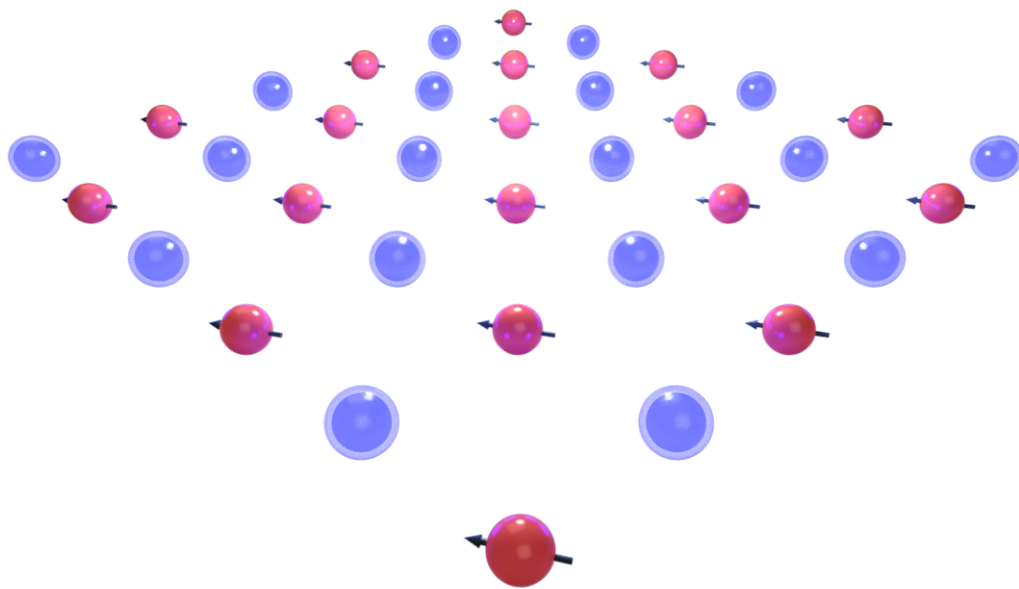


Generation of highly controllable ytterbium atom array: a hybrid system of nuclear spin and optical clock qubits towards quantum error correction



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Abstract

Neutral atoms trapped in optical tweezer arrays have emerged as a prominent platform for quantum simulation and computation. Their attractive features, such as scalable qubit arrays and flexible qubit connectivity enabled by moving tweezers, make them particularly suitable for implementing quantum error correction (QEC) toward fault-tolerant quantum computation. However, performing mid-circuit measurements—reading out ancilla qubits without disturbing data qubits—remains challenging. This difficulty arises from readout methods requiring global probe beam irradiation, which can induce heating and decoherence in the atom array.

In this study, we explore the potential of a dual-isotope ytterbium (Yb) atom array for mid-circuit measurements. The dual-Yb system consists of fermionic ^{171}Yb and bosonic ^{174}Yb isotopes. The nuclear spin qubit encoded in the ground state of ^{171}Yb serves as a data qubit, offering second-order long coherence times and MHz-scale fast qubit control. Meanwhile, the optical clock qubit in ^{174}Yb allows non-destructive readout, making it a promising candidate for the ancilla qubit.

We first achieved a dual-Yb system in a 10×10 optical tweezer array, obtaining a loading probability of 21(6) % for both isotopes while reducing dual occupancy, where both isotopes occupy the same site, to 0.2(7) % by optimizing the frequency of the light-assisted collision beam. Starting from this randomly loaded array, we constructed defect-free dual-Yb arrays using tweezer rearrangement, forming a robust starting point for quantum computation.

Next, we quantified the effect of imaging light for ^{174}Yb on the coherence of nuclear spin qubits in ^{171}Yb . By comparing Hahn-echo signals with and without imaging, we found that for a 20 ms exposure to imaging light, nuclear spin coherence was preserved at 99.1(1.8) % relative to the case without imaging. Simultaneously, we achieved a ^{174}Yb detection fidelity of 0.9992 and a survival probability after imaging of 0.988.

In addition, we developed a high-power ultraviolet laser system at a wavelength of 325 nm and output power of 800 mW. Using this laser, we observed Rabi oscillations between the metastable $(6s6p) \ ^3\text{P}_2$ and the Rydberg state $(6s71s) \ ^3\text{S}_1$, achieving a Rabi frequency of $2\pi \times 2.13(3)$ MHz. This result is a foundational step toward implementing high-fidelity two-qubit gates mediated by Rydberg interactions.

By integrating the dual-Yb atom array's non-destructive readout capabilities with fast Rydberg excitation for two-qubit gates, our findings represent significant progress toward QEC and fault-tolerant quantum computing.

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Chapter 1

Introduction

1.1 Neutral atoms in optical tweezers

Laser-cooled neutral atoms – cold atoms – [1–4] have been offering a versatile platform for quantum science and technology, owing to their high controllability and the cleanliness. Cold atoms in optical lattices have achieved significant breakthroughs, especially in quantum simulation of quantum many-body systems [5–9] and in quantum metrology [10–12], among many other fields.

In contrast to these top-down approaches, where the macroscopic behavior of the system is of primary importance, the bottom-up approach of quantum systems with individual atoms has also garnered significant interest [13]. Since the first demonstration of single-atom trapping in optical tweezers [14, 15] in 2001, the field has made rapid strides in the manipulation of individual atoms. These advancements include the cooling of single atoms to their motional ground state [16, 17], the observation of quantum phenomena [18, 19], and the implementation of quantum gate operations [20–27]. More recently, the realization of defect-free atom arrays using moving optical tweezers generated by acousto-optic deflectors [28, 29] or dynamical holographic tweezers [30] has unlocked significant potential for the optical tweezer array platform in quantum science.

The first application of neutral atoms in optical tweezer arrays is the quantum simulation of various quantum many-body systems [31]. By leveraging the long-range Rydberg interactions between atoms, it is possible to simulate spin models [32–41], topological models [42, 43], Hubbard models [44, 45], and other systems [46–50].

The second application is quantum metrology. The high controllability of individual atoms in optical tweezers enables the realization of high-precision atomic clocks [51–53]. Multibody entanglement generation [54] and spin squeezing [55, 56] using Rydberg interactions have been demonstrated, bringing quantum-enhanced metrology closer to realization.

The last, and arguably most prominent, application is quantum computing [57–61]. Neutral atoms, with inherently identical qubits encoded in atomic two-level systems, offer long coherence times and fast qubit control, making them strong candidates for quantum computing. The number of qubits in optical tweezer arrays has been rapidly increasing [62–71] (figure 1.1 (a)). Simultaneously, gate operation fidelities have improved significantly [54, 72–81] (figure 1.1 (b)), making the platform competitive with alternatives like trapped ions [82, 83] and superconducting qubits [84–87]. Moreover, coherent atom transport via moving optical tweezers [88, 89] enables flexible, all-to-all qubit connectivity, facilitating the implementation of various quantum error correction codes [90–92]. To date, logical qubit encoding and quantum error detection have been successfully demonstrated [89, 93, 94].

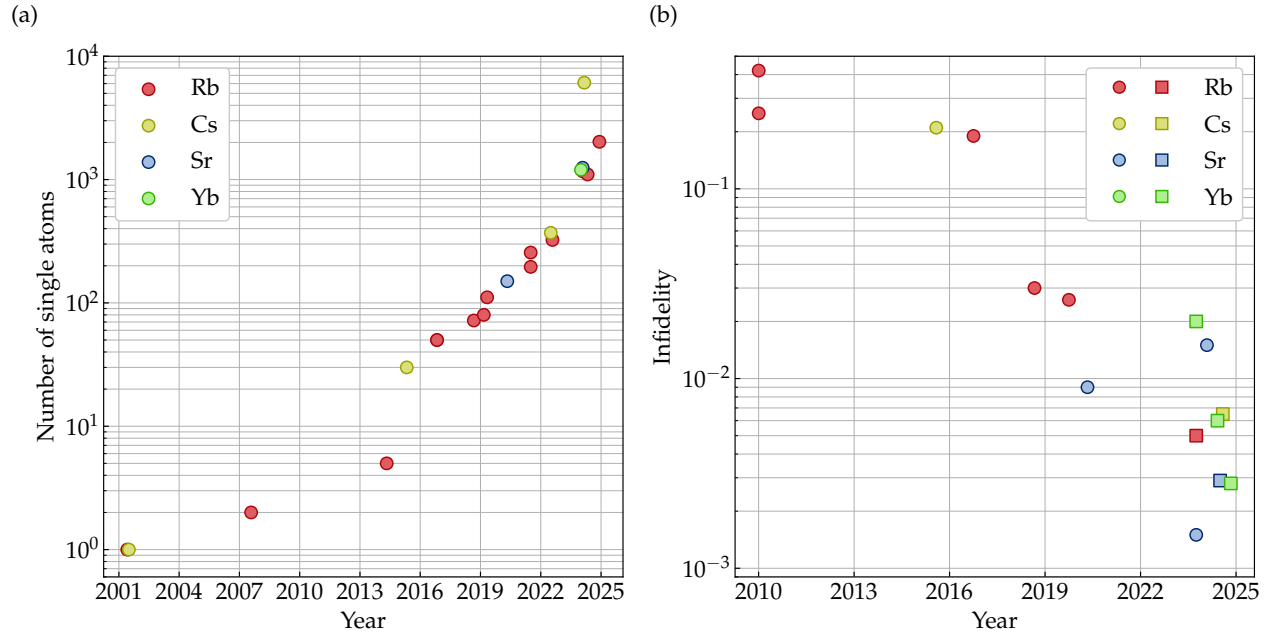


Figure 1.1 Progress in neutral atom tweezer arrays. (a) The number of single atoms in an optical tweezer array. Data are taken from Refs. [14, 15, 21, 26, 28, 29, 36, 37, 52, 62–64, 66–71, 95–97] (b) Gate fidelity in neutral atom-based quantum information processor. Circles and squares represent the Bell state fidelity and two-qubit gate fidelity, respectively. Data are taken from Refs. [22, 24, 25, 27, 54, 72–81].

1.2 Advantages of alkali-earth-like atoms

While alkali atoms, such as lithium [98, 99], sodium [100–102], potassium [103, 104], rubidium [14], and cesium [15], have been the primary choice for neutral atom experiments, alkali-earth(-like) atoms (AEA), including strontium [65, 105–110] and ytterbium [111–115], have recently emerged as attractive alternatives for optical tweezer experiments. The rich internal states of these atoms offer significant advantages for quantum computing.

The nuclear spin states of fermionic isotopes provide ideal qubits. Due to the absence of hyperfine coupling between the nuclear spin and electron—since the electronic angular momentum is canceled by two valence electrons—nuclear spin qubits exhibit coherence times that are three orders of magnitude longer than those of alkali atoms [107, 113, 116]. In particular, ytterbium-171 (^{171}Yb), with its nuclear spin of $1/2$, features the simplest nuclear spin structure, enabling long coherence times while simultaneously allowing fast qubit control by utilizing the optical transition with the excited state $^3\text{P}_1$ with a hyperfine coupling [113].

