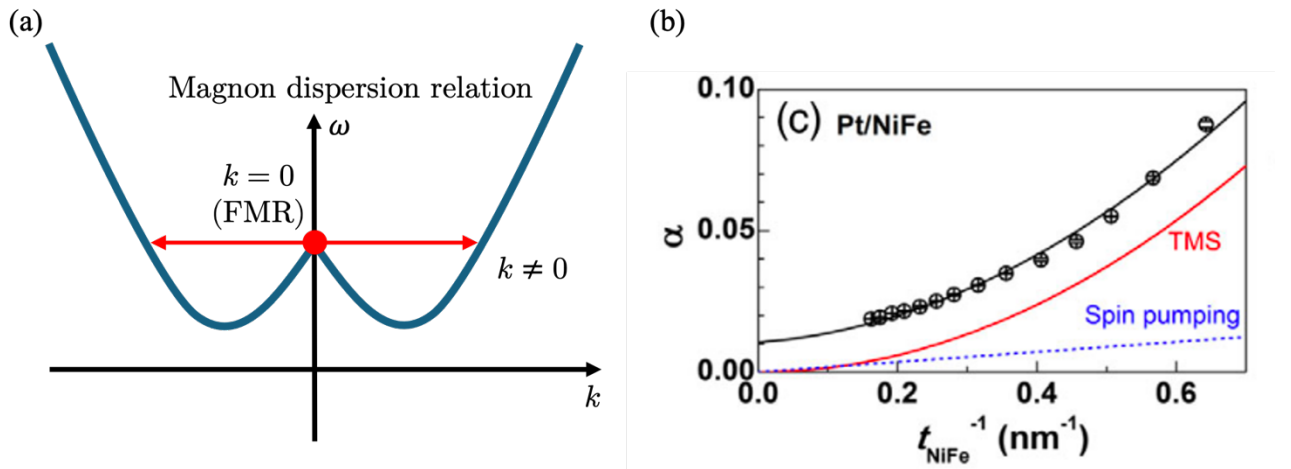


Figure 2-9. (a) Dispersion relation of magnons in ferromagnetic films. In the two-magnon scattering process, the transition of magnons from $k = 0$ to $k \neq 0$ with the same energy occurs. (b) FM thickness dependence of Gilbert damping constant, demonstrating

considerable enhancement of Gilbert damping because of $\beta_{\text{TMS}} \frac{1}{t_{\text{FM}}^2}$ in Eq. (12). Fig 2-9 (b) is quoted by [81]

In summary of this section, since a number of damping mechanisms for magnons in thin ferromagnetic films exist, more careful management of magnon damping is necessary for efficient magnonic devices in thin ferromagnetic films.

2.6 Quantum mechanical expression of magnons and photons

In modern condensed matter physics, the quantization of collective excitations is essential for understanding various physical phenomena at the microscopic level. Magnons and photons, which are quantized representations of spin waves and electromagnetic waves, respectively, both play critical roles in describing magnetic and electromagnetic interactions within solid-state systems. The necessity of treating magnons and photons in quantum mechanical way becomes even more evident when we explore magnon polaritons, where these two quasiparticles interact coherently.

This section introduces the quantization of magnons and photons and sets the stage for understanding their coupling, a key phenomenon in the emerging field of cavity magnonics and hybrid quantum systems.

Quantization of magnons

The Hamiltonian of magnon mode can be derived using second quantization by starting with the model of spins with exchange interaction and Zeeman interaction. Then, we shall apply the Holstein-Primakoff transformation, which expresses spin operators in terms of bosonic creation and annihilation operators.

The Hamiltonian for spin waves excited in a ferromagnetic material given by:

$$\mathcal{H} = -J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j - g\mu_B H \sum_i S_i^z, \quad (15)$$

where $\langle i,j \rangle$ is a sum over nearest-neighbor pairs and $\mathbf{S}_i = S_i^x \mathbf{x} + S_i^y \mathbf{y} + S_i^z \mathbf{z}$. The rectangular coordinate system is defined for external magnetic field H is parallel to z-axis. Here, the dipole-dipole interaction between spins is ignored for simplicity. Note that the sign of Zeeman term is opposite since spin wave excitation should be considered as spin-flipping state. To handle spin operators using bosonic operators, use the Holstein-Primakoff transformation:

$$S_i^+ = \sqrt{2S} a_i^\dagger \left(1 - \frac{a_i^\dagger a_i}{2S} \right)^{1/2} \approx \sqrt{2S} a_i^\dagger, \quad (16)$$

$$S_i^- = \sqrt{2S} \left(1 - \frac{a_i^\dagger a_i}{2S} \right)^{1/2} a_i \approx \sqrt{2S} a_i, \quad (17)$$

$$S_i^z = S - a_i^\dagger a_i. \quad (18)$$

Here, a_i and $a_i^\dagger$ are the annihilation and creation of magnons as bosonic particles. Then, the components of the spin operator are expressed as follows:

$$S_i^x = \frac{1}{2}(S_i^+ + S_i^-) \approx \sqrt{\frac{S}{2}}(a_i^\dagger + a_i), \quad (19)$$

$$S_i^y = \frac{1}{2}(S_i^+ - S_i^-) \approx i\sqrt{\frac{S}{2}}(a_i^\dagger - a_i), \quad (20)$$

$$S_i^z = S - a_i^\dagger a_i. \quad (21)$$

Substituting Eq. (19-21) to Eq. (15), we obtain the total Hamiltonian with bosonic operators:

$$\mathcal{H} = \sum_i \epsilon_0 a_i^\dagger a_i - JS \sum_{\langle i,j \rangle} (a_i^\dagger a_j + a_j^\dagger a_i), \quad (22)$$

where $\epsilon_0 = JSz + g\mu_B H$ and z is the number of the nearest neighbors per site. For the expression in the momentum space, introduce Fourier transforms:

$$a_i = \frac{1}{\sqrt{N}} \sum_k a_k e^{ik \cdot r_i}, \quad a_i^\dagger = \frac{1}{\sqrt{N}} \sum_k a_k^\dagger e^{-ik \cdot r_i}. \quad (23)$$

Thus, the Hamiltonian in Eq. (22) is expressed in the momentum space:

$$\mathcal{H} = \sum_k [g\mu_B H + JS(z - 2\gamma_k)] a_k^\dagger a_k, \quad (24)$$

where $\gamma_k = \sum_\delta e^{ik \cdot \delta}$ in the definition of $\delta = r_j - r_i$. For magnons with small wave vector $ka \ll 1$ in a simple cubic lattice with lattice constant a , γ_k is approximated to be $z - \frac{1}{2}a^2 k^2$ in the Taylor expansion. Then, the magnon energy dispersion relation ϵ_k without dipole interaction:

$$\epsilon_k = g\mu_B H + JSa^2 k^2. \quad (25)$$

Finally, the Hamiltonian of the quantized magnon mode with resonance angular frequency $\omega_m(k)$ is obtained:

$$\mathcal{H} = \sum_k \hbar \omega_m(k) a_k^\dagger a_k. \quad (26)$$

To consider the dipole-dipole interaction, it is necessary to add the Hamiltonian of Eq. (2) into Eq. (15). Then, the dispersion relation for the magnon mode, accounting for dipole-dipole interactions, is given by:

$$\hbar \omega_m(k) = \hbar \gamma \left[H + \frac{2zJS}{\hbar \gamma} (1 - \gamma_k) \right]^{1/2} \left[H + \frac{2zJS}{\hbar \gamma} (1 - \gamma_k) + 4\pi M_S \sin \theta_k \right]^{1/2}. \quad (27)$$

Here, θ_k is the polar angle of the wave vector when H is parallel to z-axis. This expresses the dispersion relation shown in Fig. 2-9. Refer to [1] for the details of the derivation of Eq. (27). The dipole-dipole interaction forms valleys in the dispersion relation at specific wave

numbers, which is essential for intriguing phenomena such as Bose-Einstein condensation [82–84] and the nonlinearity [52,85,86] of the magnon mode. The dipole-dipole coupling is one of the most fascinating mechanisms in recent magnonics. When substituting $k = 0$ and $\theta_k = \frac{\pi}{2}$ into Eq. (27), the dispersion relation reduces to Kittel equation, Eq. (6), which makes sure that FMR mode is $k = 0$ magnons.

Quantization of photons

The quantization of the free electromagnetic field leads to the concept of photons as quantized excitations of the field modes. In this derivation, we will start from the classical electromagnetic field, apply the Coulomb gauge, expand the field in terms of plane wave modes, and then promote the field amplitudes to operators satisfying the appropriate commutation relations. Finally, we will arrive at the Hamiltonian expressed in terms of photon creation and annihilation operators.

A solid foundation for the quantization of the electromagnetic field lies in the classical field equations, the Maxwell equations. The electromagnetic field in free space follows the Maxwell equations without any source:

$$\nabla \cdot \mathbf{B} = 0, \quad (28)$$

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}, \quad (29)$$

$$\nabla \cdot \mathbf{D} = 0, \quad (30)$$

$$\nabla \times \mathbf{H} = -\frac{\partial \mathbf{D}}{\partial t}. \quad (31)$$

Here, $\mathbf{B} = \mu_0 \mathbf{H}$ and $\mathbf{D} = \varepsilon_0 \mathbf{E}$, where ε_0 is electric permittivity of free space and $\mu_0 \varepsilon_0 = -c^2$ holds. In free space, Eqs. (28-31) can be simplified by choosing the Coulomb gauge:

$$\nabla \cdot \mathbf{A}(\mathbf{r}, t) = 0, \quad (32)$$

where $\mathbf{A}(\mathbf{r}, t)$ is the vector potential. This gauge simplifies the expressions for the electric and magnetic field:

$$\mathbf{E}(\mathbf{r}, t) = -\frac{\partial \mathbf{A}(\mathbf{r}, t)}{\partial t}, \quad (33)$$

$$\mathbf{B}(\mathbf{r}, t) = -\nabla \times \mathbf{A}(\mathbf{r}, t). \quad (34)$$

Substituting Eq. (34) into Eq. (31), we obtain the wave equation for $\mathbf{A}(\mathbf{r}, t)$:

$$\nabla^2 \mathbf{A}(\mathbf{r}, t) = \frac{1}{c^2} \frac{\partial^2 \mathbf{A}(\mathbf{r}, t)}{\partial t^2}. \quad (35)$$

Eq. (35) can be solved with the following wave equation:

$$\mathbf{A}(\mathbf{r}, t) = \sum_k (c_k \mathbf{u}_k(\mathbf{r}) e^{-i\omega_k t} + c_k^* \mathbf{u}_k^*(\mathbf{r}) e^{i\omega_k t}), \quad (36)$$

where the Fourier coefficients c_k are constant for free space. The defined vector mode functions $\mathbf{u}_k(\mathbf{r})$ corresponding to the frequency ω_k satisfies the wave equation as follows:

$$\left(\nabla^2 + \frac{\omega_k^2}{c^2}\right) \mathbf{u}_k(\mathbf{r}) = 0. \quad (37)$$

From Coulomb gauge, Eq. (32), $\mathbf{u}_k(\mathbf{r})$ is required to satisfy the transversality condition as a boundary condition and forms a complete orthonormal set,

$$\nabla \cdot \mathbf{u}_k(\mathbf{r}) = 0. \quad (38)$$

$$\int_V \mathbf{u}_k^*(\mathbf{r}) \mathbf{u}_{k'}(\mathbf{r}) d\mathbf{r} = \delta_{kk'} \quad (39)$$

In general, the mode functions depend on the boundary conditions of the physical space confining electromagnetic waves. For instance, we shall apply periodical boundary condition of a rectangle of length L with one side sufficiently large. Then, the wave vector for electromagnetic wave:

$$\mathbf{k} = \frac{2\pi}{L} (n_x, n_y, n_z), \quad (40)$$

where n_i ($i = x, y, z$) is $0, \pm 1, \pm 2, \dots$ and the mode function may be written as

$$\mathbf{u}_k(\mathbf{r}) = L^{-\frac{3}{2}} \hat{\mathbf{e}}^{(\lambda)} \exp(i\mathbf{k} \cdot \mathbf{r}), \quad (41)$$

where $\hat{\mathbf{e}}^{(\lambda)}$ is the unit polarization vector, which is required to be perpendicular to $\mathbf{k}$ by Eq. (38). Under this condition, the vector potential can be expressed as follows:

$$\mathbf{A}(\mathbf{r}, t) = \sum_k \left(\frac{\hbar}{2\omega_k \varepsilon_0} \right)^{\frac{1}{2}} (b_k \mathbf{u}_k(\mathbf{r}) e^{-i\omega_k t} + b_k^\dagger \mathbf{u}_k^*(\mathbf{r}) e^{i\omega_k t}), \quad (42)$$

where b_k and $b_k^\dagger$ are the normalization factors determined by the boundary condition. The corresponding electric field is

$$\mathbf{E}(\mathbf{r}, t) = -\frac{\partial \mathbf{A}(\mathbf{r}, t)}{\partial t} = i \sum_k \left(\frac{\hbar \omega_k}{2\varepsilon_0} \right)^{\frac{1}{2}} (b_k \mathbf{u}_k(\mathbf{r}) e^{-i\omega_k t} - b_k^\dagger \mathbf{u}_k^*(\mathbf{r}) e^{i\omega_k t}). \quad (43)$$

$$\begin{aligned} \mathbf{B}(\mathbf{r}, t) &= -\nabla \times \mathbf{A}(\mathbf{r}, t) \\ &= i \sum_k \left(\frac{\hbar}{2\varepsilon_0 \omega_k L^3} \right)^{\frac{1}{2}} (\mathbf{k} \times \hat{\mathbf{e}}^{(\lambda)}) (b_k e^{i(\mathbf{k} \cdot \mathbf{r} - \omega_k t)} - b_k^\dagger e^{-i(\mathbf{k} \cdot \mathbf{r} - \omega_k t)}). \end{aligned} \quad (44)$$

The quantization of the electromagnetic field is achieved by selecting b_k and $b_k^\dagger$ as mutually adjoint operators. Because photons are bosons, the suitable commutation relations for the operators b_k and $b_k^\dagger$ are the bosonic commutation relations:

$$[b_k, b_{k'}] = [b_k^\dagger, b_{k'}^\dagger] = 0, [b_k, b_{k'}^\dagger] = \delta_{kk'}. \quad (45)$$

The dynamical behavior of amplitude of electromagnetic field can be described by an ensemble of independent harmonic oscillators following the above commutation relations. Here, the Hamiltonian for the electromagnetic field is given by:

$$\mathcal{H} = \frac{1}{2} \int \varepsilon_0 \mathbf{E}^2 + \mu_0 \mathbf{B}^2 d\mathbf{r}. \quad (46)$$

Substituting Eq. (43) and the corresponding expression for $\mathbf{H}$ into Eq. (46) and applying the condition of Eq. (38) and (39), the Hamiltonian reduces to the form:

$$\mathcal{H} = \sum_k \hbar \omega_k \left(b_k^\dagger b_k + \frac{1}{2} \right). \quad (47)$$

This Hamiltonian represents the sum of the number of photons with the energy of $\hbar \omega_k$, where the additional $\frac{1}{2} \hbar \omega_k$ means the energy of vacuum fluctuation in each mode.

2.7 Hamiltonian of magnon polariton

In this section, we will obtain the Hamiltonian describing the coupling between magnons and photons in a ferromagnetic material. The mechanism of coupling is the dipole coupling between spins in the ferromagnetic materials and electromagnetic field via Zeeman coupling. Therefore, the convenient starting point is the Hamiltonian of Zeeman coupling. Here, both photons and magnons are assumed to be in a uniform mode with wavevector $k = 0$. This scenario is relevant to systems like ferromagnetic resonance experiments, where the uniform precession of spins interacts with a uniform alternating magnetic field. Then, the author introduces the essential concept of ultrastrong coupling regime, where the ratio of resonance frequency and coupling strength is over 0.1, and rotating wave approximation in this dissertation. Finally, the Hamiltonian of magnon polaritons with the relaxation of magnons and photons, which provides information about the influence of their relaxation on magnon polaritons.

Derivation of Hamiltonian for magnon polaritons

The Hamiltonian of the system where uniform magnon and photon coherently couple in the configuration of Fig. 2-10 is given by:

$$\mathcal{H} = \hbar \omega_m a^\dagger a + \hbar \omega_p b^\dagger b + \mathcal{H}_{\text{mp}}, \quad (48)$$

$$\mathcal{H}_{\text{mp}} = g \mu_B B_{\text{rf}}(t) \sum_i S_i^x. \quad (49)$$

The first and second terms in Eq. (48) are the Hamiltonian of magnon and photon mode, respectively. The ω_m is expressed by the Kittel mode given by Eq. (6). $\mathcal{H}_{mp}$ is the Zeeman coupling between alternating magnetic field $B_{rf}(t)$ originated from the magnetic field of photon mode and x -component of rotating spin at the site i in magnon mode S_i^x . The S_i^x and oscillating $B_{rf}(t)$ are expressed by Eq. (19) and the following equation:

$$B_{rf}(t) = B_{rms}(b + b^\dagger), \quad (50)$$

where B_{rms} is the amplitude of the alternating magnetic field. Substituting Eq. (19) and Eq. (50) into Eq. (49), we obtain:

$$\mathcal{H}_{mp} = g\mu_B B_{rms} \sum_i \sqrt{\frac{S}{2}} (a_i^\dagger + a_i)(b + b^\dagger). \quad (51)$$

The creation $a^\dagger$ and annihilation a operators of $k = 0$ magnons are given by:

$$a^\dagger = \sqrt{N} \sum_i a_i^\dagger, \quad a = \sqrt{N} \sum_i a_i. \quad (52)$$

Thus,

$$\mathcal{H}_{mp} = g\mu_B B_{rms} \sqrt{\frac{NS}{2}} (a^\dagger + a)(b + b^\dagger). \quad (53)$$

Finally, the total Hamiltonian is expressed as follows:

$$\mathcal{H} = \hbar\omega_m a^\dagger a + \hbar\omega_p b^\dagger b + g_{eff}(a^\dagger + a)(b + b^\dagger), \quad (54)$$

$$g_{eff} = g\mu_B B_{rms} \sqrt{\frac{NS}{2}}. \quad (55)$$

Here, S and N are the magnitude of spin and the number of spins in materials, respectively. The third term represents the Hamiltonian indicating the coupling between magnons and photons, where g_{eff} is the effective coupling strength. Increasing g_{eff} is essential for attaining a strongly coherent coupling between magnons and photons. Note that, according to Eq. (55), a greater number of spins benefits stronger coupling [24,27,87], favoring ferromagnetic materials due to their high spin density. Another essential factor influencing the effective g-factor is the amplitude of the oscillating magnetic field generated by the photon mode, B_{rms} which is proportional to $1/\sqrt{V}$ where V is the photon volume [29,40,88]. Therefore, confining the photons in small cavity is vital for strong

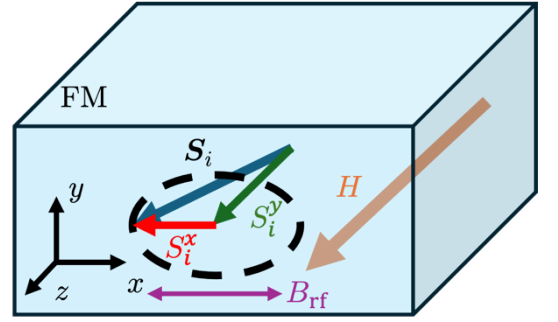


Figure 2-10. Schematic image depicting the interaction of spins and alternating magnetic field B_{rf} . The orange arrow is the external magnetic field.

magnon-photon coupling in addition to a large N . The typical way to achieve the small mode volume of photons is to use a resonator to confine photons.

To express the Hamiltonian in a form suitable for studying hybridization and energy exchange between magnons and photons, we rewrite the interaction term in the context of the rotating wave approximation (RWA), which keeps only energy-conserving terms. Under the RWA, counter-rotating terms ($a^\dagger b^\dagger$ and ab) can be disregarded because they occur at a much faster rate than energy-conserving terms, and their average expectation values is nearly zero. Thus, the total Hamiltonian becomes:

$$\mathcal{H} = \hbar\omega_m a^\dagger a + \hbar\omega_p b^\dagger b + g_{\text{eff}}(a^\dagger b + ab^\dagger). \quad (56)$$

This Hamiltonian describes a system of two coupled harmonic oscillators, where magnon mode and photon mode exchange their energy with the rate of coupling strength g_{eff} . This is referred to as the Tavis-Cummings model [89], which effectively explains magnon polariton systems by capturing the key dynamics and hybridization between magnon and photon modes.

The coupling system of magnon polaritons in superconducting resonator under ultrastrong coupling regime is known to be expressed by Hopfield model including the diamagnetic term $D(b^\dagger + b)^2 \propto \mathbf{A}^2$ with Eq. (54) [42,90–92]. The Hopfield model Hamiltonian is given by:

$$\mathcal{H} = \hbar\omega_m a^\dagger a + \hbar\omega_p b^\dagger b + g_{\text{eff}}(a^\dagger + a)(b + b^\dagger) + D(b^\dagger + b)^2. \quad (57)$$

Generally, the diamagnetic term stems from the expansion of the minimal coupling Hamiltonian of charge carriers moving in magnetic field. Whereas the electrons in superconducting resonators can govern this Hamiltonian in the system of magnon polaritons, the relationship of electrons, magnons and photons have not been fully explored, and a continuous report has been awaited.

Ultrastrong coupling (USC)

As explained in Fig. 1-4, the coupling system comes into USC regime when the ratio of g_{eff} to the resonance frequencies of magnon and photon mode (ω_p and ω_m), i.e., $g_{\text{eff}}/\{\omega_p, \omega_m\}$, is around over 0.1. Under ultrastrong coupling, RWA breaks down, and the counter-rotating terms can no longer be ignored. The Hamiltonian of magnon polaritons now includes terms like the following:

$$\mathcal{H}_{\text{CR}} = g_{\text{eff}}(a^\dagger b^\dagger + ab), \quad (58)$$

which represents processes where a magnon and a photon are created or annihilated together. These terms fundamentally alter the nature of the ground state and energy spectrum, introducing new dynamics and excitations absent in the weak or strong coupling regime. One of the key consequences of the inclusion of counter-rotating terms in the USC regime is the entanglement of the ground state [41,42]. In contrast to weak coupling, where the ground

state is typically separable and involves no excitation, the ground state in the USC regime becomes a superposition of the vacuum and virtual excitations [93]. This entangled ground state reflects virtual processes where excitations are created and annihilated in rapid succession due to the intense coupling strength. This leads to correlations between the magnon and photon fields even in the absence of real excitations. This ground-state entanglement is a hallmark of the USC regime, revealing unique quantum properties with potential applications in areas like quantum information, where ground-state entanglement can serve as a resource for various protocols.

One of the symbolic phenomena in USC regime is Bloch-Siegert Shift, the resonance shift due to the existence of counter-rotating terms [94–96]. Although the entanglement of ground state in USC regime has not been demonstrated, the observation of Bloch-Siegert Shift provides the indirect evidence of the counter-rotating terms. Thus, observing the Bloch-Siegert Shift marks a key step in utilizing the ultrastrong coupling regime.

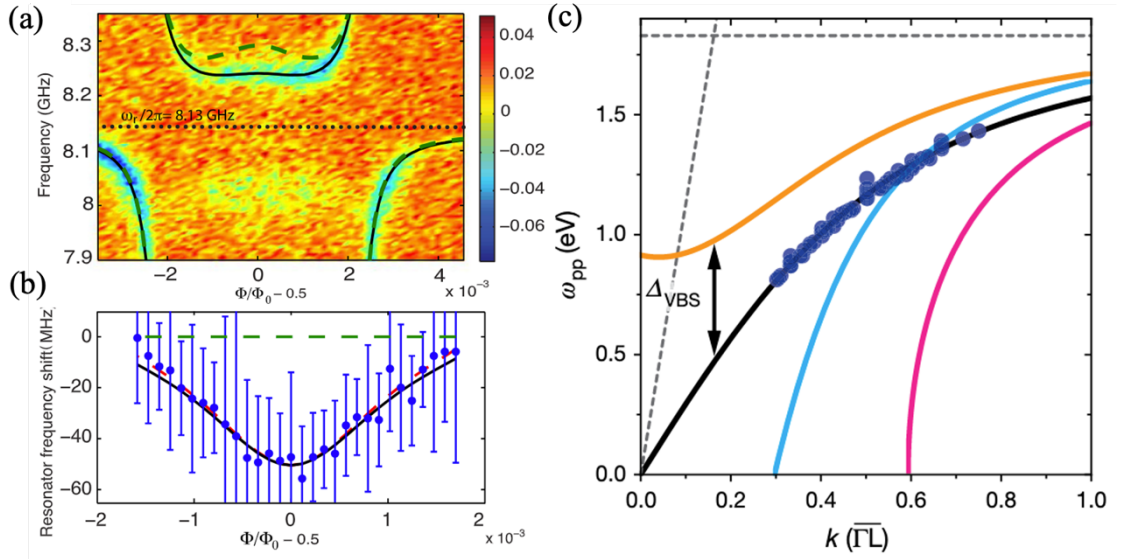


Figure 2-11. Figures showing the Bloch-Siegert shift in (a)(b) qubit-oscillator system and (c) plasmonic nanoparticle crystals. (a) Spectrum in proximity to the resonator frequency. The solid black line is the fit of a certain coupling Hamiltonian, and the dashed green line is a plot without counter-rotating terms. A distinct discrepancy between the dashed line and the data is evident in the vicinity of the qubit's symmetry point. A transition linked to the thermal population of the qubit's excited state can be discerned at approximately 8 GHz. (b) Difference between the measurement (blue dots) and the prediction of the Hamiltonian without counter-rotating terms. (c) Polariton dispersion measured and calculated with full Hamiltonian (black line), without counter-rotating terms (orange line), Dicke model (pink line) and with the RWA (blue line). These figures are quoted by [94] for Fig. 2-11. (a-b) and [96] for Fig. 2-11. (c).

2.8 Tavis-Cummings model with relaxations of magnon and photon modes

In realistic systems, both magnons and photons experience energy dissipation due to various relaxation mechanisms. Incorporating these relaxation processes into the Tavis-Cummings model is essential for accurately predicting system dynamics and interpreting experimental results in weak or strong coupling. This section presents a derivation of the eigenfrequencies and photon relaxation rates in the Tavis-Cummings model when relaxation terms are included.

First of all, introducing the relaxation rate of magnons and photons as γ and κ , the Heisenberg equations of motions for a and b are given by:

$$\frac{da}{dt} = \frac{i}{\hbar} [\mathcal{H}, a] = -i\omega_m a - g_{\text{eff}} b - \gamma a, \quad (59)$$

$$\frac{db}{dt} = \frac{i}{\hbar} [\mathcal{H}, b] = -i\omega_p b - g_{\text{eff}} a - \kappa b. \quad (60)$$

Assuming solutions of the form:

$$a(t) = a_0 e^{-i\omega t}, b(t) = b_0 e^{-i\omega t}. \quad (61)$$

Substituting Eqs. (61) into Eqs. (59-60), the static state for this system is written in matrix form:

$$\begin{pmatrix} i(\omega_m - \omega) + \gamma & g_{\text{eff}} \\ g_{\text{eff}} & i(\omega_p - \omega) + \kappa \end{pmatrix} \begin{pmatrix} a_0 \\ b_0 \end{pmatrix} = 0. \quad (62)$$

Non-trivial solutions exist if the determinant of the coefficient matrix is zero:

$$[i(\omega_m - \omega) + \gamma][i(\omega_p - \omega) + \kappa] - g_{\text{eff}}^2 = 0. \quad (63)$$

Solving this equation, the eigenfrequency $\tilde{\omega}_{\pm}$ for this system is given by:

$$\tilde{\omega}_{\pm} = \frac{\omega_m + \omega_p - i(\kappa + \gamma)}{2} \pm \frac{1}{2} \sqrt{[\omega_p - \omega_m - i(\gamma - \kappa)]^2 + 4g_{\text{eff}}^2}. \quad (64)$$

The real and imaginary part of Eq. (64) represent the resonance frequency and the relaxation rate of magnon polaritons, respectively. In the case of γ (in general, Magnis on relaxation γ is much larger than photon relaxation κ), and the resonance frequency $\omega_{\pm}$ and relaxation $\kappa_{\pm}$ are given by:

$$\omega_{\pm} = \frac{\omega_m + \omega_p}{2} \pm \frac{1}{2} \sqrt{[\omega_p - \omega_m]^2 + 4g_{\text{eff}}^2}, \quad (65)$$

$$\kappa_{\pm} = \frac{\kappa + \gamma}{2} \pm \frac{\kappa - \gamma}{2} \frac{\omega_p - \omega_m}{\sqrt{[\omega_p - \omega_m]^2 + 4g_{\text{eff}}^2}}. \quad (66)$$

Considering these equations, we can study the coupling parameter such as g_{eff} , κ , and γ in real systems in weak or strong coupling regimes, by fitting the spectra with Eq. (65) and (66). However, these calculations are not valid within the ultrastrong coupling regime since RWA breaks down. Thus, we need to derive the eigenfrequencies for the full Hamiltonian of Eq. (57).

2.8 Derivation of the resonance frequency for magnon polaritons in USC regime

Following Hopfield [90], it is convenient to introduce polariton operators $\hat{p}$ and diagonalize the Hamiltonian to derive the resonance frequencies of magnon polaritons under USC regime. The polariton operators are given by:

$$\hat{p}^{\pm} = \alpha^{\pm}\hat{a} + \beta^{\pm}\hat{b} + \gamma^{\pm}\hat{a}^{\dagger} + \delta^{\pm}\hat{b}^{\dagger}. \quad (67)$$

Then, we find the coefficient of the polariton operators that represents Eq. (57) in the diagonal form as follows:

$$\mathcal{H} = \hbar\omega^{+}\hat{p}^{+\dagger}\hat{p}^{+} + \hbar\omega^{-}\hat{p}^{-\dagger}\hat{p}^{-}. \quad (68)$$

This procedure corresponds to the establishment of the eigenvalue problem:

$$[\hat{p}^{\pm}, \mathcal{H}] = \omega^{\pm}\mathcal{H}. \quad (69)$$

The polariton frequencies corresponds with the eigenvalues of the following Hopfield matrix [91,92]:

$$M = \begin{pmatrix} \omega_m + 2D & g_{\text{eff}} & -2D & -g_{\text{eff}} \\ g_{\text{eff}} & \omega_m & -g_{\text{eff}} & 0 \\ 2D & g_{\text{eff}} & -\omega_p - 2D & -g_{\text{eff}} \\ g_{\text{eff}} & 0 & -g_{\text{eff}} & -\omega_p \end{pmatrix}. \quad (70)$$

Applying the Thomas-Reiche-Khun rule, $D = \frac{g_{\text{eff}}^2}{\omega_m}$, which is valid when the coupling is

mediated by dipole interactions, and redefining g_{eff} as $g_{\text{eff}}\sqrt{\omega_m/\omega_p}$ in Eq. (70), we

obtain the dispersion relation of magnon polaritons without any approximation as follows:

$$\omega^{\pm} = \sqrt{\frac{\omega_m^2 + \omega_p^2 + 4g_{\text{eff}}^2 \pm \sqrt{[\omega_m^2 + \omega_p^2 + 4g_{\text{eff}}^2]^2 - 4\omega_m^2\omega_p^2}}{2}}. \quad (71)$$

This dispersion relations allow for the estimation of the coupling strength between magnons and photons. Finally, the other forms such as the Hamiltonians without counter-rotating terms of $\mathcal{H}_{\text{CR}}$, the diamagnetic term and with RWA are expressed as below.

- The Hamiltonian without counter-rotating terms, $\mathcal{H}_{\text{CR}}$, gives the dispersion relations as follows:

$$\omega^{\pm} = \frac{\omega_m + \omega_p + \frac{2g_{\text{eff}}^2}{\omega_p}}{2} \pm \sqrt{\frac{\left[\omega_m + \omega_p + \frac{2g_{\text{eff}}^2}{\omega_p}\right]^2 - 4\omega_m\omega_p - 4g_{\text{eff}}^2\frac{\omega_m}{\omega_p}}{2}}. \quad (72)$$

- The Hamiltonian without the diamagnetic term, known as Dicke model, gives the dispersion relations as follows:

$$\omega^{\pm} = \sqrt{\frac{\omega_m^2 + \omega_p^2 \pm \sqrt{[\omega_m^2 - \omega_p^2]^2 + 16\omega_m^2 g_{\text{eff}}^2}}{2}}. \quad (73)$$

- The Hamiltonian with RWA gives the the dispersion relations as follows:

$$\omega^{\pm} = \frac{\omega_m + \omega_p}{2} \pm \frac{\sqrt{[\omega_m - \omega_p]^2 + 4g_{\text{eff}}^2 \frac{\omega_m}{\omega_p}}}{2}, \quad (74)$$

which is consistent with Eq. (65) in the condition of $g_{\text{eff}} \rightarrow g_{\text{eff}} \sqrt{\omega_m/\omega_p}$.

Comparing the dispersion relations with the estimated values obtained through fitting demonstrates the presence of the Bloch-Siegert shift, confirming that counter-rotating terms are validated within the system.

Chapter 3. Control of magnon damping in ultrathin ferromagnetic films

3.1 Abstract

In this chapter, the author reports on the investigation of magnon damping in ultra-thin FM film. Here, the key parameter for the control of magnon damping is the surface state of FM films, such as surface roughness and surface magnetic anisotropy. A key aspect of designing adjustable magnon damping in thin ferromagnetic films is the careful selection of both the buffer and capping layers, in addition to the thin ferromagnetic films themselves. The author carefully investigated the effect of the adjacent layer on the Gilbert damping constant, a significant indicator of magnon damping, and established the guiding principle for designing magnon damping in real devices using thin FM films.

Results presented in this chapter were published in the papers:

S. Yoshii, R. Ohshima, Y. Ando, T. Shinjo, and M. Shiraishi, “Detection of ferromagnetic resonance from 1 nm-thick Co”, *Scientific Reports* **10**, 15764 (2020).

S. Yoshii, K. Kato, E. Shigematsu, R. Ohshima, Y. Ando, K. Usami, and M. Shiraishi, “Significant suppression of two-magnon scattering in ultrathin Co by controlling the surface magnetic anisotropy at the Co/nonmagnet interfaces”, *Physical Review B* **106**, 174414 (2022).

S. Yoshii, M. Müller, H. Inoue, R. Ohshima, M. Althammer, Y. Ando, H. Huebl, and M. Shiraishi, "Significant modulation of Gilbert damping in ultrathin ferromagnetic films by altering the surface magnetic anisotropy", *Physical Review B* **109**, L020406 (2024).

3.2 Introduction and research purpose

3.2.1 Thin ferromagnetic film

The magnon, which is a quasi-particle form of a spin wave and represents the minimum excitation state of the ferromagnetic ground state, is expected to be employed as an information carrier with minimal energy consumption due to its prolonged lifetime [9,12]. Magnons have the capacity to interact with other quantum entities, including photons and phonons. The interplay between the coupling strengths and relaxation rates of the magnons and the coupled media provides a foundation for the development of novel quantum technologies. Indeed, when the coupling strength exceeds the relaxation rates of both the magnon and the coupled medium, a strong coupling state can be established, enabling the superposition of quantum states between the two quantum systems, namely, between magnons and photons [24,25,27]. The considerable attractiveness of this strong coupling state, which allows for the realization of quantum computation [14,50,97], quantum sensing [26,98,99], and novel spintronic functions [29,54], has led to the magnons and photons coupling being the subject of intense research activity over recent decades. In light of the considerable potential for utilizing strong magnon-photon coupling in quantum computing and pioneering spintronics, the capacity to reversibly and adaptively modulate the coupling strength from weak to strong state assumes considerable significance. Nevertheless, the prevailing challenge to the implementation of magnon-photon coupling is the limited tunability. Indeed, there are some studies that seek to achieve tunable magnon-photon coupling through the modulation of photon excitation [100,101]. It is important to note, however, that the direct control of a magnon state was demonstrated only via temperature control. In this way, the coupling between magnons excited in gadolinium-iron-garnet (GIG) and photons confined within a three-dimensional microwave cavity was tuned [102]. While the demonstration of tunable magnon-photon coupling was significant, it remains challenging to envisage practical applications of these temperature-controlled coupling states in either quantum technologies or spintronics. It is therefore anticipated that subsequent studies will be conducted to explore the novel processes that are required to realize tunable coupling states.

The recent discovery of gate-tunable magnetization in ultrathin Co [77,78] and in a few atomic layers of Fe [75], in addition to gate-tunable spin-orbit interaction (SOI) observations in ultrathin Pt and Pd layers [103–105], has enabled exploration of new horizons in nano-spintronics. These studies have challenged previous understandings in the research field, namely that the magnetization of ferromagnetic materials cannot be controlled using an electric field and that the magnitude of the SOI in solids is material-specific. It has been demonstrated that the application of a strong electric field enables the accumulation of carriers in ultrathin solids to a density sufficient for the efficient modulation of their Fermi levels. The successful modulation of the Fermi level of an ultrathin ferromagnetic film [75, 77, 78] offers a potential route to the realization of magnetic anisotropy modulation, which would result in the modulation of magnon excitation. It can be reasonably proposed that the

excitation of magnons in ultrathin ferromagnets represents the initial step in the process of developing gate-tunable magnon-photon coupled systems. Despite the fact that the linewidth of the ferromagnetic resonance (FMR) of ultrathin ferromagnets, which serves as an effective indicator of the magnon excitation, was too large (ca. 200 mT [106]), insertion of a Ta seeding layer beneath the 1-nm-thick Co layer produced a prominent FMR spectrum with a linewidth of 15 mT [107], marking the first demonstration of salient magnon excitation in ultrathin ferromagnets. It is also important to note that the Gilbert damping constant α of the Co (1 nm)/Ta (3 nm) system is not sufficiently small, and considerable effort is still required to suppress it. Given that α is comprised of three fundamental components, namely the intrinsic, spin pumping, and two-magnon scattering (TMS) terms, a comprehensive grasp of the interrelationship between these elements and the means of mitigating each in an ultrathin ferromagnet is of paramount importance.

