

# The temperature dependence of quantum spin pumping generated using electron spin resonance with three-magnon splittings

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**Abstract.** On the basis of the Schwinger-Keldysh formalism, we have closely investigated the temperature dependence of quantum spin pumping by electron spin resonance. We have clarified that three-magnon splittings excite non-zero modes of magnons and characterize the temperature dependence of quantum spin pumping by electron spin resonance. Our theoretical result qualitatively agrees with the experiment by Czeschka et al. that the mixing conductance is little influenced by temperature [F. D. Czeschka et al., Phys. Rev. Lett., **107**, 046601 (2011)].

*magnon, quantum spin pumping, electron spin resonance, three-magnon splittings, spintronics :*

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## 1. Introduction

The standard way to generate spin currents is spin pumping,[1, 2, 3, 4] which has already been established experimental technique at finite temperature;[5, 6] Czeschka et al.[7] have experimentally showed that the mixing conductance is little influenced by temperature. On top of this, some of technologies of spintronics are in fact going to be put into practical use;[5] they are applied to green information and communication technologies. In contrast to such experimental development, to the best of our knowledge, theoretical studies so far of quantum spin pumping at finite temperature beyond phenomenologies, unfortunately, will not be enough to explain the experimental result, in particular the above one by Czeschka et al.[7] Hence for the further development of spintronics and the application, the theoretically rigorous description of spin pumping at finite temperature is an urgent and important subject.

In order to overcome such a theoretical situation, we employ the Schwinger-Keldysh formalism[8, 9, 10, 11, 12, 13] and clarify the features of quantum spin pumping at finite temperature. This is the main aim of this paper. Let us remark that for the experimental realization of spin pumping, the time-dependent transverse magnetic field, which acts as ‘quantum fluctuations’,[14, 15] is applied and it drives the system into a non-equilibrium steady state. For the theoretical description of such systems beyond phenomenologies, one of the most suitable theoretical tools will be the Schwinger-Keldysh formalism;[8, 9, 10, 11, 12, 13] owing to the Schwinger-Keldysh closed time path,[16, 17, 18, 8] this formalism is not based on the assumption called the (well-known) Gell-Mann and Low theorem.[12, 19, 20, 21, 9] Therefore within the perturbative theory, the formalism can deal with an arbitrary time-dependent Hamiltonian[18] and can treat the system out of the equilibrium. On top of this, the Schwinger-Keldysh formalism is applicable to systems at finite temperature; the well-known Matsubara formalism[22] (i.e. the imaginary-time formalism),[19] which also can deal with thermodynamic average value, can be regarded as a simple corollary of the Schwinger-Keldysh formalism (i.e. closed time path formalism or the real-time formalism).[8] That is, the Schwinger-Keldysh formalism includes the Matsubara formalism and information about finite temperature is contained in the greater and lesser Green’s functions.[18] Consequently we can treat non-equilibrium phenomena at finite temperature owing to the Schwinger-Keldysh formalism.

Actually in our previous work,[16] we have already reformulated the quantum spin pumping theory from the viewpoint of the Schwinger-Keldysh formalism and have shown that spin pumping can be generated also by electron spin resonance (ESR)[16] as well as ferromagnetic resonance (FMR);[3, 2, 6, 25, 17] this is the natural result from the fact that the applied time-dependent transverse magnetic field (i.e. quantum fluctuations) affects conduction electrons as well as localized spins (i.e. magnons) at the interface (Fig. 1). To clarify the temperature dependence of quantum spin pumping by ESR[16] and find the microscopic origin is the final goal of this paper.

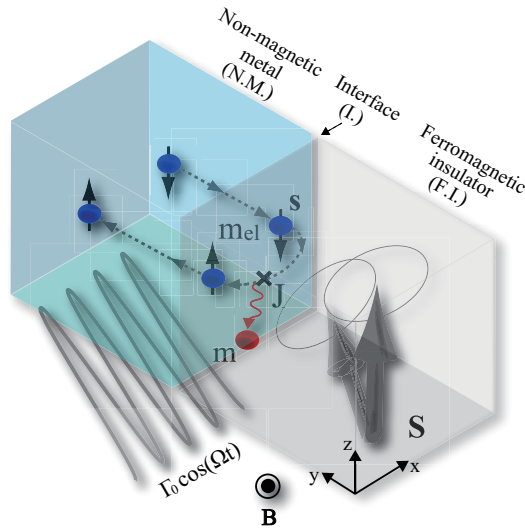
The quantum spin pumping system we had treated reads as follows;[16] we consider

a ferromagnetic insulator and non-magnetic metal junction shown in Fig. 1. At the interface, conduction electrons couple with localized spins  $\mathbf{S}(\mathbf{x}, t)$ ,  $\mathbf{x} = (x, y, z) \in \mathbb{R}^3$ ;

$$\mathcal{H}_{\text{ex}} = -2Ja_0^3 \int_{\mathbf{x} \in (\text{interface})} d\mathbf{x} \mathbf{S}(\mathbf{x}, t) \cdot \mathbf{s}(\mathbf{x}, t), \quad (1)$$

where the lattice constant of the ferromagnet is denoted as  $a_0$ . The magnitude of the interaction is supposed to be constant and it is expressed as  $2J$ . Note that owing to this exchange interaction  $\mathcal{H}_{\text{ex}}$  at the interface, the spin angular momentum can be interchanged between conduction electrons and the ferromagnet. That is, this exchange interaction  $\mathcal{H}_{\text{ex}}$  at the interface is the key to spin pumping.[5] Therefore we identify the system characterized by the exchange interaction between conduction electrons and the ferromagnet  $\mathcal{H}_{\text{ex}}$  (Hamiltonian (1)) with the spin pumping system. From now on, we exclusively focus on the dynamics at the interface.

Conduction electron spin variables are represented as  $s^j = \sum_{\eta, \zeta = \uparrow, \downarrow} c_\eta^\dagger (\sigma^j)_{\eta\zeta} c_\zeta / 2 =: c^\dagger \sigma^j c / 2$  with the  $2 \times 2$  Pauli matrices;  $[\sigma^j, \sigma^k] = 2i\epsilon_{jkl}\sigma^l$ , ( $j, k, l = x, y, z$ ). Operators  $c^\dagger/c$  are creation/annihilation operators for conduction electrons satisfying the (fermionic) anticommutation relation;  $\{c_\eta(\mathbf{x}, t), c_\zeta^\dagger(\mathbf{x}', t)\} = \delta_{\eta, \zeta} \delta(\mathbf{x} - \mathbf{x}')$ . We suppose the uniform magnetization and thus, localized spin degrees of freedom can be mapped into



**Figure 1.** The schematic picture of the quantum spin pumping system. Spheres represent magnons and those with arrows are conduction electrons. The wavy line denotes the time-dependent transverse magnetic field  $\Gamma(t)$  (i.e. the external pumping field). The interface is defined as an effective area where the Fermi gas (conduction electrons) and the Bose gas (magnons) coexist to interact;  $J \neq 0$ . The width of the interface might be supposed to be of the order of the lattice constant.[23] The interface can be regarded also as a ferromagnetic metal.[24] Conduction electrons cannot enter the ferromagnet, which is an insulator.