Metastable excited states, such as the $^3\text{P}_0$ and $^3\text{P}_2$ states, offer long-lived optical clock qubits [117] and single-photon Rydberg excitation [74]. These features enable high-fidelity two-qubit gates [54, 75, 78–81]. The optical-metastable-ground (OMG) architecture, originally proposed for trapped

ion systems [118], has been demonstrated with ytterbium atoms [75, 119], highlighting its potential for quantum error correction.

In the Rydberg states of AEAs, the inner valence electron is optically active, enabling Rydberg atom trapping [120] and light-shift-based Rydberg excitation control [121, 122]. Furthermore, fast Rydberg state detection via autoionization through excitation of the inner valence electron [74] facilitates erasure conversion [123], thereby enhancing the practicality of quantum error correction.

1.3 This work

This work focuses on advancing quantum computing with neutral atoms, a promising platform for scalable, fault-tolerant quantum computing. A key challenge in this field is the implementation of quantum error correction, which requires high-fidelity qubit operations, robust qubit initialization, and mid-circuit measurements to detect and correct errors without disturbing the quantum state. To address these challenges, we develop a hybrid atom tweezer array based on dual-isotope ytterbium atoms, leveraging the unique properties of both fermionic and bosonic isotopes to create an integrated system of nuclear spin and optical clock qubits (figure 1.2).

The fermionic isotope ^{171}Yb provides a robust nuclear spin qubit that benefits from long coherence times and fast control. These properties make ^{171}Yb an ideal candidate for data qubits that encode quantum information. In parallel, the bosonic isotope ^{174}Yb offers an optical clock qubit, which is advantageous for ancilla qubits due to its capacity for non-destructive, state-selective readout.

In the first stage of this work, we achieve efficient initialization and coherent control of nuclear spin qubits in ^{171}Yb atoms and optical clock qubits in ^{174}Yb atoms. We demonstrate coherent control

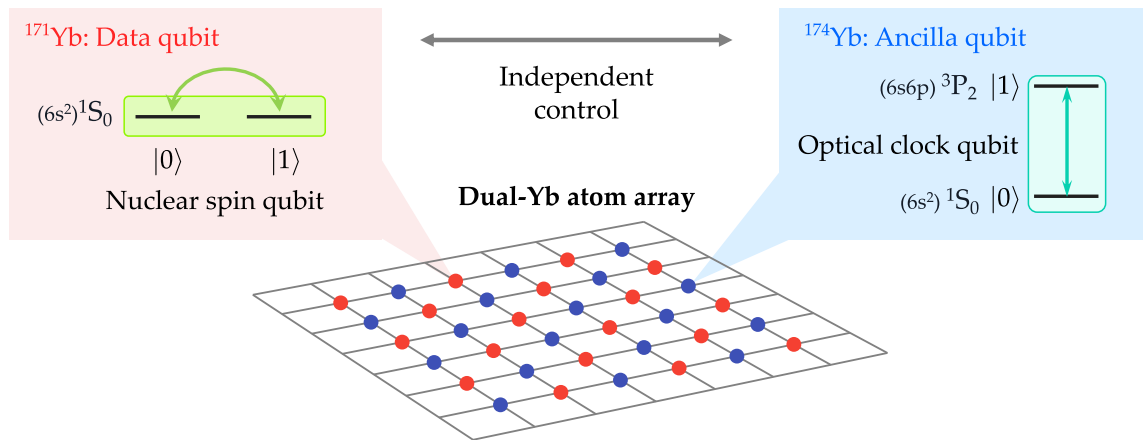


Figure 1.2 Concept of the dual-Yb atom array. The fermionic isotope ^{171}Yb provides robust nuclear spin qubits for data qubits, while the bosonic isotope ^{174}Yb offers optical clock qubits for ancilla qubits. The hybrid atom tweezer array enables low crosstalk measurements of the ancilla qubits, facilitating mid-circuit measurements for quantum error correction.

over these qubits, such as state preparation and single-qubit rotations.

Building on these capabilities, we construct a hybrid atom tweezer array composed of ^{171}Yb and ^{174}Yb atoms. This system demonstrates tunable dual occupancy of the tweezer traps, indicating a possibility of the simultaneous manipulation of data and ancilla qubits within the same experimental setup. A significant milestone is achieved by demonstrating low-crosstalk measurements of ^{174}Yb ancilla qubits while preserving the quantum state of ^{171}Yb data qubits. This represents a key step toward implementing mid-circuit measurements, a critical component for quantum error correction that allows error detection and correction in real time without collapsing the quantum state of neighboring qubits.

In parallel with the development of the hybrid tweezer array, we design and construct a high-power ultraviolet (UV) laser system tailored for fast and high-fidelity Rydberg excitation of ytterbium atoms. The UV laser is stabilized using a filtering cavity to suppress phase noise, which is essential for achieving high-fidelity qubit operations via the Rydberg interaction. Using this system, we demonstrate fast and coherent Rydberg excitation as well as a spectroscopic signature of Rydberg interactions. These results establish the foundation for implementing Rydberg-mediated two-qubit gates, which are essential for scalable quantum computing architectures.

1.4 Outline

This thesis is organized as follows:

- In chapter 2, we review the theoretical background of atom-light interactions, which play a crucial role in the coherent manipulation of nuclear spin and optical clock qubits, as well as in the trapping of ytterbium atoms.
- In chapter 3, we describe the basic properties of ytterbium atoms, including their energy levels and transition characteristics. We also summarize the properties of Rydberg atoms, which enable long-range interactions between atoms.
- In chapter 4, we present the experimental setup used in this work. This includes a description of experimental techniques such as optical tweezer generation, single-atom preparation, and detection.
- In chapter 5, we demonstrate the coherent manipulation of nuclear spin and optical clock qubits in ytterbium atoms. Specifically, we show efficient initialization and coherent control of nuclear spin qubits in a ^{171}Yb atom array, as well as narrow-line spectroscopy and coherent manipulation of optical clock qubits in a ^{174}Yb atom array.
- In chapter 6, we report on the construction of a hybrid atom tweezer array composed of ^{171}Yb and ^{174}Yb atoms. Experimental results on crosstalk measurements between the isotopes are also presented.

- In chapter 7, we describe the development of a high-power UV laser system tailored for the Rydberg excitation of ytterbium atoms and the observation of fast, coherent Rydberg excitation.
- In chapter 8, we conclude this thesis and provide an outlook on future research directions.

Journal publications

Below is a list of journal publications that have resulted from this work:

- **Hybrid atom tweezer array of nuclear spin and optical clock qubits**
Y. Nakamura, T. Kusano, R. Yokoyama, K. Saito, K. Higashi, N. Ozawa, T. Takano, Y. Takasu, and Y. Takahashi
Physical Review X **14**, 041062 (2024).
- **Development of a high-power ultraviolet laser system and observation of fast coherent Rydberg excitation of ytterbium**
Y. Nakamura, N. Ozawa, T. Kusano, R. Yokoyama, K. Shibata, T. Takano, Y. Takasu, and Y. Takahashi
Journal of Physical Society of Japan **94**, 014301 (2025). [Selected as Editors' choice]

Chapter 2

Atom-light interaction

In this chapter, we review the interaction between atoms and light. First, we discuss multipole transitions, which are essential for controlling qubits. Next, we introduce atomic polarizability and the dipole potential, which are crucial for atom trapping. Finally, we formulate the dynamics of both the internal and external degrees of freedom of atoms under light irradiation.

2.1 Multipole transitions

In this section, we review the multipole transitions between atoms and light, based on Refs. [124–126]. We consider the interaction between an atom and an electromagnetic field. The total Hamiltonian for a valence electron of an atom in an electromagnetic field $\mathbf{A}(\mathbf{r}, t)$ in the Coulomb gauge ($\nabla \cdot \mathbf{A} = 0$) is given by [127]

$$\mathcal{H} = \frac{1}{2m}(\mathbf{p} + e\mathbf{A})^2 + V(\mathbf{r}) + \frac{\epsilon_0}{2} \int d^3r (\mathbf{E}_\perp^2 + c^2 \mathbf{B}^2), \quad (2.1)$$

where e , m , $\mathbf{r}$, $\mathbf{p}$ is the charge, mass, position, and momentum of the valence electron, respectively. $V(\mathbf{r})$ is the potential energy which the electron feels from the other charged particles in the atom, and ϵ_0 , c , $\mathbf{E}_\perp = -\partial_t \mathbf{A}$, $\mathbf{B} = \nabla \times \mathbf{A}$ is the electric permittivity in vacuum, the speed of light, the transverse component of the electric field and the magnetic field, respectively. The interaction part of the Hamiltonian arises from the first term in equation (2.1) as

$$\mathcal{H}_{\text{int}} = \frac{e}{m} \mathbf{p} \cdot \mathbf{A} + \frac{e^2}{2m} \mathbf{A}^2 \simeq \frac{e}{m} \mathbf{p} \cdot \mathbf{A}. \quad (2.2)$$

Here, we have neglected the second-order term of $\mathbf{A}$ and used the fact that $[\mathbf{p}, \mathbf{A}] = 0$ in the Coulomb gauge. Additionally, we take into account the Zeeman interaction between the magnetic moment of the electron and the magnetic field, which is given by

$$\mathcal{H}_{\text{Zeeman}} = -\boldsymbol{\mu} \cdot \mathbf{B} = -\frac{eg_s}{2m} \mathbf{S} \cdot [\nabla \times \mathbf{A}], \quad (2.3)$$

where $\boldsymbol{\mu}$ is the magnetic moment of the electron, $\mathbf{S}$ is the spin operator, and g_s is the Landé g -factor. The total interaction Hamiltonian is then given by

$$\mathcal{H}_{\text{int}} = \frac{e}{m} \mathbf{p} \cdot \mathbf{A} - \frac{g_s e}{2m} \mathbf{S} \cdot [\nabla \times \mathbf{A}]. \quad (2.4)$$

The most common approach for derivation of the multipole transitions is to use Taylor expansion of the vector potential $\mathbf{A}(\mathbf{r}, t) = A_0 \hat{\mathbf{e}} \exp(i\mathbf{k} \cdot \mathbf{r} - i\omega t)$. Here, A_0 is the amplitude of the vector

potential, $\mathbf{k}$ is the wave vector, ω is the frequency of the field, and $\hat{\mathbf{e}}$ is the polarization vector. In the Coulomb gauge, the electric field is given by

$$\begin{aligned} \mathbf{E}(\mathbf{r}, t) &= -\partial_t \mathbf{A}(\mathbf{r}, t) = i\omega A_0 \hat{\mathbf{e}} \exp(i\mathbf{k} \cdot \mathbf{r} - i\omega t) \\ &= iE_0 \hat{\mathbf{e}} \exp(i\mathbf{k} \cdot \mathbf{r} - i\omega t), \end{aligned} \quad (2.5)$$

where $E_0 = \omega A_0$. Assuming that the wavelength of the light is much larger than the size of the atom ($\mathbf{k} \cdot \mathbf{r} \ll 1$), we can expand the vector potential as

$$\mathbf{A}(\mathbf{r}, t) = A_0 \hat{\mathbf{e}} \left(1 + i\mathbf{k} \cdot \mathbf{r} - \frac{1}{2}(\mathbf{k} \cdot \mathbf{r})^2 + \dots \right) \exp(-i\omega t). \quad (2.6)$$

The first term corresponds to the electric dipole (E1) transition, the second term to the electric quadrupole (E2) and magnetic dipole (M1) transitions, the third term to the electric octupole (E3) and magnetic quadrupole (M2) transitions, and so on.