3.2.2 Magnon damping in thin ferromagnetic films

The phenomenon of magnetization damping was first formulated by Gilbert [108] as the Gilbert damping constant α . Since then, it has been the subject of extensive theoretical and experimental investigation over several decades [67,109–111]. In a ferromagnetic (FM)/nonmagnetic (NM) bilayer, the Gilbert damping constant can be decomposed into three terms as a function of the FM film thickness t_{FM} , as follows [112]:

$$\alpha = \alpha_{\text{int}} + \alpha_{\text{SP}} t_{\text{FM}}^{-1} + \beta_{\text{TMS}} t_{\text{FM}}^{-2}, \quad (75)$$

where α_{int} , α_{SP} , and β_{TMS} are the thickness-independent intrinsic damping constant of the FM layer, the damping constant due to spin pumping, and the TMS coefficient, respectively. The spin-pumping contribution α_{SP} , which arises from the relaxation of spin current from the FM layer relaxes in NM materials, has been reported to be 0.055 nm for Co/Pt and 0.026 nm for Fe/Pt [113]. The TMS coefficient β_{TMS} was reported to be $2.73 \pm 0.02 \text{ nm}^2$ for Co/SiO₂ and $0.41 \pm 0.04 \text{ nm}^2$ for Co/Pt [114]. As the contribution of TMS to magnon damping is proportional to the square of $1/t_{\text{FM}}$, this contribution is significantly amplified in nanometer-thick FMs, which subsequently impedes the realization of low magnon damping in ultrathin FM films. It is therefore imperative that β_{TMS} be suppressed in order to achieve the desired low magnon damping in ultrathin FM films. Furthermore, β_{TMS} is also known to be dependent on the surface conditions of FM films, including the surface roughness and the surface magnetic anisotropy (SMA) [115,116]. Consequently, an investigation of the correlation between the TMS-induced damping and the surface state of the FM film can also provide an invaluable guiding principle to aid in achieving low magnon damping in ultrathin FM films.

In this chapter, we investigate magnetization damping in ultrathin ferromagnetic films by changing the surface state of ferromagnetic material. Equipping with a wide variety of NM seeding layers with different SMA energies, we then verify the significantly weak magnon damping that occurs in ultrathin Co by changing the seeding layer species. A scaling

relationship between the TMS and the SMA is found that provides compelling evidence that the SMA governs the physics behind our findings, which can be the guiding principle for controlling the magnon damping in ultrathin ferromagnetic films. Following this guiding principle, the author demonstrated a giant modulation of two-magnon scattering in CoFe alloys by changing the thickness of MgO capping layer on CoFe layer. In addition to the photon relaxation rate and the magnon-photon coupling strength, the magnon relaxation rate of the ultrathin Co layer with the NM seeding layer is investigated quantitatively using a cavity quantum electrodynamics (QED) system, and potential approaches to realization of the strong coupling states are proposed.

3.3 Experimental setup

This section introduces the experimental information of sample fabrication and measurement in (a) Ta buffer layer influence on magnetization damping in 1 nm-thick Co film, (b) influence of buffer layer on Gilbert damping in ultrathin Co films, (c) influence of thickness dependence of MgO capping layer on Gilbert damping in ultrathin $\text{Co}_{25}\text{Fe}_{75}$ films, and (d) estimation of magnon damping in the coupling between cavity photon and magnon in ultrathin FM films. Here, this experimental information is referred to the papers of:

- (a): S. Yoshii, R. Ohshima, Y. Ando, T. Shinjo, and M. Shiraishi, "Detection of ferromagnetic resonance from 1 nm-thick Co", Scientific Reports **10**, 15764 (2020).
- (b) + (d): S. Yoshii, K. Kato, E. Shigematsu, R. Ohshima, Y. Ando, K. Usami, and M. Shiraishi, "Significant suppression of two-magnon scattering in ultrathin Co by controlling the surface magnetic anisotropy at the Co/nonmagnet interfaces", Physical Review B **106**, 174414 (2022).
- (c) S. Yoshii, M. Müller, H. Inoue, R. Ohshima, M. Althammer, Y. Ando, H. Huebl, and M. Shiraishi, "Significant modulation of Gilbert damping in ultrathin ferromagnetic films by altering the surface magnetic anisotropy", Physical Review B **109**, L020406 (2024).

3.3.1 Sample fabrication

- **Ta buffer layer influence on magnetization damping in 1 nm-thick Co film**

Figure 3-1(a) illustrates the schematics of the sample structures. Two distinct types of samples were prepared. Figures 3-1(a) shows the schematics of sample structures. We prepared two different types of samples: One is SiO_2/Co (type-A), and the other is $\text{SiO}_2/\text{Co}/\text{Ta}$ (type-B) (see Fig. 3-1(a)). In the case of the Type-A samples, a Co thin film with a thickness of 1, 2, 3 or 5 nm was deposited onto a SiO_2 (300 nm)/Si substrate through the use of radio-frequency magnetron sputtering. In the case of the Type-B samples, a Ta buffer layer with a thickness of 3 nm was deposited beneath the 1 nm-thick Co: One is SiO_2/Co (type-A), and the other is $\text{SiO}_2/\text{Co}/\text{Ta}$ (type-B) (see Fig. 3-1(a)). In the case of the Type-A samples, a Co thin film with a thickness of 1, 2, 3 or 5 nm was deposited onto a SiO_2 (300 nm)/Si substrate using radio-frequency magnetron sputtering. In the case of the

type-B samples, a Ta buffer layer of 3 nm thickness was deposited beneath the 1 nm-thick Co.

In the type-A samples, a Co thin film of 1, 2, 3, or 5 nm in thickness layers was deposited using a radio-frequency magnetron sputtering system. The base pressure of the sputtering system was maintained at a level below 2.5×10^{-5} Pa, with the flow rate and partial pressure of the Ar gas set at 5 sccm and 0.5 Pa, respectively. The deposition temperature was kept at room temperature (RT). The deposition rate of the Co was 0.8 nm/min. A layer of SiO₂ layer (10 nm) was deposited onto the Co film in order to prevent from oxidation of the Co layer. In the case of the type-B samples, a Ta buffer layer with a thickness of 3 nm was deposited, with a deposition rate of the Ta of 3.83 nm/min. The substrate was identical to that employed for the type-A samples. During the deposition of Co and Ta, the sample holder was rotated at a rate of 20 rpm.

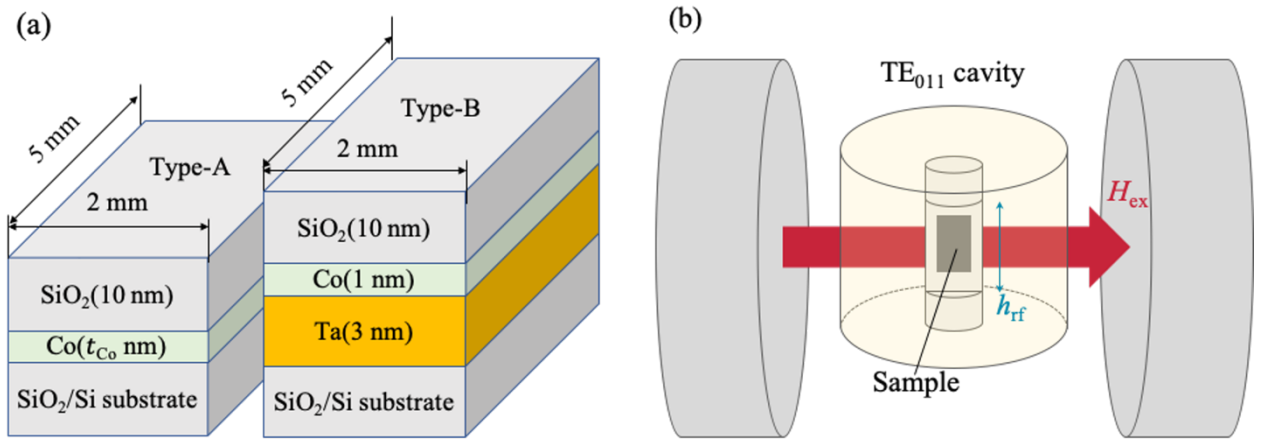


Figure 3-1. (a) Schematics of the sample structures of the type-A and the type-B samples. The thickness of the Co (t_{Co}) in the type-A samples was 1, 2, 3, or 5 nm. The samples were capped with a 10 nm-thick SiO₂ layer to prevent oxidization of the Co. (b) Schematic of the setup used for cavity FMR measurement. The figures are quoted by [107].

- **Influence of buffer layer on Gilbert damping in ultrathin Co films**

SiO₂ cap/MgO (2 nm)/Co (t_{Co})/NM (3 nm) film structures were prepared on SiO₂ substrates to investigate the correlation between TMS contribution and the uniaxial SMA field, where the Co thickness t_{Co} ranged from 2 nm to 20 nm, and the NM materials were Ti, Cu, and Al, as shown in Fig. 3-2 and Table 3-1. The Ti, Cu, Al, Co, and MgO layers were deposited using electron beam deposition (EB-depo. in the table), with the deposition pressure of 10^{-6} Pa. The SiO₂ cap and Ta seeding layers were deposited using radio frequency (RF) magnetron sputtering, with a base pressure of approximately 3×10^{-5} Pa. The Ar gas was employed for the excitation of the plasma with a gas flow rate of 5 sccm, and a deposition pressure of 0.50 Pa. Additionally, SiO₂ cap/MgO (2 nm)/Co (t_{Co}) films on SiO₂ substrates were also prepared for use as reference samples.

Sample #	Stacking order	Preparation method
1	SiO ₂ cap/MgO (2)/Co (t_{Co})/Ti (3)/SiO ₂ sub	SiO ₂ cap : Sputter MgO/Co/NM : EB-depo
2	SiO ₂ cap/MgO (2)/Co (t_{Co})/Al (3)/SiO ₂ sub	
3	SiO ₂ cap/MgO (2)/Co (t_{Co})/Cu (3)/SiO ₂ sub	
4	SiO ₂ cap/MgO (2)/Co (t_{Co})/ SiO ₂ sub	
5	SiO ₂ cap/Co (2)/Ta (3)/ SiO ₂ sub	SiO ₂ /Co/Ta : Sputter

Table 3-1. List of the prepared samples and their preparation methods.

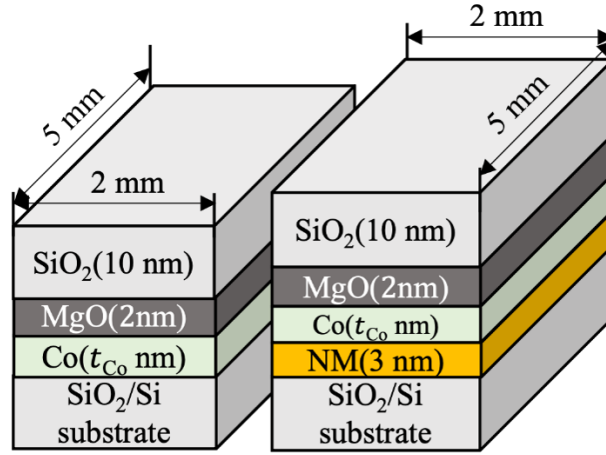


Figure 3-2. The schematics of the sample structures depicted in Table. 3-1 illustrate the variation in thickness of the Co film (t_{Co}) was altered from 2 nm to 20 nm. The samples were capped with a 2 nm-thick MgO layer and a 10 nm-thick SiO₂ layer for samples 1, 2, 3 and 4.

- Influence of thickness dependence of MgO capping layer on Gilbert damping in ultrathin Co₂₅Fe₇₅ films**

We fabricated Ta capping/MgO (t_{MgO} nm)/Co₂₅Fe₇₅ (t_{CF} nm)/Cu (3 nm)/Ta (3 nm) multi-layers, which were deposited *in-situ* on a thermally oxidized Si substrate by dc and rf magnetron sputtering at room temperature. The MgO/Co₂₅Fe₇₅ heterostructures were fabricated on thermally oxidized SiO₂/Si substrates. To optimize the growth conditions of the Co₂₅Fe₇₅ and achieve low magnetization damping, we employed Cu (3 nm)/Ta (3 nm) bilayers as a buffer layer [67]. In order to prevent the Co₂₅Fe₇₅ layers from oxidizing, we deposited Ta capping layers with a sufficient thickness (1.5 or 3 nm) on the MgO/Co₂₅Fe₇₅ bilayers. All deposition processes were conducted at room temperature utilizing 2" targets, and the base pressure in the deposition chamber was maintained below 5.0×10^{-7} Pa. The thicknesses of the MgO and Co₂₅Fe₇₅ were varied from 0.4 to 4.0 nm and from 2 to 20 nm, respectively.

Target material	Power [W]	Rate [$\text{\AA}/\text{s}$]
Ta (dc)	20	0.5
Cu (dc)	20	1.0
Co ₂₅ Fe ₇₅ (dc)	20	2.2
MgO (rf)	30	0.12

Table 3-2. Sputtering conditions for deposition. All materials are grown under an Ar gas pressure of 5.0×10^{-1} Pa.

3.3.2 Measurement

- Ta buffer layer influence on magnetization damping in 1 nm-thick Co film

Figure 3-1 (b) depicts the schematic of the FMR measurement setup for cavity FMR measurement. FMR measurements were conducted using a TE₀₁₁ (transverse electric mode) cavity of an electron spin resonance system (JEOL JES-FA 200). An external magnetic field under microwave irradiation was applied parallel to the sample plane, and the microwave frequency and power were set to be 9.12 GHz and 10 mW, respectively (see Fig. 1(b)). All of the FMR measurements were performed at RT.

The FMR spectra were deconvolved using the internal form of the FMR spectra. In addition to the Lorentzian (symmetric) component, an asymmetric component was incorporated into the analysis. The fitting function utilized is as follows:

$$\begin{aligned}
 F(\mu_0 H) = & A_{\text{sym}} \frac{\Delta\mu_0 H^2}{(\mu_0 H - \mu_0 H_{\text{res}})^2 + \Delta\mu_0 H^2} \\
 & - 2A_{\text{asym}} \frac{\Delta\mu_0 H(\mu_0 H - \mu_0 H_{\text{res}})}{(\mu_0 H - \mu_0 H_{\text{res}})^2 + \gamma\Delta\mu_0 H^2} + aH + b,
 \end{aligned} \tag{76}$$

where $\Delta\mu_0 H$ is the half-width at half-maximum, A_{sym} and A_{asym} are symmetric and asymmetric components, respectively, a and b are constants.

- Influence of buffer layer on Gilbert damping in ultrathin Co films

The objective of this investigation was to ascertain the correlation between the TMS contributions and the uniaxial SMA field at the Co/NM interfaces. To this end, two dependences of the FMR signal were examined: its microwave-frequency and magnetic-field-angular dependences. The microwave-frequency dependence FMR signals were measured with the intention of estimating the Gilbert damping constant. This was achieved through the use of a broadband FMR employing a coplanar waveguide (CPW). Following the adjustment of the microwave frequency, the intensity of microwave transition was quantified by sweeping the external magnetic field from 0 mT and 300 mT along the in-plane direction

(see Fig. 3-5(a)). Similar measurements were conducted under a range of microwave frequencies, with the aim of measuring microwave-frequency dependence of the FMR signals. The microwave frequency was varied from 5 to 15 GHz, while the input power was set at 10 dBm. To measure the angle-dependent FMR spectra, the JES-FA200 electron spin resonance (ESR) spectrometer (JEOL Resonance Inc., Japan) was utilized. Excitation was conducted in TE₀₁₁ mode, with a microwave frequency and power at the output port of 9.12 GHz and 10 dBm, respectively.

- Influence of thickness dependence of MgO capping layer on Gilbert damping in ultrathin Co₂₅Fe₇₅ films

The author conducted broadband FM resonance (bbFMR) measurements at room temperature, wherein an in-plane external magnetic field was applied. Figure 3-8(a) depicts a schematic representation of the measurement setup for bbFMR. The prepared samples were mounted facedown onto a coplanar waveguide (CPW), and the complex microwave transmission S_{21} was measured using a vector network analyzer (VNA). The microwave frequency (f) was varied between 5 and 20 GHz, while the in-plane external magnetic field was swept from 0 to 0.3 T. The microwave power was set to 0 dBm for all bbFMR measurements. Further details concerning the experimental setup can be found in Ref. [117].

3.4 Suppression of magnetization damping of 1-nm Co film by inserting Ta buffer layer

3.4.1 Co thickness dependence of FMR measurement

Figures 3-1(a) and (b) illustrate the sample structures and the measuring setup, respectively. Two distinct sample types were prepared: SiO₂/Co (type-A), and SiO₂/Co/Ta (type-B) (see Fig. 3-1(a)). In the type-A samples, a Co thin film of 1,2,3 or 5 nm thickness was deposited onto a SiO₂ (300 nm)/Si substrate using radio-frequency magnetron sputtering. In the type-B samples, a Ta buffer layer with a thickness of 3 nm was deposited beneath the 1 nm-thick Co. FMR measurements were performed using a TE₀₁₁ (transverse electric mode) cavity of an electron spin resonance system. Figure 3-3(a) depicts the FMR spectra of the four type-A samples, wherein the thickness of the Co film was varied. The resonance field and the linewidth of the spectra exhibited a significant change with variation in the thickness of the Co layer. Of particular significance was the observation that an FMR signal was undetectable when the Co thickness was less than 2 nm.

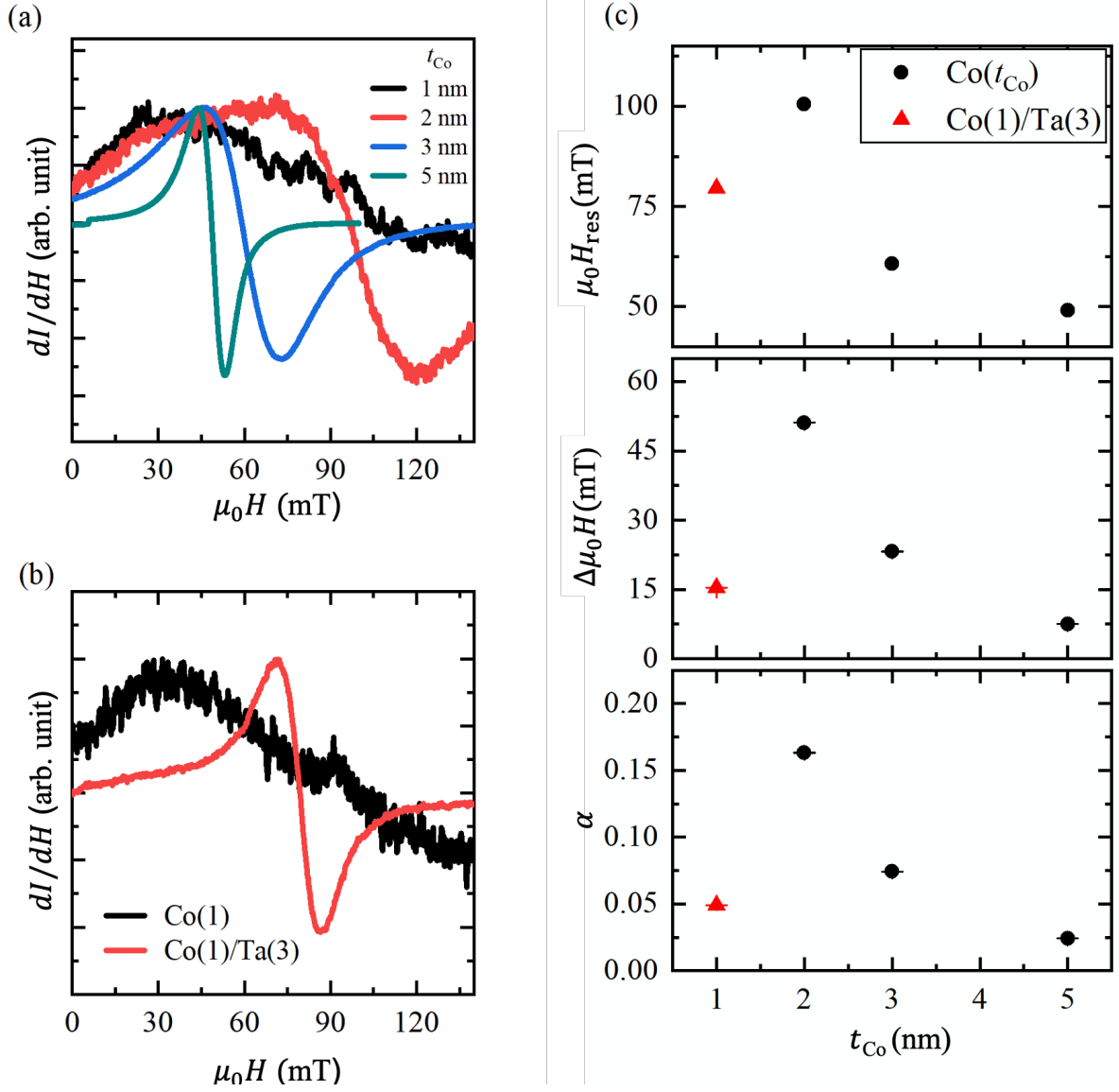


Figure 3-3. (a) FMR spectra of sample-A Co of 1, 2, 3, and 5 nm in thickness. (b) Comparison of the FMR spectra of the Co (1 nm) and Co (1 nm)/Ta (3 nm) samples. (c) The Co thickness dependence of the resonance field $\mu_0 H_{\text{res}}$ (upper panel), the half-width at half-maximum of the FMR spectra $\Delta\mu_0 H$ (middle panel), and the Gilbert damping constant α (lower panel).

To circumvent the issue of the absence of the FMR in thin Co films, we introduced Ta as a buffer layer (type-B sample). Amorphous Ta is widely recognized to possess a relatively smooth surface [118,76,77], although direct observation of the smooth surface of Ta was challenging without exposing the surface to air in our experimental setup. Additionally, structural analyses such as x-ray diffraction could not be conducted due to the thin Ta layer (3 nm). Hence, we expected that the insertion of Ta beneath ultrathin Co would facilitate the formation of a flat and continuous Co film with nm-thickness, resulting in a sharp FMR spectrum from the Co. Indeed, Chiba and co-workers observed the anomalous Hall effect from 0.4 nm-thick Co grown on 3 nm-thick Ta, which provides compelling evidence for the

formation of a continuous film [76,77]. Figure 2(b) illustrates a comparison of the FMR spectra of the Co (1 nm) and Co (1 nm)/Ta (3 nm) samples. The distinction between the FMR spectra is readily apparent, and the FMR spectrum of the Co (1 nm)/Ta (3 nm) sample is particularly evident. Given that amorphous Ta exhibits a uniform surface, the distinct FMR spectrum of the Co is attributed to the insertion of the Ta buffer layer with a smooth surface beneath the one nm-thick Co. Figure 3-2 (c) depicts the resonance fields, the half-width at half-maximum of the FMR spectra, and the Gilbert damping constant α as functions of the Co layer thickness for the type-A and -B samples. The resonance field and the $\Delta\mu_0 H$ were obtained through deconvolution of the integral form of the FMR spectra (for further detail, see Methods and Appendix). The Gilbert damping constant α was calculated using the following equation,

$$\alpha = \frac{\Delta\mu_0 H \cdot \gamma}{2\pi f_{\text{res}}}, \quad (77)$$

where $\Delta\mu_0 H$ is the half-width at half-maximum of an FMR spectrum, γ is the gyromagnetic ratio of Co, and f_{res} is the applied microwave frequency [119]. While the resonance field, the half-width at half-maximum of the FMR spectra, and the Gilbert damping constant exhibited a monotonic increase with decreasing the Co thickness, the 1 nm-thick Co layer overlying a Ta buffer layer demonstrated a notable suppression of these parameters. Therefore, it can be confirmed that the incorporation of an amorphous Ta layer beneath an ultrathin Co layer represents an efficient approach to excite FMR in the 1 nm-thick Co.

In the preceding studies, the thickness of the amorphous Ta buffer layer was fixed at 3 nm [76,77], therefore the thickness of the Ta buffer layer was varied from 2 to 5 nm in increments of 1 nm. The FMR of the 1 nm-thick Co on Ta buffer layers of various thickness was then measured. The FMR spectra of the Co layers with Ta buffer layers of various thicknesses and the Gilbert damping constants of each sample are presented in Figs. 3-3(a) and (b), respectively. Neither the FMR spectra nor the magnitude of the Gilbert damping constants exhibit a thickness-dependent behavior, indicating that the insertion of a 3 nm-thick Ta buffer layer is sufficient to induce the formation of a flat Co layer. It is noteworthy that an FMR signal was not observed from the Co (1 nm)/ Ta (1 nm) sample (see Appendix), which directly indicates that an excessively thin Ta buffer layer does not allow for the excitation of the FMR in a 1 nm-thick Co layer.

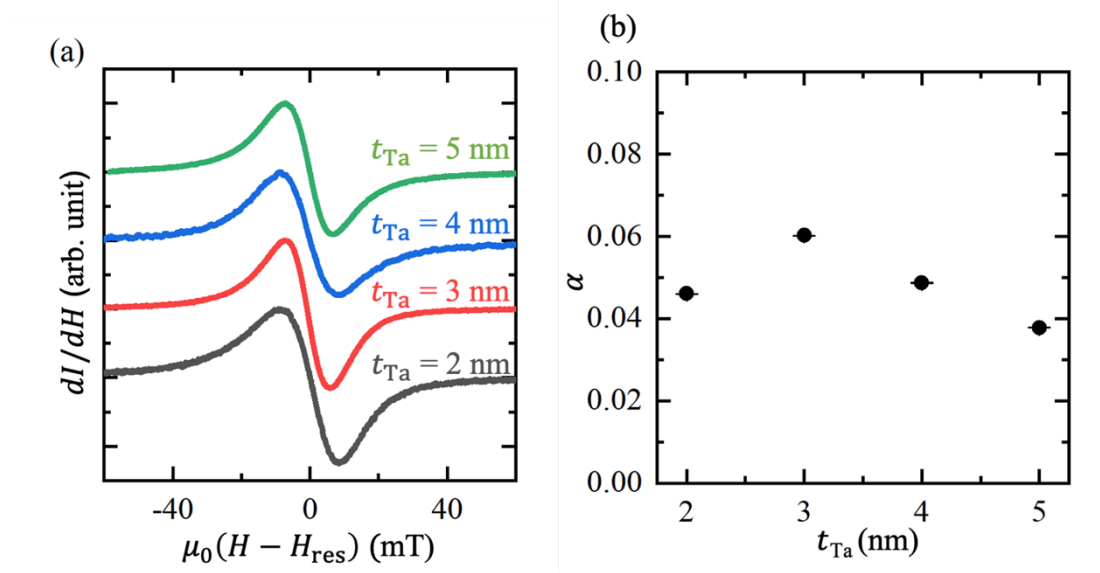


Figure 3-4. (a) FMR spectra from the Co (1 nm)/Ta (t_{Ta} nm) samples. The thickness of the Ta (t_{Ta}) was changed from 2 to 5 nm. (b) The t_{Ta} dependence of the Gilbert damping constant α .

3.4.2 Effect of oxidation of Ta buffer layer on FMR linewidth

In order to gain a deeper insight into the aforementioned phenomena, a Co (1 nm)/ $\emptyset$ /Ta (3 nm) sample was prepared, wherein the surface of the Ta was deliberately oxidized ($\emptyset$ denotes that the sample was exposed to air during fabrication process). Figure 3-5(a) presents a comparative analysis of the FMR spectra of the Co (1 nm)/Ta (3 nm) and Co (1 nm)/ $\emptyset$ /Ta (3 nm) samples. A notable discrepancy is evident in the FMR spectra with the absence of an FMR signal in the Co (1 nm)/ $\emptyset$ /Ta (3 nm) sample despite the incorporation of a 3 nm-thick Ta buffer layer. This absence in Co (1 nm)/ $\emptyset$ /Ta (3 nm) sample can be attributed to the oxidized surface of the Ta, which has resulted in the lack of an FMR signal. As previously stated, the insertion of an amorphous Ta layer beneath the 1 nm Co facilitates the formation of a flat and continuous Co film because the surface of the amorphous Ta is smooth. Meanwhile, the surface roughness of the oxidized 3 nm-thick Ta was measured to be almost the same as that of the SiO₂ substrate; a roughness of ca. 1 nm was observed for both samples by atomic force microscopy (AFM), whilst the grain sizes of these two samples slightly differed (see Figs. 3-5(b) and 3-5(c)). Here, to note is that surface of amorphous Ta cannot be measured by AFM without exposing the sample surface to air in our measuring setup. Hence, we deduce that the surface of the oxidized Ta loses sufficient smoothness, which can hinder the formation of a flat and continuous 1 nm-thick Co film. These results unequivocally rationalize that inserting a Ta buffer layer and maintaining its smooth surface play crucial roles in the growth of an ultrathin Co layer that can generate salient FMR.

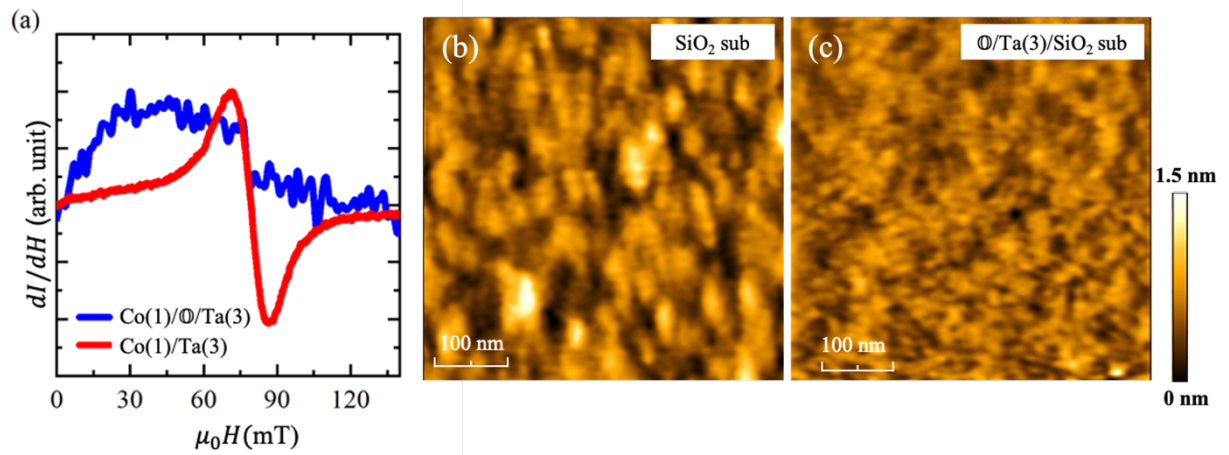


Figure 3-5. (a) Comparison of the FMR spectra of the Co (1 nm)/Ta (3 nm) and Co (1 nm)/ O / Ta (3 nm) samples. Atomic force microscopic views of the surface of (b) the SiO₂ substrates, and (c) the oxidized Ta.

3.4.3 Discussion

The shift in the resonance field as a function of the Co thickness in type-A samples (see Figs. 3-2 (a) and (c)) is attributed to a decrease in the total magnetization. Chiba and co-workers observed a significant reduction in magnetization in 1 nm-thick Co, resulting in a substantial decrease of the Curie temperature [76]. Therefore, the upshift of the resonance field can be attributed to the weaker magnetization of the ultrathin Co, which is consistent with the previous study [76]. Meanwhile, the absence of FMR spectra can be attributed to the roughness of the Co film. The SiO₂/Si substrates used in this study have a surface that is not sufficiently smooth. It has been previously demonstrated that the FMR spectra of Ni₈₀Fe₂₀ (Py) exhibit a pronounced dependence on the substrates. Specifically, the FMR linewidth of Py grown on a SiO₂ substrate has been observed to exceed the linewidths of Py grown on a SiO₂ substrate was greater than the linewidths of Py grown on yttrium-iron-garnet and non-doped diamond substrates [120]. The broader FMR spectrum of Py on a SiO₂ substrate is attributed to the substrate's roughness of an SiO₂ substrate, which impedes isotropic magnetization and uniform magnetization precession under FMR. The results obtained in the present study are consistent with those reported in the literature [120].

In chronicle of FMR studies of thin ferromagnetic films, a few studies were conducted approximately two decades ago to detect FMR spectra from ultrathin Co films [121,122]. The thickness of the Co films in the aforementioned studies is comparable to that of the films in the present study. In the meantime, the resonance field and the FMR linewidth in the aforementioned studies were approximately 500 mT and 60 mT (note that the linewidth of 60 mT is equivalent to the half-width at half-maximum of approximately 70 mT) [123], respectively. The similarly large resonance field was also reported in the literature [122], whereas the linewidth was not discussed in the study. The large resonance field and the

broad FMR linewidth are attributed to the relatively weak ferromagnetism and the lack of uniformity in the magnetization precession under FMR, which can be attributed to the inferior quality of the Co. Additionally, the FMR spectrum exhibited the single branch only when the thickness of the Co was greater than 2 nm in that study [123]. The resonance field and the half-width at half-maximum of the FMR spectra in our study are smaller and approximately 80 mT and 15 mT, respectively. Additionally, the single FMR spectrum can be observed in the 1 nm-thick Co. Consequently, the ultrathin Co utilized in our study simultaneously exhibits sufficiently strong ferromagnetism and uniform FMR, which differs from the findings of previous studies. e. Our experimental results demonstrate that the quality of the ultrathin Co with a Ta buffer layer is sufficiently high, and that the ultrathin Co is a promising material for future magnon spintronics and quantum hybrid systems.

3.5 Buffer layer dependence of magnon damping from interfacial effect

3.5.1 Broadband FMR measurement of Co thin films

Figure 3-6(b) illustrates the frequency dependence characteristics of the FMR signals from the 20-nm-thick Co sample with a Cu seeding layer. The frequency dependence of the half-width at half maximum (HWHM), designated $\mu_0\Delta H$, of the FMR spectra was estimated using the Lorentzian function [107,124] (see details in Appendix). The dependence of $\mu_0\Delta H$ is known to be expressed using the following equation [124]: $\mu_0\Delta H = 2\pi f\alpha/\gamma + \mu_0\Delta H_0$, where f and $\mu_0\Delta H_0$ represent the microwave frequency and the frequency-independent inhomogeneous linewidth, respectively. Here, the Gilbert damping constant can be extracted from the slope of the fitting function, $\mu_0\Delta H = 2\pi f\alpha/\gamma + \mu_0\Delta H_0$. The Gilbert damping constant α of this sample was estimated to be 0.026 ± 0.001 , which is comparable to that of bulk Co [125]. A similar measurement was conducted on the samples of Co (10 nm)/SiO₂, Al, Cu and Ti, and the resulting data are presented in Fig. 3-6 (c). This figure shows the microwave frequency dependence of the HWHM of these samples. The α of Co (10 nm)/SiO₂, Al, Cu and Ti were estimated to be 0.012 ± 0.001 , 0.025 ± 0.001 , 0.026 ± 0.002 and 0.018 ± 0.002 , respectively. In order to ascertain the influence of the nonmagnetic seeding layer species on the TMS coefficient, the Gilbert damping constants of prepared samples with Ti, Cu, and Al seeding layers were estimated. Figure 3-6(d) illustrates the $1/t_{\text{Co}}$ dependences of the Gilbert damping constants of the samples comprising the different NM seeding layers. For comparison, the corresponding dependence of a reference sample composed of a single ultrathin Co film deposited on a SiO₂/Si substrate is also shown. The TMS coefficients of the ultrathin Co films with the Ti, Cu, Al, and SiO₂ seeding layers were $0.01 \pm 0.07 \text{ nm}^2$, $0.35 \pm 0.08 \text{ nm}^2$, $0.42 \pm 0.05 \text{ nm}^2$, and $1.93 \pm 0.20 \text{ nm}^2$, respectively, in the fitting performed using Eq. (75). It can be seen that the ultrathin Co films with Cu, Al, and SiO₂ seeding layers demonstrate a significant contribution to the magnetization damping from TMS, whereas the corresponding films with Ta and Ti seeding layers exhibit a notable suppression of this TMS contribution. It is noteworthy that these findings align with the previously documented outcomes pertaining to the realization of minimal magnon damping

in nanometer-thick Co films through the incorporation of a Ta seeding layer [107,126]. Moreover, the potential for achieving low magnon damping in ultrathin Co films through the introduction of a Ti seeding layer is a significant observation. The findings suggest that the combination of FM and NM materials is a significant factor in the suppression of TMS. In the ultrathin region, the Gilbert damping constants of the 2-nm-thick Co films with a Ti seeding layer are 0.020, which is approximately one-fifth of those for the 2-nm-thick Co films without a Ti seeding layer. The Gilbert damping constants of the 0.111 and 0.090 nm thick Co films with Cu and Al seeding layers, respectively, are comparable to 0.018 of the Co (20 nm)/Ti sample. It can thus be concluded that the suppression of the TMS contribution by the insertion of appropriate NM seeding layers results in the occurrence of low magnon damping in nanometer-thick ferromagnetic films.