magnon[2, 26, 27] ones via the Holstein-Primakoff transformation;

$$S^+(\mathbf{x}, t) = \sqrt{2\tilde{S}} \sqrt{1 - \frac{a^\dagger(\mathbf{x}, t)a(\mathbf{x}, t)}{2\tilde{S}}} a(\mathbf{x}, t) \quad (2)$$

$$= \sqrt{2\tilde{S}} \left[ 1 - \frac{a^\dagger(\mathbf{x}, t)a(\mathbf{x}, t)}{4\tilde{S}} \right] a(\mathbf{x}, t) + \mathcal{O}(\tilde{S}^{-3/2}) \quad (3)$$

$$= (S^-)^\dagger \quad (4)$$

$$S^z(\mathbf{x}, t) = \tilde{S} - a^\dagger(\mathbf{x}, t)a(\mathbf{x}, t), \quad (5)$$

with  $\tilde{S} := S/a_0^3$ . Operators  $a^\dagger/a$  are magnon creation/annihilation operators satisfying the (bosonic) commutation relation;  $[a(\mathbf{x}, t), a^\dagger(\mathbf{x}', t)] = \delta(\mathbf{x} - \mathbf{x}')$ . Up to the  $\mathcal{O}(\tilde{S})$  terms, localized spins are reduced to free boson degrees of freedoms. Consequently, in the quadratic dispersion (i.e. long wavelength) approximation, the dynamics of localized spins with the applied magnetic field along the quantization axis (i.e.  $z$ -axis)  $B$  is described by the Hamiltonian  $\mathcal{H}_{\text{mag}}$ ;

$$\mathcal{H}_{\text{mag}} = \int_{\mathbf{x} \in (\text{interface})} d\mathbf{x} a^\dagger(\mathbf{x}, t) \left( -\frac{\nabla^2}{2m} + B \right) a(\mathbf{x}, t), \quad (6)$$

where the effective mass of magnons is denoted by  $m$ . In addition, Hamiltonian  $\mathcal{H}_{\text{ex}}$  can be rewritten as  $\mathcal{H}_{\text{ex}} = \mathcal{H}_{\text{ex}}^S + \mathcal{H}'_{\text{ex}}$  with

$$\mathcal{H}_{\text{ex}}^S = -JS \int_{\mathbf{x} \in (\text{interface})} d\mathbf{x} c^\dagger(\mathbf{x}, t) \sigma^z c(\mathbf{x}, t), \quad (7)$$

$$\begin{aligned} \mathcal{H}'_{\text{ex}} = & -Ja_0^3 \sqrt{\frac{\tilde{S}}{2}} \int_{\mathbf{x} \in (\text{interface})} d\mathbf{x} \left\{ a^\dagger(\mathbf{x}, t) \left[ 1 - \frac{a^\dagger(\mathbf{x}, t)a(\mathbf{x}, t)}{4\tilde{S}} \right] c^\dagger(\mathbf{x}, t) \sigma^+ c(\mathbf{x}, t) \right. \\ & \left. + \left[ 1 - \frac{a^\dagger(\mathbf{x}, t)a(\mathbf{x}, t)}{4\tilde{S}} \right] a(\mathbf{x}, t) c^\dagger(\mathbf{x}, t) \sigma^- c(\mathbf{x}, t) \right\} + \mathcal{O}(\tilde{S}^{-3/2}). \end{aligned} \quad (8)$$

Note that we have adsorbed the Bohr magneton and the  $g$ -factors into the definition of the magnetic field  $B$  and have taken  $\hbar = 1$  for convenience.

Here it should be stressed that according to Hamiltonian (7), the localized spin  $S$  acts as an effective magnetic field along the quantization axis for conduction electrons;

$$\mathcal{H}_{\text{ex}}^S = -JS \int_{\mathbf{x} \in (\text{interface})} d\mathbf{x} c^\dagger(\mathbf{x}, t) \sigma^z c(\mathbf{x}, t) \quad (9)$$

$$= -2JS \int_{\mathbf{x} \in (\text{interface})} d\mathbf{x} s^z(\mathbf{x}, t). \quad (10)$$

Therefore, the diagonal part of the conduction electrons is written as

$$\mathcal{H}_{\text{el}} = \int_{\mathbf{x} \in (\text{interface})} d\mathbf{x} c^\dagger(\mathbf{x}, t) \left[ -\frac{\nabla^2}{2m_{\text{el}}} - \left( JS + \frac{B}{2} \right) \sigma^z \right] c(\mathbf{x}, t), \quad (11)$$

with the effective mass of the conduction electron  $m_{\text{el}}$ .

For the experimental realization of spin pumping,[2, 28, 6] a time-dependent transverse magnetic field  $\Gamma(t)$  with an angular frequency  $\Omega$  is applied into the system as a driving energy;  $\Gamma(t) := \Gamma_0 \cos(\Omega t)$ . This applied periodic transverse magnetic field

acts as ‘quantum fluctuations’[14, 15] and drives the system into a non-equilibrium steady state.[29] Thus we identify the system described by the exchange interaction  $\mathcal{H}_{\text{ex}}$  (Hamiltonian (1)) under the applied time-dependent transverse magnetic field  $\Gamma(t)$  with the ‘quantum spin pumping system’. Note that the applied time-dependent transverse magnetic field couples with conduction electrons as well as localized spins;

$$V_{\text{el}}^{\Gamma} = \frac{\Gamma(t)}{4} \int_{\mathbf{x} \in (\text{interface})} d\mathbf{x} c^{\dagger}(\mathbf{x}, t) (\sigma^{+} + \sigma^{-}) c(\mathbf{x}, t) \quad (12)$$

$$V_{\text{mag}}^{\Gamma} = \Gamma(t) \sqrt{\frac{\tilde{S}}{2}} \int_{\mathbf{x} \in (\text{interface})} d\mathbf{x} \left\{ \left[ 1 - \frac{a^{\dagger}(\mathbf{x}, t) a(\mathbf{x}, t)}{4\tilde{S}} \right] a(\mathbf{x}, t) + a^{\dagger}(\mathbf{x}, t) \left[ 1 - \frac{a^{\dagger}(\mathbf{x}, t) a(\mathbf{x}, t)}{4\tilde{S}} \right] \right\}. \quad (13)$$

Therefore spin pumping can be generated also by ESR ( $\Omega = 2JS + B$ )[16] as well as FMR ( $\Omega = B$ ).

Finally, the total Hamiltonian of the quantum spin pumping system  $\mathcal{H}$  (i.e. the spin pumping system with  $\Gamma(t)$ ) is arranged as

$$\mathcal{H} := \mathcal{H}_{\text{mag}} + \mathcal{H}'_{\text{ex}} + \mathcal{H}_{\text{el}} + V_{\text{el}}^{\Gamma} + V_{\text{mag}}^{\Gamma}. \quad (14)$$

We investigate the features of quantum spin pumping described by this Hamiltonian (Hamiltonian (14)); we clarify the behavior of quantum spin pumping generated by ESR at finite temperature and go after the microscopic origin. This is the main aim of this paper.