As an example, we consider the electric dipole transition with this approach. The lowest order term of the interaction Hamiltonian in equation (2.4) becomes

$$\mathcal{H}_{\text{int}} = \frac{e}{m} \mathbf{p} \cdot \mathbf{A} \simeq i \frac{eE_0}{m\omega} \mathbf{p} \cdot \hat{\mathbf{e}} \exp(-i\omega t). \quad (2.7)$$

If the oscillation frequency of the light (ω) is close to the frequency of an atomic transition $|i\rangle \leftrightarrow |f\rangle$ (ω_{fi}), the coupling between the light and the atom is written as

$$i \frac{eE_0}{m\omega_{fi}} \langle f | \mathbf{p} | i \rangle \cdot \hat{\mathbf{e}} \exp(-i\omega_{fi} t). \quad (2.8)$$

We then define the coupling strength of the transition as

$$d_{fi} = \frac{1}{m\omega_{fi}} \langle f | \mathbf{p} | i \rangle \cdot \hat{\mathbf{e}}, \quad (2.9)$$

and recall that the commutation relation between the momentum operator $\mathbf{p}$ and the bare atomic Hamiltonian $\mathcal{H}_0 = \mathbf{p}^2/(2m)$ is written as $[\mathbf{r}, \mathcal{H}_0] = (i\hbar/m)\mathbf{p}$, where $\hbar = h/(2\pi)$ with the Planck constant h , the coupling strength can be rewritten as

$$\begin{aligned} d_{fi} &= \frac{1}{m\omega_{fi}} \langle f | \mathbf{p} | i \rangle \cdot \hat{\mathbf{e}} = \frac{1}{i\hbar\omega_{fi}} \langle f | [\mathbf{r}, \mathcal{H}_0] | i \rangle \cdot \hat{\mathbf{e}} \\ &= \frac{1}{i\hbar\omega_{fi}} (\langle f | \mathbf{r} \mathcal{H}_0 | i \rangle - \langle f | \mathcal{H}_0 \mathbf{r} | i \rangle) \cdot \hat{\mathbf{e}} \\ &= \frac{i}{\hbar} \langle f | \mathbf{r} | i \rangle \cdot \hat{\mathbf{e}}. \end{aligned} \quad (2.10)$$

Then, the coupling between the light and the atom (equation (2.8)) becomes

$$\begin{aligned} i \frac{eE_0}{m\omega_{fi}} \langle f | \mathbf{p} | i \rangle \cdot \hat{\mathbf{e}} \exp(-i\omega_{fi} t) &= ieE_0 \frac{i}{\hbar} \langle f | \mathbf{r} | i \rangle \cdot \hat{\mathbf{e}} \exp(-i\omega_{fi} t) \\ &= -\frac{\langle f | \mathbf{d} \cdot \hat{\mathbf{e}} | i \rangle E_0}{\hbar} \exp(-i\omega_{fi} t), \end{aligned} \quad (2.11)$$

where $\mathbf{d} = e\mathbf{r}$ is the electric dipole operator. The coupling strength $\Omega := -\langle f|\mathbf{d} \cdot \hat{\mathbf{e}}|i\rangle E_0/\hbar$ is so-called the Rabi frequency of the electric dipole transition.

We now consider the higher-order multipole transitions. Instead of using the Taylor expansion of the vector potential, we can use the vector spherical harmonics to expand the vector potential. The vector spherical harmonics $\mathbf{Y}_{K,l,q}$ are defined as

$$\mathbf{Y}_{K,l,q} = (-1)^{K-q} \sqrt{2K+1} \sum_{p=-1}^1 \begin{pmatrix} K & 1 & l \\ -q & p & q-p \end{pmatrix} Y_l^{q-p}(\theta, \phi) \hat{\mathbf{e}}_p, \quad (2.12)$$

where the parentheses denote the Wigner 3-j symbol, $Y_l^q(\theta, \phi)$ is the spherical harmonics, and $\hat{\mathbf{e}}_p$ is the unit vector of the p direction in spherical coordinate. The vector potential is then expanded as

$$\mathbf{A} = \sum_{K,l,q} \mathbf{A}_{K,l,q} \mathbf{Y}_{K,l,q}. \quad (2.13)$$

Here, $K = 1, 2, \dots$ represents the multipole order, where $K = 1$ corresponds to the dipole term, $K = 2$ to the quadrupole, and so on. The label l and q runs over $l = K-1, K, K+1$ and $|q| \leq l$, respectively. The expansion coefficients $A_{K,l,q}$ are given by

$$A_{K,l,q} = A_0 \int_0^\pi d\theta \sin \theta \int_0^{2\pi} d\phi \mathbf{Y}_{K,l,q}(\hat{\mathbf{r}}) \cdot \hat{\mathbf{e}} e^{i\mathbf{k} \cdot \mathbf{r}}. \quad (2.14)$$

Substituting the expansion of a plane wave in terms of the spherical Bessel function $j_l(x)$ and the spherical harmonics $Y_l^m(\theta, \phi)$,

$$e^{i\mathbf{k} \cdot \mathbf{r}} = 4\pi \sum_{l,q} i^l j_l(kr) Y_l^q(\hat{\mathbf{r}}) Y_l^{m*}(\hat{\mathbf{k}}) \quad (2.15)$$

with $\hat{\mathbf{r}} = \mathbf{r}/|\mathbf{r}|$ and $\hat{\mathbf{k}} = \mathbf{k}/|\mathbf{k}|$ into equation (2.14), we obtain

$$\mathbf{A}(\mathbf{r}) = 4\pi \sum_{K,l,q} i^l (\mathbf{Y}_{K,l,q}(\hat{\mathbf{k}}) \cdot \hat{\mathbf{e}}) j_l(kr) \mathbf{Y}_{K,l,q}(\hat{\mathbf{r}}). \quad (2.16)$$

Here we introduce another set of vector spherical harmonics $\mathbf{Y}_{K,q}^{(\mu)}$, which is defined as

$$\begin{aligned} \mathbf{Y}_{K,q}^{(-1)}(\hat{\mathbf{r}}) &= \sqrt{\frac{K}{2K+1}} \mathbf{Y}_{K,K-1,q}(\hat{\mathbf{r}}) - \sqrt{\frac{K+1}{2K+1}} \mathbf{Y}_{K,K+1,q}(\hat{\mathbf{r}}) = \hat{\mathbf{r}} Y_K^q(\hat{\mathbf{r}}), \\ \mathbf{Y}_{K,q}^{(0)}(\hat{\mathbf{r}}) &= \mathbf{Y}_{K,K,q}(\hat{\mathbf{r}}) = \frac{1}{\sqrt{K(K+1)}} (-i\mathbf{r} \times \nabla) Y_K^q(\hat{\mathbf{r}}), \\ \mathbf{Y}_{K,q}^{(1)}(\hat{\mathbf{r}}) &= \sqrt{\frac{K+1}{2K+1}} \mathbf{Y}_{K,K-1,q}(\hat{\mathbf{r}}) + \sqrt{\frac{K}{2K+1}} \mathbf{Y}_{K,K+1,q}(\hat{\mathbf{r}}) = \frac{r}{\sqrt{K(K+1)}} \nabla Y_K^q(\hat{\mathbf{r}}). \end{aligned} \quad (2.17)$$

Using these $\mathbf{Y}_{K,q}^{(\mu)}$, the expansion of the vector potential in equation (2.16) can be rewritten as

$$\mathbf{A}(\mathbf{r}) = 4\pi \sum_{K,q,\mu} i^{K-\mu} (\mathbf{Y}_{K,q}^{(\mu)}(\hat{\mathbf{k}}) \cdot \hat{\mathbf{e}}) \mathbf{a}_{K,q}^{(\mu)}(\hat{\mathbf{r}}), \quad (2.18)$$

with

$$\begin{aligned} \mathbf{a}_{K,q}^{(0)}(\hat{\mathbf{r}}) &= A_0 j_K(kr) \mathbf{Y}_{K,q}^{(0)}(\hat{\mathbf{r}}), \\ \mathbf{a}_{K,q}^{(1)}(\hat{\mathbf{r}}) &= A_0 \left[\sqrt{\frac{K+1}{2K+1}} j_{K-1}(kr) \mathbf{Y}_{K,K-1,q}(\hat{\mathbf{r}}) - \sqrt{\frac{K}{2K+1}} j_{K+1}(kr) \mathbf{Y}_{K,K+1,q}(\hat{\mathbf{r}}) \right]. \end{aligned} \quad (2.19)$$

Here, $\mu = 0$ and $\mu = 1$ corresponds to the magnetic and electric multipole components respectively. Note that since $\mathbf{Y}_{K,q}^{(-1)}(\hat{\mathbf{k}}) \propto \hat{\mathbf{k}}$, its contribution to the vector potential is zero under the assumption that the laser propagation axis is orthogonal to the polarization vector ($\mathbf{k} \cdot \hat{\mathbf{e}} = 0$). Substituting equation (2.19) into equation (2.4), the interaction Hamiltonian becomes

$$\begin{aligned} \mathcal{H}_{\text{int}} &= 4\pi \sum_{\mu=0,1} \sum_{K=1}^{\infty} \sum_{q=-K}^K i^{K-\mu} (\mathbf{Y}_{K,q}^{(\mu)}(\hat{\mathbf{k}}) \cdot \hat{\mathbf{e}}) \left[\frac{e}{m} \mathbf{p} \cdot \mathbf{a}_{K,q}^{(\mu)}(\hat{\mathbf{r}}) + \frac{eg_s}{2m} \mathbf{S} \cdot (\nabla \times \mathbf{a}_{K,q}^{(\mu)}(\hat{\mathbf{r}})) \right] \\ &:= 4\pi \sum_{\mu=0,1} \sum_{K=1}^{\infty} \sum_{q=-K}^K i^{K-\mu} (\mathbf{Y}_{K,q}^{(\mu)}(\hat{\mathbf{k}}) \cdot \hat{\mathbf{e}}) \mathcal{H}_{K,q}^{(\mu)}. \end{aligned} \quad (2.20)$$

2.1.1 Electric multipole transitions

The electric multipole component in the interaction Hamiltonian (equation (2.20)) is given by

$$\mathcal{H}_{K,q}^{(\text{el})} := \mathcal{H}_{K,q}^{(1)} = \frac{e}{m} \mathbf{p} \cdot \mathbf{a}_{K,q}^{(1)}(\hat{\mathbf{r}}). \quad (2.21)$$