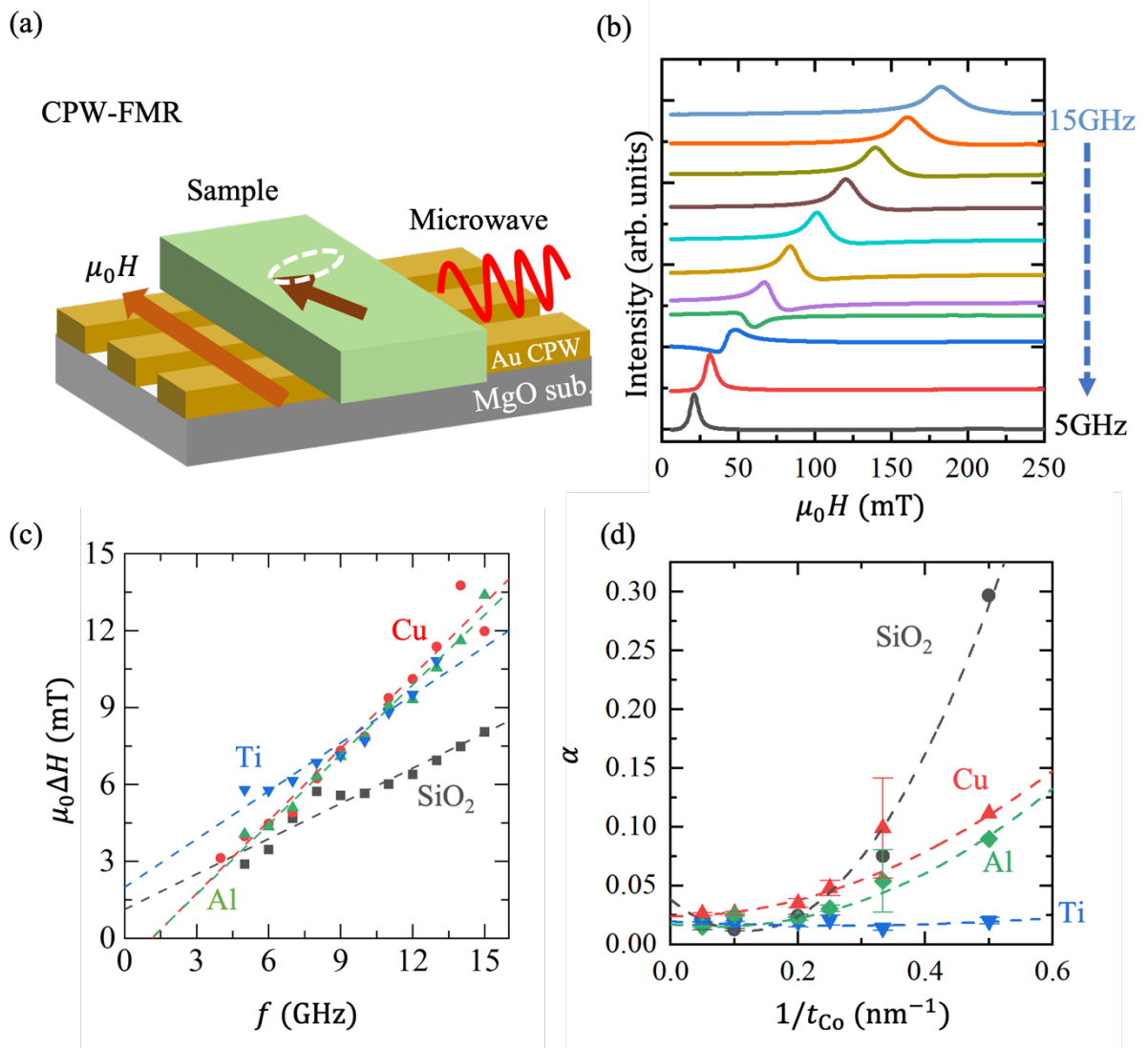


Figure 3-6. (a) Schematic of the experimental setting utilized in the CPW-FMR measurements. An external magnetic field was applied along the in-plane direction of the samples. (b) Frequency dependence of the FMR signals of Co (20 nm)/Cu, where the frequency was varied from 5 GHz to 15 GHz. (c) Frequency dependence of the HWHM in the Co (10 nm)/SiO₂, Al, Cu and Ti sample. (d) Co thickness dependence of the Gilbert

damping constant α for the samples with the various NM seeding layers and the SiO₂ underlayer (i.e., the reference sample).

3.5.2 Angular dependence of FMR measurement to estimate magnetic anisotropy

The β_{TMS} is theoretically known to be proportional to the square of the SMA field, $h_{\text{S}_u} = 2K_{\text{S}_u}/M_{\text{S}}$ [115,126], where K_{S_u} and M_{S} are defined as the uniaxial magnetic anisotropy energy and the saturation magnetization, respectively. To comprehend the NM dependence of β_{TMS} , h_{S_u} is ascertained for each ultrathin-Co/NM interface. This is achieved by estimating the t_{Co} dependence of the uniaxial magnetic anisotropy field h_u while postulating that the diamagnetization of the Co remains unaltered within the film thickness region of interest in this study. The h_u of an FM material is dependent on h_{S_u} via the thickness of the FM film and can be described as follows:

$$h_u = h_{\text{V}_u} + h_{\text{S}_u} t_{\text{Co}}^{-1}, \quad (78)$$

where h_{V_u} is the uniaxial magnetic anisotropy field of a bulk ferromagnet. A well-established method for estimating the h_{S_u} of an FM material entails the measurement of the angular dependence of the FMR spectra, which subsequently allows for determination of the effective uniaxial magnetic anisotropy field $h_{\text{eff}} (= h_u - 4\pi M_{\text{S}})$. Given that the in-plane magnetization is realized within the ultrathin Co film, the FMR frequency is expressed using the following equations, including h_{eff} [127]:

$$\left(\frac{\omega}{\gamma}\right)^2 = [\mu_0 H_{\text{res}} \cos(\theta_H - \theta) - h_{\text{eff}} \cos 2\theta][\mu_0 H_{\text{res}} \cos(\theta_H - \theta) + h_{\text{eff}} \sin^2 \theta], \quad (79)$$

$$\mu_0 H_{\text{res}} \sin(\theta_H - \theta) + \frac{h_{\text{eff}}}{2} \sin 2\theta = 0 \quad (80)$$

where $\omega = 2\pi f$ and $\mu_0 H_{\text{res}}$ are the microwave angular frequency and the resonance magnetic field of the FMR signal, respectively. Moreover, the angles θ_H and θ are the angles of the external magnetic field and the saturation magnetization with respect to the film plane, respectively, as illustrated in Fig. 3-7(a). θ_H was rotated from 0° to 180°. Figures 3-7(b) and 3-7(c) illustrate the θ_H -dependence of the FMR spectra for the sample with the Co (10 nm)/SiO₂ structure, as well as the resonance fields of the aforementioned structure. As a consequence of the in-plane magnetic anisotropy of the sample, the resonance field at $\theta_H = 0^\circ$ was observed to be the lowest. Furthermore, the upshift in the resonance field as a function of θ_H provided additional evidence in support of in-plane anisotropy of the ultrathin Co film. By solving Eqs. (79) and (80) self-consistently via substitution of the measured resonance fields (see Appendix D for details), the h_{eff} of Co (10, 5 and 3 nm)/SiO₂ samples were estimated to be -1.83×10^3 mT, -1.55×10^3 mT and -1.08×10^3 mT, respectively, which are comparable with that of the Co(100 nm)/SiO₂ sample [127]. It is important to note that our estimation method is a mathematical approach designed to identify the optimal parameters that satisfy Eqs. (79) and (80). The rationale behind this approach is that it is challenging to accurately determine the FMR field of the Co due to

the high saturation magnetization fields observed when the Co is subjected to a magnetic field perpendicular to its plane. This phenomenon hinders the numerical fitting process, as outlined Appendix D. Figure 3-7(d) illustrates the complete dataset for the $1/t_{\text{Co}}$ dependence of h_{eff} for the samples with the different NM seeding layers, and the magnitudes of h_{S_u} were determined from the slope of the linear fitting line to be 0.87 ± 0.23 Tnm (Ti), 1.3 ± 0.4 T · nm (Cu), 1.4 ± 0.1 T · nm (Al), and 3.23 ± 0.01 T · nm (SiO₂). As previously stated, β_{TMS} is proportional to the square of h_{S_u} at the Co/NM interface. Consequently, the TMS contribution should be significantly suppressed as the uniaxial SMA field of the ultrathin Co film is reduced by the insertion of the NM seeding layers. The following paragraph will demonstrate that the TMS can indeed be suppressed by controlling the SMA field.

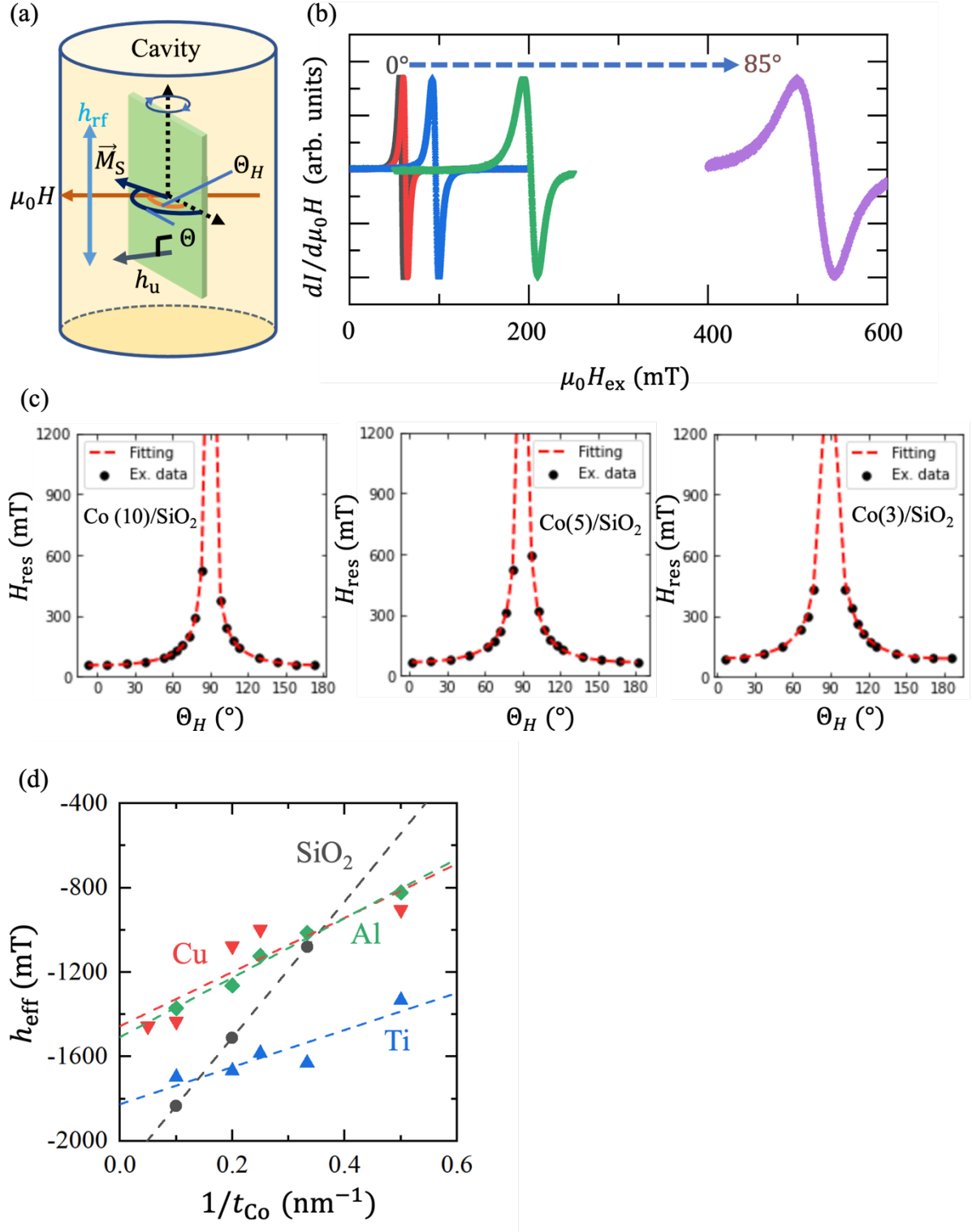


Figure 3-7. (a) Schematic of the measurement set-up used to determine the angular dependence of the FMR signals when using the ESR apparatus. (b) Angular dependence of the FMR signals of the Co (10 nm)/SiO₂ sample. (c) Angular dependence of the resonance fields of the Co (3, 5 and 10 nm)/SiO₂ samples. The broken red line represents the fitting line. (d) Co film thickness dependence of the uniaxial magnetic anisotropy field h_u , where closed lines represent the results obtained using the fitting function $h_u = h_{V_u} + h_{S_u} t_{FM}^{-1}$.

3.5.3 Correlation of two-magnon scattering contribution and surface magnetic anisotropy

As illustrated in Figs. 3-8(a) and 3-8(b), β_{TMS} is proportional to the square of h_{S_u} , i.e., $\beta_{\text{TMS}} = C_{\text{MD}}(h_{\text{S}_u})^2$, where C_{MD} is the quantity to be determined based on the roughness of the FM films (see Eq. (94) in Ref. [115]) [114,115]. C_{MD} was estimated to be $0.18 \pm 0.02 \text{ T}^{-2}$. The surface energy density dependences of selected NMs on the values of both β_{TMS} and h_{S_u} are shown in Fig. 3-8(c) and 3-8(d), respectively. In these figures, the magnitude of the NM surface energy density was cited from Ref. [128]. As illustrated in Figure 3-8(c), the h_{S_u} value at the ultrathin Co/NM interface exhibits a decline in conjunction with an augmentation in the surface energy density of the NM material. This outcome aligns with the findings presented in Ref. [128]. Figure 3-8(d) illustrates the correlation between β_{TMS} and the surface energy density of the NM materials, wherein β_{TMS} also exhibits a decline as the surface energy density of the NM materials increases. Therefore, we have successfully demonstrated that the TMS of an ultrathin FM material can be efficiently suppressed by tuning its SMA through the selection of appropriate NM seeding layers. This result indicates that the uniaxial SMA is dependent on the selection of the NM seeding layer located below the ultrathin Co film, which plays a significant role in determining the TMS contribution to magnon damping in ultrathin Co films. It is noteworthy that the β_{TMS} and h_{S_u} of the sample with $\text{SiO}_2/\text{Co}/\text{Ta}$ is also strongly suppressed, and in fact, the lowest among all of measured samples (see Appendix E). This is due to the fact that Ta has significantly high surface energy density, which allows for the growth of Co with good crystallography. This is consistent with the realization of low magnon damping in 1-nm Co film on Ta seeding layer as demonstrated in [107]. A direct comparison of the results on the $\text{SiO}_2/\text{Co}/\text{Ta}$ with those on the other samples fabricated by EB deposition was not conducted, as the $\text{SiO}_2/\text{Co}/\text{Ta}$ was fabricated by sputtering (see Table 3-1). However, the $\text{SiO}_2/\text{Co}/\text{Ta}$ sample is suitable for the following coupling measurements in section 3.7 because of its quite low β_{TMS} . Thus, the coupling measurements of the sample of SiO_2/Co (2 nm)/Ta using a rutile resonator was conducted, which is described in the section 3.7.

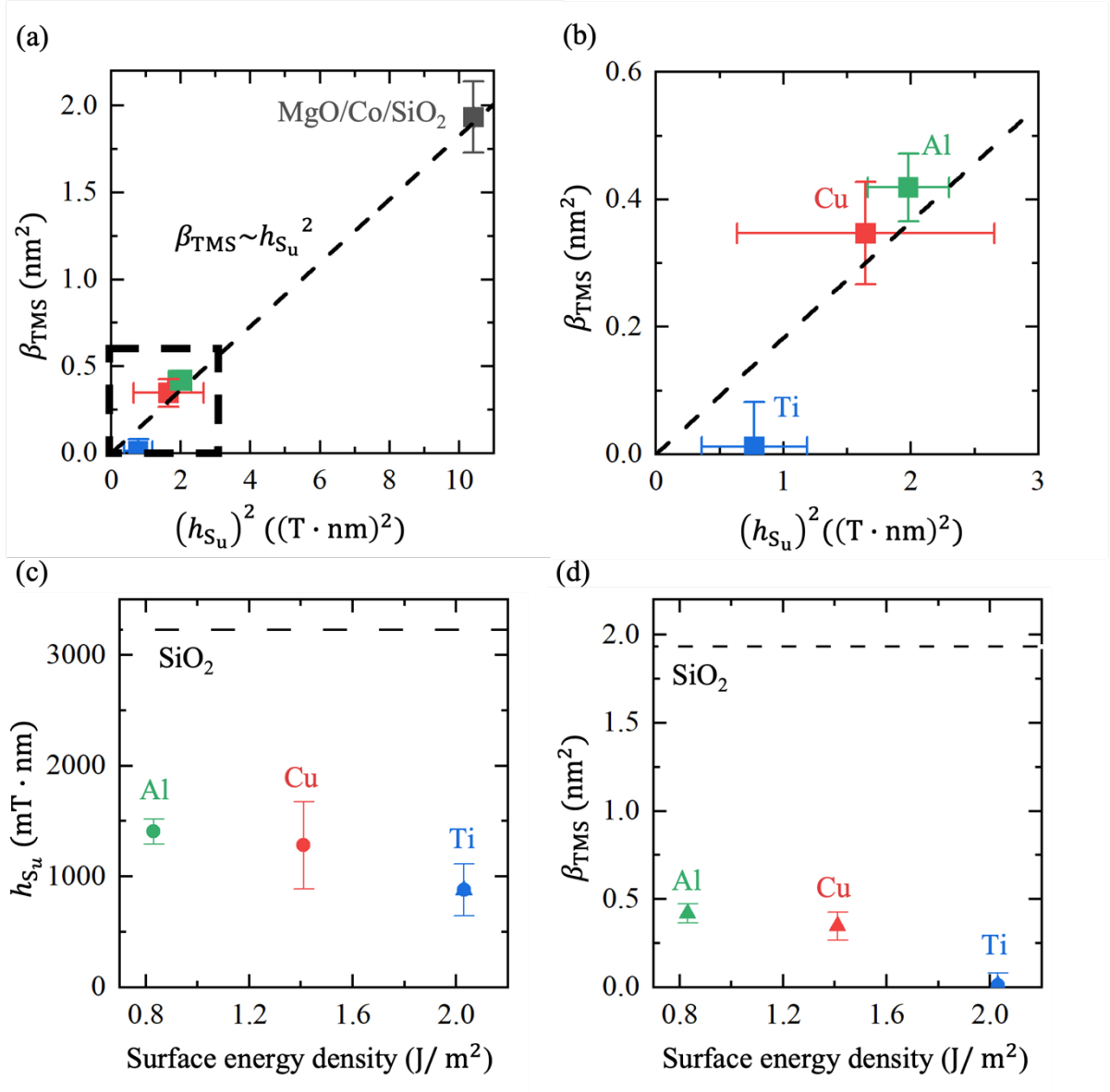


Figure 3-8. (a) Correlation diagram between the TMS coefficient β_{TMS} and the square of the SMA field $(h_{\text{Su}})^2$. The broken line represents the fitting result for the samples with the NM seeding layers. In the correlation between β_{TMS} and $(h_{\text{Su}})^2$, the data points for Co/Ti, Co/Cu, Co/Al, and Co/SiO₂ are represented by the blue, red, green, and black circles, respectively. (b) The enlarged figure within the block broken square. The surface energy density dependence of the NM materials selected as the seeding layers on (c) β_{TMS} and (d) h_{Su} , where the black broken lines in (c) and (d) show the values of the MgO/Co/SiO₂.

3.6 Manipulation of magnon damping in thin Co₂₅Fe₇₅ films by changing the thickness of MgO capping layer

3.6.1 Broadband FMR measurement of thin Co₂₅Fe₇₅ films

Figure 3-9(b) illustrates the real and imaginary components of the background-corrected transmission, designated as $\Delta S_{21} = S_{21} - S_{21}^0$, derived from the samples comprising 4 and 10 nm-thick Co₂₅Fe₇₅ layers and a 2 nm-thick MgO capping layer. In this

context, S_{21}^0 represents the background signal, which is represented by a linear function. In a thin FM film, the Gilbert damping constant and magnetic anisotropy are susceptible to alteration by changes in surface roughness and SMA, which in turn manifest as modifications to linewidth and resonance field in the bbFMR signal. Indeed, the signals of ΔS_{21} for the 4 nm-thick $\text{Co}_{25}\text{Fe}_{75}$ exhibit a larger linewidth and resonance field than those of the 10 nm-thick $\text{Co}_{25}\text{Fe}_{75}$. In order to investigate the phenomena of magnetization damping and magnetic anisotropy as observed in our field-dependent complex microwave transmission signals, we employ a fit based on the Polder susceptibility (for further details, please refer to Eq. (S13-15) in Appendix G [130,131]). This allows us to extract the resonance field, H_{res} , and the half-width at half-maximum magnetic field linewidth ΔH of the FM resonance for different fixed microwave frequencies. In accordance with the aforementioned procedure, the values of $\mu_0 H_{\text{res}}$ and $\mu_0 \Delta H$ for the 10 nm-thick (4 nm-thick) $\text{Co}_{25}\text{Fe}_{75}$ films were determined to be 56.3 mT (75.1 mT) and 3.2 mT (12.6 mT), respectively. This observation can be rationalized by considering the enhancement of the magnetization damping and effective magnetic anisotropy with the decreasing thickness of the $\text{Co}_{25}\text{Fe}_{75}$ layer, as will be discussed in the following section.

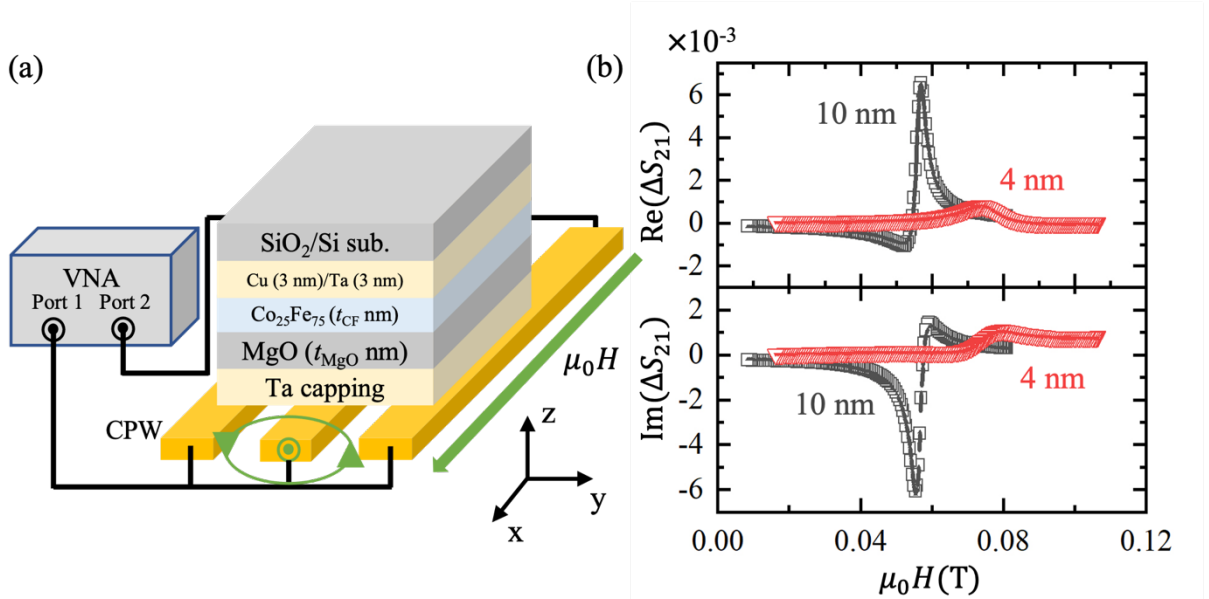


Figure 3-9. (a) Schematic image of measurement setup for bbFMR. The S_{21} of the CPW on which the sample is attached was measured by VNA. (b) Real and imaginary parts of ΔS_{21} for the samples with 10 nm (black) and 4 nm (red) thick $\text{Co}_{25}\text{Fe}_{75}$ and a 2 nm-thick MgO capping layer, using a microwave frequency of 10 GHz.

3.6.2 MgO thickness dependence of magnon damping

The Gilbert damping constant can be determined from the frequency dependence of ΔH using the following equation [119]: $\mu_0 \Delta H = \mu_0 \Delta H_0 + \frac{\alpha}{\gamma} \omega$, where $\mu_0 \Delta H_0$ and γ are the inhomogeneous linewidth and gyromagnetic ratio, respectively and $\omega = 2\pi f$. Figure 3-

10(a) illustrates the microwave frequency dependence of the extracted ΔH obtained from the bbFMR measurement for our samples with varying $\text{Co}_{25}\text{Fe}_{75}$ and MgO capping layer thicknesses. It is evident that the FMR linewidth increases with a reduction in the thickness of the $\text{Co}_{25}\text{Fe}_{75}$ layer, irrespective of the MgO layer thickness. Furthermore, it can be observed that α also exhibits an increase with the thickness of the MgO capping layer.

This enhancement is achieved by reducing the thickness of the FM layer, t_{FM} . The observed decrease in FM thickness can be attributed to two primary factors: surface magnetization damping originating from spin pumping from the FM into the adjacent nonmagnetic layer [109] and/or TMS [115]. These effects are captured by the following equation: In this case, the magnetic proximity effect (MPE) term on Gilbert damping from Ta buffer layers, as discussed in Ref. [129], can be considered negligible due to the presence of a 3 nm-thick Cu interlayer, which effectively eliminates MPE between the Ta layer and CoFe alloys [130]. To distinguish between the two effects (α_{SP} and β_{TMS}) and assess the dependence of the MgO capping layer thickness, we plot the Gilbert damping constant as function of the $\text{Co}_{25}\text{Fe}_{75}$ thickness t_{CF} (see Fig. 2(b)) and fit this data using Eq. (75) to determine α_{int} , α_{SP} and β_{TMS} (see Fig. 3-10 (c)-(e)). As illustrated in Fig. 3-10 (c), the α_{int} of the $\text{Co}_{25}\text{Fe}_{75}$ is approximately 4.0×10^{-3} for varying MgO capping layer thicknesses. This value aligns with those reported in prior studies [67,69,131], reinforcing the consistency of our findings. The α_{SP} of the 2 nm-thick $\text{Co}_{25}\text{Fe}_{75}$ is inconsequential in comparison to the other damping contributions when the MgO thickness exceeding 1 nm in thickness is zero within the margin of error (see Fig. 3-10 (d)). Meanwhile, the α_{SP} is considerable when the MgO layer is 0.4 and 0.8 nm in thickness, which may be attributed to a finite spin pumping from the $\text{Co}_{25}\text{Fe}_{75}$ into the Ta capping layer, as documented in Ref. [132]. One possible interpretation is that the observed MgO thicknesses still include pinholes that permits spin pumping into the Ta capping layer [109]. The evolution of β_{TMS} with MgO thickness is illustrated in Fig. 3-10 (e). The TMS coefficient β_{TMS} demonstrates a monotonic increase with the thickness of the MgO capping layer up to 2 nm (see Fig. 3-9(e)), which can be attributed to the modulation of TMS by alterations in USMA as previously reported in a related study [133]. It is noteworthy that the TMS contribution can be tuned by modifying the thickness of the MgO capping layer and the modulation ratio of β_{TMS} , which is approximately 300%. Additionally, the Gilbert damping α derived from TMS was modified from 0.0065 to 0.0265. Furthermore, the modulation range ($\Delta\alpha$) is analogous to that observed in a recent study utilizing 20 nm-thick FM films [134].

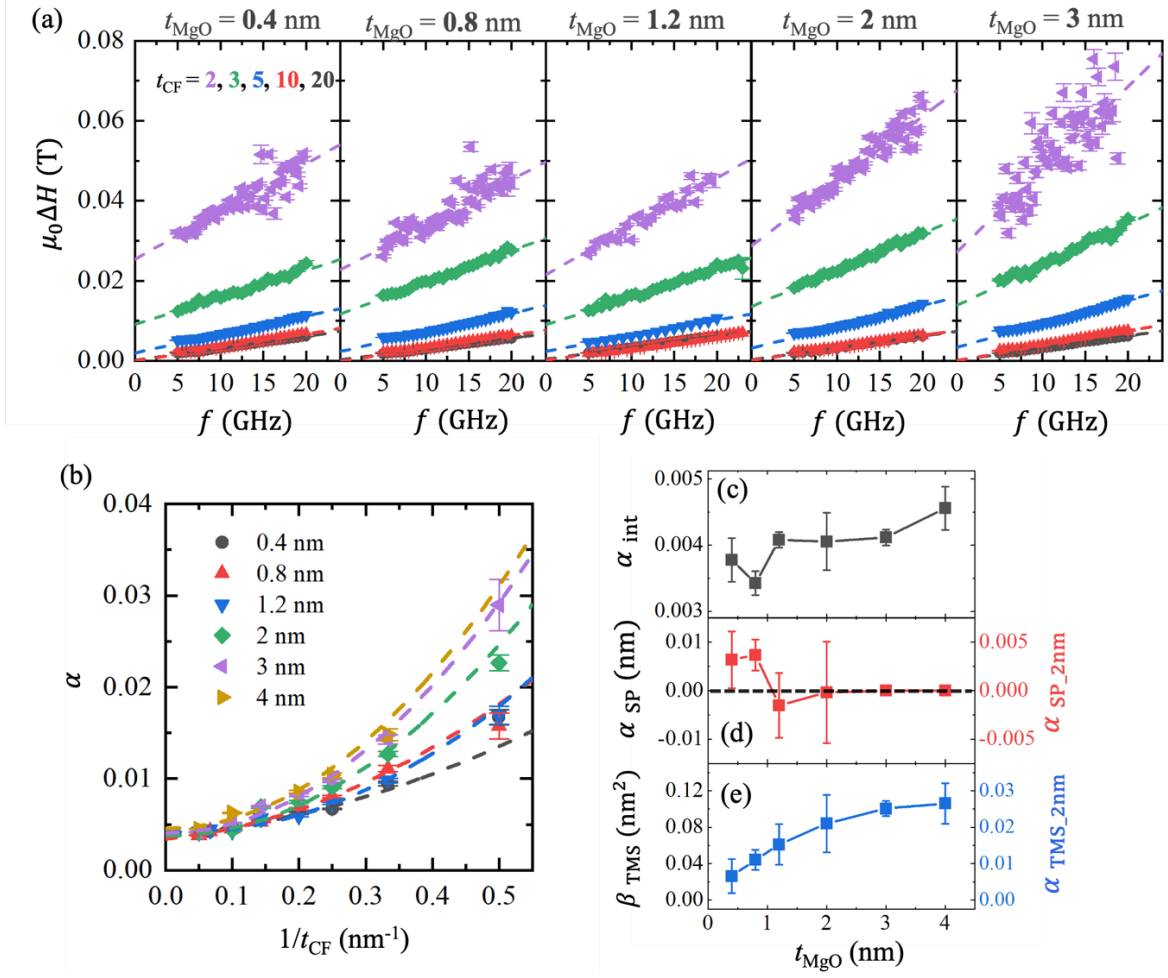


Figure 3-10. (a) Microwave frequency dependences of the HWHM obtained by fitting the FMR signals of the samples with different MgO and Co₂₅Fe₇₅ thicknesses. The broken line shows the fitting obtained using the Polder susceptibility (see details in Appendix G). (b) Co₂₅Fe₇₅ thickness dependence of the estimated Gilbert damping constant. The broken line shows the fitting obtained using Eq. (75). MgO thickness dependences of (c) α_{int} , (d) α_{SP} , and (e) β_{TMS} . The right axis of Figs. 3-9(d) and (e) show the Gilbert damping constants corresponding to that of the 2 nm-thick Co₂₅Fe₇₅ film.

Two-magnon scattering is contingent upon USMA and surface roughness [115]. Given that the magnitude of β_{TMS} is proportional to the square of the USMA field [60], the giant modulation of β_{TMS} can be construed as an augmentation of the USMA field at the MgO/Co₂₅Fe₇₅ interface in accordance with the MgO capping layer thickness. An independent quantification of the magnetic anisotropies in thin FM films can be obtained from the dispersion of the FMR which are shown in Fig. 3-11 (a). Our findings indicate that the thinner Co₂₅Fe₇₅ films exhibit a deviation from the behavior observed in thicker film ($t_{\text{CF}} = 20$ nm), suggesting the presence of an interface contribution to the total magnetic anisotropy. A comparison of MgO layer thicknesses reveals that this interface-like contribution increases (see Fig. 3-11 (a), subpanels for $t_{\text{MgO}} = 3$ nm). Therefore, the MgO

layer thickness appears to influence the USMA field, exhibiting an increasing trend with increasing t_{MgO} .

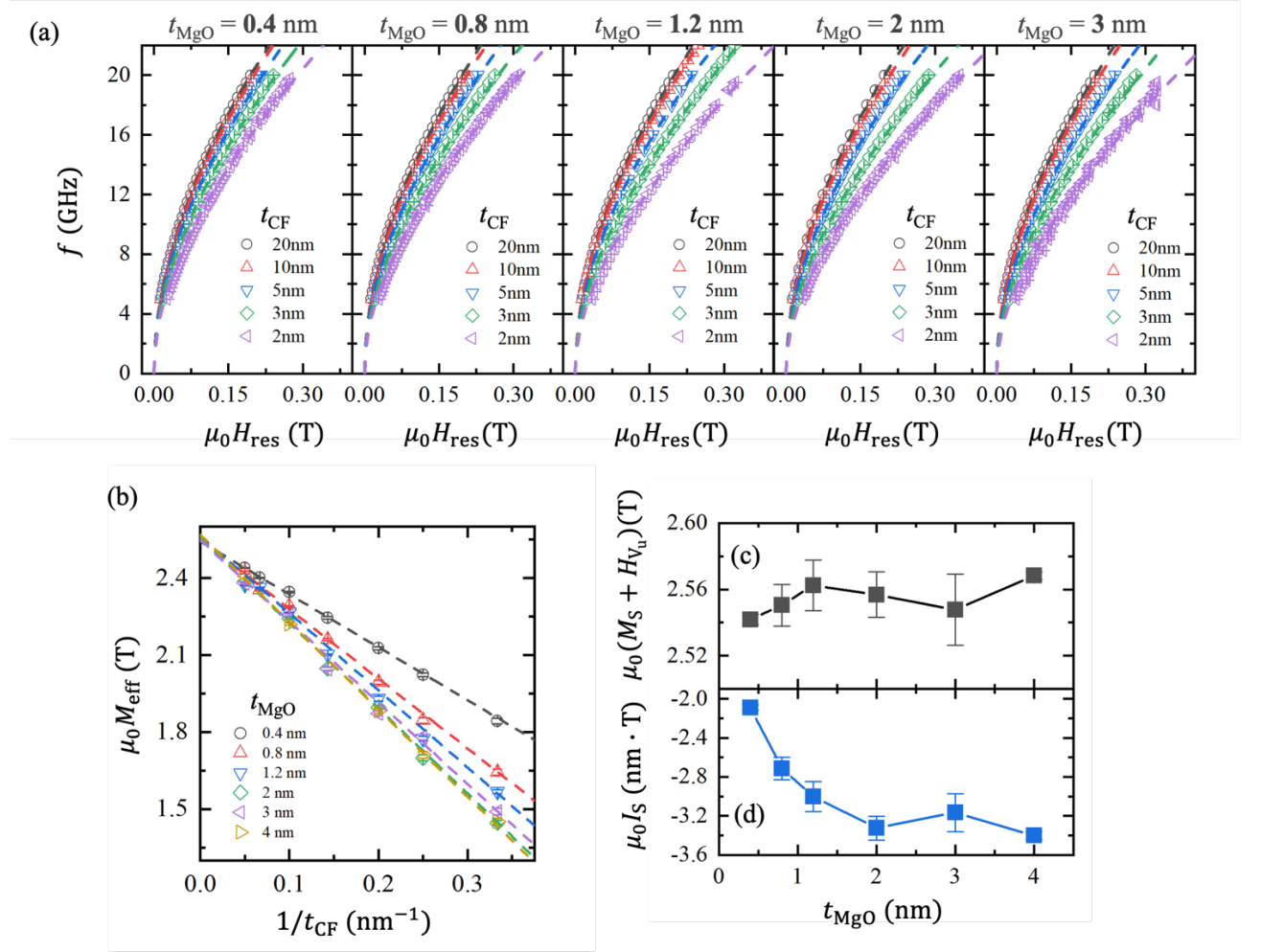


Figure 3-11. (a) Microwave frequency dependences of the resonance field obtained by fitting the FMR signals of the samples with different MgO and Co₂₅Fe₇₅ thicknesses. The broken lines depict fitting obtained using Eq. (82). (b) Co₂₅Fe₇₅ thickness dependence of the effective magnetization. MgO thickness dependences of (c) the effective magnetic anisotropy field $\mu_0 H_{\text{eff}}$ and (d) the SMA field $\mu_0 H_s$ where the black dashed line shows zero value.

3.6.3 Control of magnon damping by altering MgO capping layer thickness

An effective thickness-dependent anisotropy parameter is quantified by fitting the Kittel mode for the in-plane configuration to the data presented in Fig. 3-11 (a). In detail, we use [58]

$$f = \frac{\gamma}{2\pi} \sqrt{\mu_0 H (\mu_0 H + \mu_0 M_{\text{eff}})}, \quad (81)$$

where f and M_{eff} are the microwave frequency and effective magnetization, respectively. Figure 3-11 (b) illustrates M_{eff} as function of the inverse Co₂₅Fe₇₅ layer thickness. It should be noted that Eq. (81), already incorporates the thin film limit with the static magnetic field

applied in plane. The effective magnetization, M_{eff} , encompasses contributions such as the saturation magnetization, M_S , bulk (H_{V_u}) and interface (I_S) related anisotropies. This can be parametrized as follows [60]:

$$M_{\text{eff}} = (M_S + H_{V_u}) + I_S t_{\text{CF}}^{-1}. \quad (82)$$

Figure 3-11 (c) shows the MgO capping layer thickness dependence for $\mu_0(M_S + H_{V_u})$, which is determined from the value of M_{eff} at $1/t_{\text{CF}} = 0$, which we obtain by fitting Eq. (82) to the data. As illustrated in Figure 3-11 (c), the MgO thickness has a relatively minor impact on $M_S + H_{V_u}$. It can be inferred that the introduction of the MgO capping layer does not affect the intrinsic magnetic properties ($M_S + H_{V_u}, \alpha_{\text{int}}$) of $\text{Co}_{25}\text{Fe}_{75}$. The absolute value of the estimated I_S , which contains the USMA, increases with increasing MgO thickness and reaches a saturation point when the thickness exceeds 2 nm (see Fig. 3-11(d)), indicating that the modulation of the USMA field at the MgO/ $\text{Co}_{25}\text{Fe}_{75}$ interface is governed by the thickness of the MgO capping layer. Given that β_{TMS} displays a comparable dependency with respect to MgO thickness, it can be postulated that the control of USMA facilitates the tunability of α in the ultrathin $\text{Co}_{25}\text{Fe}_{75}$ film. To substantiate this assertion, the correlation between β_{TMS} and the square of the USMA field, represented via I_S^2 , is illustrated in Fig. 3-12, thereby providing empirical evidence to support the proposed connection between β_{TMS} and the USMA field. By subtracting α_{SP} from α , the modulation of α is estimated to range from $\Delta\alpha = 0.0105 - 0.0305$, which is in close proximity to that observed in previous work utilizing 20 nm-thick FM film, with a range of $\Delta\alpha = 0.0054 - 0.0240$ [17]. However, in this study, we studied an ultrathin FM film. Our findings indicate that the results obtained for a 20 nm-thick FM film can be extended to the regime of ultra-thin magnetic layers. This thickness regime is particularly suited for gate-tunable Gilbert damping, which is an advantageous property.