The remain of the paper is organized as follows; we quickly review our quantum spin pumping theory[16] and stress the point in sec. 2. The distinction from the standard spin pumping theory by Tserkovnyak et al. is also clarified. The readers who are familiar with our formalism can skip sec. 2. The temperature dependence of quantum spin pumping by ESR is revealed in sec. 3. We go after the microscopic origin and qualitatively understand the behavior from the viewpoint of three-magnon splittings.

## 2. Quantum spin pumping theory based on Schwinger-Keldysh formalism

Before getting straight to the explanation of our quantum spin pumping theory, let us remark a point. In the last section, localized spin degrees of freedom have been mapped into magnon ones via the Holstein-Primakoff transformation; we have expanded it up to the  $\mathcal{O}(\tilde{S}^{-1/2})$  terms (see eqs. (3) and (4)). Therefore the magnon-magnon interaction,  $a^{\dagger} a^{\dagger} a a = \mathcal{O}(\tilde{S}^0)$ , and the magnon-electron interaction,  $a^{\dagger} a c^{\dagger} \sigma^z c = \mathcal{O}(\tilde{S}^0)$ , may emerge as well as Hamiltonian (6), (7) and (14). Nevertheless, we have omitted the terms. The reason reads as follows; in sharp contrast to  $\mathcal{H}'_{\text{ex}}$ ,  $V_{\text{mag}}^{\Gamma}$  and  $V_{\text{el}}^{\Gamma}$ , (see Hamiltonian (8), (13) and (12)), the magnon-magnon interaction and the magnon-electron interaction do not include the spin-flip operators  $(\sigma^{\pm})_{\dagger}$  and on top of this, they consist of the even

‡ As the result, they commute with the z-component of the spin density  $\rho_s^z := c^{\dagger} \sigma^z c / 2$  and hence, they do not directly contribute to the SRT defined in eq. (15).

number in respect to magnon creation/annihilation operators.§ Consequently the terms can not directly contribute to spin pumping;[30] as shown in Fig. 1, spin-flip processes described by spin-flip operators  $\sigma^\pm$  are essential to spin pumping. Therefore, within the approximation of eq. (20), we are allowed to omit the effects of the magnon-magnon interaction and the magnon-electron interaction.||

### 2.1. Breaking of spin conservation law

We have formulated the spin pumping theory on the basis of the spin continuity equation for conduction electrons:[16, 18, 31]

$$\dot{\rho}_s^z + \nabla \cdot \mathbf{j}_s^z = \mathcal{T}_s^z, \quad (15)$$

where the dot denotes the time derivative of the z-component for the spin density defined as  $\rho_s^z := c^\dagger \sigma^z c / 2$ , and  $\mathbf{j}_s$  is the spin current density. Let us emphasize that in sharp contrast to the case of charges, the spin conservation law is broken and it is represented by the spin relaxation torque (SRT)[16, 31]  $\mathcal{T}_s$ , which appears in the spin continuity equation. Through the Heisenberg equation of motion, the z-component of the SRT can be explicitly written down as

$$\begin{aligned} \mathcal{T}_s^z = & iJa_0^3 \sqrt{\frac{\tilde{S}}{2}} \left\{ a^\dagger(\mathbf{x}, t) \left[ 1 - \frac{a^\dagger(\mathbf{x}, t)a(\mathbf{x}, t)}{4\tilde{S}} \right] c^\dagger(\mathbf{x}, t) \sigma^+ c(\mathbf{x}, t) \right. \\ & - \left. \left[ 1 - \frac{a^\dagger(\mathbf{x}, t)a(\mathbf{x}, t)}{4\tilde{S}} \right] a(\mathbf{x}, t) c^\dagger(\mathbf{x}, t) \sigma^- c(\mathbf{x}, t) \right\} \\ & + \frac{\Gamma(t)}{4i} \left[ c^\dagger(\mathbf{x}, t) \sigma^+ c(\mathbf{x}, t) - c^\dagger(\mathbf{x}, t) \sigma^- c(\mathbf{x}, t) \right]. \end{aligned} \quad (16)$$

Note that the SRT has arisen from  $\mathcal{H}'_{\text{ex}}$  and  $V_{\text{el}}^\Gamma$ , which consist of spin-flip operators ( $\sigma^\pm$ ) and quantum fluctuations ( $\Gamma(t)$ );  $\mathcal{T}_s^z = [\rho_s^z, \mathcal{H}'_{\text{ex}} + V_{\text{el}}^\Gamma] / i$ . Let me emphasize that though each spin conservation law for conduction electrons and magnons is broken, the total spin angular momentum is conserved in the spin pumping systems.[16, 31]

### 2.2. Pumped net spin current

One can easily see that the expectation value of the spin density for conduction electrons reads  $\langle \rho_s^z \rangle = \sum_{n=0, \pm 1} \langle \rho_s^z(n) \rangle e^{2in\Omega t} + \mathcal{O}(\Gamma^4)$ , where  $\rho_s^z(n)$  represents the (time-independent) expansion coefficient of each angular frequency mode. Thus the time-

§ Now, we have focused on the SRT accompanied by the exchange interaction  $J$  between conduction electrons and magnons (eqs. (16) and (20)). Although  $V_{\text{mag}}^\Gamma$  does not include spin-flip operators ( $\sigma^\pm$ ), it consists of the odd number in respect to magnon creation/annihilation operators and hence, the term  $V_{\text{mag}}^\Gamma$  is essential to spin pumping; from the viewpoint of the calculation based on the perturbation theory (i.e. Wick's theorem), one can easily see that it directly contributes to the SRT represented by eq. (16) (i.e. spin pumping). In other words, it is clear, from the viewpoint of Wick's theorem, that the SRT becomes zero without  $V_{\text{mag}}^\Gamma$ .

|| The magnon-magnon interaction and the magnon-electron interaction indirectly lead to higher-terms than eq. (20), which are out of the aim of the present work; they give  $\mathcal{O}(J^2)$ -terms for instance.

average of the time-derivative becomes zero (note that  $\langle \dot{\rho}_s^z \rangle := \langle \partial_t \rho_s^z \rangle = \partial_t \langle \rho_s^z \rangle$ );

$$\overline{\langle \dot{\rho}_s^z \rangle} = 0. \quad (17)$$

As the result, the spin continuity equation for conduction electrons, eq. (15), reads  $\overline{\langle \nabla \cdot \mathbf{j}_s^z \rangle} = \overline{\langle \mathcal{T}_s^z \rangle}$ . Here it should be noted that conduction electrons cannot enter the ferromagnet,[26] which is an insulator (see Fig. 1 (b)). Consequently by integrating over the volume of the interface and adopting the Gauss's divergence theorem, the time-average of the net spin current pumped into the adjacent non-magnetic metal (i.e.  $\int \mathbf{j}_s^z \cdot d\mathbf{S}_{\text{interface}}$  with the surface of the interface  $\mathbf{S}_{\text{interface}}$ ) can be evaluated as

$$\overline{\left\langle \int \mathbf{j}_s^z \cdot d\mathbf{S}_{\text{interface}} \right\rangle} = \int_{\mathbf{x} \in (\text{interface})} d\mathbf{x} \overline{\langle \mathcal{T}_s^z \rangle}. \quad (18)$$

Experimentally, this pumped spin current can be detected via the inverse spin Hall effect[6] in the non-magnetic metal.