Note that since $\nabla \times \mathbf{a}_{K,q}^{(1)} \propto \nabla \times (\hat{\mathbf{r}} + \nabla Y_K^q) = 0$, the electric components does not contain the coupling to the spin operator $\mathbf{S}$. Using the equations (2.17) and (2.20), the electric multipole component is rewritten as

$$\begin{aligned} \mathbf{a}_{K,q}^{(1)}(\hat{\mathbf{r}}) &= A_0 \left[\sqrt{\frac{K+1}{2K+1}} j_{K-1}(kr) \mathbf{Y}_{K,K-1,q}(\hat{\mathbf{r}}) - \sqrt{\frac{K}{2K+1}} j_{K+1}(kr) \mathbf{Y}_{K,K+1,q}(\hat{\mathbf{r}}) \right] \\ &= A_0 \frac{\sqrt{K(K+1)}}{2K+1} \mathbf{Y}_{K,q}^{(-1)}(\hat{\mathbf{r}}) [j_{K-1}(kr) + j_{K+1}(kr)] \\ &\quad + A_0 \frac{1}{2K+1} \mathbf{Y}_{K,q}^{(1)}(\hat{\mathbf{r}}) [(K+1)j_{K-1}(kr) - Kj_{K+1}(kr)] \\ &= A_0 \sqrt{K(K+1)} \frac{j_K(kr)}{kr} \mathbf{Y}_{K,q}^{(-1)}(\hat{\mathbf{r}}) + A_0 \left[\frac{j_K(kr)}{kr} + \frac{\partial j_K(kr)}{\partial(kr)} \right] \mathbf{Y}_{K,q}^{(1)}(\hat{\mathbf{r}}). \end{aligned} \quad (2.22)$$

Here we used the recurrence relation of the spherical Bessel function as

$$\begin{aligned} j_{K-1}(kr) &= \frac{K+1}{kr} j_K(kr) + \frac{\partial j_K(kr)}{\partial(kr)}, \\ j_{K+1}(kr) &= \frac{K}{kr} j_K(kr) - \frac{\partial j_K(kr)}{\partial(kr)}. \end{aligned} \quad (2.23)$$

To further proceed the calculation, we introduce a gauge transformation of the vector potential as

$$\begin{aligned} \mathbf{A}(\mathbf{r}, t) &\rightarrow \mathbf{A}'(\mathbf{r}, t) = \mathbf{A}(\mathbf{r}, t) + \nabla \chi(\mathbf{r}, t), \\ \Phi(\mathbf{r}, t) &\rightarrow \Phi'(\mathbf{r}, t) = \Phi(\mathbf{r}, t) - \partial_t \chi(\mathbf{r}, t), \end{aligned} \quad (2.24)$$

with a gauge function $\chi(\mathbf{r}, t)$ of

$$\chi(\mathbf{r}, t) = -\frac{A_0}{k} \sqrt{\frac{K+1}{K}} j_K(kr) Y_K^q(\hat{\mathbf{r}}) e^{i\omega t}. \quad (2.25)$$

This gauge transformation results in the following vector and scalar potentials as

$$\begin{aligned} \mathbf{a}_{K,q}^{(1)}(\hat{\mathbf{r}}) &\rightarrow -A_0 j_{K+1}(kr) \left[\mathbf{Y}_{K,q}^{(1)}(\hat{\mathbf{r}}) - \sqrt{\frac{K+1}{K}} \mathbf{Y}_{K,q}^{(-1)}(\hat{\mathbf{r}}) \right], \\ \Phi(\mathbf{r}) &\rightarrow -i\omega\chi(\mathbf{r}, t), \end{aligned} \quad (2.26)$$

and thus the interaction Hamiltonian of equation (2.21) becomes

$$\begin{aligned} \mathcal{H}_{K,q}^{(\text{el})} &= \frac{e}{m} \mathbf{p} \cdot \mathbf{a}_{K,q}^{(1)}(\hat{\mathbf{r}}) + e\Phi(\mathbf{r}) \\ &= -\frac{eA_0}{m} j_{K+1}(kr) \mathbf{p} \cdot \left[\mathbf{Y}_{K,q}^{(1)}(\hat{\mathbf{r}}) - \sqrt{\frac{K+1}{K}} \mathbf{Y}_{K,q}^{(-1)}(\hat{\mathbf{r}}) \right] + i\frac{\omega e A_0}{k} \sqrt{\frac{K+1}{K}} j_K(kr) Y_K^q(\hat{\mathbf{r}}). \end{aligned} \quad (2.27)$$

By assuming the condition of $kr \ll 1$ and using the asymptotic form of the spherical Bessel function as $j_K(kr) \simeq (kr)^K / (2K+1)!!$, the lowest order term of the electric multipole Hamiltonian is given by

$$\begin{aligned} \mathcal{H}_{K,q}^{(\text{el})} &\simeq i\frac{\omega e A_0}{k} \sqrt{\frac{K+1}{K}} \frac{(kr)^K}{(2K+1)!!} Y_K^q(\hat{\mathbf{r}}) \\ &= icA_0 \sqrt{\frac{(2K+1)(K+1)}{4\pi K}} \frac{k^K}{(2K+1)!!} e \sqrt{\frac{4\pi}{2K+1}} r^K Y_K^q(\hat{\mathbf{r}}) \\ &= icA_0 \sqrt{\frac{(2K+1)(K+1)}{4\pi K}} \frac{k^K}{(2K+1)!!} Q_{K,q}^{(\text{el})}, \end{aligned} \quad (2.28)$$

with $c = \omega/k$ is the speed of light, and $Q_{K,q}^{(\text{el})}$ in the last line is the electric multipole transition operator defined as

$$Q_{K,q}^{(\text{el})} = e \sqrt{\frac{4\pi}{2K+1}} r^K Y_K^q(\hat{\mathbf{r}}). \quad (2.29)$$

This operator is a tensor operator with rank K .

The obtained electric multipole Hamiltonian $\mathcal{H}_{K,q}^{(\text{el})}$ determines the transition rules of the electric multipole transitions. Using a parity operator $\mathcal{P}$, we find that

$$\mathcal{P} \mathcal{H}_{K,q}^{(\text{el})} = \mathcal{P} [r^K Y_K^q(\hat{\mathbf{r}})] = r^K Y_K^q(-\hat{\mathbf{r}}) = (-1)^K r^K Y_K^q(\hat{\mathbf{r}}), \quad (2.30)$$

and thus the Hamiltonian has the parity of $(-1)^K$. Considering that the parity of the atomic state is determined by the angular momentum L as $(-1)^L$, and the parity must be conserved in the transition, the E1 transition ($K = 1$) is allowed only when $\Delta L = \pm 1$, and the E2 transition ($K = 2$) is allowed only when $\Delta L = 0, \pm 2$ and so on, where ΔL is the change of the angular momentum in the transition.

Additional transition rules arises from the coupling to the angular momentum of the light. The matrix element of the electric multipole transition operator $Q_{K,q}^{(\text{el})}$ between the atomic states $|i\rangle = |\gamma, J, m\rangle$ and $|f\rangle = |\gamma', J', m'\rangle$ is obtained by employing the Wigner-Eckart theorem as

$$\langle \gamma', J', m' | Q_{K,q}^{(\text{el})} | \gamma, J, m \rangle = (-1)^{J'-m'} \begin{pmatrix} J' & K & J \\ -m' & q & m \end{pmatrix} \langle \gamma', J' || Q_{K,q}^{(\text{el})} || \gamma, J \rangle. \quad (2.31)$$

Here, γ is the radial wavefunction, J and m is the total angular momentum and its projection onto the quantization axis, respectively. The parentheses represent the Wigner 3j-symbol, and $\langle \gamma', J' || Q_{K,q}^{(\text{el})} || \gamma, J \rangle$ is the reduced matrix element. The Wigner 3j-symbols follow the triangle condition $|J' - K| \leq J \leq J' + K$, which results in the additional selection rules. For example, the E1 transition ($K = 1$) prohibits $J = 0 \leftrightarrow J' = 0$, and the E2 transition ($K = 2$) prohibits $J = 0 \leftrightarrow J' = 0, 1$. We also find the restriction to K and q as $K \geq |\Delta J| = |J' - J|$ and $q = \Delta m = m' - m$.

2.1.2 Magnetic multipole transitions

The magnetic multipole component in the interaction Hamiltonian (equation (2.20)) is given by

$$\mathcal{H}_{K,q}^{(\text{mg})} := \mathcal{H}_{K,q}^{(0)} = \frac{e}{m} \mathbf{p} \cdot \mathbf{a}_{K,q}^{(0)}(\hat{\mathbf{r}}) + \frac{e g_s}{2m} \mathbf{S} \cdot (\nabla \times \mathbf{a}_{K,q}^{(0)}(\hat{\mathbf{r}})). \quad (2.32)$$

Using the following relations

$$\mathbf{L} = \mathbf{r} \times \mathbf{p} = -i\hbar \mathbf{r} \times \nabla, \quad \nabla = \hat{\mathbf{r}} \frac{\partial}{\partial r} - \frac{i}{\hbar r^2} \mathbf{r} \times \mathbf{L}, \quad (2.33)$$

the first term of the magnetic multipole Hamiltonian is written as

$$\begin{aligned} \mathbf{p} \cdot \mathbf{a}_{K,q}^{(0)}(\hat{\mathbf{r}}) &= -i\hbar A_0 \left(\hat{\mathbf{r}} \frac{\partial}{\partial r} - \frac{i}{\hbar r^2} \mathbf{r} \times \mathbf{L} \right) \cdot [j_K(kr) \mathbf{Y}_{K,q}^{(0)}(\hat{\mathbf{r}})] \\ &= -\hbar A_0 \hat{\mathbf{r}} \cdot \frac{\mathbf{r} \times \nabla}{\sqrt{K(K+1)}} Y_K^q(\hat{\mathbf{r}}) \frac{\partial j_K(kr)}{\partial r} - A_0 \frac{j_K(kr)}{r^2} (\mathbf{r} \times \mathbf{L}) \cdot \mathbf{Y}_{K,q}^{(0)}(\hat{\mathbf{r}}) \\ &= -iA_0 \hat{\mathbf{r}} \cdot \frac{\mathbf{L}}{\sqrt{K(K+1)}} Y_K^q(\hat{\mathbf{r}}) \frac{\partial j_K(kr)}{\partial r} - A_0 \frac{j_K(kr)}{r^2} (\mathbf{r} \times \mathbf{Y}_{K,q}^{(0)}(\hat{\mathbf{r}})) \cdot \mathbf{L} \\ &= -A_0 \frac{j_K(kr)}{r^2} (\mathbf{r} \times \mathbf{Y}_{K,q}^{(0)}(\hat{\mathbf{r}})) \cdot \mathbf{L}. \end{aligned} \quad (2.34)$$