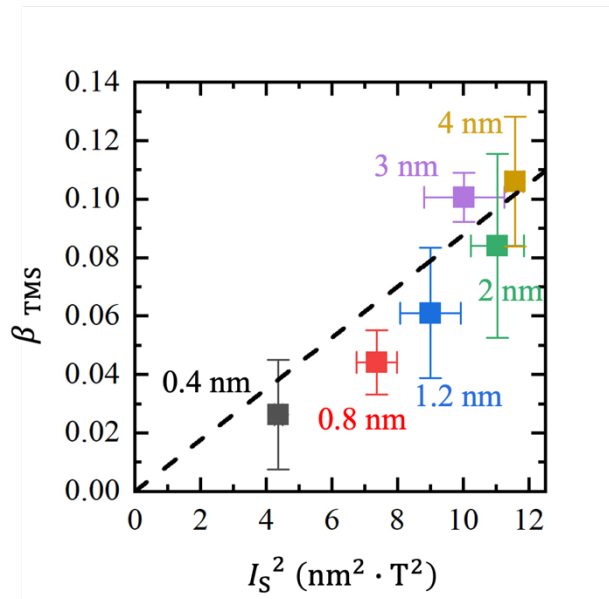


Figure 3-12. Correlation between the TMS coefficient and the square of the USMA field represented by I_S^2 .

In conclusion, our findings demonstrate a notable modulation of the Gilbert damping constant in ultrathin $\text{Co}_{25}\text{Fe}_{75}$ films through alternations the thickness of the MgO layer. Furthermore, our findings suggest a correlation between the observed increase in Gilbert damping and the alternation in the USMA field at the $\text{Co}_{25}\text{Fe}_{75}/\text{MgO}$ interface, which both depend on the MgO thickness. The modulation amplitude of the Gilbert damping constant ($\Delta\alpha$) in the 2 nm-thick $\text{Co}_{25}\text{Fe}_{75}$ film was approximately 0.02, without altering the bulk magnetic properties. The modulation principle of the Gilbert damping constant identified in this study is versatile and applicable to gate-tunable magnon systems comprising ultrathin conducting FMs, offering high tunability.

3.7 Estimation of coupling strength and damping rate in thin ferromagnetic films

As a preliminary step towards realization of magnon-photon strong coupling using ultrathin FM films, measurements were conducted to estimate the coupling strengths of magnons and photons, in addition to their dampings, in an ultrathin Co film on a Ta seeding layer using a rutile resonator with a high-quality factor. The full width at half-maximum, κ_i , represents the microwave photon relaxation rate in the rutile resonator. This value was extracted by fitting the microwave frequency dependence of the input reflection coefficient of the scattering parameter S_{11} at each external magnetic field as illustrated in Fig. 3-13 (b). The data shown in this figure was obtained by subtracting linear and weak background signals from the raw data. The fitting function is expressed as following equation:

$$S_{11} = \frac{i(\omega - \omega_c) + \frac{\kappa_e - \kappa_i}{2}}{i(\omega - \omega_c) - \frac{\kappa_e + \kappa_i}{2}}, \quad (83)$$

where ω_c and κ_e are the resonance angular frequency of the rutile resonator and the relaxation rate of microwave photons passing through a coaxial cable connected to the coil, respectively. Figure 3-13 (c) depicts κ_i as a function of the external magnetic field, exhibiting and a clear resonance peak was observed successfully, even from the 2-nm-thick Co film. In input-output theory [135,136], κ_i is expressed using the following equation (see also the Appendix for a more detailed description):

$$\kappa_i = \kappa_{i0} + \gamma_m \frac{g_{\text{eff}}^2}{(\omega - \omega_b)^2 + \left(\frac{\gamma_m}{2}\right)^2}, \quad (84)$$

$$\omega_b = \gamma \sqrt{\mu_0 H (\mu_0 H + M_{\text{eff}})}, \quad (85)$$

where κ_{i0} , γ_m , and ω_b are the linewidth without FMR excitation, the relaxation rate of an excited magnon, and the FMR angular frequency with in-plane magnetization, respectively. The relaxation rate corresponds to the linewidth of the magnetostatic mode. The red broken line shown in Fig. 3-13 (c) depicts the fitting result obtained using Eq. (84), where $\kappa_{i0} = 2\pi \times 22.7$ MHz, $g_{\text{eff}} = 2\pi \times 3.88$ MHz, and $\gamma_m = 2\pi \times 745$ MHz. Furthermore, the effective magnetization, M_{eff} , was estimated to be 0.792 T, which is comparable to the magnitude of the effective magnetization in an ultrathin Co film [106]. The photon decay rate in the rutile resonator with the sample was enhanced by a factor of 20 compared with the rate without the sample ($2\pi \times 1$ MHz), which is most likely attributed to microwave absorption occurring in the metallic sample (see Appendix C).

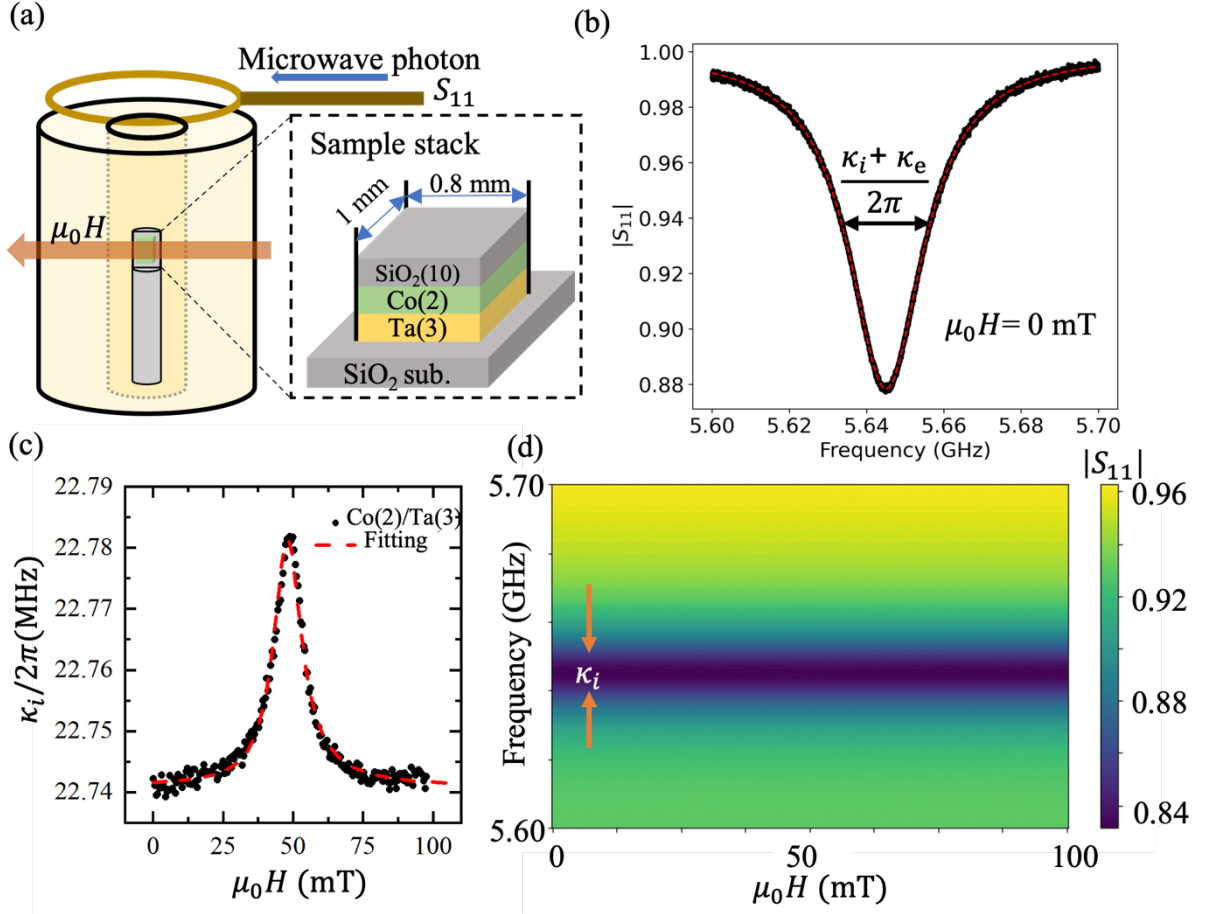


Figure 3-13. (a) Schematic of the measurement set-up used to perform the FMR measurements using the rutile cavity. The external magnetic field was applied in-plane along the sample and the microwave signal was applied via the coil. (b) Frequency dependence of $|S_{11}|$ at a zero external magnetic field, where the red line shows the fitting result obtained using Eq. (5). (c) External magnetic field dependence of κ_i , where the red broken line represents the fitting result obtained using Eq. (6). (d) Color map of $|S_{11}|$ for the external magnetic fields and microwave frequencies.

The coupling strength between the single spin and the photon mode can be expressed as $g_{0,\text{Co}} = g_{\text{Co}}^* \mu_B B_{\text{zpf}} / \sqrt{2} \hbar$ (see Appendix B for the derivation), where g_{Co}^* and B_{zpf} are

the g-factor of Co and the vacuum fluctuation of the microwave magnetic field in the rutile resonator when applied to the single spin in the Co atom. The estimated value for B_{zpf} was 5.4 pT, and the calculated value for $g_{0,\text{Co}}$ was $2\pi \times 133$ mHz in a COMSOL simulation. Given that the effective coupling strength is given by $g_{\text{eff}} = g_{0,\text{Co}}\sqrt{N}$ [24] and the number of spins in a 2-nm-thick Co film is calculated to be 6.192×10^{14} , the estimated effective coupling strength of the 2-nm-thick Co film in the rutile resonator is thus $2\pi \times 3.31$ MHz. The magnon-photon coupling state of the rutile resonator and the Co (2 nm)/Ta system is weak, as evidenced by the estimated parameters g_{eff} , γ_{m} and κ_{i0} , which satisfy the following relation: $g_{\text{eff}} < \gamma_{\text{m}}, \kappa_{\text{i0}}$. Figure 3-13 (d) depicts a color map of the $|S_{11}|$ parameter, which clearly illustrates that the magnon-photon hybrid system is in a weak coupling regime. It is noteworthy that the photon mode linewidth exhibited a slight increase at around the external magnetic field, where a magnon was excited (see Fig. 3-13(c)). It can be demonstrated that microwave photon and the excited magnon are capable of interacting with each other, thereby creating an additional decay path for the photon mode. Although estimation of coupling parameters in a weakly coupled regime in ultrathin metallic films has not been straightforward thus far, our approach allows for the quantitative estimation of such ultrathin films by measuring the additional decay rate in the 3D resonator with high-quality factor, which is significance of this study.

Although the tunability of the strong magnon-photon coupling system based on $\text{Y}_3\text{Fe}_5\text{O}_{12}$ (YIG) has been demonstrate by controlling microwave power [100,101], the control of the magnon-photon coupling state via electric modulation of magnon remains a significant challenge. The realization of this control expands the potential applications of quantum coupling systems by means of conventional electronic approaches, such as those used in field-effect transistors. It can be reasonably deduced that magnon-photon coupling between magnons excited in ultrathin FM films and microwave photons may facilitate the development of a gate-tunable magnon-photon coupling system. To realize a gate-tunable strong coupling regime using ultrathin FMs, it is necessary to implement a mechanism for weaker and tunable magnon damping, which will enable the coupling strength to be adjusted. The TMS contribution in such an ultrathin FM film can be excluded successfully by inserting Ta or Ti layers with small SMA fields. In contrast, the intrinsic contribution α_{int} of Co is still insufficient ($\sim 2.0 \times 10^{-2}$) to achieve strong magnon-photon coupling due to the electron-magnon damping in the Co. Accordingly, a potential solution to circumvent this issue may be the utilization of a ferromagnetic metal that mitigates the electron-magnon damping, e.g., $\text{Co}_{25}\text{Fe}_{75}$ [67], given that the α of this metal is 2.1×10^{-3} . Indeed, despite the α -value of $\text{Ni}_{81}\text{Fe}_{19}$ (Py) being 2.2×10^{-2} , which is analogous to the damping constant of the ultrathin-Co/Ti sample, the realization of strong magnon-photon coupling was achieved through the use of a superconducting LC circuit [28,29]. The combination of the aforementioned approaches allows for the realization of strong magnon-photon coupling that is gate-tunable.

3.8 Summary

In summary, the author studied on the effect of surface and interfacial state of ferromagnetic thin film on its magnon damping. To date, spin pumping damping, magnetization damping induced by generating the spin current to adjacent layer, has been enthusiastically studied. However, this section demonstrated it is also crucial to design the surface state such as roughness and surface magnetic anisotropy to control magnon damping in thin ferromagnetic films. The guiding principle suggested in this chapter to decrease magnon damping in thin ferromagnetic films is applicable not only to magnon polaritons in thin ferromagnetic films studied in the chapter 5, but also to general magnon study in thin ferromagnetic films. In addition, the author demonstrated the measurement method to estimate coupling strength, magnon damping and photon damping in weak coupling of magnon and photon, which would be applied to the fundamental estimation method to improve each coupling parameter.

Chapter 4. Superconducting resonator made of high- T_C YBCO

4.1 Abstract

In this chapter, the author reports the establishment of a superconducting resonator made of high- T_c superconductor, $\text{YBa}_2\text{Cu}_3\text{O}_7$ (YBCO). Superconducting resonator fabricated on chip is promising media for photon mode coupling with magnons in thin ferromagnetic films since its strong confinement of photons on the surface of resonators, which allows for the high AC magnetic field of photons coupling with magnons in the mounted ferromagnetic thin films. The excited photons are characterized by the design of LC resonator circuit, significantly modifying the coupling strength with magnons. Therefore, the optimized design of superconducting resonators is essential for the achievement of magnon polaritons in thin ferromagnetic films. The author demonstrated the establishment of YBCO LC resonator, showing high quality factor around 4000. In addition, the robustness of YBCO resonator to magnetic field and environment temperature, which may promise significant advantages to measurement of magnon polaritons.

Results presented in this chapter will be published in the paper:

S. Yoshii, M. Müller, R. Ohshima, M. Althammer, Y. Ando, H. Huebl, and M. Shiraishi, in preparation.

4.2 Introduction and research purpose

4.2.1 Superconducting resonator

Superconducting resonators have emerged as a cornerstone technology in quantum information processing, providing a pathway for building coherent, low-loss quantum circuits [137,138]. Unlike conventional resonators, superconducting resonators utilize materials that exhibit zero electrical resistance below a critical temperature, effectively eliminating Joule heating and enabling the preservation of quantum coherence over extended periods. This property is invaluable for applications in quantum computing and quantum communication, where decoherence and energy loss are significant challenges. By minimizing energy dissipation, superconducting resonators allow for the storage and manipulation of quantum information with high fidelity, making them indispensable in the field of quantum technology [139,140].

In a superconducting resonator, electromagnetic fields are confined within a narrow, high-quality resonant cavity, where photons can be excited at specific microwave frequencies. The design of these resonators enables the creation of high-Q (quality factor) circuits, where the electromagnetic energy can oscillate between the electric and magnetic fields multiple times before dissipating. This high-Q characteristic is essential for quantum applications, as it permits the storage of microwave photons with minimal energy loss, thereby preserving the integrity of quantum states. The ability to sustain coherent photon excitations over long timescales also facilitates strong coupling with other quantum systems, such as qubits [141–145] or spins [146–149], allowing for efficient quantum state transfer. Furthermore, the multiple quantum coupling of numbers of quanta such as superconducting qubit, photons and spin ensembles [150] as shown in Fig. 4-1(a) makes the coupling system with various quantum systems, called as “quantum hybrid system”, expanding the functionalities of superconducting resonator via photon coupling. Recently, quantum excitation of spin waves, magnons, achieved strong coupling with photons excited in a superconducting resonator [24,40,151], where magnons have jumped into the candidates composing of quantum hybrid systems [26,98,99] as shown in Fig. 4-1(b).

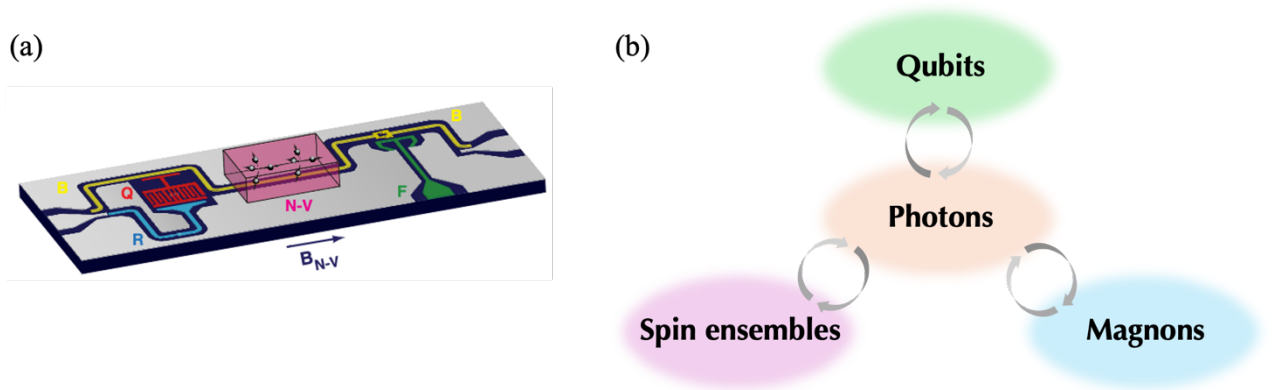


Figure 4-1. (a) An example of a quantum hybrid system that integrates qubits, photons, and spin ensembles within NV centers in diamond. This is the first demonstration of quantum hybrid systems utilizing the superconducting resonator. The figure is quoted

by [150]. (b) Schematic of quantum hybrid systems, where spins-photons-qubits coupling and magnons-photons-qubits are demonstrated by ref. [150] and refs. [26,98,99], respectively.

One of the greatest advantages of superconducting resonators is that highly confined photon mode can be excited on the surface of superconducting resonators. The ability to confine and control photons within a small spatial region in superconducting resonators provides a critical advantage for coupling with other quantum excitations excited in thin films with a thickness of around a few tens of nanometers. In particular, thin-film ferromagnetic materials benefit from superconducting resonators for exploring magnon-photon interactions, as they can govern photons strongly coupling with magnons within nanometer dimensions. By placing a thin-film ferromagnetic material into the photon modes triggered on the surface of a superconducting resonator allows for strong coupling between the resonator's confined photons and the magnons in the ferromagnetic films [39,40,151], forming a hybrid quantum state known as a magnon polariton. This magnon-photon coupling holds significant potential for advancing magnonics as a part of quantum hybrid systems. Traditional approaches to magnon polaritons in thin ferromagnetic films face challenges related to energy dissipation and coherence and exclude the selection of ferromagnetic materials except for YIG. Researchers can now observe magnon polaritons in thin ferromagnetic metallic films by utilizing superconducting resonators. This breakthrough removes barriers to leveraging the rich physics of magnonics in hybrid quantum systems. This hybrid approach combines the advantages of both photons and magnons, enabling the coherent transfer of information between electromagnetic and spin systems, which is crucial for developing highly efficient magnonic devices.

Furthermore, the narrow confinement of photons within a superconducting resonator enhances the magnon-photon coupling strength, a parameter essential for observing coherent magnon polariton behavior and extremely strong coupling regimes such as ultrastrong and deep strong coupling. Ultrastrong coupling regimes, where the ratio of the interaction energy over resonance energy is more than 0.1, are critical for creating robust magnon polaritons that can be utilized in quantum technologies. This regime allows for coherent quantum entanglement between magnons and photons, opening up possibilities for developing quantum memory, non-reciprocal devices, and even quantum transducers that bridge microwave and optical frequencies

4.2.2 YBCO as a material for superconducting resonator

The Nb or Al, superconducting materials, with a critical temperature of approximately 10 K and 4 K, respectively, have been used as the main materials for superconducting resonators because of the ease of deposition and patterning [152]. In addition, there are a lot of established studies for Al to design Josephson Junction [153], which has been a fundamental technology for recent quantum information technologies. However, regarding the interaction between superconducting resonators and ferromagnetic

materials, the ferromagnetic materials can generate stray magnetic fields that disrupt the superconducting state and the quality factors of resonators, in addition to the external magnetic field necessary to excite magnons. Thus, identifying a strong superconducting material in a magnetic field is crucial for creating the optimal conditions to investigate magnon polaritons in thin ferromagnetic films. Manuel et al. established a superconducting resonator using NbTiN with a higher critical temperature (T_c) of up to 17 K to develop an optimized photon system coupling with ferromagnets, exhibiting greater resilience to magnetic fields (approximately 200 mT) [154].

The author predicted that $\text{YBa}_2\text{Cu}_3\text{O}_7$ (YBCO), one of the type-II superconductor, holds great potential as a material for superconducting resonators. YBCO is a widely studied high-temperature superconductor with the perovskite structure (see Fig. 4-2) from when it was reported in 1987 [155], because of the higher T_c around 90 K. Since its discovery as one of the first materials to exhibit superconductivity above the boiling point of liquid nitrogen, YBCO has opened new avenues in superconducting technology by allowing operations at temperatures far above those required by traditional superconductors. This higher critical temperature (T_c), around 90 K, enables the use of more practical cooling systems, making YBCO-based circuits more accessible and manageable in experimental setups.

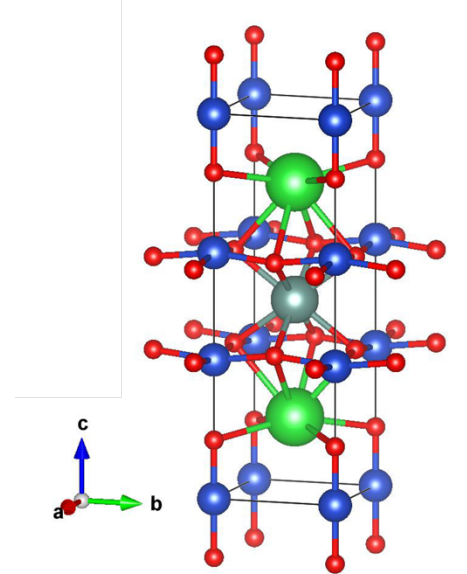


Figure 4-2. Crystal structure of YBCO.

A key advantage of YBCO in resonator applications is its resilience to environmental variations, especially temperature and magnetic field fluctuations. The essential features for superconducting resonators such as resonance frequency f_r and quality factor of resonator itself Q_r are characterized as follows [156,157]:

$$\frac{1}{Q_r} + 2i \frac{\delta f}{f_r} = \frac{\alpha}{\mu_0 \omega_r \lambda_L(0)} (R_s(T) + i\delta X_s(T)), \quad (86)$$

where μ_0 , α , and $\lambda_L(T)$ are vacuum permeability, the kinetic inductance fraction to the total inductance and London penetration depth, referring to how far a magnetic field penetrates into a superconductor, respectively. Here, $R_s(T)$ and $X_s(T)$ are the surface resistance and surface reactance with temperature dependence expressed as follows [154]:

$$R_s(T) = \frac{1}{2} \mu_0^2 \lambda_L^3(T) \sigma_1(\omega, T), \quad (87)$$

$$\delta X_s(T) = \mu_0 \omega_r [\lambda_L(T) - \lambda_L(0)]. \quad (88)$$

Here, the $\lambda_L(T)$ and $\sigma_1(\omega, T)$ are the London's penetration depth and surface conductivity with the temperature and the resonance frequency dependence, respectively. Since $R_s(T)$ and $X_s(T)$ are directly related to the Q_r and f_r , respectively,

Superconductors that exhibit high critical temperatures and low penetration depths λ_L are promising candidates for superconducting resonators to observe magnon polaritons. Unlike conventional superconductors that require ultra-low temperatures, YBCO maintains its superconducting properties across a broader range of temperatures below its T_c . This thermal stability offers a practical benefit for quantum systems where minor temperature shifts are inevitable. In experiments involving magnon-photon coupling, such as magnon polariton formation in ferromagnetic materials, this robustness ensures that the resonator can operate consistently without significant changes in performance due to temperature drift. Such stability is essential for keeping a high-quality factor in the resonator, which directly impacts the coherence and strength of the magnon-photon interaction. Additionally, YBCO exhibits an impressive tolerance to magnetic fields, a property that enhances its suitability for integration with magnetic materials. This robustness allows YBCO-based resonators to maintain their superconducting state even when exposed to stray magnetic fields generated by nearby ferromagnetic elements. This magnetic field tolerance is particularly beneficial for research into magnon polaritons, where the resonator must be positioned close to a ferromagnetic thin film to facilitate interaction with magnons. YBCO's high critical magnetic field threshold enables the resonator to function reliably without degradation, supporting strong and coherent magnon-photon coupling required for observing hybrid quantum phenomena.

The exceptional properties of YBCO are not limited to its high transition temperature and field tolerance. Advances in thin-film deposition and microfabrication techniques have facilitated the precise patterning of YBCO films, thereby enabling the design of superconducting resonators with highly controlled geometries and performance characteristics. This control enables researchers to optimize the resonator's photon confinement and coupling conditions, thereby maximizing the interaction with magnons in thin ferromagnetic films. The creation of tailored resonators provides the optimal conditions for the study of magnon polaritons, wherein the attainment of strong coupling regimes hinges on the resonator's capacity to confine photons in a stable, low-loss environment.

4.2.3 Purpose of this chapter

The objective of this chapter is to present the establishment of a YBCO-based superconducting resonator. The author examines the loss mechanism in YBCO superconducting resonators, in addition to the morphological characteristics, particularly surface roughness, influenced by the fabrication processes. The objective is to enhance the magnon properties in thin ferromagnetic films. Moreover, the author presents a discussion of the dependence of resonance peaks on temperature and magnetic field, as well as an analysis of the properties of superconducting resonators.

4.3 Experimental preparation

This section presents the fabrication method for a YBCO superconducting resonator used in this dissertation.

4.3.1 Sample fabrication

- **YBCO films**

The YBCO films are deposited onto a 6 mm $\times$ 10 mm sapphire substrate using a sputtering system that incorporates flowing O₂ gas, sourced from Ceraco ceramic coating GmbH. The composition of YBCO can be adjusted with the applications, where there are two types of YBCO with rich Cu and Y, called S-type and M-type, respectively. The difference the author is concerned with is the surface flatness since the surface state largely could influence the confinement of photon and magnon damping in thin ferromagnetic films mounted on the YBCO resonator. Figure 4-3 shows the AFM images of S-type and M-type YBCO films, where the thicknesses of YBCO are 150 nm-meter. While M-type YBCO films have the advantages of higher critical current density and low surface resistance ($< 300 \mu\Omega$), where the surface of YBCO appears porous, possibly violating the magnetization dynamics, the flat surface of S-type YBCO helps the smooth growth of thin ferromagnetic films on YBCO resonators. In Chapter 5, the author mainly used S-type YBCO for the measurement of magnon polaritons. All samples have Au layers on the YBCO layer to prevent oxygen from escaping and achieve good electrical contact.

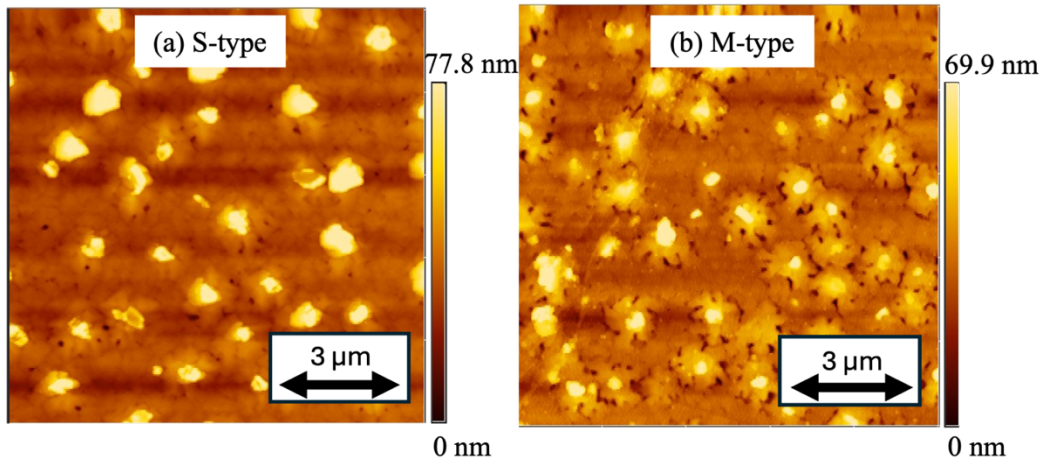


Figure 4-3. AFM images of the surface on (a) S-type and (b) M-type YBCO.

- **Superconducting resonator structure**

Defining the resonator structure on demand is crucial as a first step for establishing a superconducting resonator. The common resonator structures on a chip, the coplanar waveguide resonator, and the lumped element resonator, both capacitively coupled with a feedline, are shown in Figs. 4-4 (a) and (b). They serve as essential resonators for building today's quantum information devices, utilized in various ways to enhance specific features [158]. In the realm of quantum coupling involving superconducting resonators and magnons, it is crucial to enhance the coupling strength within the system following Eq. (55),

providing a clear direction to compress the photon mode as much as possible for stronger coupling between magnons and photons.

Before setting up the structure, we will see the fundamental physics of superconducting resonators. Figure 4-4. (c) depicts the schematic image of the LC(R) circuit, where Q_{int} is the internal quality factor characterizing the coherence time. When LC resonance is excited by applying A.C. current from a feedline, the uniform photon modes with the same Hamiltonian as Eq. (47) are excited in the circuit resonator [159]. Superconducting resonators allow for the photon mode's long coherence time since the LCR circuit's resistance is almost zero under the superconducting state, which strongly suppresses the energy damping in the LC circuit. Considering the energy conservation in the LC circuit, the L part generates an A.C. magnetic field, enabling dipole coupling with magnons. In the system of magnon-photon coupling governed by superconducting resonator, the effective coupling strength is expressed as follows [40,60,151]:

$$g_{\text{eff}} = g_s \sqrt{N} = g \mu_B B_{\text{rms}} \omega_r \sqrt{\frac{N}{8\hbar Z_r}}, \quad (89)$$

where ω_r , Z_r and g_s are the resonance frequency of LC circuit, $1/\sqrt{LC}$, the characteristic impedance, $\sqrt{L/C}$, the coupling strength between photons and individual spins in ferromagnetic materials, $g_s = g \mu_B B_{\text{rms}} \omega_r / \sqrt{8\hbar Z_r}$, respectively. From the viewpoint of designing superconducting resonator, the lumped element style with the freedom of manipulating L and C produces the room for the improvement of g_s . Expecting the coupling between homogenous photons and large amount of thin ferromagnetic films discussed in Chapter 5, the author adopted the lumped element structure with meandered capacitance and inductor to excite highly confined photon mode on the surface of the inductive part with a large area of around $400 \mu\text{m} \times 1 \text{mm}$ (see Fig. 4-5). This resonator structure increases the homogeneity of the microwave magnetic field on the superconducting resonator with a low relaxation rate, allowing for uniform coupling with magnons in the mounted thin ferromagnetic films [160,161]. The uniform feature of photon mode confined in the small volume is essential to enhance the numbers of spins coupled with photon mode with maintaining high g_s .

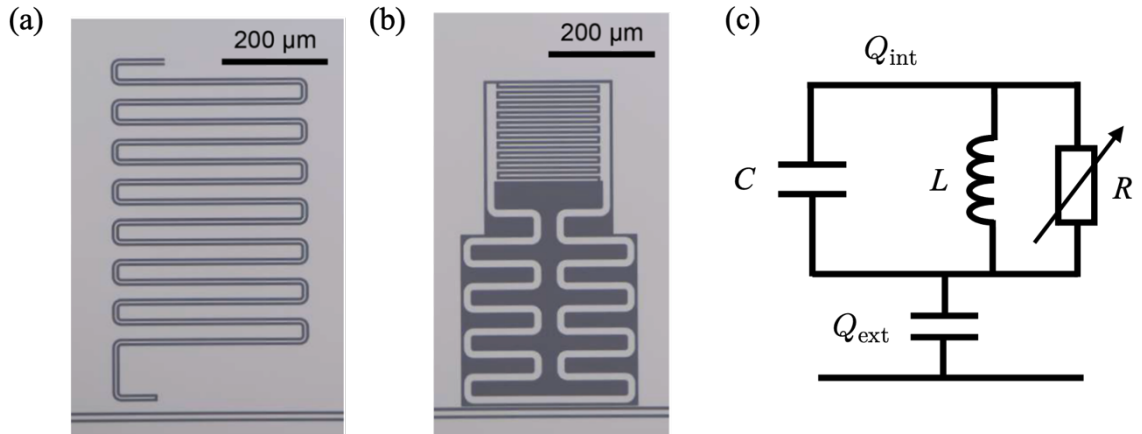


Figure 4-4. Typical superconducting resonators. (a) CPW resonator is capacitively coupled to a feedline and (b) lumped element resonator with a meandered capacitive and inductive element, which are connected and compose an LC resonator circuit. Figures (a) and (b) are quoted by [157]. (c) The schematic circuit of an LCR resonator capacitively coupled with a feedline. Here, Q_{int} and Q_{ext} are the quality factors of the LC resonator and capacitive coupling between a feedline and resonator.

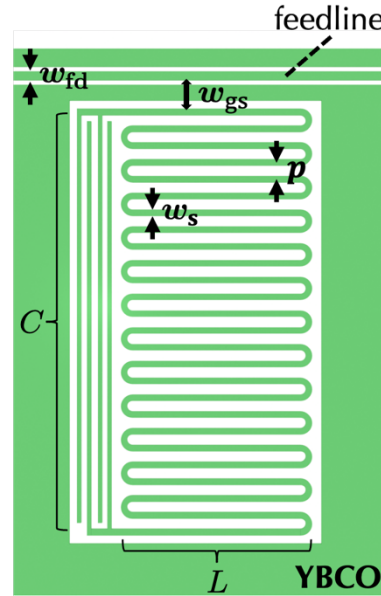


Figure 4-5. Schematic of lumped element structure with the meandered capacitance and inductance capacitively coupled to a feedline. Here, w_{fd} , w_s , w_{gs} , and p are the linewidth of a feedline, a linewidth of inductive part, a distance between feedline and resonator, and the pitch of inductive lines, respectively.

- **Fabrication process of YBCO superconducting resonator**

The prepared YBCO films are etched to pattern superconducting restores by using an Ar milling system and electron beam lithography. The author screened PMMA double layers to protect the YBCO layer, which is not supposed to be etched. After electron beam lithography of resonator structure, the PMMA resist is developed by dipping the sample into the compound liquid of MIBK and IPA with the portion of 3:1, respectively. Since etching 40 nm-thick Au + 150 nm-thick YBCO layers takes a pretty long time, the reduction of oxygen in YBCO can take place at the surface of YBCO due the o annealing effect during Ar milling, leading to the degradation of YBCO's LC resonator. Thus, the milling process takes an interval cooling time of about 5 min with 5 min milling, i.e., intermittent etching, to prevent oxygen escaping [162,163]. The total etching time without the intermittent etching is 15 min with the acceleration voltage (V_{ac}) of 200 V for 40 nm-thick Au layer and 90 min with the V_{ac} of 300 V 150 nm-thick YBCO layer. After etching, the resist of PMMA was removed by dipping the sample into the acetone and the O₂ ashing in the reactive ion

etching system. The sample is cooled down from the mounting stage, where water flows backside. In this chapter, the author utilized 3 samples, M-type#1, S-type#1 and S-type#2, which are used for the following sections. Table 4-1 illustrates the correlation between the sample and the section in which it was utilized.

Sample	Section
YBCO-M#1	4.4.1, 4.4.2, 4.4.3
YBCO-S#1	4.4.3, 4.5.1
YBCO-S#2	4.5.2

Table 4-1. The summary of the correlation between the sample and the section in which it was utilized.

Resonator parameter	
w_{fd}	20 μm
w_s	15 μm
w_{gs}	72 μm
p	25 μm

Table 4-2. The summary of the structure parameters of the prepared resonators.