Let us emphasize that the time-average of the pumped net spin current,  $\overline{\langle \int \mathbf{j}_s^z \cdot d\mathbf{S}_{\text{interface}} \rangle}$ , is expressed only in terms of the SRT (see eq. (18)); note that calculating  $\overline{\langle \dot{\rho}_s^z \rangle}$  has no relation with evaluating the pumped net spin current even when the total spin angular momentum is conserved. That is, the spin density for conduction electrons,  $\rho_s^z$ , is not relevant to quantum spin pumping mediated by magnon.[27] This is one of the main results from our formalism. Thus from now on, we focus on  $\mathcal{T}_s^z$  and qualitatively clarify the features of quantum spin pumping mediated by magnons.

### 2.3. Spin relaxation torque

It is also easy to see that the expectation value of the SRT reads[16]  $\langle \mathcal{T}_s^z \rangle = \sum_{n=0,\pm 1} \langle \mathcal{T}_s^z(n) \rangle e^{2in\Omega t} + \mathcal{O}(\Gamma^4)$ , where  $\mathcal{T}_s^z(n)$  represents the (time-independent) expansion coefficient of each angular frequency mode. Thus the time-average becomes

$$\overline{\langle \mathcal{T}_s^z \rangle} = \langle \mathcal{T}_s^z(n=0) \rangle. \quad (19)$$

The interface is, in general, a weak coupling regime;[28] the exchange interaction  $J$  (see Hamiltonian (8)) is supposed to be smaller than the Fermi energy and the exchange interaction among ferromagnets. In addition, a weak transverse magnetic field  $\Gamma$  is applied. Therefore we are allowed to treat  $\mathcal{H}'_{\text{ex}}$ ,  $V_{\text{el}}^\Gamma$ , and  $V_{\text{mag}}^\Gamma$  as perturbative terms to evaluate the SRT.

Through the standard procedure of the Schwinger-Keldysh (or contour-ordered) Green's function[32, 12, 9] and the Langreth method,[11, 10, 13, 8] the SRT,  $\langle \mathcal{T}_s^z(n=0) \rangle$ , is evaluated as follows (see also Fig. 3 (b)). The detail of the calculation had been shown in our previous work.[16];

$$\begin{aligned} \langle \mathcal{T}_s^z(n=0) \rangle &= \frac{J}{2} \left( \frac{\Gamma_0}{2} \right)^2 S \left[ 1 - \frac{i}{\tilde{S}} \int \frac{d\mathbf{k}'}{(2\pi)^3} \int \frac{d\omega'}{2\pi} G_{\mathbf{k}',\omega'}^< \right] \int \frac{d\mathbf{k}}{(2\pi)^3} \int \frac{d\omega}{2\pi} \\ &\quad \times \left[ (G_{0,-\Omega}^a + G_{0,-\Omega}^r) (\mathcal{G}_{\downarrow,\mathbf{k},\omega-\Omega}^t \mathcal{G}_{\uparrow,\mathbf{k},\omega}^t - \mathcal{G}_{\downarrow,\mathbf{k},\omega-\Omega}^< \mathcal{G}_{\uparrow,\mathbf{k},\omega}^>) \right. \\ &\quad + (G_{0,\Omega}^a + G_{0,\Omega}^r) (\mathcal{G}_{\downarrow,\mathbf{k},\omega+\Omega}^t \mathcal{G}_{\uparrow,\mathbf{k},\omega}^t - \mathcal{G}_{\downarrow,\mathbf{k},\omega+\Omega}^< \mathcal{G}_{\uparrow,\mathbf{k},\omega}^>) \\ &\quad \left. - (G_{0,-\Omega}^r + G_{0,-\Omega}^a) (\mathcal{G}_{\uparrow,\mathbf{k},\omega-\Omega}^t \mathcal{G}_{\downarrow,\mathbf{k},\omega}^t - \mathcal{G}_{\uparrow,\mathbf{k},\omega-\Omega}^< \mathcal{G}_{\downarrow,\mathbf{k},\omega}^>) \right] \end{aligned}$$

$$\begin{aligned}
 & - (\mathcal{G}_{0,\Omega}^r + \mathcal{G}_{0,\Omega}^a)(\mathcal{G}_{\uparrow,\mathbf{k},\omega+\Omega}^t \mathcal{G}_{\downarrow,\mathbf{k},\omega}^t - \mathcal{G}_{\uparrow,\mathbf{k},\omega+\Omega}^< \mathcal{G}_{\downarrow,\mathbf{k},\omega}^>) \\
 & + \mathcal{O}(J^0) + \mathcal{O}(J^2) + \mathcal{O}(\Gamma^4) + \mathcal{O}(JS^{-1}).
 \end{aligned} \tag{20}$$

The variables  $\mathcal{G}^{t(r,a,<,>)} / \mathcal{G}^{t(r,a,<,>)}$  are the fermionic/bosonic time-ordered, retarded, advanced, lesser, and greater Green's functions, respectively.[16] We here have taken the extended time (i.e. the contour variable) defined on the Schwinger-Keldysh closed time path,[8, 9, 10, 11, 13]  $c$ , on the forward path  $c_{\rightarrow}$ ;  $c = c_{\rightarrow} + c_{\leftarrow}$ . Even when the time is located on the backward path  $c_{\leftarrow}$ , the result of the calculation does not change because each Green's function is not independent;[9, 12, 10, 13]  $\mathcal{G}^r - \mathcal{G}^a = \mathcal{G}^> - \mathcal{G}^<$ . This relation comes into effect also in the bosonic case.[9, 12, 10, 13]

The SRT (eq. (20)) has become proportional to  $\Gamma_0^2$ ;  $\langle \mathcal{T}_s^z(n=0) \rangle \propto \Gamma_0^2$ . Thus the SRT (i.e. the pumped net spin current) can be interpreted as the non-linear response to the applied time-dependent transverse magnetic field (i.e. quantum fluctuations). This is one of the main features of our quantum spin pumping theory; our theory well describes the experimental features of quantum spin pumping that quantum fluctuations are essential.[16]

Now, let us introduce the dimensionless SRT,  $\overline{\langle \mathcal{T}_s^z \rangle} / \Lambda$ , and the one in the wavenumber-space for conduction electrons,  $\langle \widetilde{\mathcal{T}}_s^z(n=0) \rangle$ , as follows;

$$\overline{\langle \mathcal{T}_s^z \rangle} := \Lambda \int_0^\infty \left( \sqrt{\frac{F}{\epsilon_F}} dk \right) \langle \widetilde{\mathcal{T}}_s^z(n=0) \rangle \tag{21}$$

with

$$\Lambda := \frac{\sqrt{\epsilon_F} S \Gamma_0^2}{4(2\pi\sqrt{F})^3}. \tag{22}$$

We have denoted as  $F := (2m_{\text{el}})^{-1}$  and the parameters,  $\epsilon_F$  and  $k$ , represent the Fermi energy and the wavenumber of conduction electrons.