Here, we used the fact that $\mathbf{Y}_{K,q}^{(0)}(\hat{\mathbf{r}}) \propto \mathbf{L}$ and thus $(\mathbf{r} \times \mathbf{L}) \cdot \mathbf{Y}_{K,q}^{(0)}(\hat{\mathbf{r}}) = (\mathbf{r} \times \mathbf{Y}_{K,q}^{(0)}(\hat{\mathbf{r}})) \cdot \mathbf{L}$ in the third line, and $\hat{\mathbf{r}} \cdot \mathbf{L} = 0$ in the last line. This expression is further simplified as

$$\begin{aligned} \mathbf{p} \cdot \mathbf{a}_{K,q}^{(0)}(\hat{\mathbf{r}}) &= iA_0 \frac{j_K(kr)}{r^2} \left[\mathbf{r} \times \frac{\mathbf{r} \times \nabla}{\sqrt{K(K+1)}} Y_K^q(\hat{\mathbf{r}}) \right] \cdot \mathbf{L} \\ &= iA_0 \frac{j_K(kr)}{r^2} \left[(\mathbf{r} - r^2 \nabla) \frac{1}{\sqrt{K(K+1)}} Y_K^q(\hat{\mathbf{r}}) \right] \cdot \mathbf{L} \\ &= iA_0 \left[\frac{\partial j_K(kr)}{\partial r} \frac{\hat{\mathbf{r}}}{\sqrt{K(K+1)}} Y_K^q(\hat{\mathbf{r}}) - \frac{j_K(kr)}{\sqrt{K(K+1)}} \nabla Y_K^q(\hat{\mathbf{r}}) \right] \cdot \mathbf{L} \\ &= i \frac{A_0}{\sqrt{K(K+1)}} \nabla [j_K(kr) Y_K^q(\hat{\mathbf{r}})] \cdot \mathbf{L}. \end{aligned} \quad (2.35)$$

The second term in equation (2.32) is calculated as

$$\begin{aligned}
 \nabla \times \mathbf{a}_{K,q}^{(0)}(\hat{\mathbf{r}}) &= -iA_0(\nabla \times \mathbf{r} \times \nabla)j_K(kr)\frac{1}{\sqrt{K(K+1)}}Y_K^q(\hat{\mathbf{r}}) \\
 &= -iA_0(\mathbf{r}\nabla^2 - \nabla(\mathbf{r} \cdot \nabla + 1))j_K(kr)\frac{1}{\sqrt{K(K+1)}}Y_K^q(\hat{\mathbf{r}}) \\
 &= iA_0\sqrt{\frac{K+1}{K}}\nabla[j_K(kr)Y_K^q(\hat{\mathbf{r}})].
 \end{aligned} \tag{2.36}$$

In the above calculation, we used $\nabla^2 \mathbf{a}_{K,q}^{(0)} = 0$, and the first term in the second line vanishes. Finally, we obtain the magnetic multipole Hamiltonian as

$$\mathcal{H}_{K,q}^{\text{mg}} = i\frac{eA_0}{m}\frac{1}{\sqrt{K(K+1)}}\nabla[j_K(kr)Y_K^q(\hat{\mathbf{r}})] \cdot \mathbf{L} + i\frac{eg_sA_0}{2m}\sqrt{\frac{K+1}{K}}\nabla[j_K(kr)Y_K^q(\hat{\mathbf{r}})] \cdot \mathbf{S}. \tag{2.37}$$

We again assume $kr \ll 1$ and use the asymptotic form of the spherical Bessel function to obtain the lowest order term of the magnetic multipole Hamiltonian as

$$\begin{aligned}
 \mathcal{H}_{K,q}^{\text{mg}} &= iA_0\sqrt{\frac{(2K+1)(K+1)}{4\pi K}}\frac{k^K}{(2K+1)!!}\frac{e\hbar}{2m}\sqrt{\frac{4\pi}{2K+1}}\nabla[r^K Y_K^q(\hat{\mathbf{r}})] \cdot \left[\frac{2}{K+1}\frac{\mathbf{L}}{\hbar} + g_s\frac{\mathbf{S}}{\hbar}\right] \\
 &= iA_0\sqrt{\frac{(2K+1)(K+1)}{4\pi K}}\frac{k^K}{(2K+1)!!}Q_{K,q}^{(\text{mg})},
 \end{aligned} \tag{2.38}$$

where $Q_{K,q}^{(\text{mg})}$ is the magnetic multipole transition operator defined as

$$Q_{K,q}^{(\text{mg})} = \frac{e\hbar}{2m}\sqrt{\frac{4\pi}{2K+1}}\nabla[r^K Y_K^q(\hat{\mathbf{r}})] \cdot \left[\frac{2}{K+1}\frac{\mathbf{L}}{\hbar} + g_s\frac{\mathbf{S}}{\hbar}\right]. \tag{2.39}$$

This operator is also a tensor operator with rank K .

Similar to the electric multipole transitions, the magnetic multipole Hamiltonian $\mathcal{H}_{K,q}^{\text{mg}}$ determines the transition rules of the magnetic multipole transitions. Using a parity operator $\mathcal{P}$, we find that

$$\mathcal{P}\mathcal{H}_{K,q}^{\text{mg}} = \mathcal{P}\left[\nabla[r^K Y_K^q(\hat{\mathbf{r}})] \cdot (\mathbf{L} + \mathbf{S})\right] = (-1)^{K+1}\nabla[r^K Y_K^q(\hat{\mathbf{r}})] \cdot (\mathbf{L} + \mathbf{S}), \tag{2.40}$$

and the Hamiltonian has the parity of $(-1)^{K+1}$, which is opposite to the electric multipole Hamiltonian. Therefore, the M1 transition ($K = 1$) is allowed only when $\Delta L = 0$, and the M2 transition ($K = 2$) is allowed only when $\Delta L = \pm 1$ and so on.

The matrix element of the magnetic multipole transition operator $Q_{K,q}^{(\text{mg})}$ between the atomic states $|i\rangle = |\gamma, J, m\rangle$ and $|f\rangle = |\gamma', J', m'\rangle$ is obtained by employing the Wigner-Eckart theorem as

$$\langle \gamma', J', m' | Q_{K,q}^{(\text{mg})} | \gamma, J, m \rangle = (-1)^{J'-m'} \begin{pmatrix} J' & K & J \\ -m' & q & m \end{pmatrix} \langle \gamma', J' || Q_{K,q}^{(\text{mg})} || \gamma, J \rangle. \tag{2.41}$$

This form is the same as the electric multipole transition matrix element in equation (2.31), and thus the selection rules on the total angular momentum of the magnetic multipole transitions are the same as those of the electric multipole transitions.

2.1.3 Selection rules and relative transition strength of multipole transitions

In the previous sections, we obtained the selection rules of the electric and magnetic multipole transitions. These are determined by equations (2.30), (2.31), (2.40) and (2.41), and summarized in table 2.1.

Table 2.1 Selection rules of multipole transitions.

Transition	Parity	L	J	Example
E1	$\mathcal{P}_i = -\mathcal{P}_f$	$\Delta L = \pm 1$	$\Delta J = 0, \pm 1 (0 \nleftrightarrow 0)$	$^1S_0 \leftrightarrow ^1P_1$
M1	$\mathcal{P}_i = \mathcal{P}_f$	$\Delta L = 0$	$\Delta J = 0, \pm 1 (0 \nleftrightarrow 0)$	$^3P_1 \leftrightarrow ^3P_2$
E2	$\mathcal{P}_i = \mathcal{P}_f$	$\Delta L = 0, \pm 2$	$\Delta J = 0, \pm 1, \pm 2 (0 \nleftrightarrow 0, 1)$	$^2S_{1/2} \leftrightarrow ^2D_{5/2}$
M2	$\mathcal{P}_i = -\mathcal{P}_f$	$\Delta L = \pm 1$	$\Delta J = 0, \pm 1, \pm 2 (0 \nleftrightarrow 0, 1)$	$^1S_0 \leftrightarrow ^3P_2$

The relative transition strength between the electric and magnetic multipole transitions is estimated by comparing the Hamiltonians of equations (2.28) and (2.38). The strength of each transitions is proportional to the square of the matrix element as

$$\frac{\Gamma_{K,q}^{(\text{mg})}}{\Gamma_{K,q}^{(\text{el})}} = \frac{|\mathcal{H}_{K,q}^{(\text{mg})}|^2}{|\mathcal{H}_{K,q}^{(\text{el})}|^2} \propto \left(\frac{e\hbar}{2m} r^{K-1} \right)^2 = \left(\frac{\hbar}{2mcr} \right)^2. \quad (2.42)$$

In atomic systems, the radius r is approximately expressed as a_0/Z_{eff} , where $a_0 = \hbar/(mc\alpha)$ is the Bohr radius, Z_{eff} is the effective charge of the nucleus, and α is the fine structure constant. Therefore, the relative strength of the magnetic multipole transitions to the electric multipole transitions is estimated as

$$\frac{\Gamma_{K,q}^{(\text{mg})}}{\Gamma_{K,q}^{(\text{el})}} \propto (\alpha Z_{\text{eff}})^2. \quad (2.43)$$

Since the fine structure constant $\alpha \simeq 1/137$ is a small number, the magnetic multipole transitions are much weaker than the electric multipole transitions. Furthermore, the relative strength of the electric multipole transitions with different K is estimated as

$$\frac{\Gamma_{K+1,q}^{(\text{el})}}{\Gamma_{K,q}^{(\text{el})}} = \frac{|\mathcal{H}_{K,q}^{(\text{mg})}|^2}{|\mathcal{H}_{K,q}^{(\text{el})}|^2} \propto (kr)^2. \quad (2.44)$$

The wavenumber $k = E/(\hbar c)$ is propotional to the transition energy E , which is approximately expressed as $E \propto Z_{\text{eff}}^2 \alpha^2 mc^2$. Therefore, the relative strength of the electric multipole transitions with different K is estimated as

$$\frac{\Gamma_{K+1,q}^{(\text{el})}}{\Gamma_{K,q}^{(\text{el})}} \propto \left(Z_{\text{eff}}^2 \alpha^2 \frac{mc}{\hbar} \cdot \frac{\hbar}{Z_{\text{eff}} mc \alpha} \right)^2 = (\alpha Z_{\text{eff}})^2. \quad (2.45)$$

Equations (2.43) and (2.45) is the reason why the electric dipole transitions are the most dominant transitions in atomic systems.