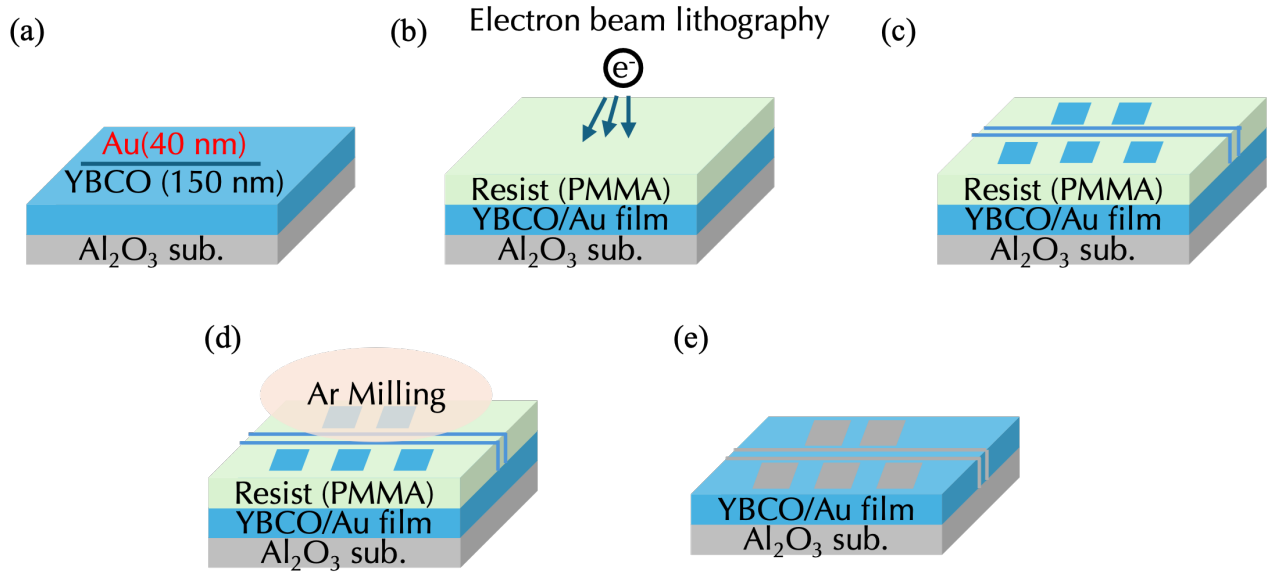


Figure 4-6. The fabrication process of the YBCO LC resonator. (a) The 150 nm-thick YBCO is sputtered on a 6 mm $\times$ 10 mm sapphire substrate. (b) Writing by electron beam lithography. (c) Development of positive resist of PMMA. (d) Ar milling to etch YBCO film. (e) Removal of positive resist. Here, the line in the middle of the sample and the square box depicts the feedline and lumped element resonators, respectively.

In this dissertation, the author adapted the same structure shown in Figs. 4-7. Figure 4-7 (a) shows the optical image for one example of the prepared YBCO chips with lumped

element resonators with different resonance frequencies characterized by $1/\sqrt{LC}$. In this structure, the capacitive component in the resonators is differently designed to control the length of the comb in LC resonators, which enables the measurement of the transmission of all the resonators just by changing the frequency of AC current flowing in a feedline.

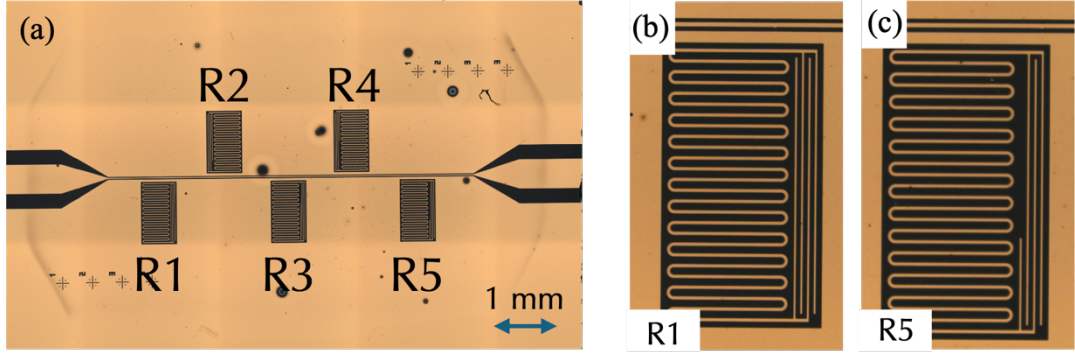


Figure 4-7. (a) The optical image for one example of the prepared YBCO chips with lumped element resonators. Five resonators with different resonance frequencies, named R1-5, are coupled with a feedline in the middle of the sample. The enlarged images of (a) R1 and (b) R5. R1 has a higher capacitive component than R5, leading to a lower resonance frequency for R1 than R5. R2-R4 has the middle values between R1 and R5, and the order of resonance frequency follows the same order as the index.

4.3.2 Measurement

- **Two-terminal measurement to evaluate T_C of the prepared chip**

The two-terminal measurement was conducted in physical properties measurement system (PPMS), where the temperature goes down to 4K. The author measured the temperature dependence of two-terminal resistance R_{2T} , where the dc voltage was applied from -5mV to +5mV and the temperature was swept from 150 K to 30 K.

- **Cu box attaching YBCO resonator**

The author prepared an SMA end launch connector and Cu box, attaching the prepared YBCO chip to install the sample into the cryostat measurement system, as shown in Fig. 4-8. The resonator chip is fixed with conductive silver glue around the edges, forming the ground level of the LC resonator. SMA end launch connectors are inserted from the hole of the Cu sample box to connect the center pin of the SMA connector to the feedline of the YBCO chip using silver glue. The Cu cap is attached to the Cu box to restrict the photon mode in the free space of the Cu box, preventing the excited photon from relaxing into the free space [164].

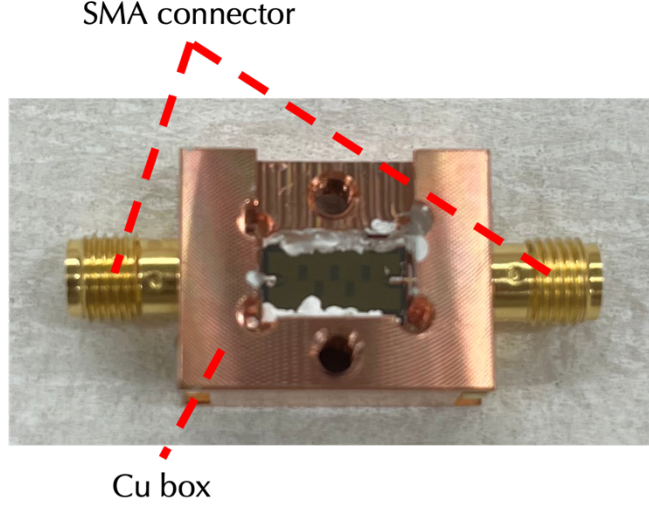


Figure 4-8. Image of the sample box containing the fabricated YBCO chip equipped with an SMA connector.

- **Transmission measurement of superconducting resonator**

The sample box containing the YBCO chip is placed inside the cryostat chamber (purchased from Thermal Block Inc.), and the SMA end connectors link to the RF cable that connects to the vector network analyzer (VNA). The schematic circuit of the measurement system is shown in Fig. 4-9. First, the cryostat chamber is vacuumed at around 5×10^{-3} Pa. The shield and cooling stage are cooled by the He compressor. For the transmission measurement with applying an external in-plane magnetic field $\mu_0 H$ along the feedline of the chip.

The microwave is generated by the VNA, with a power level set at 0 dBm and a frequency range spanning from 300 kHz to 20 GHz, where the author just swept between 4 GHz and 6 GHz since the resonance frequency is designed to be around 5 GHz. The resonance peaks are observed by measuring the transmission spectra S_{21} , a part of the S-parameter (scattering parameter) matrix, where S_{21} is defined as the transmission coefficient from port 1 to port 2 of two-port VNA. The S_{21} in lumped element superconducting resonator is quantified as follows [165]:

$$S_{21}(f) = \frac{\tilde{V}_2}{\tilde{V}_1} = ae^{i\beta}e^{-2\pi if\tau} \left[1 - \frac{(Q/|Q_{\text{ext}}|)e^{i\phi}}{1 + 2iQ(\frac{f}{f_r} - 1)} \right], \quad (90)$$

$$Q = \left(\frac{1}{Q_{\text{ext}}} + \frac{1}{Q_{\text{int}}} \right)^{-1}, \quad (91)$$

where the factor of $ae^{i\beta}e^{-2\pi if\tau}$ is the environment coefficient originated from the measurement system excluding the resonators, the inside component of the square brackets expresses the component of ideal resonators including an additional correction $e^{i\phi}$ and f_r is the resonance frequency formulated by $1/\sqrt{LC}$. As defined in Fig. 4-4 (c), Q_{int} and

Q_{ext} are the quality factors of resonator itself and capacitive coupling between a resonator and a feedline, respectively. Since this dissertation focuses on the features of superconducting resonators, the author neglects the environment coefficient. In addition, f denotes the microwave frequency derived by VNA.

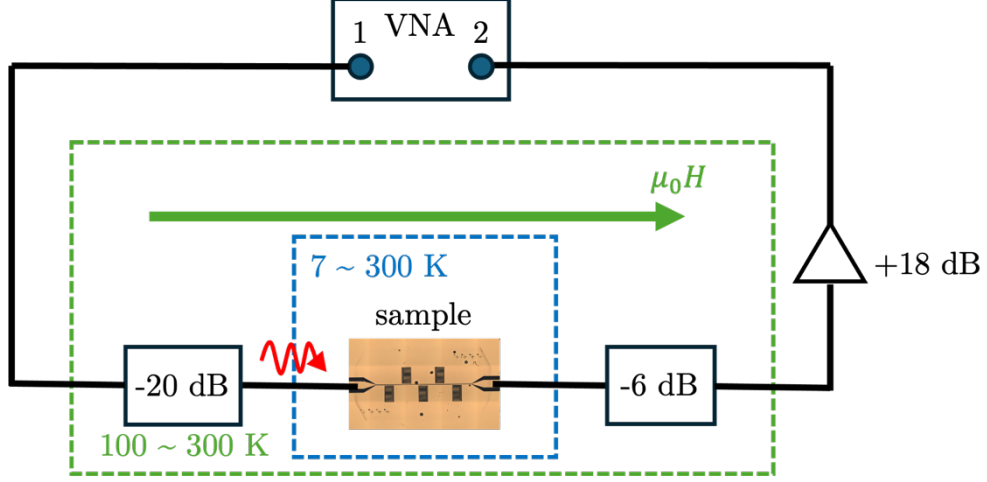


Figure 4-9. Schematic of the microwave circuit of the transmission measurement setup. The broken green square represents the cooling shield within the cryostat chamber, capable of reaching temperatures as low as 100 K. Meanwhile, the broken blue square denotes the sample box connected to the cooling stage, which can be cooled to approximately 7 K. The external magnetic field $\mu_0 H$ is applied along the feedline of the chip.

- **Temperature and magnetic field dependence of f_r and Q_{int}**

For the temperature and magnetic field dependence, the temperature was swept from 10 K to 75 K and the external magnetic field was swept between ± 145 mT. The input power was set to be 0 dBm and the frequency range was from 4 GHz to 6 GHz.

4.4 Fundamental features of YBCO-LC resonator

4.4.1 Evaluation of the superconducting resonator

The milling of patterns in YBCO resonators requires a considerable investment of time, which could potentially compromise the superconductivity of the patterned YBCO with narrow linewidth [162,163]. It is of paramount importance to exercise caution and consideration when it comes to the potential damage that could be caused to the YBCO resonator during the fabrication process. A two-terminal measurement was therefore conducted with the objective of evaluating the critical temperature, T_C , in a feedline of the prepared YBCO chip. Figure 4-10(a) illustrates the two-terminal measurement circuit for investigating the temperature dependence of two-terminal resistance (R_{2T}), which demonstrates the superconducting transition at 87 K. This outcome substantiates the quality of the superconducting state of the fabricated YBCO chip.

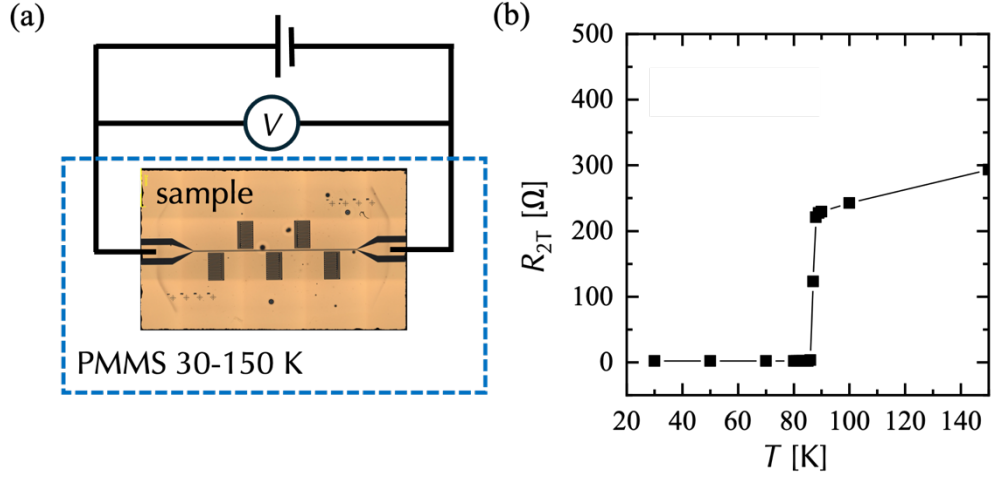


Figure 4-10. (a) Schematic circuit for the two-terminal measurement of a feedline of YBCO chip at low temperature in PPMS. The sample is situated within the PPMS system, which is represented by the blue broken square. (b) The temperature dependence of the two-terminal resistance of a feedline. A pronounced shift is observed at approximately 90 K, indicative of the critical temperature for superconductivity.

Then, the transmission measurement of the prepared M-type YBCO resonator was carried out to examine the properties such as the resonance frequency f_r , the internal quality factor Q_{int} and the external quality factor Q_{ext} . Figure 4-10 (a) shows the magnitude of transmission spectra, S_{21} , where there are the five peaks at different resonance frequencies. The measurement temperature was 10 K and the microwave power was 0 dBm. Fitting each peak spectra with Eq. (89) and (90) provides the estimated f_r , Q_{int} and Q_{ext} , where Fig. 4-10 (b) shows the enlarged spectra around the peak corresponding with R1 resonator with the fitting curve. The fitting function of Eq. (89) nicely reproduces the result of the resonance peak for R1. Table. 4-3 summarizes the fitting results for all the resonators of R1-5. In the condition of $Q_{\text{int}} \ll Q_{\text{ext}}$, the coupling between a resonator and a feedline is called undercoupling, where the small number of photons can be excited from a feedline and it shows the relatively small peaks against the previous works [154,161]. The resonance frequencies are varied following $1/\sqrt{LC}$, being consistent with the magnitude order of the length capacitance component of resonators. Note that the internal quality factor, Q_{int} , significant to consider coupling systems with magnon mode coupled with AC magnetic field excited in inductive component, is around one thousand, and the resonance peak's height is about 0.10 dB. These properties are not sufficient compared with the previous research of magnon polaritons in superconducting resonator.

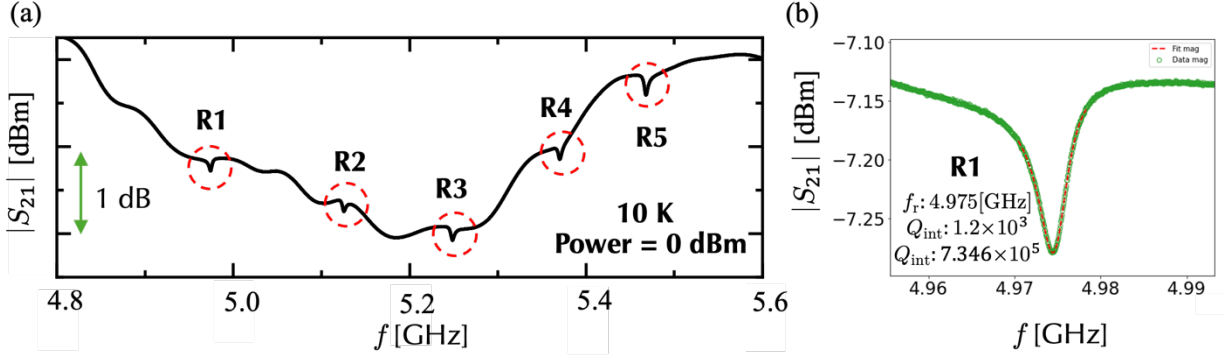


Figure 4-11. (a) The magnitude of transmission spectra S_{21} at 10 K with the microwave power of 0 dBm. (b) The enlarged spectra of the peak corresponding with R1 resonator with the fitting function of Eq. (90).

	f_r [GHz]	$Q_{int} (\times 10^3)$	$Q_{ext} (\times 10^4)$
R1	4.975	1.200 ± 0.003	7.346 ± 0.028
R2	5.125	1.153 ± 0.005	7.437 ± 0.051
R3	5.248	1.105 ± 0.003	6.021 ± 0.024
R4	5.371	1.067 ± 0.018	5.167 ± 0.015
R5	5.468	1.092 ± 0.003	3.899 ± 0.015

Table 4-3. Summary of the fitting results for YBCO resonators of R1-5. f_r , Q_{int} and Q_{ext} are the resonance frequency of photon, the internal quality of factor for the resonator itself, and the external quality of factor for the capacitive coupling between a feedline and a resonator.

4.4.2 Loss mechanism in the superconducting resonator

A comprehensive understanding of the photon loss mechanism in superconducting resonators is essential for optimizing the functionality of photon resonators for coupling with magnons in thin ferromagnetic films [164,166,157]. The investigation of the various loss mechanisms has constituted a significant area of research, with the objective of improving the superconducting resonator in quantum information processing. This section provides a concise overview of the material losses of photons in superconducting resonators, with the aim of offering a useful reference for optimizing YBCO resonators. The primary loss channels for photons excited in superconducting resonators are as follows:

- (a) Two-level systems (TLS) losses induced by absorbing a photons into TLS and generating a phonon into the bath [166,167].
- (b) Thermal quasiparticle loss that is characterized by the surface quality of the material defined by the surface reactance X_s and the surface resistance R_s , leading to the resonator losses formulated by Eq. (86) [168].
- (c) Eddy current loss excited in adjacent metallic layer, where AC current is induced,

leading to the loss channel [164].

- (d) External losses such as radiative loss [169] and damping into parasitic modes of the used sample boxes [170,171].
- (e) Vortex losses induced by trapped magnetic flux [172].

The primary objective of this section is to suppress (c) eddy current losses. This can be achieved by optimizing the fabrication processes and the structure of the samples. In our samples, a 40-nm-thick gold layer is deposited in situ to prevent the escape of oxidation and to facilitate a good electronic contact with a 150-nm-thick YBCO layer. However, the incorporation of an Au layer within the resonator structure gives rise to eddy current loss. This phenomenon can be attributed to the generation of eddy currents in the Au layer by the excited AC magnetic and electronic fields, which subsequently result in photon loss (see Fig. 4-12). It is thus anticipated that the etching of the Au layer on the resonator structure will result in an enhancement of the internal quality factor. This objective was accomplished by the author through the use of Ar ion milling, which was employed to remove the Au layer. It is important to note that the annealing effect during etching can induce the diffusion of Au particles into YBCO layers, which can ultimately lead to the destruction of the superconducting state [173]. Therefore, it is essential to utilize an intermittent etching method to prevent this phenomenon.

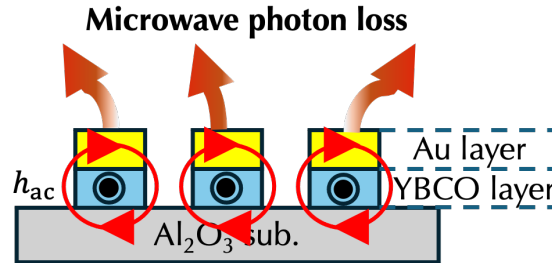


Figure 4-12. Schematic of microwave photon mitigation due to the Eddy current loss.

Figures 4-13 (a) and (b) show the comparison of the transmission spectra S_{21} for the YBCO resonators with and without 40 nm-thick Au layer on the resonator structures. Despite the elevated temperature measurement for the Au-layer-containing sample, the removal of the Au layer resulted in the observation of more pronounced resonance peaks. This phenomenon can be attributed to the suppression of eddy current loss of photons in the Au layer. The fitting results for the YBCO resonator without Au layer is shown in Table 4-4. The estimated resonance frequencies are not so different after the removal of Au layer. It is noteworthy that the internal quality factor, Q_{int} , after the removal of the Au layer reaches 4000, which is approximately four times larger than the value observed prior to removal. This observation indicates that the 40 nm-thick Au layer was a significant contributor to the overall loss in this system. The quality factor of 4000 is comparable to that reported in another study on strong magnon-photon coupling in superconducting

resonators. [40,151]. In this dissertation, the Au layer on the YBCO resonators is basically removed without noting that.

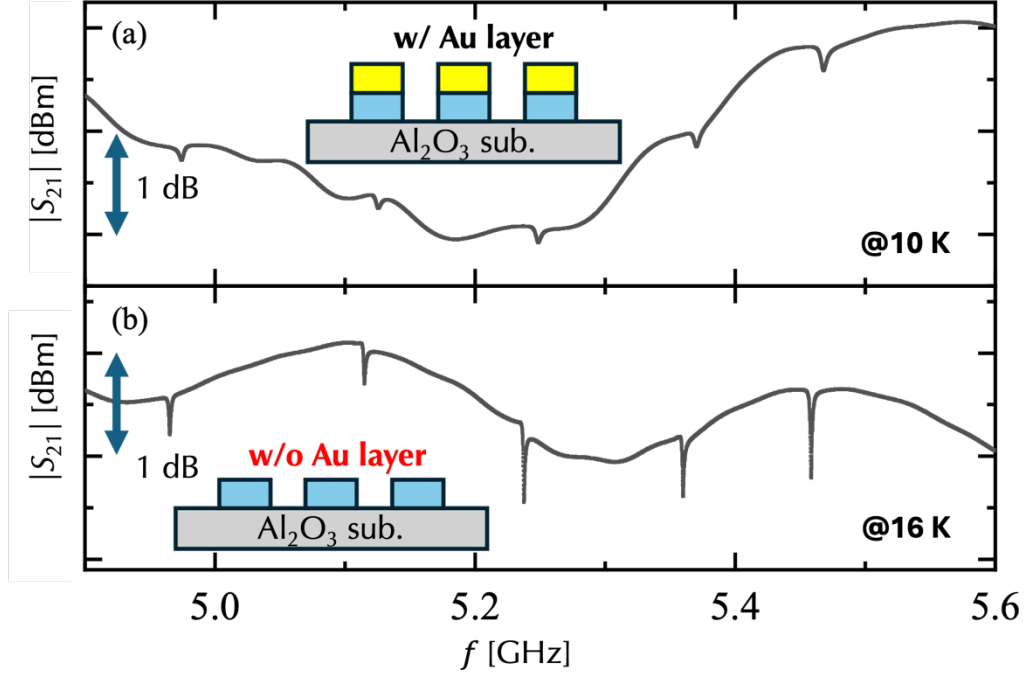


Figure 4-13. The magnitude of transmission spectra $|S_{21}|$ of the same YBCO resonator (a) with and (b) without 40 nm-thick Au layer. The resonators without Au layer on them shows the sharper resonance peaks than them with Au layer.

	w/ Au			w/o Au		
	f_r [GHz]	$Q_{\text{int}} (\times 10^3)$	$Q_{\text{ext}} (\times 10^4)$	f_r [GHz]	$Q_{\text{int}} (\times 10^3)$	$Q_{\text{ext}} (\times 10^4)$
R1	4.975	1.200 ± 0.003	7.346 ± 0.028	4.965	3.039 ± 0.025	6.758 ± 0.069
R2	5.125	1.153 ± 0.005	7.437 ± 0.051	5.115	3.423 ± 0.016	7.411 ± 0.033
R3	5.248	1.105 ± 0.003	6.021 ± 0.024	5.238	4.062 ± 0.026	4.459 ± 0.026
R4	5.371	1.067 ± 0.018	5.167 ± 0.015	5.360	4.003 ± 0.018	5.701 ± 0.023
R5	5.468	1.092 ± 0.003	3.899 ± 0.015	5.459	3.682 ± 0.005	3.553 ± 0.046

Table 4-4. The comparison of the estimated f_r , Q_{int} , and Q_{ext} for the sample with and without Au layer.

In conclusion of this section, the author wishes to highlight the significant potential for optimization in the fabrication process for YBCO resonators. Fortunately, a substantial body of research on the fabrication of YBCO devices has been conducted since the 1990s, with the initial focus being on the fabrication of Josephson junctions and SQUIDs using YBCO. While these insights are not frequently utilized in the fabrication of superconducting resonators, the author anticipates that this discovery will stimulate considerable interest in the application of YBCO manufacturing expertise to the investigation of magnon polaritons in conjunction with superconducting resonators.

4.4.3 Evaluation of resonators made of S-type YBCO

As mentioned in the experimental part, the YBCO composition can be modulated during the co-evaporation, which are Cu and Y rich called as S-type and M-type. Considering effects of the surface state on magnetization dynamics in thin films, S-type YBCO having relatively flat surface should be favorable as for the superconducting resonator to compose of magnon polaritons in thin ferromagnetic films combined with YBCO resonators. The company has indicated that the critical current density of S-type YBCO is inferior to that of M-type YBCO, which may present a challenge in the exploitation of S-type YBCO for superconducting resonators. In this section, the author fabricated the resonators using S-type YBCO and investigate the properties with that of the M-type YBCO.

Figures 4-14. show the comparison of $|S_{21}|$ for YBCO resonators made of M-type and S-type, where the measurement temperature and the microwave power were set to be 10 K and 0 dBm for both measurements, respectively. The measurement for M-type YBCO resonator was carried out after remounting the sample, where the $|S_{21}|$ is slightly different from the result for Figs. 4-13. The S-type YBCO resonators also show the clear resonance peaks corresponding with the five resonances. The fitting results of the resonance features for S-type YBCO resonators are shown in Table. 4-5. All the resonators have the internal quality factors Q_{int} comparable with these of M-type YBCO resonators, showing the promising feature for the observation of magnon polaritons in thin ferromagnetic films. The fabrication of YBCO resonators is a well-established process for both M-type and S-type YBCO films. In Chapter 5, the author employs only the S-type YBCO resonator to maintain relatively flat surfaces for suppressing the additional magnon damping in thin ferromagnetic films.

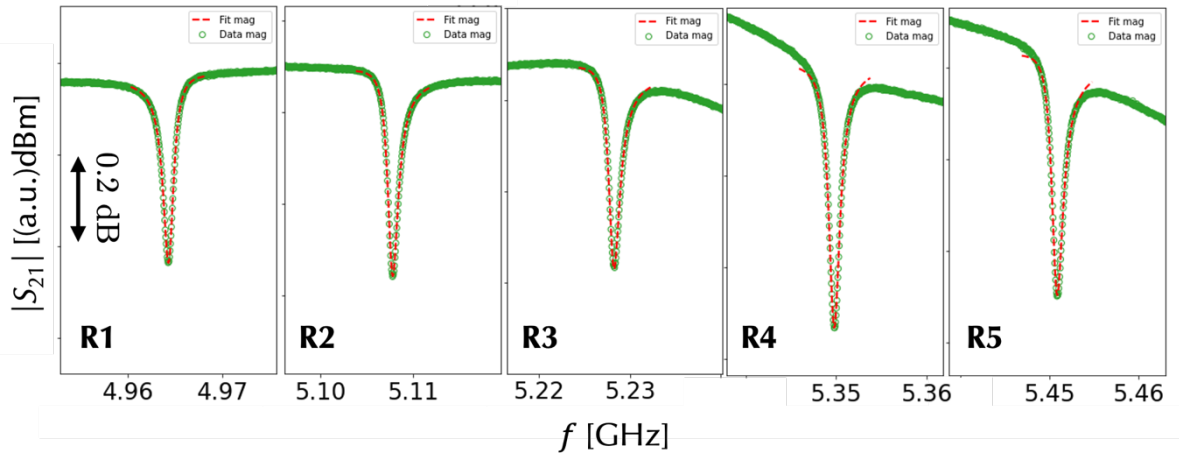


Figure 4-14. The magnitude of transmission spectra $|S_{21}|$ for all the resonators made of S-type YBCO. The measurement temperature was 10 K and the microwave power was 0 dBm. The red broken lines depict the fitting function of Eq. (90). Here, the y-axis is kept being the same range and the one scale is 0.2 dB

	YBCO-S#1		
	f_r [GHz]	$Q_{\text{int}} (\times 10^3)$	$Q_{\text{ext}} (\times 10^4)$
R1	4.964	3.747 ± 0.007	7.752 ± 0.014
R2	5.111	3.810 ± 0.008	7.186 ± 0.014
R3	5.228	3.593 ± 0.026	6.880 ± 0.026
R4	5.350	3.663 ± 0.028	5.343 ± 0.039
R5	5.451	3.643 ± 0.031	5.860 ± 0.048

Table 4-5. The summary of fitting results for S-type YBCO resonators estimated by Eq. (90).

4.5 Temperature and magnetic field dependence of resonance frequency and photon damping

The resilience of the YBCO resonator to temperature and magnetic field fluctuations is anticipated to be a compelling attribute, facilitating the corroboration of observations in magnon polaritons. In this section, the author investigated the temperature and magnetic field dependence of the f_r and the Q_{int} for R1-5 in the S-type YBCO chip to prepare for the observation of magnon polaritons.

4.5.1 Temperature dependence of resonance frequency and internal quality factor

The initial investigation involved examining the temperature dependence of resonance peaks in the absence of an applied magnetic field. Figures 4-15 show the resonance spectra for R1 in YBCO chip at different temperature from 10 K to 70 K. It is noteworthy that the resonance peak persists up to 70 K, exhibiting high quality factors of approximately 1000 even at this temperature. This demonstrates the resilience of YBCO resonators against environmental temperature fluctuations. According to equations Eq. (86-88), this remarkable stability of YBCO resonators can be attributed to the suppression of thermal excitation quasiparticles in YBCO, even at elevated temperatures.

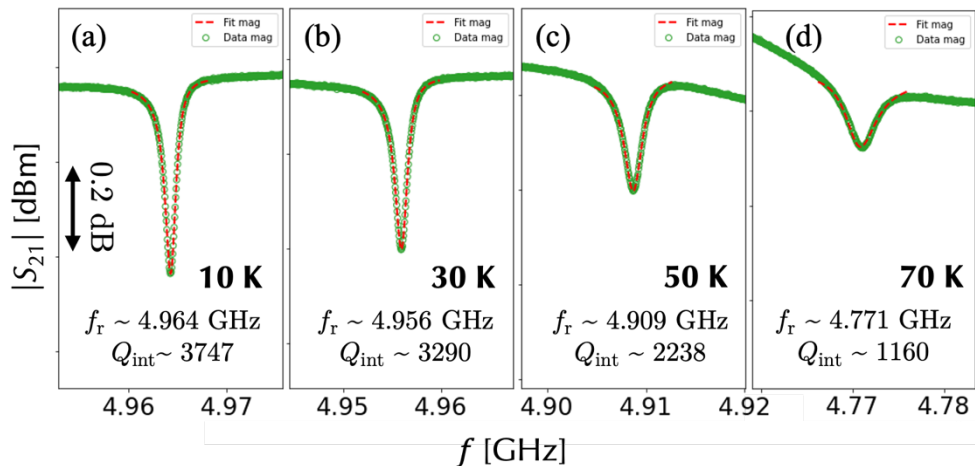


Figure 4-15. The resonance spectra for R1 at various temperature of (a) 10 K, (b) 30 K, (c) 50 K and (d) 70 K. The broken lines in the figure depict the fitting curves of Eq. (90).

Figures 4-16 show the temperature dependence of the f_r and the Q_{int} for all the resonator on the chip (R1-5). The f_r and the Q_{int} decreases with increasing the temperature, these trends can be characterized by Eq. (86). The resonance frequency f_r is parameterized by the surface reactance of YBCO film formulated by Eq. (88), where the London's penetration depth $\lambda_L(T)$ is the definitive parameter to determine the resonance frequency. The $\lambda_L(T, H)$ in YBCO is approximately expressed as follows [174]:

$$\lambda_L(T, H) \sim \frac{\lambda_L(0, H)}{\sqrt{1 - \left(\frac{T}{T_C}\right)^4}}, \quad (92)$$

where T_C is the critical temperature of YBCO films. While $T/T_C \ll 1$, $\lambda_L(T, H) \sim \lambda_L(0, H)$ and we assume the $\lambda_L(0, 0)$ is equal to around 150 nm [175]. This formulation and the experiment results support that high- T_C superconductor is promising for the strong resilience to the ambient temperature. As for Q_{int} , the surface conductivity $\sigma_1(\omega, T)$ is also crucial to parameterize Q_{int} of the superconducting resonators in addition to $\lambda_L(T, H)$. The temperature dependence of $\sigma_1(\omega, T)$ has been carefully investigated in the ref. [175] as shown in Fig. 4-17. Although the bump around 30 K in Fig. 4-16(b) may be attributed to the trend illustrated in Fig. 4-17, we will refrain from a detailed investigation of the origin of the temperature dependence, as we have already identified a promising resonator with robust performance against ambient temperature fluctuations. Anyway, we confirmed the feature of robustness to the temperature fluctuations in the prepared YBCO resonators.

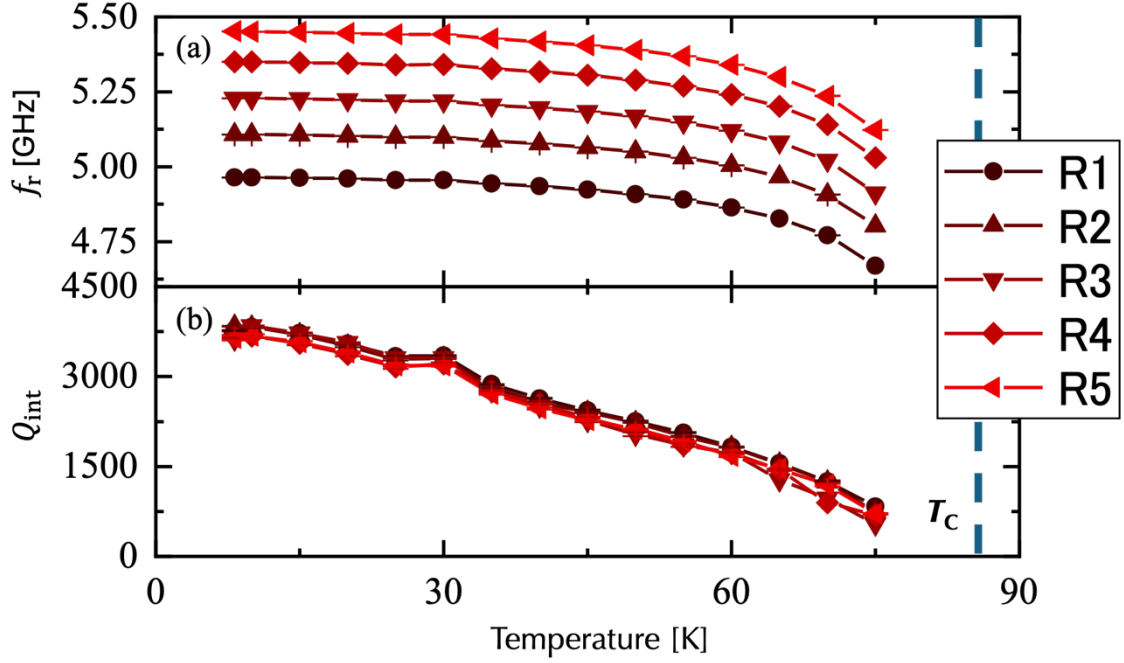


Figure 4-16. The temperature dependence of (a) resonance frequency f_r and (b) internal quality factor Q_{int} for R1-R5 in S-type YBCO resonators. The blue broken line depicts the point of the critical temperature of a feedline in this YBCO chip.

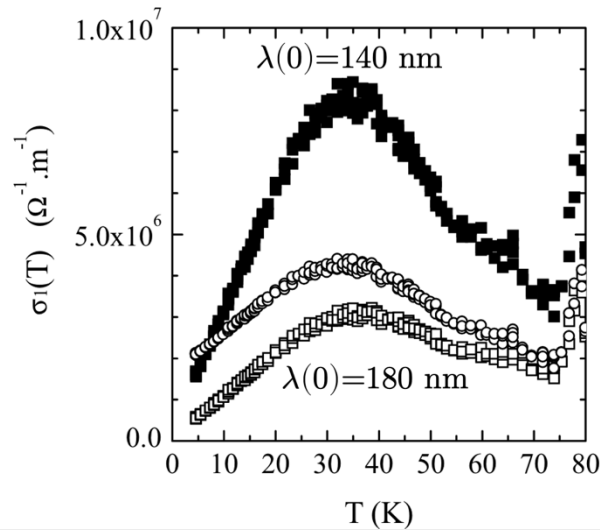


Figure 4-17. The temperature dependence of $\sigma_1(\omega, T)$ in 400 nm-thick YBCO film. Squares show $\sigma_1(\omega, T)$ when taking for the value of the penetration depth $\lambda_L(0, 0) = 130$ nm (fully symbols) and 180 nm (open symbols). This figure is quoted by ref. [175].

4.5.2 Magnetic field dependence of resonance frequency and internal quality factor

For the investigation of performance in magnon-polariton measurements, the subsequent section of this study examines the influence of the external in-plane magnetic field $\mu_0 H$ on the f_r and the Q_{int} for the YBCO resonators at 10 K. Figure 4-18 illustrates the magnetic field dependence of the magnitude of the transmission spectra $|S_{21}|$,

which exhibits five distinct absorption lines that are independent of the magnetic field. The resilience against the external magnetic field within ± 145 mT can be confirmed at glance.