According to Hamiltonian (11) and (6), the effective magnetic field along the quantization axis for conduction electrons,  $s^z = c^\dagger \sigma^z c / 2$ , reads  $2JS + B$  and that for magnons does  $B$  at the interface. On top of this, the applied time-dependent transverse magnetic field  $\Gamma(t) = \Gamma_0 \cos(\Omega t)$  (i.e. quantum fluctuations) affects conduction electrons as well as magnons at the interface; Fig. 1. Thus, the SRT (eq. (20)) becomes a non-zero value around the points[16]  $\Omega = 2JS + B$  and  $\Omega = B$ , which are generated by ESR and FMR, respectively. That is (eq. (18)), spin pumping is generated by ESR ( $\Omega = 2JS + B$ ) as well as FMR ( $\Omega = B$ ).¶

As you know, concerning spin pumping by FMR ( $\Omega = B$ ), Tserkovnyak et al. have already revealed the dynamics and have given clear explanations.[3, 25, 4] Therefore from now on, we exclusively focus on quantum spin pumping by ESR ( $\Omega = 2JS + B$ ).

¶ Our theory based on Schwinger-Keldysh formalism describes spin pumping by ESR and FMR; see Fig. A1.



#### 2.4. Comparison with the pioneering theory proposed by Tserkovnyak et al.

Czeschka et al. has employed the spin pumping theory proposed by Tserkovnyak et al.[3, 33] as the standard theoretical model to analyze their experiment.[7] Hence, it might be helpful to remark the distinction[16] between our quantum spin pumping theory and the pioneering theory proposed by Tserkovnyak et al.; they have phenomenologically treated the spin-flip scattering processes and their theory has now become the standard one.

##### 2.4.1. Pumped spin current gained by the theory proposed by Tserkovnyak et al.

According to the phenomenological[3, 33] spin pumping theory by Tserkovnyak et al. and their notation,[34, 3] the pumped spin current  $\mathbf{I}_{\text{s-pump}}$  reads

$$\mathbf{I}_{\text{s-pump}} = G_{\perp}^{(\text{R})} \mathbf{m} \times \dot{\mathbf{m}} + G_{\perp}^{(\text{I})} \dot{\mathbf{m}}, \quad (23)$$

where the dot denotes the time derivative and the variable  $G_{\perp}$  is the complex-valued mixing conductance that depends on the material;[35, 36]  $G_{\perp} = G_{\perp}^{(\text{R})} + iG_{\perp}^{(\text{I})}$ . We have taken  $e = 1$ , and  $\mathbf{m}(\mathbf{x}, t)$  denotes a unit vector along the magnetization direction. It should be noted that, in sharp contrast to our present treatment (i.e. magnons), they have treated  $\mathbf{m}(\mathbf{x}, t)$  as classical variables which can be described by the Landau-Lifshitz-Gilbert (LLG) eq.;

$$\dot{\mathbf{m}} = \gamma \mathbf{H}_{\text{eff}} \times \mathbf{m} + \alpha \mathbf{m} \times \dot{\mathbf{m}}, \quad (24)$$

where  $\gamma$  is the gyro-magnetic ratio and  $\alpha$  is the Gilbert damping constant[37, 38] that determines the magnetization dissipation rate. The effective magnetic field is set as  $\mathbf{H}_{\text{eff}} = (\Gamma(t), 0, B)$  with the time-dependent transverse magnetic field  $\Gamma(t)$ . Then the LLG eq., eq. (24), gives

$$\dot{m}^x = -\gamma B m^y + \alpha(m^y \dot{m}^z - \dot{m}^y m^z), \quad (25)$$

$$\dot{m}^y = \gamma(B m^x - \Gamma m^z) + \alpha(m^z \dot{m}^x - \dot{m}^z m^x), \quad (26)$$

$$\dot{m}^z = \gamma \Gamma m^y + \alpha(m^x \dot{m}^y - \dot{m}^x m^y). \quad (27)$$

Eqs. (25)-(27) is substituted into  $I_{\text{s-pump}}^z$ , eq. (23). After that, we include the contribution of the Gilbert damping term, which depends on the materials, up to  $\mathcal{O}(\alpha)$ ;  $\alpha \sim 10^{-3}, 10^{-2}$  for  $\text{Ni}_{81}\text{Fe}_{19}$  (metal),[28] and  $\alpha \sim 10^{-5}$  for  $\text{Y}_3\text{Fe}_5\text{O}_{12}$  (insulator),[26] as examples. <sup>+</sup> Consequently, the z-component of the pumped spin current which the theory by Tserkovnyak et al. gives reads

$$\begin{aligned} I_{\text{s-pump}}^z = & G_{\perp}^{(\text{R})} \left\{ \gamma B [(m^x)^2 + (m^y)^2] - \gamma \Gamma m^x m^z \right. \\ & \left. - \alpha \gamma \Gamma [m^x m^y m^x + (m^y)^3 + (m^z)^2 m^y] \right\} \\ & + G_{\perp}^{(\text{I})} \left\{ \alpha \gamma \{ B [(m^x)^2 + (m^y)^2] - \Gamma m^x m^z \} + \gamma \Gamma m^y \right\} + \mathcal{O}(\alpha^2) \end{aligned} \quad (28)$$

$$\xrightarrow{\Gamma \rightarrow 0} [G_{\perp}^{(\text{R})} + \alpha G_{\perp}^{(\text{I})}] \gamma B [(m^x)^2 + (m^y)^2]. \quad (29)$$

$$\xrightarrow{G_{\perp}^{(\text{I})} \rightarrow 0} G_{\perp}^{(\text{R})} \gamma B [(m^x)^2 + (m^y)^2]. \quad (30)$$

<sup>+</sup> Their theory is believed to be applicable to both ferromagnetic metals and insulators.[25]

### 2.4.2. Distinction

At finite temperature, the magnetization is thermally activated;  $\mathbf{m} \neq 0$ . [39] Then the time derivative of the z-component means

$$\dot{m}^z = \gamma\Gamma m^y + \alpha\gamma\{B[(m^x)^2 + (m^y)^2] - \Gamma m^x m^z\} + \mathcal{O}(\alpha^2). \quad (31)$$

$$\xrightarrow{\Gamma \rightarrow 0} \alpha\gamma B[(m^x)^2 + (m^y)^2]. \quad (32)$$

$$\neq 0. \quad (33)$$

Eqs. (29), (30) and (33) show that, within the standard spin pumping theory proposed by Tserkovnyak et al. with the LLG eq., they may gain pumped spin currents at finite temperature if only the magnetic field along the z-axis,  $B$ , is applied;

$$I_{\text{s-pump}}^z \xrightarrow{\Gamma \rightarrow 0 (B \neq 0)} 0. \quad (34)$$

That is, the spin pumping theory by Tserkovnyak et al. [3, 34, 25] with the LLG eq. concludes that spin currents may be pumped at finite temperature without time-dependent transverse magnetic fields.