2.1.4 Angular dependence of multipole transition strength

As we derived in the previous sections, the interaction Hamiltonian of the multipole transitions is written as

$$\mathcal{H}_{\text{int}} = 4\pi \sum_{\mu=0,1} \sum_{K=1}^{\infty} \sum_{q=-K}^K i^{K-\mu} (\mathbf{Y}_{K,q}^{(\mu)}(\hat{\mathbf{k}}) \cdot \hat{\mathbf{e}}) \mathcal{H}_{K,q}^{(\mu)}. \quad (2.46)$$

In the above expression, we can find that the multipole transition also depends on the polarization and propagation axis of the light via the term of

$$C_{K,q}^{(\mu)} = \mathbf{Y}_{K,q}^{(\mu)}(\hat{\mathbf{k}}) \cdot \hat{\mathbf{e}}. \quad (2.47)$$

The transition strength of the multipole transitions is determined by $A_{K,q}^{(\mu)} = |C_{K,q}^{(\mu)}|^2$. To obtain explicit form of the angular dependence of the transition strength, we adopt following coordinates as

$$\begin{aligned} \hat{\mathbf{B}} &= \hat{\mathbf{z}} \\ \hat{\mathbf{k}} &= \sin(\theta)\hat{\mathbf{x}} + \cos(\theta)\hat{\mathbf{z}} \\ \hat{\mathbf{e}}_1 &= \hat{\mathbf{B}} \times \hat{\mathbf{k}} = \hat{\mathbf{y}} \\ \hat{\mathbf{e}}_2 &= \hat{\mathbf{k}} \times \hat{\mathbf{e}}_1 = -\cos(\theta)\hat{\mathbf{x}} + \sin(\theta)\hat{\mathbf{z}}, \end{aligned} \quad (2.48)$$

where $\hat{\mathbf{B}}$ is the quantization axis (figure 2.1 (a)). The polarization of the light is represented using a normalized Jones vector (α, β) as $\hat{\mathbf{e}} = \alpha\hat{\mathbf{e}}_1 + \beta\hat{\mathbf{e}}_2$ with $|\alpha|^2 + |\beta|^2 = 1$. Note that in this coordinate the polarization and propagation axis of the light are orthogonal to each other ($\hat{\mathbf{k}} \cdot \hat{\mathbf{e}} = 0$), and the azimuthal angle $\phi = 0$, that is $\mathbf{Y}_{K,q}^{(\mu)}(\hat{\mathbf{k}}) = \mathbf{Y}_{K,q}^{(\mu)}(\theta, 0)$. Recalling equations (2.12) and (2.17), $\mathbf{Y}_{K,q}^{(\mu)}(\theta, 0)$ is written as

$$\begin{aligned} \mathbf{Y}_{K,q}^{(1)}(\theta, 0) &= \sum_{p=-1}^1 \left[c_{K,q,p}^{(+)} Y_{K+1}^{q-p}(\theta, 0) + c_{K,q,p}^{(-)} Y_{K-1}^{q-p}(\theta, 0) \right] \hat{\mathbf{e}}_p, \\ \mathbf{Y}_{K,q}^{(0)}(\theta, 0) &= \sum_{p=-1}^1 c_{K,q,p}^{(0)} Y_K^{q-p}(\theta, 0) \hat{\mathbf{e}}_p, \end{aligned} \quad (2.49)$$

where the coefficients $c_{K,q,p}^{(\pm,0)}$ is the weight of the projection of the spherical harmonics onto the p direction and can be obtained from the definition of the vector spherical harmonics (equation (2.12))

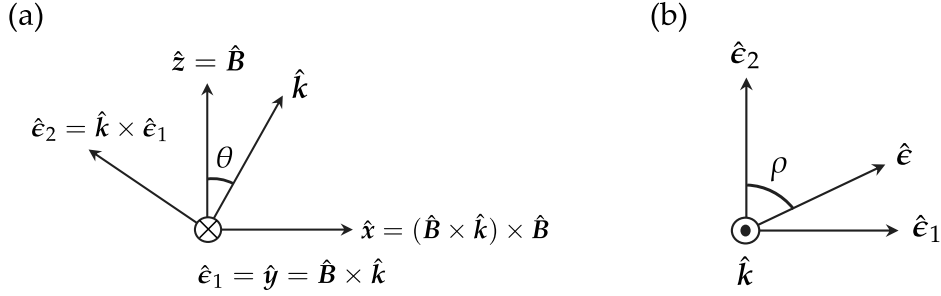


Figure 2.1 Coordinates for angular dependency calculation of the multipole transition.

as

$$\begin{aligned}
 c_{K,q,p}^{(+)} &= (-1)^{K-q} \sqrt{K} \begin{pmatrix} K & 1 & K+1 \\ -q & p & q-p \end{pmatrix} \\
 c_{K,q,p}^{(-)} &= (-1)^{K-q} \sqrt{K+1} \begin{pmatrix} K & 1 & K-1 \\ -q & p & q-p \end{pmatrix} \\
 c_{K,q,p}^{(0)} &= (-1)^{K-q} \sqrt{2K+1} \begin{pmatrix} K & 1 & K \\ -q & p & q-p \end{pmatrix}.
 \end{aligned} \tag{2.50}$$

The unit vectors $\hat{e}_{\pm}$ and $\hat{e}_0$ correspond to $\sigma^{\pm}$ - and π -transitions, respectively. Equation (2.49) means that in the multipole transitions the light can provide the angular momentum of $m_s = 0, \pm 1$ by the photon spin, which is represented by $\hat{e}_p$, and additional angular momentum of $m_l = q, q \pm 1$, which is represented by the spherical harmonics. Considering the angular momentum conservation, the transition must satisfy $\Delta m = m_s + m_l$, where Δm is the difference between the projection of the angular momentum of the initial and final states.

In the following, we visualize the angular dependence of the transition strength based on equations (2.47) and (2.49). For simplicity, we consider linearly polarized light with a polarization vector of

$$\begin{aligned}
 \hat{e}_{\text{lin}} &= \sin(\rho) \hat{e}_1 + \cos(\rho) \hat{e}_2 \\
 &= \sin(\rho) \hat{y} + \cos(\rho) (-\cos(\theta) \hat{x} + \sin(\theta) \hat{z}) \\
 &= \frac{i \sin(\rho) + \cos(\rho) \cos(\theta)}{\sqrt{2}} \hat{e}_1 + \cos(\rho) \sin(\theta) \hat{e}_0 + \frac{i \sin(\rho) - \cos(\rho) \cos(\theta)}{\sqrt{2}} \hat{e}_{-1},
 \end{aligned} \tag{2.51}$$

where ρ is the angle between the polarization vector and its projection onto the x - z plane (figure 2.1 (b)). Figure 2.2 shows the angular dependence of the transition strength for the E1, E2, M1, and M2 transitions. We can find that the transition strength of the electric and magnetic multipole transitions of the same order has the same angular dependence, up to shift of the angle ρ by $\pi/2$. This is because the electric and magnetic fields are orthogonal to each other.

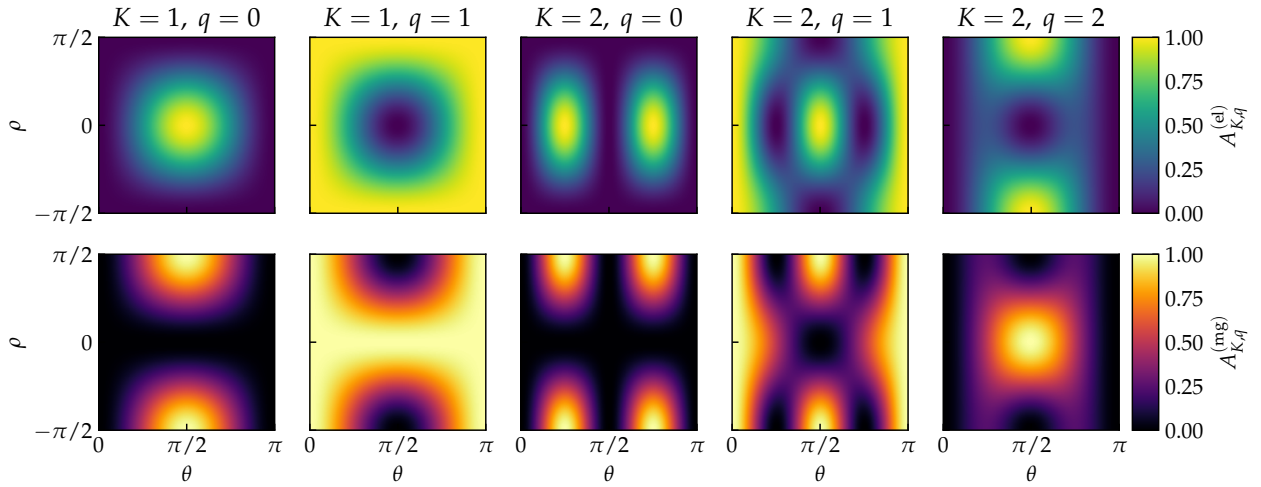


Figure 2.2 Angular dependence of the multipole transition strength calculated from equations (2.47) and (2.49). The top and bottom row represents electric and magnetic multipole transition, respectively.

2.2 Atomic polarizability and dipole potential

As we discussed in section 2.1.3, the atom-light interaction is approximated by the electric dipole transitions in most cases. The induced electric dipole $\mathbf{d}$ by the oscillating electric field $\mathbf{E}(\mathbf{r}, t) = \hat{\mathbf{e}}E(\mathbf{r})\exp(-i\omega t) + \text{c.c}$ is given by

$$\mathbf{d}(\mathbf{r}, t) = \alpha(\omega)\mathbf{E}(\mathbf{r}, t). \quad (2.52)$$

Here, $\alpha(\omega)$ is the atomic polarizability, which depends on the frequency of the light. As we will see later, the real and imaginary part of the polarizability is related to the dipole potential and the scattering rate of the off-resonant light, respectively.

In this section, we will calculate the atomic polarizability in a two-level system and introduce optical dipole traps, which are essential in this study. After that we will extend it to a multi-level system.

2.2.1 Two-level system

The polarizability in a two-level system can be understood by the Lorentz-oscillator model, where the electron is bound to the core with an oscillation frequency ω_0 and interacts with the light field with a frequency ω . This classical model gives the polarizability as [128]

$$\alpha(\omega) = 6\pi\epsilon_0 c^3 \frac{\Gamma}{\omega_0^2 \left(\omega_0^2 - \omega^2 - i\Gamma \frac{\omega^3}{\omega_0^2} \right)}, \quad (2.53)$$

where Γ is the decay rate of the excited state.

The induced dipole potential (also referred as light shift) U_{dip} can be calculated as

$$U_{\text{dip}}(\mathbf{r}) = -\frac{1}{2} \frac{1}{T} \int_0^T \mathbf{d}(\mathbf{r}, t) \cdot \mathbf{E}(\mathbf{r}, t) dt = -\frac{1}{2} \text{Re}[\alpha(\omega)] |\mathbf{E}(\mathbf{r})|^2 = -\frac{1}{2\epsilon_0 c} \text{Re}[\alpha(\omega)] I(\mathbf{r}), \quad (2.54)$$

where ϵ_0 is the vacuum permittivity, c is the speed of light, and $I(\mathbf{r})$ is the intensity of the light. The factor of $1/2$ means that the dipole is induced by the electric field. The corresponding dipole force is then calculated as

$$\mathbf{F}(\mathbf{r}) = -\nabla U_{\text{dip}}(\mathbf{r}) = \frac{1}{2\epsilon_0 c} \nabla \text{Re}[\alpha(\omega)] I(\mathbf{r}). \quad (2.55)$$

If the polarizability is positive, the force is attractive, and if the polarizability is negative, the force is repulsive. The associated scattering rate γ_{sc} , which corresponds to the absorbed power per photon, is calculated as

$$\gamma_{\text{sc}} = \frac{1}{\hbar\omega} \frac{1}{T} \int_0^T \left[\frac{\partial}{\partial t} \mathbf{d}(\mathbf{r}, t) \right] \cdot \mathbf{E}(\mathbf{r}, t) dt = \frac{1}{\hbar\epsilon_0 c} \text{Im}[\alpha(\omega)] I(\mathbf{r}). \quad (2.56)$$