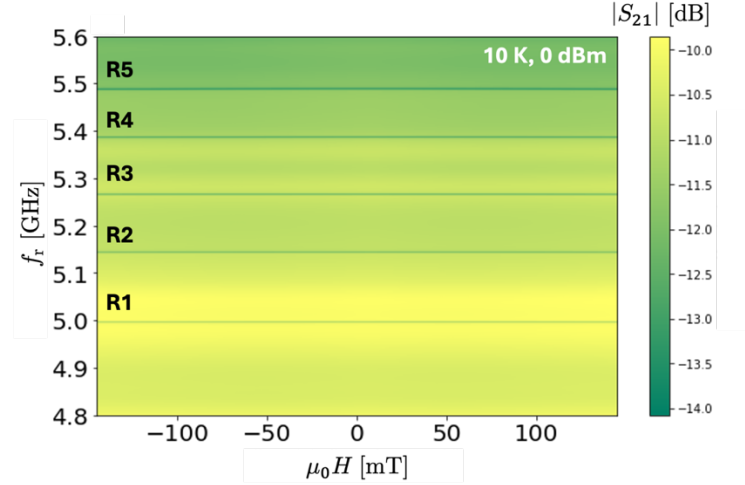


Figure 4-18. The magnetic field dependence of the magnitude of transmission spectra $|S_{21}|$ for R1-R5 resonators, where there are the five horizontal lines independent on magnetic field corresponding with the resonance peaks for YBCO resonators.

For the precise analysis of the influence of magnetic field on the resonance peaks, the fitting of the resonance peaks is carried out for all the magnetic field by using Eq. (90). Figure 4-18 shows the relative change in the resonance frequency and the internal quality factor for all the resonator of the YBCO-S#2. The observed reduction of resonance frequency is within 0.02 % under 145 mT, which shows the stronger resilience of frequency against the external magnetic field than the case of NbTiN that around 0.3 % reduction of resonance frequency at 150 mT was reported [154]. As for the reduction of internal quality factor, the internal quality of factor keeps over 95 % for all the resonators even at the external magnetic field of 145 mT. This robustness is superior to the previous report of NbTiN [154] and Nb resonators [176]. In conclusion, the established YBCO resonator demonstrates remarkable resilience against temperature and magnetic field fluctuations, making it an ideal platform for observing stabilized magnon-polaritons.

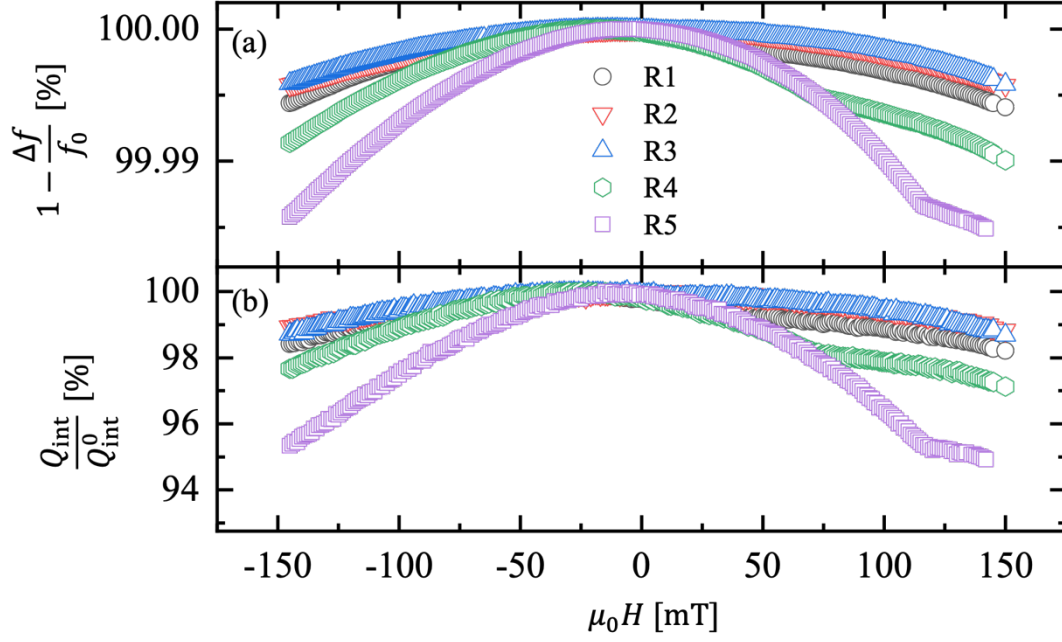


Figure 4-19. Characteristic features of YBCO-S#2 resonators of R1 – R5 in the swept magnetic field. (a) Normalized change in resonance frequency, $1 - \Delta f/f_0$ and (b) normalized internal quality factor by the value at 0 mT, as a function of an external in-plane magnetic field $\mu_0 H$.

4.6 Summary

In conclusion, the author demonstrated the fabrication of superconducting resonators utilizing a high- T_C superconductor, YBCO, with a quality factor of approximately 4000. The resilience of the high- T_C superconductor to an external magnetic field, including the stray field from ferromagnetic materials, is of great importance for the observation of phenomena in magnon polaritons with a large magnon volume. This chapter demonstrates the robust functionality of the resonator in the presence of temperature and external in-plane magnetic fields, with a maximum operating temperature of 70 K and an external magnetic field of 145 mT, respectively. While the maximum external magnetic field is constrained by the experimental setup, the YBCO resonator is capable of functioning with higher magnetic fields, establishing a promising platform for measuring magnon polaritons.

Chapter 5. Magnon polaritons in thin ferromagnetic films

5.1 Abstract

In this chapter, the author investigates strong magnon-photon coupling state, magnon polaritons, in thin ferromagnetic films using YBCO superconducting resonator established in Chapter 4. The energy splitting characterizing strong coupling is observed by mounting a NiFe alloy (Py) stripe on a YBCO resonator. Increasing the number of the ferromagnetic stripes mounted on the YBCO resonator, the coupling state goes into ultrastrong coupling state in 30 nm-thick Py films. The robustness of YBCO superconducting resonator against temperature and magnetic field allows for the clear observation of magnon polaritons' feature in ultrastrong coupling regime, leading to the breakdown of rotating wave approximation and Bloch-Siegert shift originated from the existence of counter-rotating term in coupling Hamiltonian. In addition, the effective coupling strength dependence of Bloch-Siegert shift is observed by changing the number of Py stripes. This report opens up the door to quantum information process in quantum electrodynamics of magnon polaritons.

Results presented in this chapter will be published in the paper:

S. Yoshii, M. Müller, R. Ohshima, M. Althammer, Y. Ando, T. Hioki, H. Huebl, and M. Shiraishi, in preparation.

5.2 Introduction and research purpose

5.2.1 Strong magnon-photon coupling in thin ferromagnetic films mounted on superconducting resonator

The study of hybrid quantum systems has seen significant progress in recent years, particularly in the exploration of strong coupling regimes where distinct quantum excitations interact coherently [177–179]. One such system involves the coupling of magnons—quasiparticles representing collective spin excitations in ferromagnetic materials—and photons, the quanta of electromagnetic waves. When magnons and photons are strongly coupled, they form hybrid excitations known as magnon polaritons [24–27,39,40,56,85,151,180–182]. This regime of strong coupling is distinguished by the coherent exchange of energy between the magnon and photon modes at a rate exceeding their individual decay rates. The formation of magnon polaritons has opened up new avenues for quantum information processing, spintronics, and magnonics. These hybrid quasiparticles leverage the properties of both magnons and photons, enabling efficient and coherent information transfer between magnetic and optical systems.

In magnon-photon coupling, the strength of the interaction is contingent upon the overlap of the electromagnetic field with the collective spin dynamics of the ferromagnetic material. Historically, strong magnon-photon coupling in thin ferromagnetic films has been a challenging phenomenon to achieve, due to a number of factors. These include the limited confinement of the electromagnetic field, the relatively weak interaction between photons and magnons, and the larger magnon damping that occurs in thin ferromagnetic films. However, the use of high-quality superconducting resonators has transformed this field, providing a means to confine photons within a narrow spatial region and markedly enhance the coupling strength [39,40,151]. Superconducting resonators, with their low energy dissipation and high-quality factors, provide an optimal platform for observing and manipulating magnon-photon interactions in the strong coupling regime.

The utilization of superconducting resonators for the attainment of magnon-photon strong coupling in thin-film ferromagnets proffers a number of advantages. First, the high-quality factor of superconducting resonators ensures minimal photon losses, thereby maintaining coherence and enabling the clear observation of hybrid magnon-polariton modes. Secondly, a superconducting resonator is capable of exciting a photon mode that is confined within a small volume on the surface of the resonator, thereby enabling a large filling factor for magnon-photon coupling. Third, the capacity to meticulously regulate the resonator's configuration and the thin film's magnetic characteristics enables researchers to precisely calibrate the coupling strength and investigate a range of magnon-photon coupling scenarios. This flexibility is of critical importance for advancing the understanding of hybrid quantum systems and for developing practical applications in quantum information processing, including quantum transducers, memory elements, and non-reciprocal devices.

The integration of thin-film ferromagnetic materials with superconducting resonators has facilitated the robust realization of magnon-photon strong coupling. This

accomplishment marks a pivotal point in the advancement of magnonics and hybrid quantum systems, paving the way for the development of novel technologies that leverage the distinctive attributes of these hybrid quasiparticles for coherent information transfer and quantum communication.

5.2.2 Ultrastrong coupling in magnon polariton

In the field of light-matter interaction, research has increasingly focused on achieving the ultrastrong or deep coupling regime. This is due to the potential of such regimes to unveil new quantum phenomena beyond those observable in the conventional strong coupling regime [41,42]. Ultra-strong coupling is defined as a coupling strength between light and matter that is comparable to or even exceeds the resonant frequencies of the system itself. In this regime, the rotating wave approximation (RWA), which is typically employed to simplify the Hamiltonian of coupled systems, is no longer applicable. This breakdown leads to the emergence of novel hybrid states and unconventional quantum effects, such as Bloch-Siegert shift, ground-state energy shifts due to the existence of counter-rotating term [94–96]. The realization of the ultrastrong coupling regime has significant implications for developing advanced quantum technologies, including the dynamical quantum entanglement robust against the dissipation [93,183].

The extension of the concept of ultrastrong coupling to magnon-polariton systems represents a significant advancement in the field of hybrid quantum physics. In conventional magnon-photon coupling, the coupling Hamiltonian offers the rotating wave approximation, whereby the counter-rotating term is deemed to be negligible. However, the exploration of ultrastrong coupling in magnon-polariton systems pushes the boundaries of this interaction, enabling the study of a new quantum regime where it is theoretically predicted that counter-rotating terms in a coupling system generates the quantum entanglement of ground state [184,185]. Achieving ultrastrong coupling in magnon-polariton systems is particularly exciting because it opens up opportunities on magnonics to observe effects that are not accessible in the standard strong coupling regime. Moreover, the plenty physics in magnonics, such as tunability of magnetic properties [16,34,60,76,77,186–190], non-reciprocity [43–49,191], non-linearity [35,50–52,82,85,86,192–194] and non-Hermitian [195–201] can be introduced into the field of light-matter interaction.

The ultrastrong coupling regime in magnon polaritons represents a remarkable achievement, yet achieving a magnon-photon coupling strength comparable to the resonance frequency remains challenging. To date, the ultrastrong coupling regime in magnon polaritons is facilitated by the use of superconducting resonators [91,92,202] and magnetochiral metamolecules [203], opening up avenues to the intriguing physics caused by the counter-rotating term. Although the frontier of the ultrastrong coupling regime in magnon polaritons has been started to be explored, the choice of ferromagnetic materials remains limited, which hampers the investigation of magnon polaritons under an ultrastrong coupling regime and the full realization of the potential of magnonics. The primary

constraints are a consequence of the trade-off between the volume of the photon mode and the number of spins in ferromagnetic materials. The restricted volume of the photon mode, which offers the potential for achieving a large coupling strength with each spins g_s , limits the volume of the magnon mode capable of overlapping with photons, and vice versa.

5.2.3 Purpose of this chapter

The purpose of this chapter is to explore the magnon polaritons in thin ferromagnetic films operating under an ultrastrong coupling regime. The author achieved this ultrastrong coupling of magnon-photon coupling between the YBCO resonator and 30 nm-thick Py films. This system enables adjustment of magnon-photon coupling strength by changing the number of Py strips on a resonator, which are coherently interconnected via a uniform photon mode. YBCO resonators allow for the clear observation of coupling spectra, demonstrating the Bloch-Siegert Shift. In addition, the author demonstrated the modulation of Bloch-Siegert Shift by changing the number of Py strips.

5.3 Experimental setup

This section introduces the experimental details for the observation of magnon polaritons in thin ferromagnetic films. In this chapter, the YBCO-S#1 or #2 are used for the measurement.

5.3.1 Sample fabrication

- **Fabrication of FM strips**

In this chapter, the author selected $\text{Ni}_{81}\text{Fe}_{19}$ alloy (Py) thin films as ferromagnetic material for magnon excitation since Py is very common ferromagnetic material with relatively low intrinsic magnon damping ($\alpha_{\text{int}} \sim 0.005$ [1,2]) and low crystalline magnetic anisotropy. First, the author patterned PMMA resist for the FM strip structure with electron beam lithography. A 20 nm-thick SiO_2 layer was deposited on the YBCO resonators to prevent direct electrical contact with the ferromagnetic film and YBCO. As shown in Fig. 5-1(a), a Ti/Py/MgO stacking layer with a strip was deposited on a SiO_2 separation layer within the inductive part of YBCO resonator by electron beam deposition. The thicknesses of Ti, Py and MgO were 3 nm, 30 nm, and 2 nm, respectively. A 10 nm-thick SiO_2 layer was added to protect the Py layer from oxidation. Finally, the deposited films is removed by the lift-off process in acetone. Figure 5-1 (b) shows the optical image of the fabricated single Py strip on a YBCO resonator without Au layer. The multiple Py strips are fabricated in the same way as the case of single Py strip.

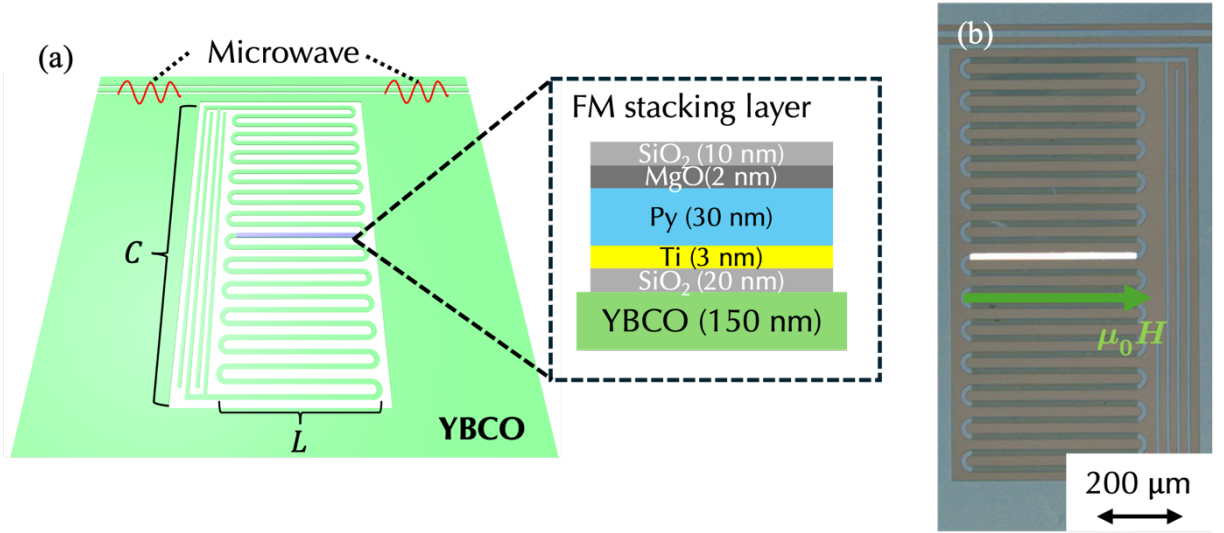


Figure 5-1. Illustration of the fabricated FM strip on YBCO resonator. (a) Schematic of the system with YBCO resonator and Py thin film of the strip structure, where the layer stacking is SiO₂ (20)/Ti (3)/Py (30)/MgO (2)/SiO₂ (10). (b) Optical image of the resonator (R1) with a single FM strip. The external magnetic field $\mu_0 H$ is applied along the Py strip. We see the YBCO layer (light blue area), YBCO/SiO₂ cap (grey area) and FM strip (white area).

- **Fabrication of FM strips**

The multiple Py strips with the geometry of $400 \mu\text{m} \times 15 \mu\text{m}$ are fabricated in the same way as the case of single Py strip. The total number of strips in the inductive part of the resonator is 26, which is equal to the maximum number of FM strips that can be accommodated. In order to investigate the Py strip number dependence of coupling strength and the phenomena that occur under ultrastrong coupling strength, the author altered the number of Py strips on a resonator. In this section, the author examined the YBCO resonators with the Py strips' number n of 1, 8, 16 and 26. As for the YBCO-S#1, it has a single and 26 Py strips on R5 and R4, respectively. The sample of YBCO-S#2 has 1 (R5), 8 (R4), 16 (R3), and 26 (R1) Py strips on the each resonator.

5.3.2 Measurement

- **Transmission measurement as a function of external magnetic field**

The transmission spectra S_{21} is measured with altering the external magnetic field to examine magnon polaritons in thin Py films mounted on YBCO resonator. The measurement setup was the same as the measurement in the section of 4.5.2. The measurement temperature and input microwave power were set to be 10 K and 0 dBm, respectively. The external magnetic field is swept from 145 mT to -145 mT. For analyzation of spectra, we obtain ΔS_{21} in which the background spectra were extracted from the magnetic field dependence of the spectra by dividing with the spectra at a specific magnetic field ($\mu_0 H =$

30 mT). This field was selected due to the minimal absorption observed in the spectra, which is attributed to anti-crossing.

5.4 Magnon polaritons in thin ferromagnetic films

In this section, the author investigates the coupling state of photons excited by YBCO resonator and magnons excited in 30 nm-thick Py films. The polariton mode exhibits a splitting into two excitation states due to the strong exchange of energy between magnons and photons. This results in an anti-crossing of the transmission spectra as a function of the external magnetic field, which is observed as a variation in the resonance frequency of YBCO resonators.

5.4.1 Evaluation of magnon-photon coupling state

First, we see the transmission spectra for the resonator with a single Py strip as a function of the external magnetic field and evaluate the coupling parameters such as the effective coupling strength g_{eff} , the resonance frequency of photon mode, and that of magnon mode by analyzing the transmission spectra. Figures 5-2 (a) and (b) show the magnitude of background subtracted transmission spectra $|\Delta S_{21}|$ as a function of the external magnetic field for the resonator of R5 with a single Py strip and the spectra with the fitting by Eq. (90), respectively. Figure 5-2(a) provides a striking illustration of the splitting of polariton resonances around 30 mT, a defining feature of the strong coupling regime. To evaluate the coupling parameters via Eqs. (65),(66) and (71-74), it is necessary to determine the magnetic field dependence of the resonance peaks of magnon polaritons, f_{mp} , and the internal relaxation rate ($\kappa_{\text{int}} = \pi f_{\text{mp}}/Q_{\text{int}}$), which is defined as the half-width at half-maximum of resonance peaks in the coupling state. Since magnon mode coupled with a resonator is just an additional environment to the resonator with a ferromagnetic material, Eq. (90) is valid at the different magnetic field. Thus, the estimation of the f_{mp} and κ_{int} for all the magnetic field was carried out by fitting the spectra with Eq. (90) as shown in Fig. 5-2(b). Using the obtained f_{mp} and κ_{int} as a function of $\mu_0 H$ and Eqs. (65),(66) and (71-74), the coupling parameters such as g_{eff} , f_p , M_{eff} , κ_p and κ_m can be discussed.

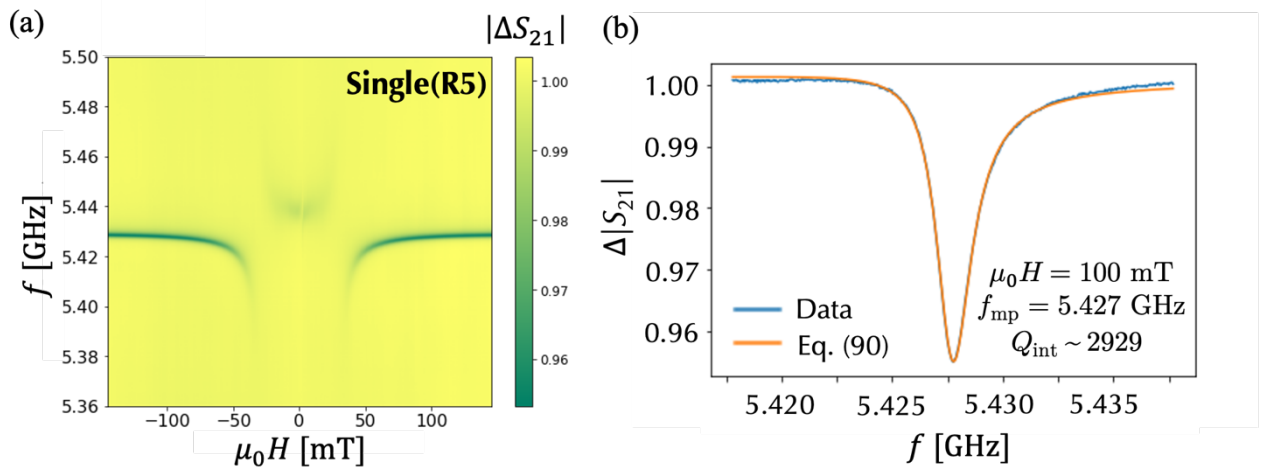


Figure 5-2. (a) The magnitude of subtracted transmission spectra $|\Delta S_{21}|$ as a function of the external magnetic field $\mu_0 H$ for the resonator (R5) with a single Py strip. (b) The spectra data at $\mu_0 H = 100$ mT (blue) with the fitting curve (orange) by Eq. (90). Here, f_{mp} is defined as the magnon polariton frequency.

Next, we start with the non-dissipative Hamiltonian with rotating-wave approximation, i.e., Tavis-Cummings model, defined as Eq. (56). The eigen-frequency of this system is formulated by Eq. (74), which corresponds with the estimated frequency of the magnon polariton, f_{mp} , as a function of the external magnetic field. Figure 5-3. shows the magnitude of the transmission spectra $|\Delta S_{21}|$ with the fitting results, where $\omega_m = 2\pi f_m = \gamma\mu_0\sqrt{H(H + M_{\text{eff}})}$ in Eq. (74). The f_p , $\mu_0 M_{\text{eff}}$ and $g_{\text{eff}}/2\pi$ are estimated to be 5.434 GHz, 1.376 T, and 136 MHz, respectively, which are comparable with the coupling strength with NbTiN resonator and 30 nm-thick CoFe strip with the same structure studied in Müller's PhD thesis [204]. Here, the ratio of g_{eff} and ω_p , $\eta = g_{\text{eff}}/\omega_p$ is equal to 0.025, which is a bit far from ultrastrong coupling. While the fitting function nicely reproduces the lower branch of magnon polariton mode, the fitting of upper branch doesn't match well. This can be attributed to the influence of magnon damping on the coupling state. As discussed in Chapter 2, when the coupling strength is below the relaxation rate of the magnons or photons, the energy splitting breaks down and the coupling state enters the weak coupling regime or the Purcell regime [27]. The established way for considering the damping parameter in magnon polaritons under the strong coupling regime is to introduce the damping parameters of magnons κ_m and photons κ_p into the Hamiltonian, resulting in Eq. (65) and (66) by approximately solving the eigen-value problem [40,204]. Then, the f_{mp} and κ_{mp} as a function of the external magnetic field are fitted by Eq. (65) and (66) to estimate all the coupling parameters.

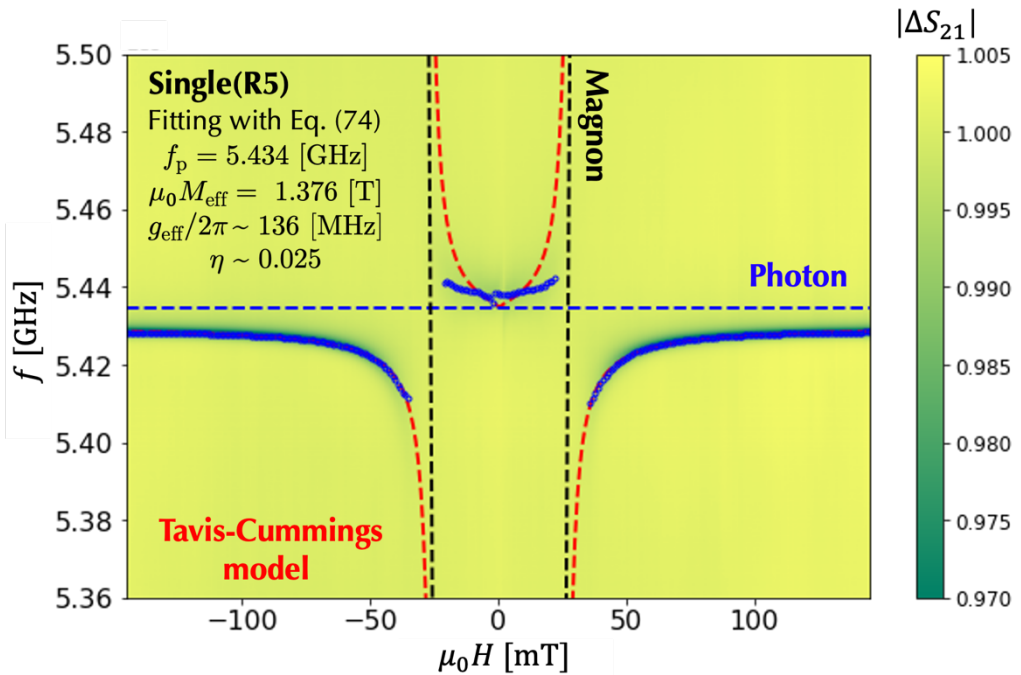


Figure 5-3. The magnitude of the transmission spectra $|\Delta S_{21}|$ with the fitting results. Here, the blue circle points show the f_{mp} estimated by Eq. (90). The broken black and blue lines show the magnon mode formulated by Kittel mode of Eq. (6) and the photon mode of the YBCO resonator of R5. The red broken curves show the magnon polariton mode formulated by Eq. (56).

Fitting the transmission spectra considering the damping in magnons and photons under strong coupling, we evaluate the damping of magnon and photon mode in addition to the f_p , $\mu_0 M_{\text{eff}}$ and g_{eff} . Figures 5-4 (a) and (b) show the f_{mp} and κ_{mp} as a function of the external magnetic field with the fitting results with Eqs. (65) and (66). The fitting provides the estimated coupling parameters: $f_p = 5.431$ GHz, $\mu_0 M_{\text{eff}} = 1.333$ T, $g_{\text{eff}}/2\pi = 129$ MHz, $\kappa_p/2\pi = 0.53$ MHz and $\kappa_m/2\pi = 461$ MHz, where the cooperativity $C = g_{\text{eff}}^2/\kappa_p\kappa_m = 67.4$. The coupling state is in the Percell regime ($\kappa_m > g_{\text{eff}} > \kappa_p$) [27], which may originate from the large magnon damping in these ferromagnetic films due to the pole on the YBCO surface as discussed in section 4.3. Note that the fitting curves for the upper branch does not reproduce the experimental data even if we introduce the damping parameters in Hamiltonian. Considering the high cooperativity of this system, the enhancement of photon damping around 0 mT can be attributed to the Purcell effect due to the existence of the dissipative magnons, where a lot of photons dissipates into magnon mode [27,205]. The coupling strength of g_s with a single Bohr magneton is estimated from $g_s = g_{\text{eff}}/\sqrt{N}$, where N is the number of spins in the Py ($\text{Ni}_{80}\text{Fe}_{20}$) strip with the volume of $400 \times 15 \times 0.03 \mu\text{m}^3$. Measuring the magnetic moment per volume in 10 nm-thick Py films (see Appendix K), the number of spins for the Py strip is estimated to be 1.92×10^{13} . Thus, we obtained the g_s of 29.5 Hz, which is larger than the CPW resonator in previous studies [40,151].

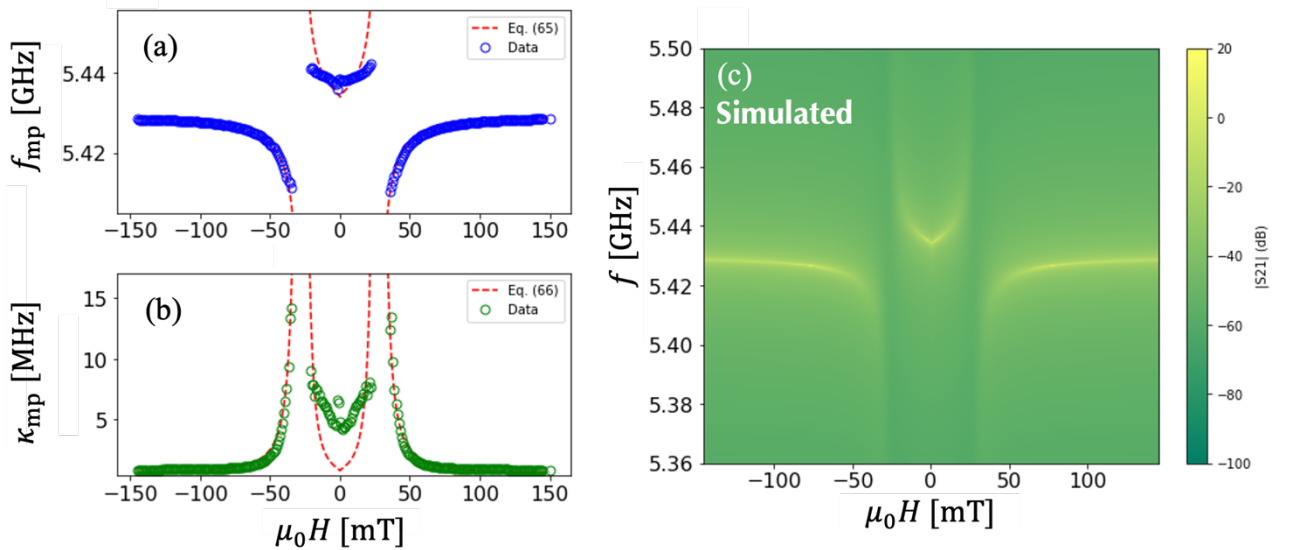


Figure 5-4. The evaluation of the coupling parameter considering the damping of magnon and photon mode. The fitting results using Eqs. (65) and (65) for (a) the resonance frequency

f_{mp} and (b) the damping κ_{mp} of the magnon polariton, where the red broken curves show the fitted curve for upper and lower branches. (c) The simulated magnitude of transmission spectra $|\Delta S_{21}|$ simulated by Eq. (94).

Considering the input-output theory [206], we can add the driving Hamiltonian of photon operators given by:

$$H_{\text{drive}} = -i\hbar\zeta(a - a^\dagger), \quad (93)$$

where ζ is the coupling rate between a feedline and a superconducting resonator, to Eq. (56). Solving the modified Hamiltonian, we obtain the result that the transmission spectra of this system is proportional to the term as follows [24,40,151]:

$$|S_{21}| \propto \frac{\kappa_{\text{ext}}/2\pi}{i(f - f_r) - \kappa_p/2\pi + \frac{[g_{\text{eff}}/2\pi]^2}{i(f - f_m) + \kappa_m/2\pi}}. \quad (94)$$

Figure 5-4 (c) shows the simulated spectra with Eq. (94) using the obtained parameters by Eqs. (65) and (66), which does not nicely reproduce the resonance frequency and the damping of the upper branch. This may be because the coupling system is in the Purcell regime and Eq. (94) is not fully valid in this system.

5.4.2 Enhancement of coupling strength with multiple Py strips

The effective coupling strength of magnon polaritons in superconducting resonator is formulated by:

$$g_{\text{eff}} = g_s \sqrt{N} = \frac{\xi}{2} \sqrt{\frac{\mu_0 \hbar \omega_p}{V_p}} \gamma \sqrt{2Ns}, \quad (95)$$

where V_p and ξ are the volume of photon mode and the filling factor of magnon mode in the photon mode excited on the resonator ($0 < \xi < 1$), respectively. As the parameters ω_p and V_p are dependent on the structure of the superconducting resonator, the only accessible parameter is the number of spins N coupled with the microwave resonator. Aiming for the enhancement of N , we start to think of increasing the number of strips, n , on a YBCO resonator. Given placing the separated Py stripes on the inductive component in a resonator, it can be seen that the excited magnons in each strip exchange energy with the photon mode separately. Consequently, the magnon polariton mode simply degenerates, and the effective number of the total Bohr magneton should remain unaltered. Nevertheless, the separated ferromagnet is capable of exhibiting behavior akin to that of a single magnon mode, as a consequence of magnon bright mode when they are excited in a phase. This has been evidenced in both magnons in magnon-magnon couplings via a photon resonators [14,207,208]. The concept of utilizing magnon bright mode is anticipated to facilitate the enhancement of coupling strength in magnon polaritons. Figure 5-5(a) illustrates the schematic of the YBCO resonator comprising multiple Py stripes. As

previously discussed in the preceding sections, the excited microwave photon mode can excite magnons and exchange their energy with one another. While this system has the separated Py stripes, the excited magnon modes in each Py strips can behave like one magnon mode due to the magnon-magnon coupling via the photon resonators as shown in Fig. 5-5 (b).

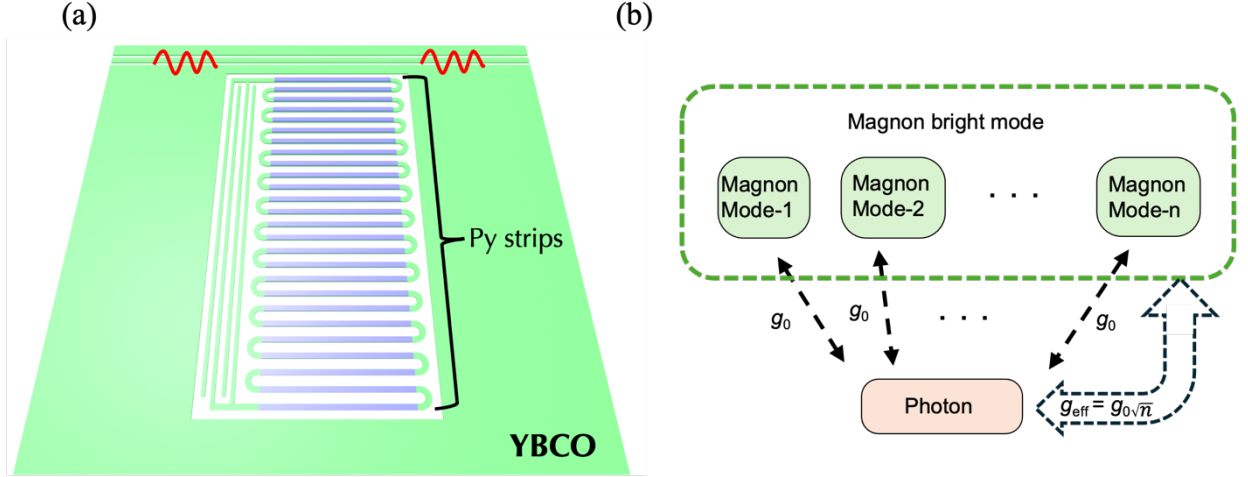


Figure 5-5. (a) Schematic of the YBCO resonator with multiple Py stripes. The maximum number of multiple Py stripes we can place in this structure is 26. (b) The coupling between photon mode and magnon bright mode. The excited magnons in multiple Py strips couples with each other via the uniform photon mode and behave like one magnon mode with the effective coupling strength, $g_{\text{eff}} = g_0\sqrt{n}$.

Aiming for the enhancement of the coupling strength, the author prepared the multiple Py stripes with the same stacking order as single one on a YBCO resonator of R4 in the YBCO-S#1. Here, the number of strips n is 26 and the resonators of R1-R3 are the empty resonators without the Py stripes. Figure 5-6. shows $|\Delta S_{21}|$ as a function of the external magnetic field, where the background is subtracted by dividing the whole spectra with the spectra at $\mu_0 H = 30$ mT. We see the splitting curves also in R5 with multiple Py stripes in addition to R4 with a single one. Notably, the bending of the dispersion relation is stronger than that for a single Py strip, which indicates the coupling strength of magnon polariton is enhanced by increasing the number of Py stripes n . First, we think of the Hamiltonian with rotating wave approximation, deriving the polaritons frequency of Eq. (74) to estimate the coupling strength as shown in Fig. 5-6(b). Then, we obtained the fitting results; $f_p = 5.396$ GHz, $\mu_0 M_{\text{eff}} = 1.306$ T, and $g_{\text{eff}}/2\pi = 614$ MHz. The coupling strength is largely enhanced by increasing the Py stripes on a YBCO resonator. According to Eq. (95), since the normalized coupling strength, $\epsilon \equiv g_{\text{eff}}/2\pi\sqrt{f_p}$, is proportional to the $\sqrt{N}$, the estimated $\epsilon(n)$ should be enhanced by $\sqrt{n}$. The estimated $\frac{\epsilon(26)}{\epsilon(1)} = 5.16$ is comparable with $\sqrt{26} \sim 5.09$. This $\sqrt{n}$ rule will be discussed in detail in Section 5.4.4 when investigating the n -dependence of the coupling strength.

Here, the fraction of g_{eff} and $2\pi f_p$, $\eta = g_{\text{eff}}/2\pi f_p \sim 0.114 > 0.1$, entering the ultrastrong coupling regime [41,42]. In this regime, the rotating wave approximation breaks down and the counter-rotating term in the coupling Hamiltonian becomes non-negligible. We need the analysis considering counter-rotating term and A^2 term in the system Hamiltonian, i.e., Eq. (77) for the precise investigation [91,209].