On the other hand, our quantum spin pumping theory based on the Schwinger-Keldysh formalism, which explicitly captures the non-equilibrium spin-flip processes generated by the applied transverse magnetic field, gives different result;

$$\langle \mathcal{I}_s^z(n=0) \rangle \propto (\Gamma_0)^2 \quad (35)$$

$$\xrightarrow{\Gamma_0 \rightarrow 0} 0. \quad (36)$$

That is, our quantum spin pumping theory shows that spin currents mediated by magnons cannot be pumped without quantum fluctuations (i.e. time-dependent transverse magnetic fields,  $\Gamma(t)$ ). That is, according to our theory, quantum fluctuations are essential to spin pumping mediated by magnons.

This (eqs. (36) and (34)) is the significant distinction between our quantum spin pumping theory and the one proposed by Tserkovnyak et al. Here, let us mention that our theoretical result qualitatively agrees with the one obtained by Takeuchi et al.; [40] although their analysis is restricted to the system at zero-temperature, they have adopted the same approach with ours (i.e. Schwinger-Keldysh formalism) and have reached the same conclusion with ours.

## 3. Temperature dependence of quantum spin pumping by ESR

From now on, we investigate the features of quantum spin pumping mediated by magnons under ESR ( $\Omega = 2JS + B$ ), in particular the temperature dependence.

Fig. 2 shows the temperature dependence of the SRT by ESR; it is clear that when temperature rises, the SRT becomes smaller. To find the reason and the microscopic origin is the main aim of this section (sec. 3.1 and 3.2). The resulting features of quantum spin pumping by ESR is shown in sec. 3.3.

Here, it would be helpful to remark that our present theory is applicable to the quantum spin pumping systems where the following conditions, (i) and (ii), are satisfied;

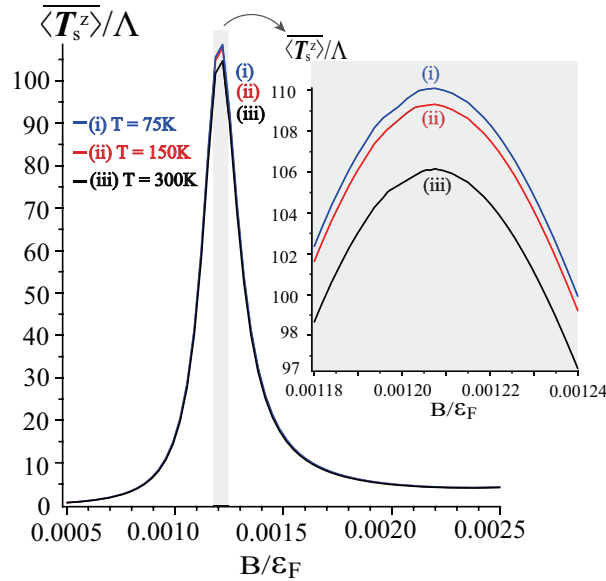
(i) the magnitude of the applied transverse magnetic field  $|\Gamma|$  and the exchange interaction between conduction electrons and magnons are smaller than the Fermi energy and the exchange interaction among ferromagnets, (ii) temperature of the system is lower than Fermi temperature and Curie temperature.

### 3.1. Effective magnetic field for conduction electrons

As mentioned (Hamiltonian (10)), localized spins  $S$  act as effective magnetic fields along the quantization axis for conduction electrons. According to eq. (20), the effective magnetic field at finite temperature  $S_{\text{eff}}(T)$  becomes

$$S_{\text{eff}}(T) = S \left[ 1 - \frac{i}{\tilde{S}} \int \frac{d\mathbf{k}'}{(2\pi)^3} \int \frac{d\omega'}{2\pi} G_{\mathbf{k}',\omega'}^< \right], \quad (37)$$

and it is the monotone decreasing function in respect to temperature  $T$  around the ESR point ( $\Omega = 2JS + B$ , Fig. 3 (b-iii));  $dS_{\text{eff}}(T)/(dT) < 0$ . In addition, the SRT is proportional to  $S_{\text{eff}}(T)$  (see eq. (20));  $\langle T_s^z(n=0) \rangle \propto JS_{\text{eff}}(T)\Gamma_0^2$ . As the result, the SRT by ESR (see Fig. 2. See also sec. 3.3 in advance) is also the monotone decreasing function in respect to temperature  $T$ ;  $d\langle T_s^z(n=0) \rangle/(dT) < 0$ . This behavior of quantum spin pumping by ESR ( $\Omega = 2JS + B$ ) at finite temperature is the main distinction from that of the standard spin pumping by FMR ( $\Omega = B$ ).[28, 42, 7]



**Figure 2.** The temperature dependence of the SRT around the ESR point ( $\Omega = 2JS + B$ ). When temperature rises, the SRT becomes smaller. As a typical case,[16] each parameter is set as follows:[28, 39, 26, 41, 16]  $\epsilon_F = 5.6$  eV,  $T = 300$  K,  $F := (2m_{\text{el}})^{-1} = 4$  eV  $\text{\AA}^2$ ,  $D := (2m)^{-1} = 0.3$  eV  $\text{\AA}^2$ ,  $a_0 = 3$   $\text{\AA}$ ,  $S = 1/2$ ,  $\Omega/\epsilon_F = 0.0032$ , and  $J/\epsilon_F = 0.002$ .

### 3.2. Three-magnon splittings

We microscopically go after the origin of the effective magnetic field at finite temperature  $S_{\text{eff}}(T)$ . As shown in eqs. (3) and (4), we have rewritten the localized spin degrees of freedom into magnon ones via the Holstein-Primakoff transformation;  $S^+(\mathbf{x}, t) = \sqrt{2\tilde{S}}[1 - a^\dagger(\mathbf{x}, t)a(\mathbf{x}, t)/(4\tilde{S})]a(\mathbf{x}, t) + \mathcal{O}(\tilde{S}^{-3/2}) = (S^-)^\dagger$ . Note that we have expanded up to  $\mathcal{O}(\tilde{S}^{-1/2})$  and have included the effects of three-magnon splittings[27, 42] (Fig. 3 (b-ii));

$$\frac{a^\dagger(\mathbf{x}, t)a(\mathbf{x}, t)a(\mathbf{x}, t)}{\tilde{S}} \quad \text{and} \quad \frac{a^\dagger(\mathbf{x}, t)a^\dagger(\mathbf{x}, t)a(\mathbf{x}, t)}{\tilde{S}}, \quad (38)$$

which lead to the loop effects[20] (i.e. quantum effects) expressed as (see eq. (37) and Fig. 3 (b))

$$S \left[ -\frac{i}{\tilde{S}} \int \frac{d\mathbf{k}'}{(2\pi)^3} \int \frac{d\omega'}{2\pi} \mathbf{G}_{\mathbf{k}', \omega'}^< \right] =: -\Lambda' \int_0^\infty \left( \sqrt{\frac{D}{\epsilon_F}} dk' \right) \tilde{n}_{k'} \quad (39)$$

with

$$\Lambda' := \frac{S}{4\pi^3 \tilde{S}} \left( \frac{\epsilon_F}{D} \right)^{3/2}. \quad (40)$$

We have denoted as  $D := (2m)^{-1}$  and the variable  $\tilde{n}_{k'}$  in eq. (39) represents the dimensionless distribution function of magnons in the dimensionless wavenumber space for magnons ( $\sqrt{D/\epsilon_F} dk'$ ); Fig. 3 (b-iv). It is apparent from Fig. 3 (b-iv) and eqs. (37) and (39) that the three-magnon splittings excite the non-zero modes of magnons ( $\mathbf{k}' \neq 0$ ); when temperature rises, the wavenumber of excited magnons becomes larger and the number of excited magnons also increases. Consequently, the magnitude of localized spins along the quantization axis become smaller and it leads to the behavior of quantum spin pumping by ESR at finite temperature which is discussed in sec. 3.1. That is, the three-magnon splittings characterize the effective magnetic field at finite temperature  $S_{\text{eff}}(T)$  and the temperature dependence of quantum spin pumping by ESR.