By substituting equation (2.53) into equations (2.54) and (2.56), we can obtain the dipole potential and the scattering rate in a two-level system as

$$U_{\text{dip}}(\mathbf{r}) = -\frac{3\pi c^2 \Gamma}{2\omega_0^2} \frac{\omega_0^2 - \omega^2}{(\omega_0^2 - \omega^2)^2 - \Gamma^2 \frac{\omega^6}{\omega_0^4}} I(\mathbf{r}) \simeq -\frac{3\pi c^2}{2\omega_0^3} \left(\frac{\Gamma}{\omega_0 - \omega} + \frac{\Gamma}{\omega_0 + \omega} \right) I(\mathbf{r}), \quad (2.57)$$

$$\gamma_{\text{sc}} = \frac{6\pi c^2 \Gamma^2}{\hbar\omega_0^2} \frac{\frac{\omega^3}{\omega_0^2}}{(\omega_0^2 - \omega^2)^2 - \Gamma^2 \frac{\omega^6}{\omega_0^4}} I(\mathbf{r}) \simeq \frac{3\pi c^2}{2\hbar\omega_0^3} \left(\frac{\omega}{\omega_0} \right)^3 \left(\frac{\Gamma}{\omega_0 - \omega} + \frac{\Gamma}{\omega_0 + \omega} \right)^2 I(\mathbf{r}). \quad (2.58)$$

In the last step, we neglected the term of $\Gamma^2 \omega^6 / \omega_0^4$ in the denominator, which is valid for far off-resonant light. We introduce the detuning $\Delta = \omega - \omega_0$ and apply the rotating wave approximation ($|\omega_0 - \omega| \ll \omega_0 + \omega$, and $\omega \sim \omega_0$) to obtain simpler expressions as

$$U_{\text{dip}}(\mathbf{r}) = -\frac{3\pi c^2 \Gamma}{2\omega_0^3 \Delta} I(\mathbf{r}), \quad (2.59)$$

$$\gamma_{\text{sc}} = \frac{3\pi c^2}{2\hbar\omega_0^3} \left(\frac{\Gamma}{\Delta} \right)^2 I(\mathbf{r}). \quad (2.60)$$

We can see that the dipole potential is attractive for red-detuned light ($\Delta > 0$), which is used for optical trapping of the atoms. A gaussian beam is usually used for this purpose (optical dipole trap), and the intensity of the gaussian beam is given by

$$I(\rho, z) = I_0 \left(\frac{w_0^2}{w(z)^2} \right) \exp \left(-\frac{2\rho^2}{w(z)^2} \right), \quad (2.61)$$

where ρ is the distance from the beam center, z is the position along the beam propagation axis, w_0 is the beam waist at the focus ($z = 0$), $w(z) = w_0 \sqrt{1 + (z/z_R)^2}$ is the beam waist at the position z , and $z_R = \pi w_0^2 / \lambda$ is the Rayleigh length. I_0 is the peak intensity of the beam defined as $I_0 = 2P / (\pi w_0^2)$

with beam power P . By substituting equation (2.61) into equation (2.59) and using Taylor expansion around $z = 0$, we can obtain the dipole potential as a harmonic trap as

$$U_{\text{dip}}(\rho, z) = -U_0 \left[1 - 2 \left(\frac{\rho}{w_0} \right)^2 - \left(\frac{z}{z_R} \right)^2 \right] = -U_0 + \frac{1}{2} m \omega_\rho^2 r^2 + \frac{1}{2} m \omega_z z^2, \quad (2.62)$$

where $U_0 = 3\pi c^2 \Gamma / (2\omega_0^3 \Delta) I_0$ is the potential depth, m is the atomic mass, and ω_ρ and ω_z are the trap frequencies along the radial and axial direction of the trap, which are defined as

$$\omega_\rho := \sqrt{\frac{4U_0}{mw_0^2}}, \quad \omega_z := \sqrt{\frac{2U_0}{mz_R^2}}. \quad (2.63)$$

Figure 2.3 shows the spatial distribution of the dipole potential made by a gaussian beam. The potential is harmonic along the radial axial directions.

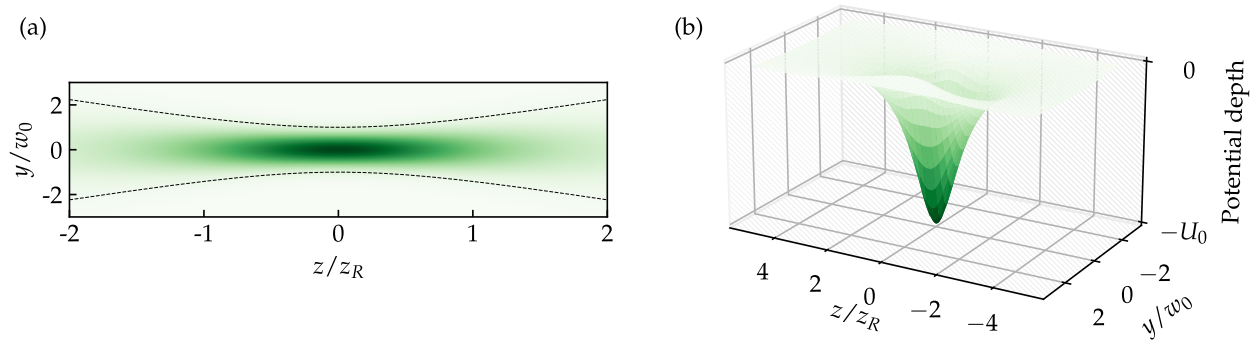


Figure 2.3 Dipole potential of a gaussian beam. (a) Two-dimensional plot. The dashed lines represent $1/e^2$ radius of the trapping beam. (b) Three-dimensional plot.

2.2.2 Multi-level system

In reality, the atom has multiple energy levels, and thus we must take into account these levels to calculate the polarizability. Hereafter, we refer to the polarizability as the real part of the complex polarizability. The atomic polarizability is rank 2 tensor arising from the ac Stark shift Hamiltonian, and can be decomposed into the scalar, vector, and tensor parts, which are given by [129]

$$\alpha_{nJ}^S = \frac{1}{\sqrt{3(2J+1)}} \alpha_{nJ}^{(0)}, \quad (2.64)$$

$$\alpha_{nJ}^V = -\sqrt{\frac{2J}{(J+1)(2J+1)}} \alpha_{nJ}^{(1)}, \quad (2.65)$$

$$\alpha_{nJ}^T = -\sqrt{\frac{2J(2J-1)}{3(J+1)(2J+1)(2J+3)}} \alpha_{nJ}^{(2)}, \quad (2.66)$$

$$(2.67)$$

with

$$\alpha_{nJ}^{(K)} = (-1)^{K+J+1} \sqrt{2K+1} \sum_{n', J'} (-1)^{J'} \begin{Bmatrix} 1 & K & 1 \\ J & J' & J \end{Bmatrix} |\langle n' J' || \mathbf{d} || n J \rangle|^2 \quad (2.68)$$

$$\times \frac{2\pi}{\epsilon_0 c \hbar^2} \text{Re} \left[\frac{1}{\omega_{n' J' n J} - \omega - i \gamma_{n' J' n J} / 2} + \frac{(-1)^K}{\omega_{n' J' n J} + \omega - i \gamma_{n' J' n J} / 2} \right].$$

Here, n, J is the principal quantum number and total angular momentum of the atomic state $|n, J\rangle$, $\omega_{n' J' n J}$ is the transition frequency between the states $|n', J'\rangle$ and $|n, J\rangle$, $\gamma_{n' J' n J}$ is the decay rate of the state $|n', J'\rangle$, $\{\cdot\}$ is the Wigner 6j-symbol, and $\langle n' J' || \mathbf{d} || n J \rangle$ is the reduced dipole matrix element. With these polarizabilities, the dipole potential operator is written as

$$\hat{\mathcal{U}}_{\text{dip}} = -\frac{\hbar}{4} \left[\alpha_{nJ}^S - i \alpha_{nJ}^V \frac{(\boldsymbol{\epsilon}^* \times \boldsymbol{\epsilon}) \cdot \mathbf{J}}{2J} + \alpha_{nJ}^T \frac{3[(\boldsymbol{\epsilon}^* \cdot \mathbf{J})(\boldsymbol{\epsilon} \cdot \mathbf{J}) + (\boldsymbol{\epsilon}^* \cdot \mathbf{J})(\boldsymbol{\epsilon} \cdot \mathbf{J})] - 2J^2}{2J(2J-1)} \right] I(\mathbf{r}), \quad (2.69)$$

and the dipole potential is calculated as $U_{\text{dip}}(\mathbf{r}) = \langle n, J | \hat{\mathcal{U}}_{\text{dip}} | n, J \rangle$. Note that we conventionally chose the coefficient of the dipole potential as $\hbar/4$.

If the atomic levels have hyperfine structures $\mathbf{F} = \mathbf{J} + \mathbf{I}$, where $\mathbf{I}$ is the nuclear spin, the polarizability and dipole potential are written as

$$\alpha_{nJF}^S = \alpha_{nJ}^S = \frac{1}{\sqrt{3(2J+1)}} \alpha_{nJ}^{(0)}, \quad (2.70)$$

$$\alpha_{nJF}^V = (-1)^{J+I+F} \sqrt{\frac{2F(2F+1)}{F+1}} \begin{Bmatrix} F & 1 & F \\ J & I & J \end{Bmatrix} \alpha_{nJ}^{(1)}, \quad (2.71)$$

$$\alpha_{nJF}^T = -(-1)^{J+I+F} \sqrt{\frac{2F(2F-1)(2F+1)}{3(F+1)(2F+3)}} \begin{Bmatrix} F & 2 & F \\ J & I & J \end{Bmatrix} \alpha_{nJ}^{(2)}, \quad (2.72)$$

and

$$\hat{\mathcal{U}}_{\text{dip}} = -\frac{\hbar}{4} \left[\alpha_{nJF}^S - i \alpha_{nJF}^V \frac{(\boldsymbol{\epsilon}^* \times \boldsymbol{\epsilon}) \cdot \mathbf{F}}{2F} + \alpha_{nJF}^T \frac{3[(\boldsymbol{\epsilon}^* \cdot \mathbf{F})(\boldsymbol{\epsilon} \cdot \mathbf{F}) + (\boldsymbol{\epsilon}^* \cdot \mathbf{F})(\boldsymbol{\epsilon} \cdot \mathbf{F})] - 2F^2}{2F(2F-1)} \right] I(\mathbf{r}). \quad (2.73)$$

If the off-diagonal coupling is much smaller than the Zeeman coupling, and the energy levels are well described as the Zeeman sublevels $|F, m_F\rangle$, the dipole potential is further simplified as

$$U_{\text{dip}}(\mathbf{r}) = -\frac{\hbar}{4} \left[\alpha_{nJF}^S - 2\text{Im}(\epsilon_x^* \epsilon_y) \alpha_{nJF}^V \frac{m_F}{2F} - (1 - 3|\epsilon_z|^2) \alpha_{nJF}^T \frac{3m_F^2 - F(F+1)}{2F(2F-1)} \right] I(\mathbf{r}) \quad (2.74)$$

$$= -\frac{\hbar}{4} \left[\alpha_{nJF}^S - \sin(2\gamma) \alpha_{nJF}^V \frac{m_F}{2F} - (1 - 3\cos^2 \theta) \alpha_{nJF}^T \frac{3m_F^2 - F(F+1)}{2F(2F-1)} \right] I(\mathbf{r}),$$

where θ is the angle between the light polarization axis and the quantization axis, and γ is the ellipticity angle of the light, which represents the elliptical polarization as

$$\boldsymbol{\epsilon} = \cos(\gamma) \hat{\mathbf{x}} + i \sin(\gamma) \hat{\mathbf{y}}. \quad (2.75)$$

As seen in the above expression, while the scalar term is isotropic, the vector and tensor terms depend on the polarization of the light. The vector term is vanished when the light is linearly polarized ($\gamma = 0$), and is maximized when the light is circularly polarized ($\gamma = \pi/4$). The tensor term also has the tunability of the light shift via the angle between the polarization and quantization axis.