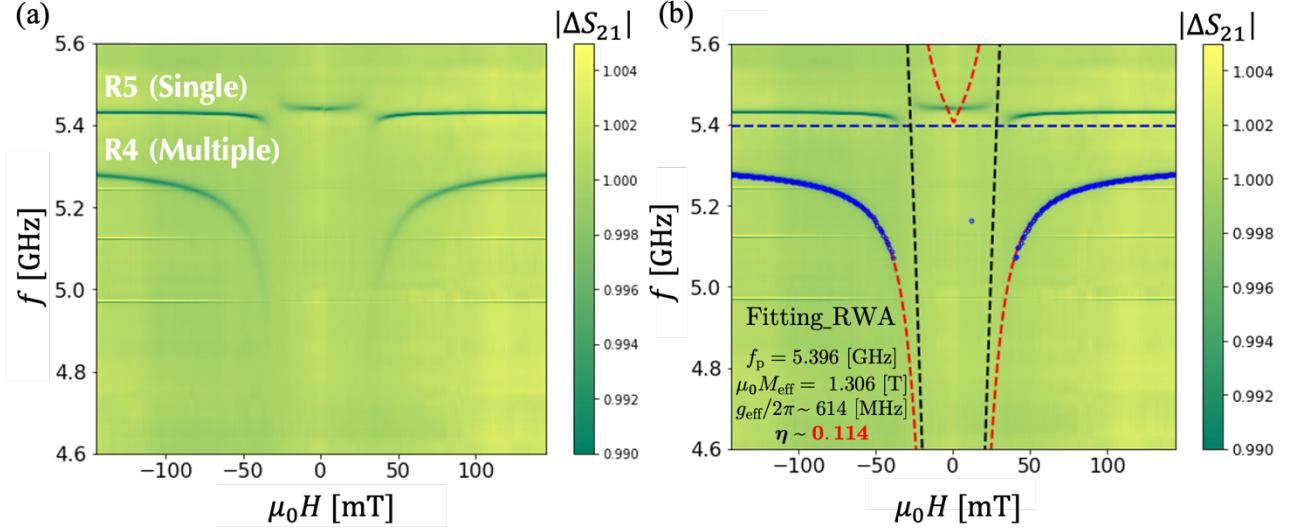


Figure 5-6. (a) The $|\Delta S_{21}|$ as a function of the external magnetic field for the YBCO resonators with single ($n = 1$) and multiple ($n = 26$) Py stripes, which are placed on R5 and R4, respectively. The three lines under R4 are the peaks of R1-3, where the resonators also without Py strips have the slight magnetic field dependence of resonance as discussed in Chapter 4. (b) Fitting results by Eq. (74) with rotating wave approximation (RWA). Blue circles show the estimated f_{mp} from the spectra.

5.4.3 Breakdown of rotating wave approximation in magnon polaritons

In the ultrastrong coupling regime, the rotating wave approximation (RWA) breaks down and the analyzation with the full Hamiltonian considering the counter-rotating term and A^2 term is inevitable [95,96]. Here, we will analyze the transmission spectra of the magnon polariton frequency with Eq. (77) and compare the fitting results with and without the rotating wave approximation. The estimated parameters with RWA (Tavis-Cummings model for Eq. (56)) and without RWA (Hopfield model for Eq. (57)) are summarized in Table 5-1. It is noteworthy the estimated f_p in the multiple Py stripes under ultrastrong coupling exhibits a considerable discrepancy depending on the model applied. Conversely, the f_p in a single Py strip remains largely consistent. Furthermore, the f_p in magnon polariton coupling should be comparable with or lower than the resonance frequency in the absence of Py stripes. In this context, the estimated f_p in the resonator with multiple Py stripes under RWA is greater than the f_p in the absence of Py, as illustrated in Table 4-5. Figures 5-7. demonstrate the $|\Delta S_{21}|$ as a function of the external magnetic field with the fitting curves expressed by the eigen-frequency in the Tavis-Cummings and Hopfield model.

The parameters in these models were estimated using Eq. (77). It is notable that the Tavis-Cummings model (the model with RWA) demonstrates good agreement with the experimental data when the coupling is conventional. In contrast, when the coupling is ultrastrong, the model with RWA exhibits a markedly different behavior, which leads to the breakdown of RWA in magnon polariton systems. In the following sections, we will use the full Hamiltonian of Hopfield model to evaluate the parameters. The analysis of counter-rotating term and A^2 term is discussed in detail in section 5.5.

	Single Py strip on R5		Multiple Py stripes on R4	
	w/ RWA	w/o RWA	w/ RWA	w/o RWA
f_p [GHz]	5.431	5.430	5.396	5.310
$\mu_0 M_{eff}$ [T]	1.333	1.411	1.306	1.401
$g_{eff}/2\pi$ [MHz]	129	145	614	674

Table 5-1. The comparison of the estimated parameters, f_p , $\mu_0 M_{eff}$, and $g_{eff}/2\pi$ for the Hamiltonian with and without RWA, which are named as Tavis-Cummings model and Hopfield model.

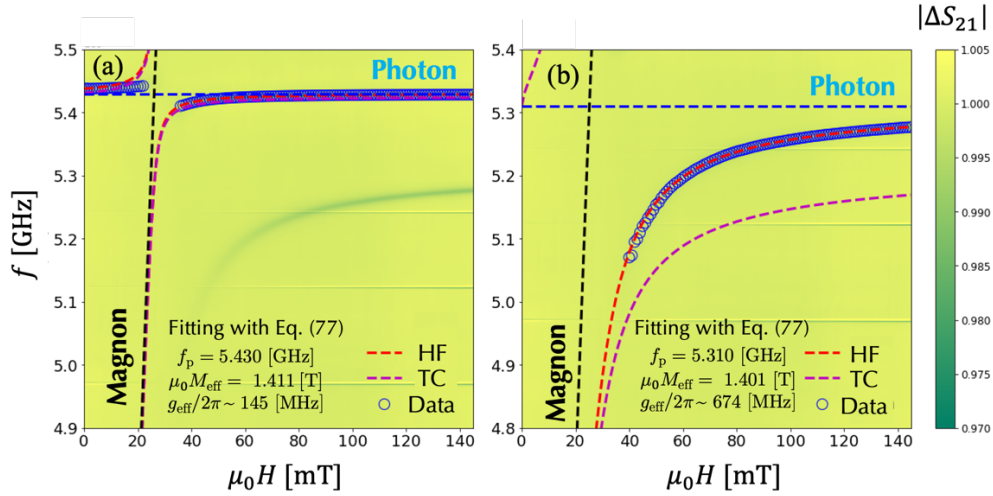


Figure 5-7. The $|\Delta S_{21}|$ as a function of the external magnetic field with the fitting results by Eq. (77) for (a) single Py strip and (b) multiple Py stripes. Here, the red and purple broken lines show the magnon polaritons frequency for Hopfield (HF) Hamiltonian and Tavis-Cummings (TC) Hamiltonian, which are calculated using the parameters estimated by Eq. (77). The black and blue broken lines depict the uncoupled magnon and photon mode calculated by the estimated f_p and $\mu_0 M_{eff}$.

5.4.4 Py strip number dependence of coupling strength

Expecting the modulation of coupling strength in magnon polaritons, we change the number of Py strips, n , on a YBCO resonator. The 30 nm-thick Py stripes with the different number of strips ($n = 1, 8, 16, 26$) are mounted on the resonators in a YBCO-S#2 chip.

Figures 5-8 illustrate the dependence of the $|\Delta S_{21}|$ on the external magnetic field for resonators with varying numbers of Py strips, together with the corresponding fitting results. In this section, Eq. (77) has been employed to estimate the relevant parameters, showing the good agreement with the observed magnon polariton modes. The estimated parameters of f_p , $\mu_0 M_{\text{eff}}$, and $g_{\text{eff}}/2\pi$ are summarized in Table 5-2, demonstrating the increase of the effective coupling strength with increasing the number of Py strips. As for the case of $n = 26$, the coupling state enters the ultrastrong coupling regime. Considering Eq. (95), the normalized coupling strength, $\epsilon \equiv g_{\text{eff}}/2\pi\sqrt{f_p}$, should be proportional to $\sqrt{n}$ since the number of spins, N , are proportional to the number of Py stripes, n , when the strips couple with each other. Figure 5-9. shows the ϵ as a function of $\sqrt{n}$. The $\epsilon(n)$ is defined as follows:

$$\epsilon(n) = \alpha D \sqrt{n}, \quad (96)$$

where D is the phenomenological coefficient of coupling strength between Py strips in this stripe structure and the coupling coefficient for a single Py strip. In addition, the α is formulated by $g_s/2\pi\sqrt{N_s/f_p}$, where g_s and N_s are the coupling strength with a single Bohr magneto and the number of spins in single Py strip. The α in the resonators of YBCO-S#1 and YBCO-S#2 are calculated to 1.91×10^3 and 1.63×10^3 , respectively, from the ϵ for a single stripe. Moreover, the αD is estimated to be $1786\sqrt{\text{Hz}}$ for YBCO-S#1 and $1556\sqrt{\text{Hz}}$ for YBCO-S#2 from the slope of ϵ , providing D is 0.935 and 0.955, respectively.

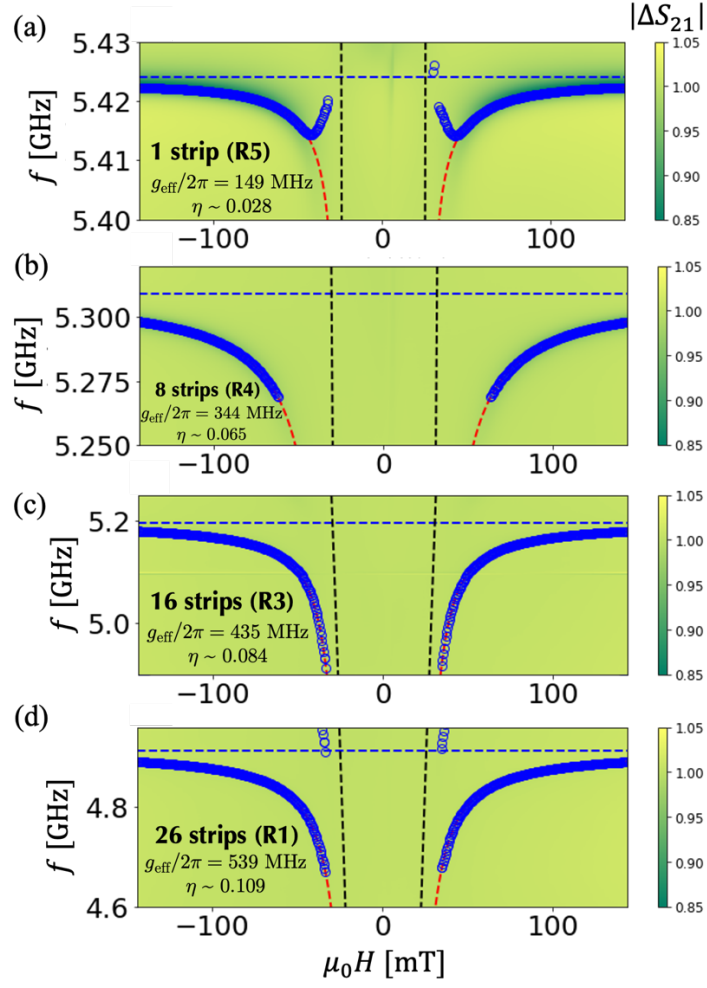


Figure 5-8. The $|\Delta S_{21}|$ as a function of the external magnetic field, $\mu_0 H$, for the resonators with the different number of Py strips. Here, the broken red, black and blue lines depict the fitting curve of Eq. (77), magnon mode expressed by Kittel mode, and photon mode of YBCO resonators.

n	1	8	16	26
f_p [GHz]	5.424	5.309	5.196	4.913
$\mu_0 M_{eff}$ [T]	1.478	1.124	1.113	1.192
$g_{eff}/2\pi$ [MHz]	149	344	435	538
η	0.028	0.065	0.084	0.109

Table 5-2. Summary of the estimated parameters for the different number of Py strips.

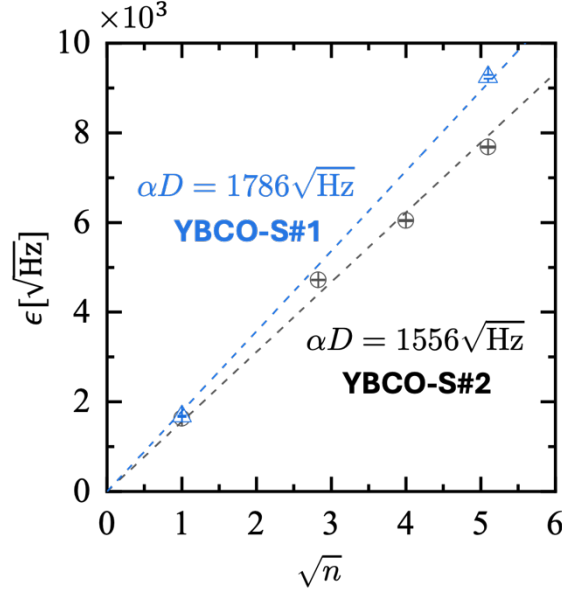


Figure 5-9. The n dependence of $\epsilon = g_{\text{eff}}/2\pi\sqrt{f_p}$.

5.5 Ultrastrong coupling in magnon polaritons

When the coupling state enters the ultrastrong coupling regime, the rotating wave approximation breaks down. This breakdown leads to the existence of non-negligible counter rotating term, $\mathcal{H}_{\text{CR}}$, in the magnon polariton system, which is given by:

$$\mathcal{H}_{\text{CR}} = g_{\text{eff}}(a^\dagger b^\dagger + ab). \quad (97)$$

The counter-rotating term causes the non-conservation of magnons and photons, i.e., virtual excitation of photons [41,42,95]. The presence of counter-rotating terms in magnon polaritons enables the quantum mechanical effects of squeezing and entanglement of magnons and photons [210] containing a finite virtual photons, which have been the subject of considerable interest as a potential avenue for novel quantum information processing. However, to date, the experimental demonstration of the existence of counter-rotating term in the magnon polariton system has not been reported and the symbolic phenomena on magnon polaritons in ultrastrong coupling regime has not been explored. Therefore, this section reports on one of the symbolic phenomena in ultrastrong coupling regime, Bloch-Siegert Shift, and its magnetic field dependence, where the Bloch-Siegert shift is the frequency shift due to the existence of the counter-rotating term. In addition to the Bloch-Siegert Shift, the author investigates the A^2 term as a function of the external magnetic field.

5.5.1. Observation of Bloch-Siegert Shift and A^2 shift

The Bloch-Siegert shift and A^2 shift in the ultrastrong coupling regime are observed by quantitatively evaluating the contribution from the counter-rotating term and the diamagnetic term in Eq. (57), which can be separated by solving the eigen-frequency of

the Hamiltonian with and without these terms as shown in Eq. (71-73) [94–96]. Figure 5-10 illustrates the magnetic field dependence of $|\Delta S_{21}|$, contrasting the theoretical calculation of the polariton frequency as outlined in Eqs. (71-73) for the various model Hamiltonians. In this instance, the estimated parameters derived from the fitting of the spectra by the full Hamiltonian are employed in the calculation. The discrepancy between Eqs. (71) and (72) demonstrates the impact of the A^2 term on the photon energy shift, whereas the discrepancy between Eqs. (71) and (73) illustrates the influence of the counter-rotating term. Therefore, the observation of Δf_{BS} and Δf_{A^2} is the direct evidence of the presence of non-negligible counter-rotating term and A^2 term in this magnon polariton system, where the magnitude of them parametrizes the energy portion of counter-rotating term and A^2 term in the system.

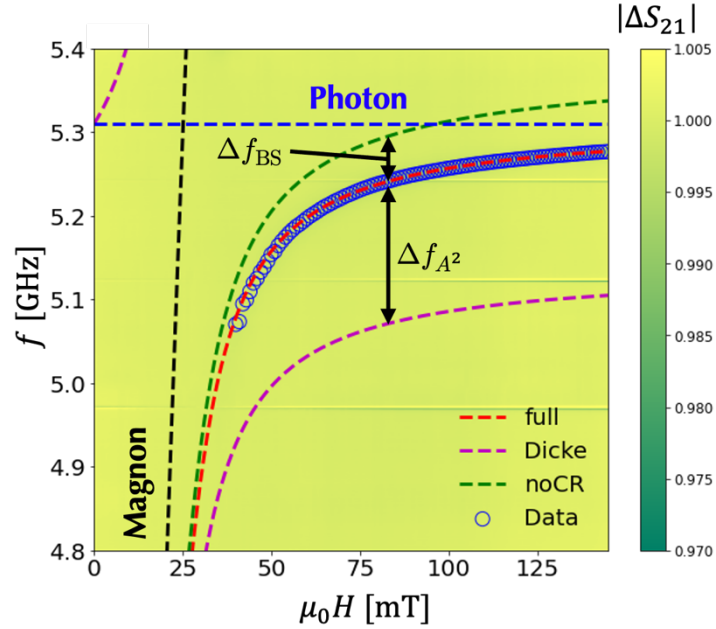


Figure 5-10. Comparison of the numerical calculation of dispersion relation on the Hamiltonian of Eq. (71-73). The full, Dicke and no CR in the caption represent the polariton frequency of Eq. (71) for full Hamiltonian, Eq. (73) for Dicke Hamiltonian, and Eq. (72) for the Hamiltonian without the counter-rotating term, respectively. The Δf_{BS} and Δf_{A^2} are the frequency shift of photon mode due to the existence of the counter-rotating term and A^2 term.

We see that the tuning of coupling strength by changing the number of Py strips enables the modulation of the Δf_{BS} and Δf_{A^2} . Figures 5-11 show the comparisons of (a) Δf_{BS} and (b) Δf_{A^2} as a function of magnetic field depending on the prepared YBCO resonators with the different number of Py strips. It should be noted that, as for the resonator comprising a multitude of Py strips, specifically in the context of strong coupling situated in proximity to ultrastrong coupling, the magnitude of both Δf_{BS} and Δf_{A^2} is observed to be markedly larger. As the perturbative energy of the counter-rotating term and

the A^2 term in the coupling system is reflected in both Δf_{BS} and Δf_{A^2} , the ratios of the energy against the resonance frequency $\Delta f_{\text{BS}}/f_p$ and $\Delta f_{A^2}/f_p$ are approximately 1% and 3%, respectively. With regard to the counter-rotating term, this result shows the non-conserved excitation of magnons and photons with virtual excitation represents a 1% energy communication in total systems, whilst the counter-rotating process in the resonator with the single stripe is negligibly small ($\sim 0.04\%$). The A^2 term is derived from minimal coupling replacement in the presence of charge in the AC electromagnetic field [42,95,96]. Consequently, the potential sources of the A^2 term in this system can be classified as follows: (a) electrons in metallic ferromagnetic films under microwave photon mode and (b) Cooper pairs in YBCO resonators under AC electromagnetic field. It can be deduced that this considerable A^2 shift may be attributed to these two components.

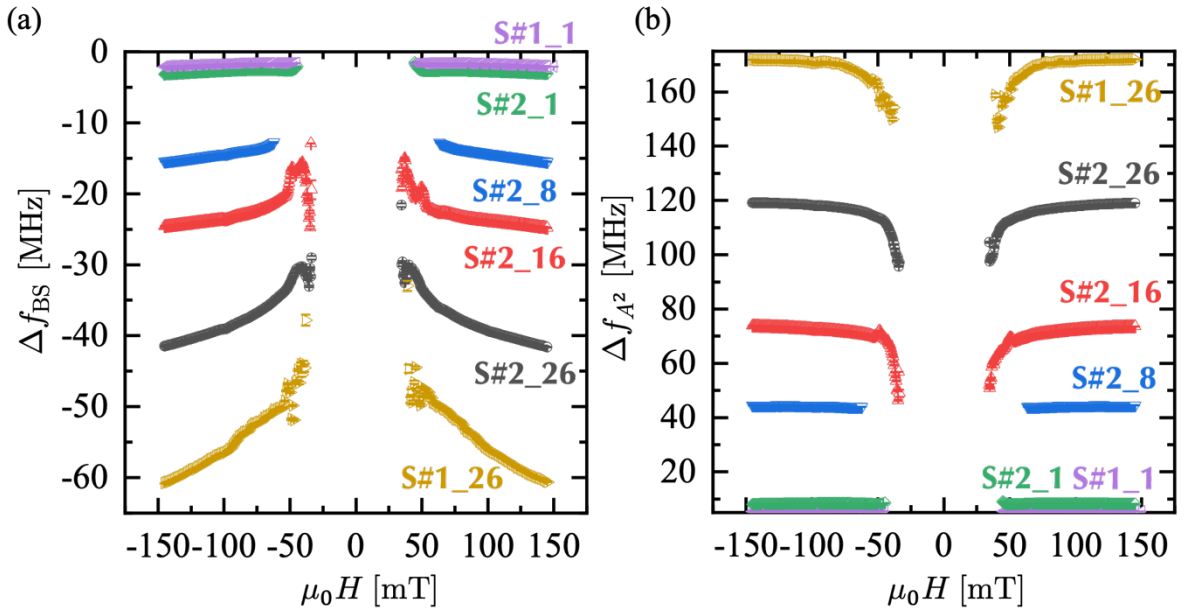


Figure 5-11. The comparison of the magnetic field dependence of (a) Δf_{BS} and (b) Δf_{A^2} for the different resonators with various number of Py stripes, where for instance S#1_1 means the resonator with 1 Py strip in YBCO-S#1 chip.

5.5.2. Coupling strength dependence of Bloch-Siegert Shift

The lowest order-term of Bloch-Siegert Shift is theoretically derived to be the order of $g_{\text{eff}}^2/\omega_p$ [1,2], i.e., the square of the normalized coupling strength, ϵ^2 . Figure 5-12 shows the Δf_{BS} as a function of the square of normalized coupling strength, ϵ^2 at 60 mT, 100 mT and 140 mT, showing the excellent agreement with the linear fitting. We obtain the following relationship of the energy ratio of Bloch-Siegert Shift, $\Delta f_{\text{BS}}/f_p$:

$$\frac{\Delta f_{\text{BS}}}{f_p} = C \frac{\epsilon^2}{f_p} = C \left(\frac{g_{\text{eff}}/2\pi}{f_p} \right)^2. \quad (98)$$

Here, the C is the coefficient of ϵ^2 , which are estimated to be -0.617 ± 0.012 (65 mT), -0.655 ± 0.006 (100 mT), and -0.694 ± 0.005 (140 mT). These results experimentally

demonstrate the Bloch-Siegert Shift follows the theoretical prediction to be proportional to the square of the normalized coupling strength ϵ^2 . Furthermore, this analyzation also demonstrates the energy ratio of Δf_{BS} against the photon resonance f_p is proportional to the square of $\eta = g_{\text{eff}}/2\pi f_p$, which has been thought to be the historically conventional indicator for determining the ultrastrong coupling regime [42]. However, when Eq. (98) is considered, the conventional criteria for determining the ultrastrong coupling regime, $\eta > 0.1$, is the border of $\Delta f_{\text{BS}}/f_p > 0.01$. This implies the energy ratio of the counter-rotating term process in the coupling system start to be non-negligible as theoretically predicted [211]. Thus, this result experimentally verifies the rotating wave approximation to ignore the counter-rotating term in magnon polariton system.

It is worth noting that the ϵ^2 law of Δf_{BS} is valid in the very dissipative magnon system ($\kappa_m/2\pi = 461 \text{ MHz} \sim g_{\text{eff}}/2\pi$), supporting the theoretical work that the virtual excitation is protected in the dissipative system [93].

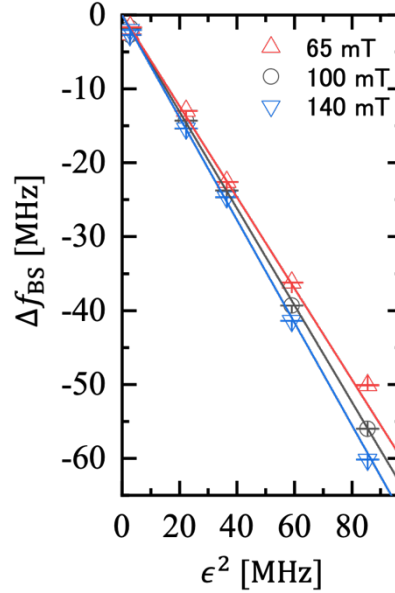


Figure 5-12. The Δf_{BS} as a function of the square of normalized coupling strength, ϵ^2 for the prepared resonators at certain magnetic field. The plots include the data of 65 mT (red triangle), 100 mT (black circle) and 140 mT (blue triangle).

5.5.3. Upper branch of the dispersion relation for magnon polariton in this system

Before summarizing the chapter, this section introduces the deviation of the polariton frequency by considering the counter-rotating term and the A^2 term. As previously discussed, since the magnon mode is a significant dissipative system that governs the Purcell effect. Consequently, the spectra for the upper branch of magnon polariton are markedly suppressed due to the intensification of photon loss via the magnon relaxation. As a consequence of the Purcell effect, the enhancement of photon loss is proportional to the cooperativity factor $g_{\text{eff}}^2/\kappa_p\kappa_m$ [205], which makes the upper branch to be more challenging

to observe in the context of stronger coupling. However, enhancing the photon resonance peak can be one solution of this challenge. According to Table. 4-5, the coupling between resonators and a feedline in YBCO chip is under-coupling ($Q_{\text{int}} < Q_{\text{ext}}$), which suppresses the resonance peak [154]. Decreasing the distance between resonators and a feedline, the author investigated the upper branch of magnon polaritons for the R1 with 26 Py strips in YBCO-S#2. Figure 5-13 show the clear deviation between Eqs. (71-73), which is the evidence of breakdown of rotating wave approximation.

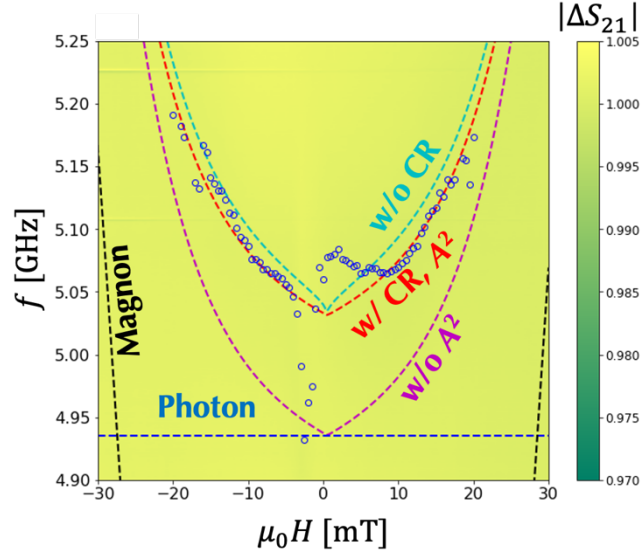


Figure 5-13. The $|\Delta S_{21}|$ as a function of the external magnetic field for the R1 with 26 Py stripes in YBCO-S#1. Here, the broken red, purple and green lines show the numerical Eq. (71) for the full Hamiltonian, Eq. (73) for Dicke Hamiltonian and Eq. (72) for the Hamiltonian without counter-rotating term. The parameters used for the numerical calculation are estimated from Eq. (71).

5.6 Summary

In summary, the author demonstrated the ultrastrong coupling regime of magnon polaritons in 30 nm-thick Py stripes by using the YBCO resonators, which allows for the observation of breakdown of rotating wave approximation in the magnon polariton system. The author also investigated the Bloch-Siegert shift and A^2 shift in the magnon polaritons in thin Py stripes on YBCO resonators. Modulating magnon volume coupled with each Py stripe via the uniform photon mode, the coupling strength is tuned, leading to the manipulating the magnitude of Bloch-Siegert Shift and A^2 shift, providing the crucial information of the ratio of counter-rotating term in the system with the dependence of $g_{\text{eff}}^2/\omega_p$. These findings would allow for the further understanding of the phenomena and physics for the magnon polaritons in thin ferromagnetic films under ultrastrong coupling regime. From the point of quantum hybrid systems, the magnon polariton is the quasiparticle with the property of both magnons and photons. Thus, the author hopes that many kinds

of magnonic features such as non-reciprocity, tunability, and nonlinearity, will be integrated into the physics of magnon polaritons under the ultrastrong coupling regime.

Chapter 6. Conclusion

In this dissertation, the author studied the magnon state in thin ferromagnetic films with microwave interaction in thin ferromagnetic films in broad range of coupling strength g_{eff} . Depending on the g_{eff} between magnon and photon mode, the coupling state indicates completely different physics. When the g_{eff} is under the damping parameters, the coupling state is called weak coupling state, where the non-coherent coupling from photon to magnon induce ferromagnetic resonance in ferromagnet. The ferromagnetic resonance is pivotal tool to investigate the magnetic damping in ferromagnets, providing the crucial information of magnon damping aiming for the design of coupling systems. After the g_{eff} exceeds the damping parameters of magnons and photons, the state enters strong coupling regime, where the magnon and photon mode coherently couples with each other. Furthermore, when g_{eff} is comparable with the resonance frequency, the rotating wave approximation breaks down and the counter-rotating process is not negligible, inducing the squeezing and entanglement of magnons and photons at the ground state. The YBCO resonator system represents a promising photon system, exhibiting robust behavior with respect to both temperature and magnetic field, which is conducive to the observation of phenomena in the ultrastrong coupling regime. The tunability of g_{eff} in this YBCO system with FM stripes provides valuable insight into the behaviour of magnon polaritons in the presence of a counter-rotating term.

The author summarizes conclusions in this dissertation as below:

Control of magnon damping in ultrathin ferromagnetic films (Chapter 3)

- The author demonstrates the control of two-magnon scattering, magnon scattering to magnon mode with the different wave number, by modulating the surface magnetic anisotropy in ultrathin ferromagnetic films (< 2 nm).
- This chapter produces the crucial guiding principle for designing of magnon damping in ultrathin ferromagnetic films.

Superconducting resonator made of high- T_c YBCO (Chapter 4)

- The superconducting resonators of YBCO with the strong robustness against the magnetic field and temperature were achieved, which allows for the promising platform to observe the magnon polariton with strong coupling strength.
- The high-quality factor (~ 4000) of uniform photon mode is obtained in the lumped element structure of YBCO resonator.

Magnon polaritons in thin ferromagnetic films (Chapter 5)

- The coupling strength between the YBCO resonators and 30 nm-thick Py films can be enhanced by increasing the number of Py strips n , which are coupled with each other strips on the resonators. The scaling law of $\sqrt{n}$ is confirmed in this system.
- The ultrastrong coupling of magnon polaritons in 30 nm-thick Py films are realized, demonstrating the Bloch-Siegert Shift due to the counter-rotating term, the symbolic phenomena in ultrastrong coupling regime.
- The Bloch-Siegert Shift in this system can be tuned by changing the number of Py strips, and the theoretical prediction that Bloch-Siegert Shift is proportional to the $\epsilon^2 \propto g_{\text{eff}}^2/\omega_p$ is experimentally confirmed.
- The coupling system with a large dissipation governs the ϵ^2 law for the Bloch-Siegert Shift, implying the counter-rotating process is protected from the dissipation as theoretically suggested in ref. [1].

Appendix

A. Derivation of the photon mode linewidth with the coupling FMR mode

In input-output theory [206], the operators that express the magnon mode, denoted by a , and the resonator mode, denoted by b , which couple with each other, are given by:

$$\frac{da}{dt} = -i\omega_c a - \frac{\kappa_{i0} + \kappa_e}{2} a - igb - \sqrt{\kappa_e} a_{\text{in}}, \quad (\text{A1})$$

$$\frac{db}{dt} = -i\omega_b b - \frac{\gamma_m}{2} b - iga, \quad (\text{A2})$$

where ω_c and ω_b are the microwave angular frequency and the magnon angular frequency, respectively. In addition, g , γ_m , κ_{i0} , κ_e , and a_{in} denote the coupling coefficient of the magnon mode and the resonator mode, the magnon damping coefficient, the microwave photon damping coefficient, the transmission line decay, and the microwave signal input operators, respectively. The operator expression rotating with angular frequencies, $a = \tilde{a}e^{-i\omega t}$ and $b = \tilde{b}e^{-i\omega_b t}$, are substituted into Eqs. (A1) and (A2),

$$\left[-i(\omega - \omega_c) + \frac{\kappa_{i0} + \kappa_e}{2} \right] \tilde{a} = -ig\tilde{b} - \sqrt{\kappa_e} \tilde{a}_{\text{in}}, \quad (\text{A3})$$

$$\left[-i(\omega - \omega_b) + \frac{\gamma_m}{2} \right] \tilde{b} = -ig\tilde{a}. \quad (\text{A4})$$

We solve Eq. (A4) for $\tilde{b}$ and substitute the result into (A3), which gives us the equation for the microwave input operator and the operator of the resonator mode. By defining the susceptibility as χ , the result can be calculated using the following equation:

$$\chi \tilde{a} = -\sqrt{\kappa_e} \tilde{a}_{\text{in}}, \quad (\text{A5})$$

$$\chi = \left[-i(\omega - \omega_c) + \frac{\kappa_{i0} + \kappa_e}{2} + \frac{g^2}{-i(\omega - \omega_b) + \frac{\gamma_m}{2}} \right], \quad (\text{A6})$$

Around $\omega = \omega_c$, the real part of χ represents the HWHM of $|S_{11}|$, and then the damping coefficient of the resonator mode coupled with the magnon mode can be expressed as κ_i :

$$\kappa_i = 2\text{Re}(\chi) = \kappa_{i0} + \gamma_m \frac{g^2}{(\omega_c - \omega_b)^2 + (\frac{\gamma_m}{2})^2}. \quad (\text{A7})$$

B. Derivation of coupling strength between single spin in ferromagnet and photon mode

The Hamiltonian of the magnon-photon interaction is expressed as the following product of magnetic z-component $\hat{B}_z$ due to the microwave exciting photon mode $\text{TE}_{01\delta}$ in the resonator and the z-component of the magnetic moment $\hat{\mu}_z$ in ferromagnet,

$$H = \hat{B}_z \hat{\mu}_z. \quad (\text{B1})$$

Here, the z direction is defined along the direction of the ac magnetic field. The ac magnetic field generated by the microwave in the resonator is represented by the following equation,

$$\hat{B}_z = B_{\text{zpf}}(\hat{a} + \hat{a}^\dagger), \quad (\text{B2})$$

where the B_{zpf} , $\hat{a}$ and $\hat{a}^\dagger$ are zero-point fluctuation of the magnetic field in the resonator, creation, and annihilation operators of the photon, respectively.

The magnetic moment in the ferromagnet is expressed as the summation of the contribution of each magnetic spin, $\hat{S}_i$, as shown in the following equation, $\hat{\mu}_x = g\mu_B \sum \hat{S}_i$. Each magnetic spin operator of $\hat{S}_i$ in ferromagnet is expressed by the Holstein-Primakoff transformation as follows,

$$\hat{S}_i^+ = \sqrt{2S} \left(1 - \frac{\hat{b}_i^\dagger \hat{b}_i}{2S} \right)^{\frac{1}{2}} \hat{b}_i, \quad (\text{B3})$$

$$\hat{b}_i = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} e^{-i\mathbf{k} \cdot \mathbf{R}_i} \hat{b}_{\mathbf{k}}, \quad (\text{B4})$$

where $\hat{b}_i$ and $\hat{b}_{\mathbf{k}}$ are bosonic operators expressing magnon excitation in the real space and in the wavevector space, respectively. The S and N are the number of spins in each atom and that of atoms in the ferromagnet, respectively. The excitation numbers of magnon, $\hat{b}_i^\dagger \hat{b}_i$, are assumed to be sufficiently small and the first order of $\hat{b}_{\mathbf{k}}$ is remained in Eq. (B3). In addition, since only the $\mathbf{k} = 0$ magnon mode is excited with uniform ac magnetic field, Eq. (B3) reduces to the following equation,

$$\hat{S}_i^+ = \sqrt{2S/N} \hat{b}. \quad (\text{B5})$$

Here, the magnon mode at the $\mathbf{k} = 0$, $\hat{b}_0$, is defined to be the $\hat{b}$ and each magnetic spin operator is considered to be the same. Thus, the magnetic moment of the $\mathbf{k} = 0$ magnon mode, $\hat{\mu}_x$, is written as,

$$\begin{aligned} \hat{\mu}_x &= g\mu_B N \hat{S}_{ix} \\ &= \frac{g\mu_B N}{2} (\hat{S}_i^+ + \hat{S}_i^-) \\ &= \frac{g\mu_B}{2} \sqrt{\frac{NS}{2}} (\hat{b} + \hat{b}^\dagger). \end{aligned} \quad (\text{B6})$$

Finally, Eqs. (B2) and (B6) were substituted into Eq. (B1) and the magnon-photon coupling Hamiltonian was obtained by applying the rotating wave approximation,

$$\hat{H} = \frac{g\mu_B B_{\text{zpf}}}{\sqrt{2}} \sqrt{NS} (\hat{a}^\dagger \hat{b} + \hat{a} \hat{b}^\dagger) = \hbar g_{0,\text{Co}} \sqrt{NS} (\hat{a}^\dagger \hat{b} + \hat{a} \hat{b}^\dagger), \quad (\text{B7})$$

where $g_{0,\text{Co}}$ is defined as the coupling strength between single spin in ferromagnet and photon mode and described as,

$$g_{0,\text{Co}} = \frac{g\mu_B B_{\text{zpf}}}{\sqrt{2\hbar}}. \quad (\text{B8})$$

C. Absorption curve of empty rutile resonator at room temperature

While the quality factor of 10^6 was realized under very low temperature of a few kelvins as described in Ref., our measurement temperature is room temperature, which is the reason of the deviation of quality factors between in the previous report and our study. The $|S_{11}|$ of the rutile resonator without a sample at room temperature in our study is shown in Fig. S1. From the model fitting of the $|S_{11}|$ by using Eq. (5) in the main text, f_c , κ_e and κ_i are estimated to be 5.62 GHz, 0.53 MHz and 0.51 MHz, respectively, which are comparable to photon loss in the 3D Cu resonator conventionally used at a few kelvins [25,212]. Here, the quality factor (Q) of the rutile resonator at room temperature is calculated by the equation, $Q = f_c/\kappa_i = 1.06 \times 10^4$. Therefore, this quality factor of the rutile resonator we measured is sufficiently high at room temperature.