If we expand up to only  $\mathcal{O}(\sqrt{\tilde{S}})$ ;  $S^+(\mathbf{x}, t) = \sqrt{2\tilde{S}}a(\mathbf{x}, t) + \mathcal{O}(\tilde{S}^{-1/2}) = (S^-)^\dagger$ , and neglect three-magnon splittings, the effective magnetic field is reduced to  $S$  (Fig. 3 (a));  $S_{\text{eff}} \rightarrow S$ . This corresponds to the large- $S$  limit.[43] In this case, only the zero mode of magnons is relevant to the SRT (see eqs. (20) and (39)) and non-zero modes of magnons are not excited.

### 3.3. Contribution of conduction electrons

The SRT at finite temperature,  $\overline{\langle \mathcal{T}_s^z \rangle} |_{T=}$   $\langle \mathcal{T}_s^z(n=0) \rangle$ , can be expressed also as follows (see eqs. (20) and (37));

$$\overline{\langle \mathcal{T}_s^z \rangle} |_{T=} = \overline{\langle \mathcal{T}_s^z \rangle} |_{T=300[\text{K}]} \times \eta^{\text{ratio}}(T) \times S_{\text{eff}}^{\text{ratio}}(T) \quad (41)$$

with

$$\eta^{\text{ratio}}(T) := \frac{\mathcal{T}_s^{z\text{-ratio}}(T)}{S_{\text{eff}}^{\text{ratio}}(T)}, \quad (42)$$

$$\mathcal{T}_s^{z\text{-ratio}}(T) := \frac{\langle \mathcal{T}_s^z \rangle |T}{\langle \mathcal{T}_s^z \rangle |T=300[\text{K}]}, \quad (43)$$

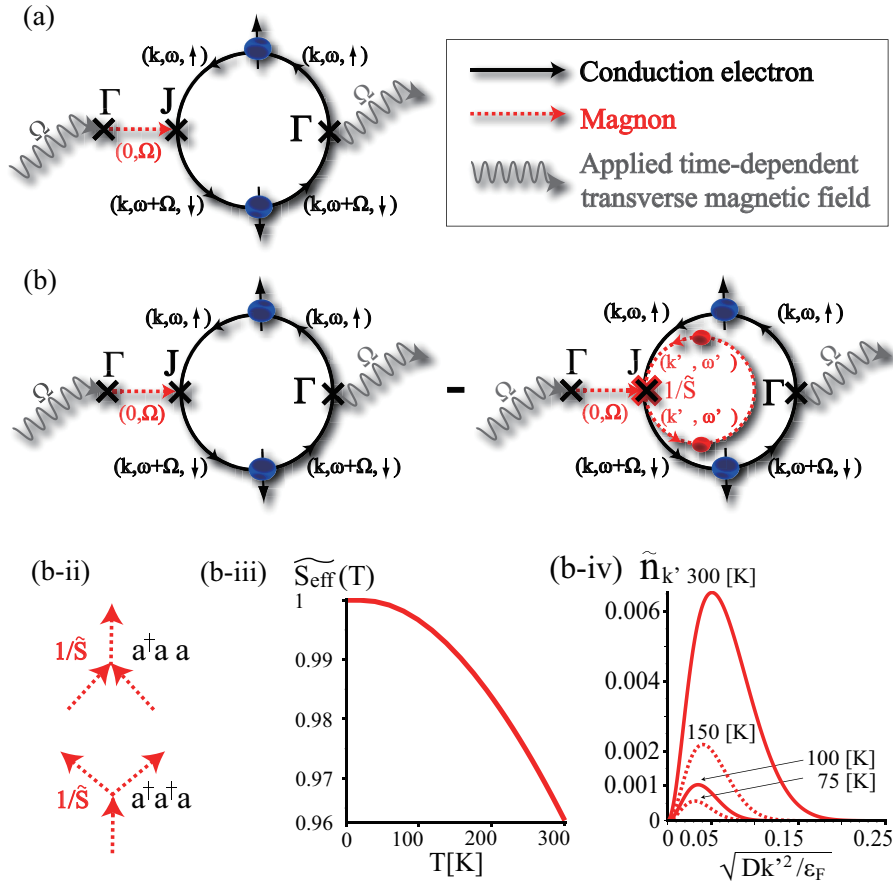
and

$$S_{\text{eff}}^{\text{ratio}}(T) := \frac{\widetilde{S}_{\text{eff}}(T)}{\widetilde{S}_{\text{eff}}(T = 300[\text{K}])}. \quad (44)$$

By using eq. (42), the dimensionless SRT at finite temperature can be rewritten as  $\mathcal{T}_s^{z\text{-ratio}}(T) = \eta^{\text{ratio}}(T) \times S_{\text{eff}}^{\text{ratio}}(T)$ . Consequently, it is clear that the variable  $\eta^{\text{ratio}}(T)$  represents the contribution of conduction electrons to spin pumping (see also eqs. (20) and (37)) and thus, it can be interpreted to correspond to the mixing conductance in the spin pumping theory proposed by Tserkovnyak et al.[3, 25, 4]

One can easily see from Fig. 4 that  $\eta^{\text{ratio}}(T)$  is little influenced by temperature;

$$\eta^{\text{ratio}}(T) \sim 1. \quad (45)$$



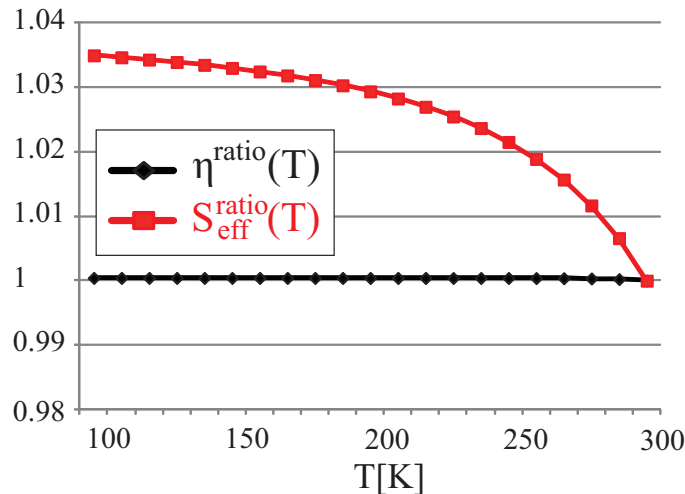
**Figure 3.** (a) A Feynman diagram of the SRT in the large- $S$  limit. (b) A Feynman diagram of the SRT with three-magnon splittings;  $\langle \mathcal{T}_s^z(n=0) \rangle$ . (b-ii) The schematic picture of three-magnon splittings. (b-iii) The plot of the function  $\widetilde{S}_{\text{eff}}(T) := S_{\text{eff}}(T)/S$  as a function of temperature  $T$  on the ESR point ( $\Omega = 2JS + B$ ). (b-iv) The plot of the dimensionless distribution function of magnons in the dimensionless wavenumber space on the ESR point ( $\Omega = 2JS + B$ );  $\tilde{n}_{k'}$ .