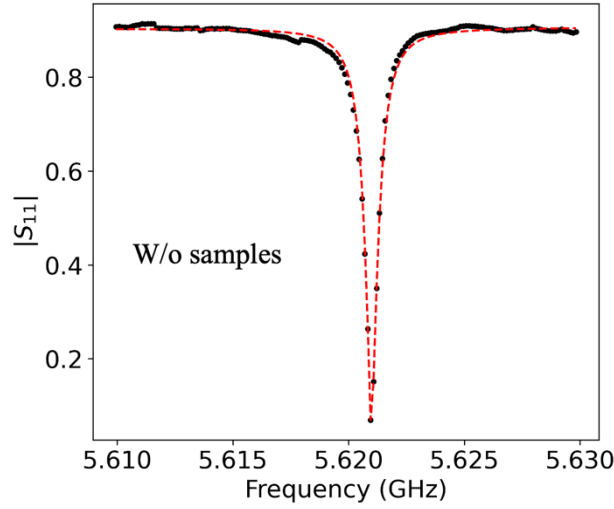


Figure C1. The absolute value of S_{11} of the empty rutile resonator without any samples.

D. The evaluation of estimated effective uniaxial magnetic anisotropy

The Eq. (3) is solved about $\mu_0 H_{\text{res}}$ and the following equation is obtained,

$$\mu_0 H_{\text{res}} = \frac{1}{2 \cos(\Theta_H - \Theta)} \left\{ h_{\text{eff}} (\sin^2 \Theta - \cos 2\Theta) + \sqrt{h_{\text{eff}}^2 \cos^4 \Theta + 4 \left(\frac{\omega}{\gamma} \right)^2} \right\}. \quad (\text{D1})$$

The Θ_H dependence of magnetization angle Θ can be found from the Θ_H angular dependence of FMR resonance fields. So, we can determine the values of $\mu_0 H_{\text{res}}$, Θ_H and Θ when a h_{eff} is fixed. Then, the calculated resonance fields, $\mu_0 H$, in Eq. (S2) can be obtained for certain h_{eff} . The square sum, Δ , of the difference between $\mu_0 H_{\text{res}}$ and $\mu_0 H_{\text{cal}}$ can be calculated by the following equation with weight,

$$\Delta = \sum \frac{(\mu_0 H_{\text{res}} - \mu_0 H_{\text{cal}})^2}{|\mu_0 H_{\text{cal}}|}, \quad (\text{D2})$$

where $|\mu_0 H_{\text{cal}}|$ is the weight for the optimization of h_{eff} , which is because the change of $\mu_0 H_{\text{cal}}$ is relatively large around $\Theta_H = 90^\circ$. Finally, we can determine the optimal h_{eff} to

make Δ the smallest, where the optimal h_{eff} is the estimated effective magnetic anisotropy field.

To underscore the validity of our estimation of h_{eff} , the fitting lines of $h_{\text{eff}} = -1.83 \times 10^3$ mT (the estimated value) and, $h_{\text{eff}} = -1.5, -1.7, -2.0$ and -2.2×10^3 mT (for comparison) for the Co (10)/SiO₂ sample are shown in Fig. S3. The deviation of the fitting line for the experimental data becomes more discernable around $\Theta_H = 0^\circ$. From these results, we concluded that $h_{\text{eff}} = -1.83 \times 10^3$ mT is the most precise value in the estimation.

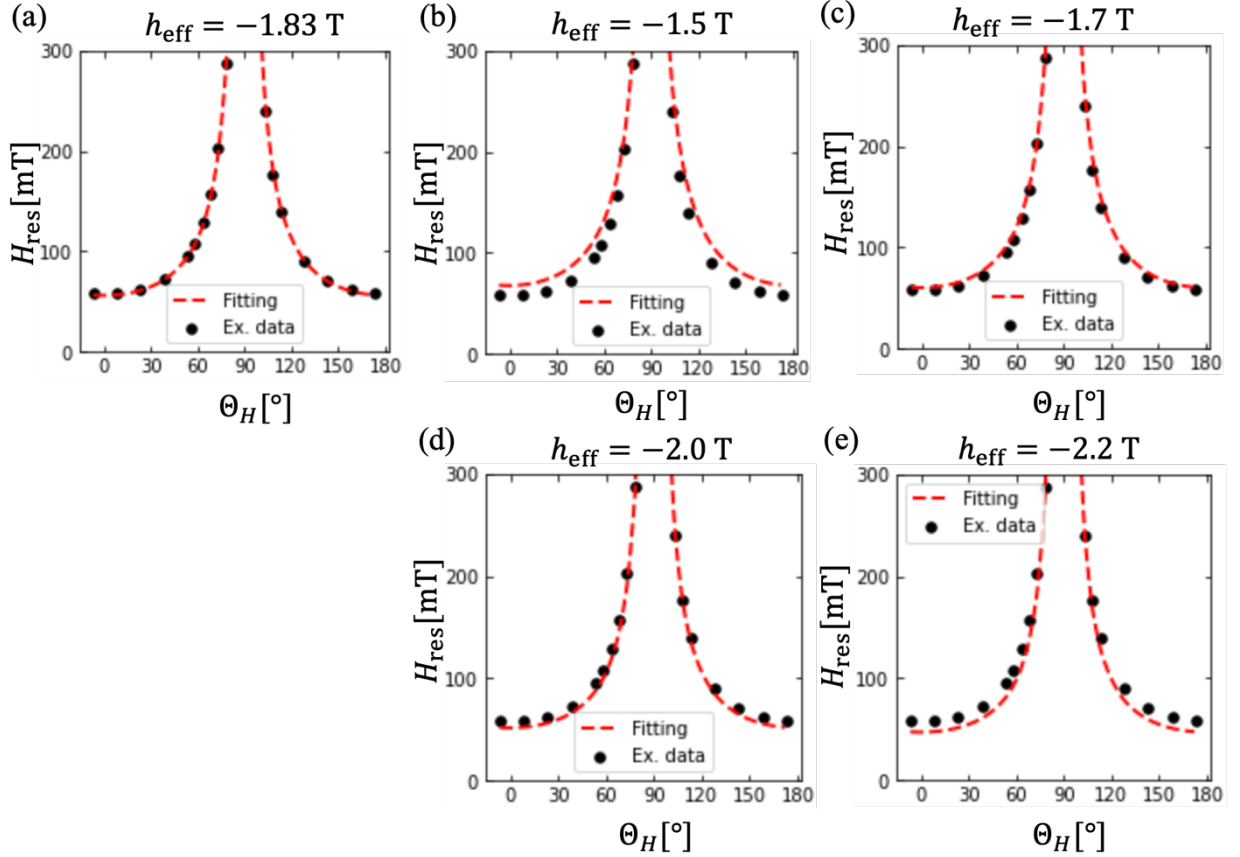


Figure D1. Angular dependence of the resonance fields of the MgO/Co (10)/SiO₂ sample. The broken red lines represent the fitting lines when (a) $h_{\text{eff}} = -1.83 \times 10^3$ mT (the estimated value), (b) $h_{\text{eff}} = -1.5 \times 10^3$ mT, (c) $h_{\text{eff}} = -1.7 \times 10^3$ mT, (d) $h_{\text{eff}} = -2.0 \times 10^3$ mT and (e) $h_{\text{eff}} = -2.2 \times 10^3$ mT.

E. Correlation between TMS contribution and SMA field of samples with Co/SiO₂ and Co/Ta deposited by sputtering systems

In our previous study [107], the Ta seeding layer beneath 1-nm-thick Co exhibited sizable suppression of the Gilbert damping constant. Although Ta is not grown in the EB deposition system due to restriction of material growth setups, we fabricated a SiO₂/Co/Ta sample by sputtering and examine the TMS contribution and uniaxial SMA field of the SiO₂/Co/Ta samples based on our previous study. We prepared the samples with SiO₂(10 nm)/Co (t_{Co})/SiO₂-substrate and SiO₂(10 nm)/Co(t_{Co})/Ta(3 nm)/SiO₂-substrate. The Co thickness, t_{Co} , ranged from 2 nm to 20 nm as described in the main text. The Co thickness

dependence of the Gilbert damping constants is shown in Fig. S3(a), which was obtained by the same manner as shown in the main text. The TMS contribution, β_{TMS} , of prepared samples were estimated to be $0.48 \pm 0.12 \text{ nm}^2$ (SiO_2) and $0.05 \pm 0.13 \text{ nm}^2$ (Ta), where the value of the sample with Co/Ta interface is comparable to the β_{TMS} of MgO/Co/Ti. Therefore, the insertion of Ta buffer layer suppresses the TMS contribution and help ultrathin ferromagnetic film realize low-magnon damping, which is consistent with the previous report [107]. The Co thickness dependence of the effective uniaxial magnetic anisotropy field was obtained as shown in Fig. S4(b). From these results, the uniaxial SMA fields, h_{S_u} , of the samples of SiO_2 (10 nm)/Co (t_{Co})/ SiO_2 -substrate and SiO_2 (10 nm)/Co(t_{Co})/Ta(3 nm)/ SiO_2 -substrate were estimated to be $1.2 \pm 0.1 \text{ T} \cdot \text{nm}$ (SiO_2) and $0.26 \pm 0.50 \text{ T} \cdot \text{nm}$ (Ta), respectively, which signifies the insertion of Ta buffer layer suppresses the uniaxial SMA field.

Correlation between the TMS contribution and the square of the uniaxial SMA field is shown in Figs. S5(a) and (b). The results of the SiO_2 (10 nm)/Co(t_{Co})/ SiO_2 -substrate and SiO_2 (10 nm)/Co (t_{Co})/Ta (3 nm)/ SiO_2 -substrate are nicely fitted by linear relationship, where the SiO_2 /Co/Ta exhibits quite small TMS contribution that is comparable to that of SiO_2 /MgO/Co/Ti. Furthermore, the β_{TMS} decreases with increasing the surface energy density of the NM materials as shown in Figs. S5 (c) and (d). This tendency is consistent with the guiding principle obtained in the present study.

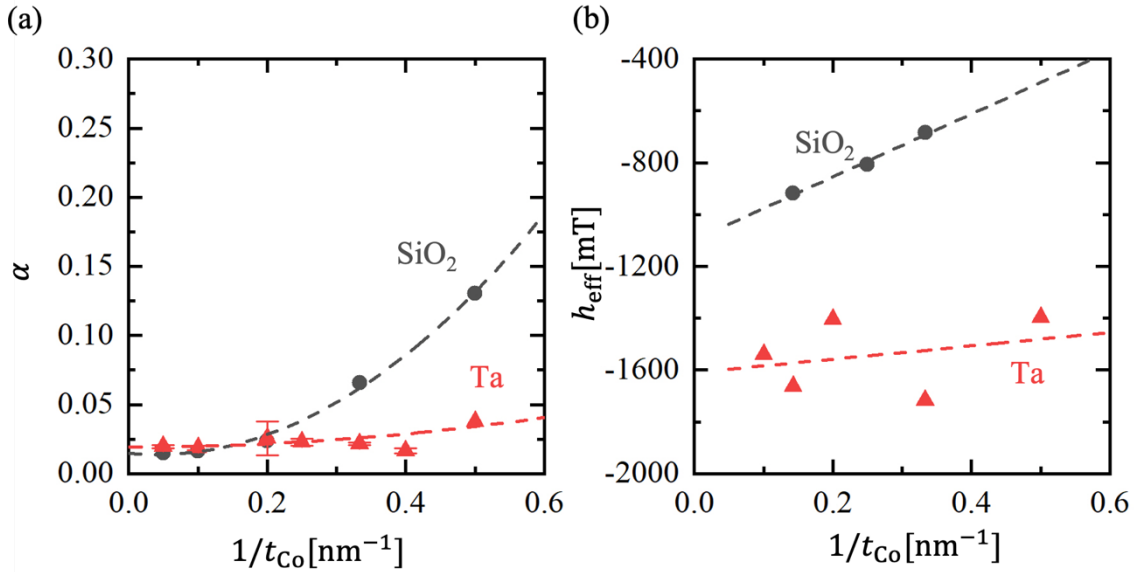


Figure E-1. The Co thickness dependence of (a) Gilbert damping constant and (b) effective magnetic anisotropy field of samples with SiO_2 (10 nm)/Co(t_{Co})/ SiO_2 substrate and SiO_2 (10 nm)/Co(t_{Co})/Ta(3 nm)/ SiO_2 substrate.

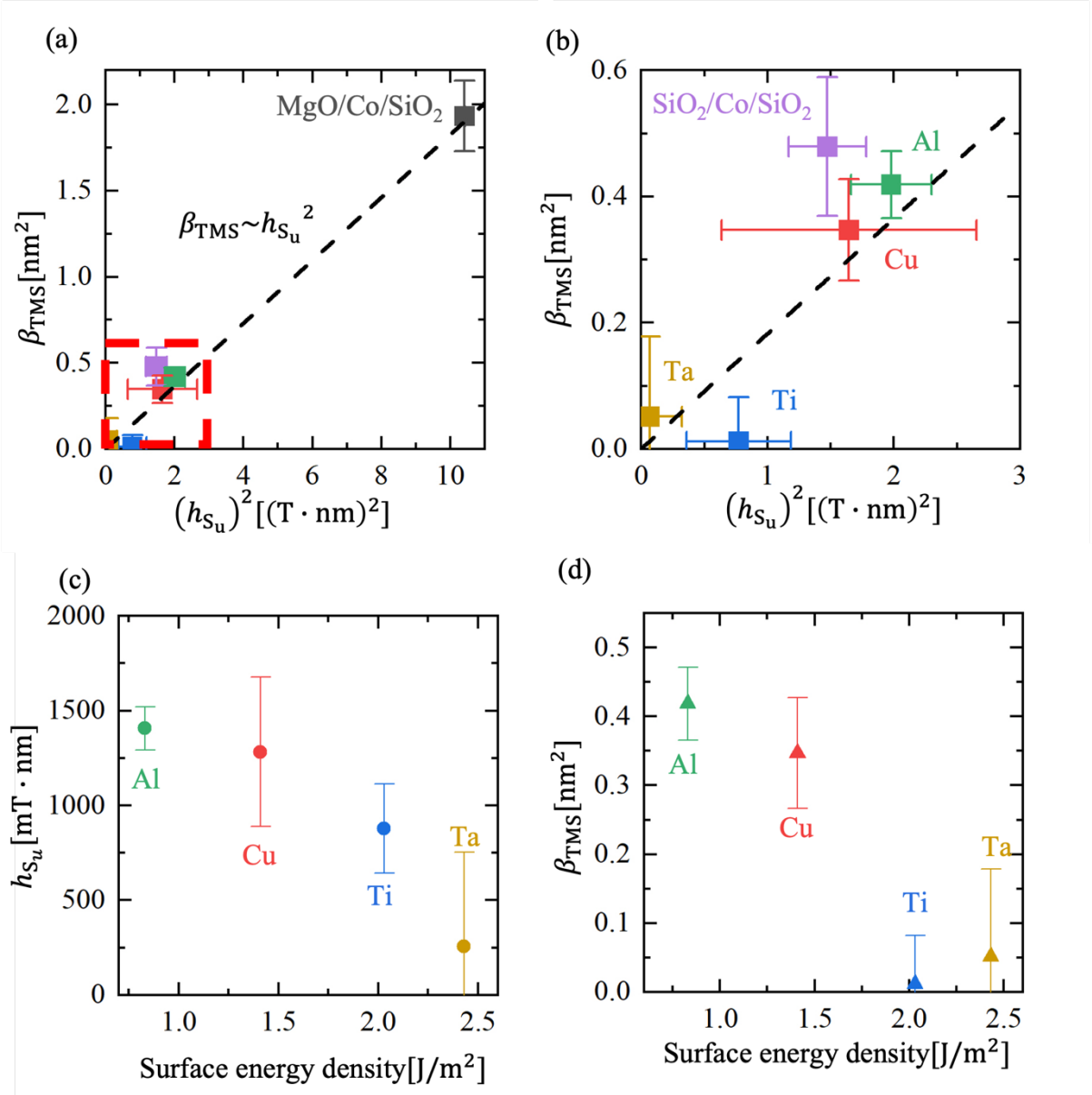


Figure E-2. Correlation diagram between TMS coefficient β_{TMS} and the square of the SMA field $(h_{\text{su}})^2$ for (a) all prepared samples. (b) The enlarged figure in the red broken square. Surface energy density dependence of the NM materials selected as the seeding layers on (c) β_{TMS} and (d) h_{su} .

F. Simulation of the amplitude of ac magnetic field excited in rutile resonator

The schematic image of the rutile resonator used in this study and the amplitude of the ac magnetic field calculated in the rutile resonator by using RF module of COMSOL 5.6 are shown in Fig. S7, and the ac magnetic field is almost uniform at the position the sample was placed.

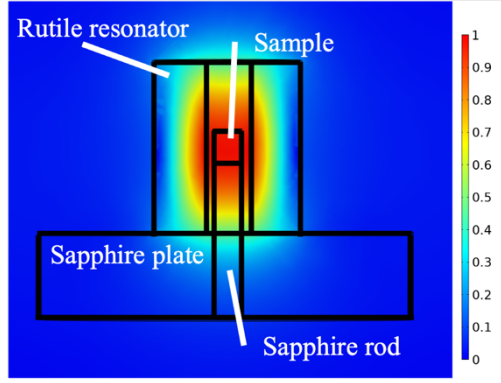


Figure F-1. The schematic image of rutile resonator and the amplitude ac magnetic field calculated by COMSOL. The side color bar shows the normalized amplitude of ac magnetic field originated from $TE_{01\delta}$ photon mode.

G. Simulation of the amplitude of ac magnetic field excited in rutile resonator

In our setup for measuring broadband ferromagnetic resonance (bbFMR), the prepared samples are positioned face-down on a coplanar waveguide (CPW). FMR of the samples is excited using an AC magnetic field generated by an AC electric current flowing through the signal line of the CPW. When microwave radiation interacts with the ferromagnetic layer in the samples, it excites magnetization dynamics, which lead to changes in the transmission signal during the FMR measurements. Specifically, when an external magnetic field $\mu_0 H$ is applied along the in-plane direction, the scattering parameter measured from port 1 to port 2 of the VNA S_{21} can be calculated using the following equation [119,213,214]:

$$S_{21}(\mu_0 H) = C_0 + C_1 \cdot \mu_0 H - iC_3 \frac{\chi_{yy}(\omega, \mu_0 H) + \chi_{zz}(\omega, \mu_0 H)}{\mu_0 M_S}, \quad (G1)$$

where $C_0 + C_1 \cdot \mu_0 H$, C_3 , $\mu_0 M_S$, and $\chi_{yy(zz)}(\omega, \mu_0 H)$ are parameters that account for the background contribution from the measurement setup, complex amplitude including phase information, saturation magnetization, and diagonal susceptibilities components to the ferromagnetic response, respectively. By analyzing the changes in S_{21} and the transmission signal, we can obtain

$$\Delta S_{21} = -iC_3 \frac{\chi_{yy}(\omega, \mu_0 H) + \chi_{zz}(\omega, \mu_0 H)}{\mu_0 M_S}, \quad (G2)$$

which is used for fitting the bbFMR data to determine the resonance field and the half-width at half-maximum (HWHM) for a fixed frequency ω . In the following paragraphs, we calculate the susceptibilities $\chi_{xx(yy)}$ by utilizing the Landau–Lifshitz–Gilbert (LLG) equation

$$\frac{d\mathbf{M}}{dt} = -\gamma \mathbf{M} \times \mu_0 \mathbf{H}_{\text{eff}} + \frac{\alpha}{M_S} \mathbf{M} \times \frac{d\mathbf{M}}{dt}, \quad (G3)$$

where γ and α are the gyromagnetic ratio and Gilbert damping constant, respectively. $\mathbf{H}_{\text{eff}}$ is the effective magnetic field that the magnetization experiences. $\mathbf{M} \times \frac{d\mathbf{M}}{dt}$ is directed toward the equilibrium orientation of the magnetization and the magnitude of Gilbert

damping constant α determines how fast the magnetization excitation relaxes to the equilibrium state.

When the external AC magnetic field is applied to the samples, $\mathbf{H}_{\text{eff}}$ and $\mathbf{M}$ are expressed using an oscillatory field $\mathbf{h}_{\text{rf}}(t)$ and magnetization $\mathbf{m}(t)$ as follows:

$$\mathbf{H}_{\text{eff}} = \mathbf{H} - \mathbf{H}_{\text{de}} + \mathbf{h}_{\text{rf}}(t) = (H - N_x M_s) \mathbf{e}_x + \mathbf{h}_{\text{rf}}(t), \quad (\text{G4})$$

$$\mathbf{M} = \mathbf{M}_s + \mathbf{m}(t) = M_s \mathbf{e}_x + \mathbf{m}(t). \quad (\text{G5})$$

Here, $\mathbf{H}$ is the static magnetic field. In addition, we introduce the demagnetization field $\mathbf{H}_{\text{de}}$ in (S4), which accounts for the thin-film geometry. In particular, we parametrize the demagnetization tensor $\mathbf{H}_{\text{de}} = N_x M_x \mathbf{e}_x + N_y M_y \mathbf{e}_y + N_z M_z \mathbf{e}_z$ [58] using N_x , N_y , N_z .

Using the following ansatz for $\mathbf{h}_{\text{rf}}(t)$ and $\mathbf{m}(t)$

$$\mathbf{h}_{\text{rf}}(t) = (h_{\text{rf},y} \mathbf{e}_y + h_{\text{rf},z} \mathbf{e}_z) \cdot e^{i\omega t}, \quad (\text{G6})$$

$$\mathbf{m}(t) = (m_y \mathbf{e}_y + m_z \mathbf{e}_z) \cdot e^{i\omega t}. \quad (\text{G7})$$

and substituting Eqs. (G4) and (G5) into the LLG equation (G3), we can rewrite the dynamical part as follows:

$$\begin{pmatrix} m_y \\ m_z \end{pmatrix} = \hat{\chi}_P \begin{pmatrix} h_{\text{rf},y} \\ h_{\text{rf},z} \end{pmatrix}. \quad (\text{G8})$$

Here, we have introduced the Polder-susceptibility

$$\hat{\chi}_P = \frac{\mu_0 M_s}{\text{Det}(\hat{A})} \hat{A}, \quad (\text{G9})$$

with

$$\hat{A} = \begin{pmatrix} H + M_s \cdot (N_y - N_z) + \frac{i\omega\alpha}{\gamma\mu_0} & \frac{i\omega}{\gamma\mu_0} \\ -\frac{i\omega}{\gamma\mu_0} & H + M_s \cdot (N_x - N_z) + \frac{i\omega\alpha}{\gamma\mu_0} \end{pmatrix}. \quad (\text{G10})$$

In a thin film satisfying the condition $[N_x = 1, N_z = N_y = 0]$ (see also the coordinate system presented in Fig. 1 (a) in the main text), the susceptibility takes the following form:

$$\hat{\chi}_P = \begin{pmatrix} \chi_{yy} & \chi_{yz} \\ \chi_{zy} & \chi_{zz} \end{pmatrix} = \frac{\mu_0 M_s}{D} \begin{pmatrix} H + i\frac{\Delta H}{2} & \frac{i\omega}{\gamma\mu_0} \\ -\frac{i\omega}{\gamma\mu_0} & H + M_s + i\frac{\Delta H}{2} \end{pmatrix}. \quad (\text{G11})$$

$$D = \left(\mu_0 H + \mu_0 M_s + i\frac{\mu_0 \Delta H}{2} \right) \left(\mu_0 H + i\frac{\mu_0 \Delta H}{2} \right) - \left(\frac{\omega}{\gamma} \right)^2, \quad (\text{G12})$$

From this derivation, we obtain the following equations for the fitting of the Polder susceptibility,

$$\Delta S_{21} = -i \frac{C_3}{\mu_0 M_s} \{ \chi_{yy}(\mu_0 H) + \chi_{zz}(\mu_0 H) \}, \quad (\text{G13})$$

$$\chi_{yy}(\mu_0 H) = \frac{\mu_0 M_s}{D} \left(\mu_0 H + i\frac{\mu_0 \Delta H}{2} \right), \quad (\text{G14})$$

$$\chi_{zz}(\mu_0 H) = \frac{\mu_0 M_s}{D} \left(\mu_0 H + \mu_0 M_s + i\frac{\mu_0 \Delta H}{2} \right). \quad (\text{G15})$$

H. Co₂₅Fe₇₅ thickness dependences of inhomogeneous linewidth

The inhomogeneous linewidth of the prepared samples can be estimated by fitting the microwave frequency dependence of the HWHM using the equation $\mu_0\Delta H = \mu_0\Delta H_0 + \frac{\alpha}{\gamma}\omega$, where $\mu_0\Delta H$ is the HWHM, α is the Gilbert damping constant, γ is the gyromagnetic ratio, and $\omega = 2\pi f$ is the angular frequency of the microwave excitation field. The inhomogeneous linewidth is influenced by various factors such as applied field, magnetization, surface anisotropy, surface roughness, and magneto-crystalline anisotropy of the ferromagnetic films [62].

The inhomogeneous linewidth $\mu_0\Delta H_0$ increases with decreasing Co₂₅Fe₇₅ thickness, as shown in Fig. S2, suggesting that surface inhomogeneity is dominant in the Co₂₅Fe₇₅ film. Note that, whereas significant MgO thickness dependence of β_{TMS} exists, there is almost no impact of the MgO thickness dependence on the inhomogeneous linewidth $\mu_0\Delta H_0$, which implies that the surface magnetic anisotropy field dominantly impacts the Gilbert damping α .

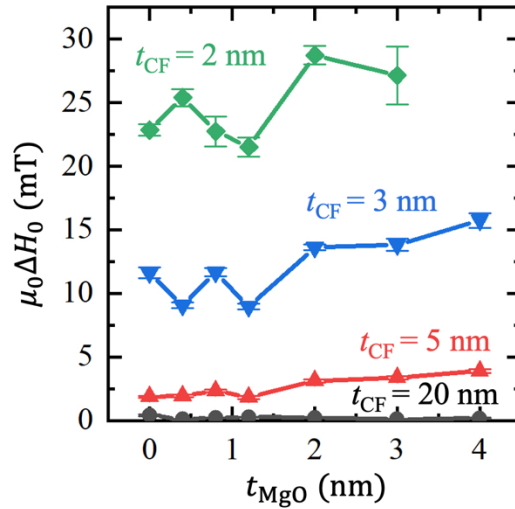


Figure H-1. MgO thickness dependence of inhomogeneous linewidth in FMR signals for fabricated samples with different Co₂₅Fe₇₅ layer thickness.

I. Surface roughness of the Co₂₅Fe₇₅ films

As mentioned in the main text, TMS is affected by the surface state of FM layer such as the surface magnetic anisotropy and the surface roughness. The author investigated the top surface state of the sample with the stack of TaO_x/Co₂₅Fe₇₅(10)/Cu/Ta and TaO_x/MgO (2)/Co₂₅Fe₇₅ (10)/Cu/Ta via atomic force microscopy (AFM) experiments to indirectly address the surface state of Co₂₅Fe₇₅. The root mean square (RMS) roughness of these AFM images is estimated to be 645.8 pm (TaO_x/Co₂₅Fe₇₅) and 759.0 pm (TaO_x/MgO(2)/Co₂₅Fe₇₅).

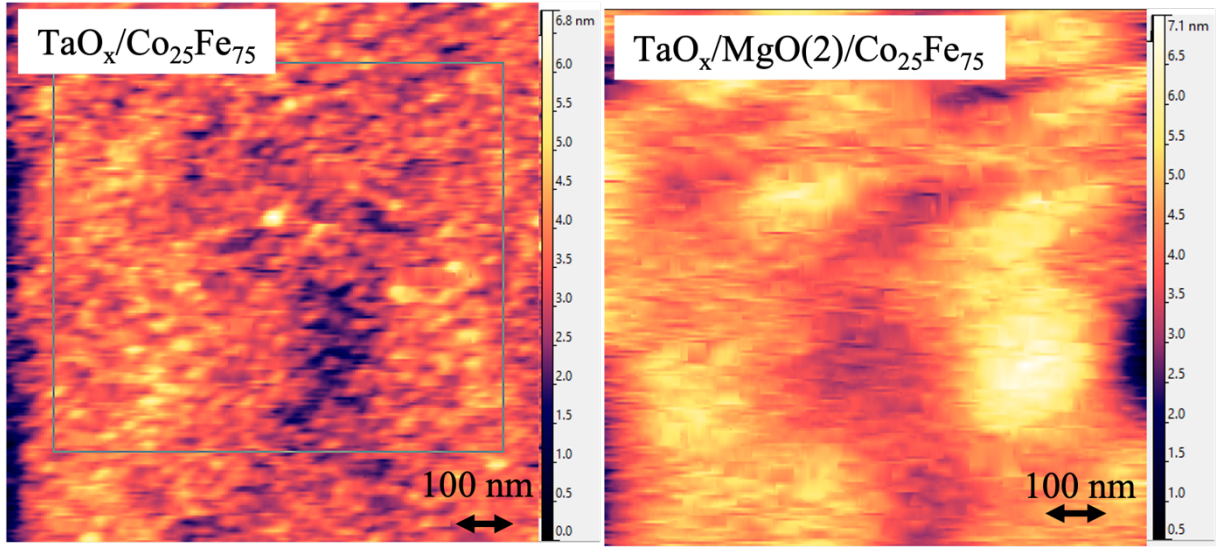


Figure I-1. The surface image of the sample w/ and w/o 2 nm thick MgO capping layer measured by AFM experiments.

J. M-H loops of $\text{Co}_{25}\text{Fe}_{75}$ films beneath MgO capping layer in in-plane geometry

The author measured M-H loops of $\text{Co}_{25}\text{Fe}_{75}$ layers of 0.4 nm-thick MgO capping layer by vibrating sample magnetometer in in-plane geometry as shown in Fig. S3. The coercivity of the MH loop increases and deviate from the rectangular shape with decreasing the $\text{Co}_{25}\text{Fe}_{75}$ thickness, which can be attributed to emergent of perpendicular magnetic anisotropy (PMA) due to surface magnetic anisotropy (SMA) of $\text{Co}_{25}\text{Fe}_{75}$. Thus, we measure MgO capping layer thickness dependence of MH loops for thin $\text{Co}_{25}\text{Fe}_{75}$ films to investigate the MgO capping layer effect on SMA of $\text{Co}_{25}\text{Fe}_{75}$. Figure S4 shows IP MH loops for the samples of 3 nm- and 5 nm-thick $\text{Co}_{25}\text{Fe}_{75}$ films beneath 0.4 nm-, 1.2 nm- and 3 nm-thick MgO capping layer. Whilst there is no large difference in 5 nm-thick $\text{Co}_{25}\text{Fe}_{75}$, the result of 3 nm-thick $\text{Co}_{25}\text{Fe}_{75}$ have a MgO thickness dependence of the MH loop, e.g. 3 nm-thick $\text{Co}_{25}\text{Fe}_{75}$ with 3 nm-thick MgO capping layer thicker MgO shows higher coercivity than that with 0.4 nm-thick MgO. The deviation of the shape of MH loops in 3 nm-thick $\text{Co}_{25}\text{Fe}_{75}$ films get larger in thicker MgO films can be also explained by the modulation of SMA by changing MgO capping layer thickness as shown in Fig. 3d in the main text.

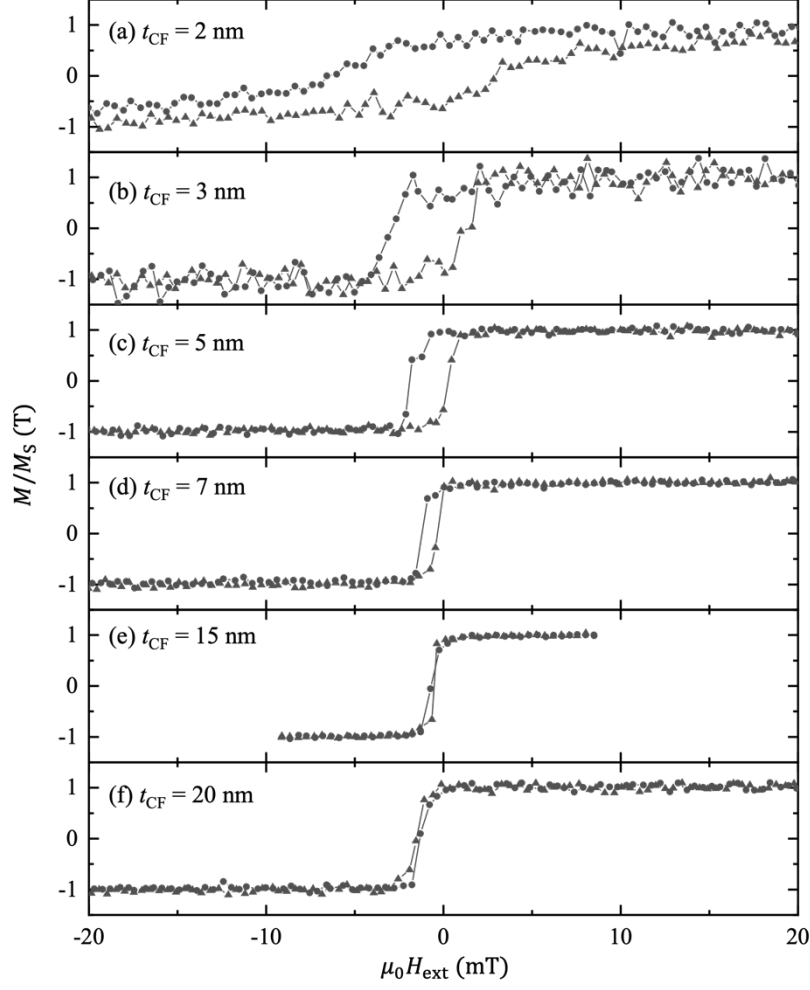


Figure J-1. M-H loops for different thick $\text{Co}_{25}\text{Fe}_{75}$ films beneath 0.4 nm-thick MgO capping layer measured by VSM in IP geometry.

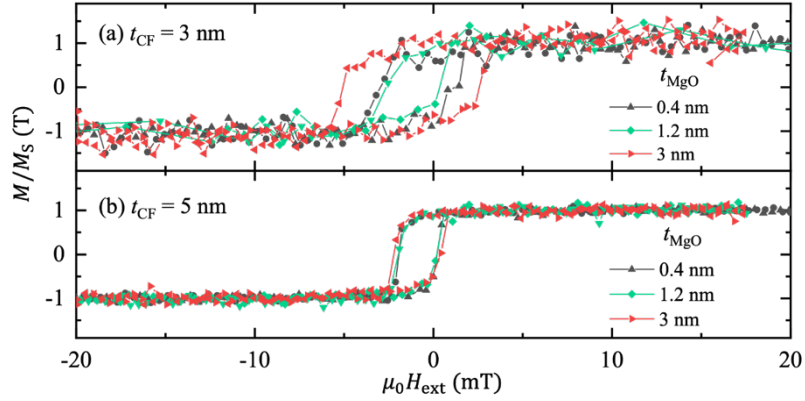


Figure J-2. IP MH loops for the samples of (a) 3 nm- and (b) 5 nm- thick $\text{Co}_{25}\text{Fe}_{75}$ film beneath 0.4 nm, 1.2 nm and 3 nm thick MgO capping layer.

K. Evaluation of the number of spins in unit volume for Py

The estimation of g_s needs the evaluation of the number of spins in unit volume for Py deposited by electron beam deposition. The deposited Py is typically amorphous and it is difficult to estimate the number of spins in the unit cell. The author carried out the VSM

measurement to measure the saturated magnetic moment m_s in thin Py films. Here, the 10 nm-thick Py film is prepared on $1\text{ cm} \times 1\text{ cm}$ SiO_2 substrate by electron beam deposition. The external magnetic field is applied in the plane of the Py film. Figure K-1 shows the M - m_s curve for the 10 nm-thick Py film with the area of $1\text{ cm} \times 1\text{ cm}$. The saturated magnetic moment is $1.793 \times 10^{-3}\text{ emu}$, which is converted to $1.933 \times 10^{17}\text{ } \mu_B$. From this value, the number of spins in the unit volume of Py is calculated.

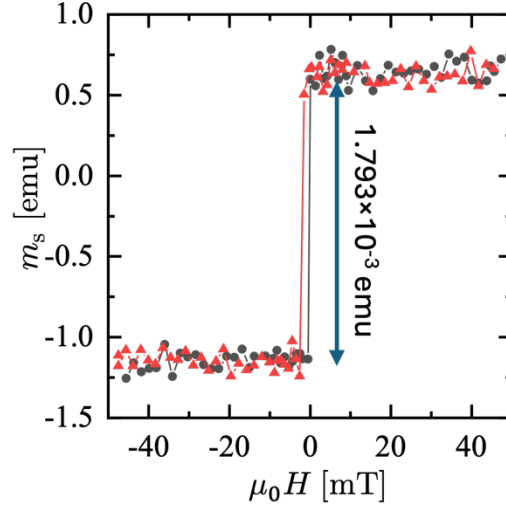


Figure K-1. The magnetic moment of 10 nm-thick Py films with the area of $1\text{ cm} \times 1\text{ cm}$ as a function of the external magnetic field.

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2. ○S. Yoshii, E. Shigematsu, R. Ohshima, Y. Ando and M. Shiraishi, "Modulation of magnon damping in ultrathin Co by introducing nonmagnetic buffer layer", International Symposium on Creation of Advanced Photonic and Electronic Devices 2021, March, 2021, Online. [Oral]
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