This temperature dependence of  $\eta^{\text{ratio}}(T)$  qualitatively shows the good agreement with the experimental result by Czeschka et al.[7] (i.e. the measurement of the mixing conductance under the standard spin pumping by FMR) that the mixing conductance is little influenced by temperature. That is, this temperature dependence (eq. (45)) is the common properties of spin pumping by FMR and ESR.

In conclusion, the temperature dependence of quantum spin pumping by ESR is determined mainly by  $S_{\text{eff}}(T)$ , which is governed by three-magnon splittings. On top of this,  $\eta^{\text{ratio}}(T)$ , which represents the contribution of conduction electrons to spin pumping and corresponds to the mixing conductance in the spin pumping theory proposed by Tserkovnyak et al., is little influenced by temperature. This qualitatively shows the good correspondence with the experiment by Czeschka et al.;[7] this temperature dependence (eq. (45)) is the common properties of spin pumping by FMR and ESR.

#### 4. Summary and discussion

We have clarified the temperature dependence of quantum spin pumping generated by ESR and have found the microscopic origin. When temperature rises, the pumped net spin current under ESR decreases; this is our theoretical prediction. This temperature dependence is governed by three-magnon splittings, which excite non-zero modes of magnons. On top of this,  $\eta^{\text{ratio}}(T)$ , which represents the contribution of conduction electrons to spin pumping and corresponds to the mixing conductance in the spin pumping theory proposed by Tserkovnyak et al., is little influenced by temperature. This qualitatively shows the good correspondence with the experiment by Czeschka et al.[7] That is, the temperature dependence (i.e.  $\eta^{\text{ratio}}(T) \sim 1$ ) is the common properties of spin pumping by FMR and ESR.



**Figure 4.** Plot of  $\eta^{\text{ratio}}(T)$  and  $S_{\text{eff}}^{\text{ratio}}(T)$  as a function of temperature  $T$  on the ESR point ( $\Omega = 2JS + B$ ). Compared with  $S_{\text{eff}}^{\text{ratio}}(T)$ , the function  $\eta^{\text{ratio}}(T)$  is little influenced by temperature;  $\eta^{\text{ratio}}(T) \sim 1$ .

Let us remark that we have theoretically predicted that the pumped net spin current by ESR decreases when temperature rises; this temperature dependence of quantum spin pumping by ESR will be experimentally confirmed by the inverse spin Hall effect.[6] Although external pumping magnetic fields are supposed to be applied to the whole of the sample as well as the interface due to the restriction of experimental techniques[24] (see Fig. 1), fortunately only the ESR at the interface occurs when the angular frequency is tuned to  $\Omega = 2JS + B$ . Other resonances take place in other regime; ESR at the non-magnetic metal and FMR at the interface and the ferromagnetic metal occur when  $\Omega = B$ . Thus by adjusting the angular frequency of the applied magnetic field, the temperature dependence of the pumped spin current purely by ESR at the interface will be observable.

On the other hand, to clarify the effects of the unusual energy dispersion of the lowest magnon mode in YIG, which is a relevant material to the experiment of magnon BEC[44, 45, 46] and spin pumping,[26, 2, 27] is a significant theoretical issue.

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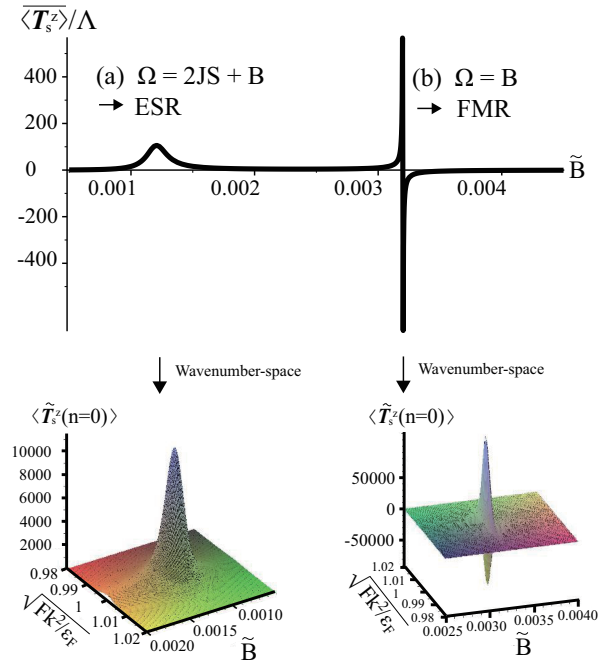
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## Appendix A. Quantum spin pumping by FMR and ESR

Fig. A1 shows  $\overline{\langle T_s^z \rangle} / \Lambda$  and  $\langle \widetilde{T}_s^z(n=0) \rangle$ . It is clear that sharp peaks exist on the point[42] (a)  $\Omega = 2JS + B$  and (b)  $\Omega = B$ . The effective magnetic field along the quantization axis for conduction electrons reads  $2JS + B$  and that for magnons (i.e. localized spins) does  $B$ . Therefore it can be concluded that the sharp peak on the point (a)  $\Omega = 2JS + B$  has originated from ESR and that on the point (b)  $\Omega = B$  from FMR. This is the natural result from the fact that at the interface quantum fluctuations (i.e. time-dependent transverse magnetic fields) affect conduction electrons as well as

localized spins (i.e. magnons) which is acting as effective magnetic fields for conduction electrons;  $2JS$ .

Last, let us stress that although in the present manuscript we have explicitly clarified that the mixing conductance under spin pumping by ESR is little influenced by temperature, our quantum spin pumping theory also shows that the mixing conductance under spin pumping by FMR[7] is little influenced by temperature;\* this theoretical result agrees with the experiment by Czeschka et al. [F. D. Czeschka et al., Phys. Rev. Lett., **107**, 046601 (2011)].



**Figure A1.** Plot of the SRT as a function of  $\tilde{B} := B/\epsilon_F$ ;  $\langle \tilde{T}_s^z \rangle / \Lambda$  and  $\langle \tilde{T}_s^z(n=0) \rangle$ . Sharp peaks exist on the point; (a)  $\Omega = 2JS + B$  and (b)  $\Omega = B$ , which has resulted from (a) ESR and (b) FMR.

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