These properties act as “tuning knobs” of the dipole potential. One can utilize these to make a magic condition for a certain atomic transition, where total differential light shift between the transition states is canceled out. An elliptic polarization, which makes a magic condition, is called magic polarization [130–132]. Similarly, if the magic condition is satisfied by adjusting the angle between the polarization and quantization axis, the angle is called magic angle [109, 119, 133–135]. These magic conditions are useful for high-fidelity excitation, cooling, and imaging of the atoms.

2.3 Coherent dynamics in atom-light interaction

Coherent manipulation of the atomic internal state by light is essential in quantum information processing in neutral atom systems. In this section, we will discuss the coherent dynamics of the atom-light interaction. For simplicity, we consider a two-level system consisting of the ground state $|g\rangle$ and the excited state $|e\rangle$ with a energy difference of $\hbar\omega_0$. The total Hamiltonian of the system is given by

$$\begin{aligned}\mathcal{H} &= \hbar\omega_0 |e\rangle\langle e| + \langle g| \mathcal{H}_{\text{int}} |e\rangle |g\rangle\langle e| + \langle e| \mathcal{H}_{\text{int}} |g\rangle |e\rangle\langle g| \\ &= \hbar\omega_0 |e\rangle\langle e| + \frac{\hbar\Omega}{2} \left(e^{-i\omega t} |e\rangle\langle g| + e^{i\omega t} |g\rangle\langle e| \right),\end{aligned}\tag{2.76}$$

where $\mathcal{H}_{\text{int}}$ is given as equation (2.4), Ω is the Rabi frequency, and ω is the frequency of the light. In the rotating frame with an angular frequency ω_0 , the Hamiltonian is given by the transformation with a unitary operator $U = \exp(i\omega t |e\rangle\langle e|)$ as

$$\tilde{\mathcal{H}} = U\mathcal{H}U^\dagger + i\hbar(\partial_t U)U^\dagger = -\hbar\Delta |e\rangle\langle e| + \frac{\hbar\Omega}{2}(|e\rangle\langle g| + |g\rangle\langle e|).\tag{2.77}$$

Here, $\Delta = \omega_0 - \omega$ is the detuning of the light.

The time-evolution of the system $\rho(t) = \rho_{gg} |g\rangle\langle g| + \rho_{ee} |e\rangle\langle e| + \rho_{ge} |g\rangle\langle e| + \rho_{eg} |e\rangle\langle g|$ is described by the master equation as

$$\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar}[\tilde{\mathcal{H}}, \rho] + \Gamma \mathcal{D}[|g\rangle\langle e|]\rho,\tag{2.78}$$

where Γ is the decay rate of the excited state, and $\mathcal{D}[c]\rho$ is the Lindblad superoperator defined as

$$\mathcal{D}[c]\rho = c\rho c^\dagger - \frac{1}{2}(c^\dagger c\rho + \rho c^\dagger c).\tag{2.79}$$

The time evolution of each component is then written as

$$\begin{aligned}
\partial_t \rho_{gg} &= -i \frac{\Omega}{2} (\rho_{eg} - \rho_{ge}) + \Gamma \rho_{ee}, \\
\partial_t \rho_{ee} &= i \frac{\Omega}{2} (\rho_{eg} - \rho_{ge}) - \Gamma \rho_{ee}, \\
\partial_t \rho_{ge} &= -\left(\frac{\Gamma}{2} + i\Delta\right) \rho_{ge} - i \frac{\Omega}{2} (\rho_{ee} - \rho_{gg}), \\
\partial_t \rho_{eg} &= -\left(\frac{\Gamma}{2} - i\Delta\right) \rho_{eg} + i \frac{\Omega}{2} (\rho_{ee} - \rho_{gg}).
\end{aligned} \tag{2.80}$$

These equations are called optical Bloch equations. When the atom is in the ground state, and the light is on resonance of the transition, the dynamics of $\langle \sigma_z \rangle = \rho_{ee} - \rho_{gg}$ is written as [127]

$$\langle \sigma_z(t) \rangle = -1 + \frac{\Omega^2}{\Omega^2 + \Gamma^2/2} \left[1 - e^{-(3\Gamma/4)t} \left(\cos(\Omega_\Gamma t) + \frac{3\Gamma}{4\Omega_\Gamma} \sin(\Omega_\Gamma t) \right) \right], \tag{2.81}$$

with $\Omega_\Gamma = \sqrt{\Omega^2 - (\Gamma/4)^2}$ as shown in figure 2.4. The steady-state solution is given by

$$\rho_{ee}(t \rightarrow \infty) = 1 - \rho_{gg}(t \rightarrow \infty) = \frac{\frac{\Omega^2}{\Gamma^2}}{1 + \left(\frac{2\Delta}{\Gamma}\right)^2 + \frac{2\Omega^2}{\Gamma^2}} = \frac{1}{2} \frac{I/I_{\text{sat}}}{1 + (2\Delta/\Gamma)^2 + I/I_{\text{sat}}}. \tag{2.82}$$

Here, we introduced the saturation intensity $I_{\text{sat}} = \pi \hbar c \Gamma / (3\lambda^3)$, where λ is the wavelength of the light, and used the relation $I/I_{\text{sat}} = 2\Omega^2/\Gamma^2$.

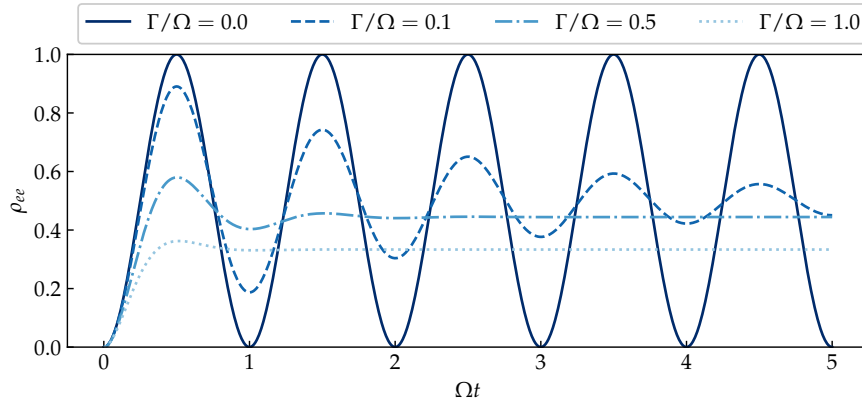


Figure 2.4 Rabi oscillations for various Rabi frequencies with zero detuning.

2.4 Center-of-mass motion of atoms in optical transitions

In addition to the coherent dynamics of the atom-light interaction, the mechanical effects of the light on the atom are also pivotal in cooling and trapping of the atoms.

2.4.1 Atoms in free space

First, we consider the center-of-mass motion of the atom moving in one-dimensional free space with a velocity v and momentum $p = mv$. The scattering rate during irradiation of near-resonant light with a detuning Δ and an intensity I is given by

$$R_{\text{sc}}(v) = \Gamma \rho_{ee}(t \rightarrow \infty) = \frac{\Gamma}{2} \frac{I/I_{\text{sat}}}{1 + (2(\Delta - kv)/\Gamma)^2 + I/I_{\text{sat}}}. \quad (2.83)$$

Here, we take into account the Doppler shift of $-kv$ in the detuning. The corresponding radiation force acting on the atom is given by

$$F_{\text{rad}} = \hbar k R_{\text{sc}}. \quad (2.84)$$

When two counter-propagating beams are irradiated, the energy change of the atom $(dE)_{\text{cool}}$ in time dt , which is induced by the light irradiation, is given by

$$(dE)_{\text{cool}} = F_{\text{rad}} v dt = \hbar k (R_{\text{sc}}(v) - R_{\text{sc}}(-v)) dt. \quad (2.85)$$

When the Doppler shift is much smaller than the detuning, the scattering rate is approximated as

$$R_{\text{sc}}(v) = R_{\text{sc}}(0) + v \left. \frac{dR_{\text{sc}}}{dv} \right|_{v=0} = R_{\text{sc}}(0) + kv \frac{4(\Delta/\Gamma)I/I_{\text{sat}}}{(1 + (2\Delta/\Gamma)^2 + I/I_{\text{sat}})^2}. \quad (2.86)$$

Substituting equation (2.86) into equation (2.88), we obtain

$$(dE)_{\text{cool}} = \frac{32(\Delta/\Gamma)I/I_{\text{sat}}}{(1 + (2\Delta/\Gamma)^2 + I/I_{\text{sat}})^2} \frac{E_R}{\hbar} E dt, \quad (2.87)$$

where $E_R = \hbar^2 k^2 / (2m)$ is the recoil energy. When the beams are red-detuned ($\Delta < 0$), the energy change is negative, meaning that the beams cool the atom. On the other hand, the atom absorbs and emits a photon with a momentum $\hbar k$ randomly, resulting in a random walk in momentum space and energy increase of the atom $(dE)_{\text{heat}}$ as

$$(dE)_{\text{heat}} = 2E_R (R_{\text{sc}}(v) + R_{\text{sc}}(-v)) dt = 4E_R \frac{\Gamma}{2} \frac{I/I_{\text{sat}}}{1 + (2\Delta/\Gamma)^2 + I/I_{\text{sat}}} dt. \quad (2.88)$$

The factor of 2 arises from the random process of the absorption and emission of photons. We finally obtain the net energy change of the atom from equations (2.85) and (2.88) as

$$\frac{dE}{dt} = \frac{(dE)_{\text{heat}} + (dE)_{\text{cool}}}{dt} = 2E_R \frac{I/I_{\text{sat}}}{1 + (2\Delta/\Gamma)^2 + I/I_{\text{sat}}} \left(\Gamma + \frac{16(\Delta/\Gamma)I/I_{\text{sat}}}{1 + (2\Delta/\Gamma)^2 + I/I_{\text{sat}}} \frac{E}{\hbar} \right). \quad (2.89)$$

In the steady state and low intensity limit ($I/I_{\text{sat}} \rightarrow 0$), the energy becomes

$$E = -\frac{\hbar \Gamma}{16} \frac{1 + (2\Delta/\Gamma)^2}{\Delta/\Gamma}, \quad (2.90)$$

which has a minimum at $E_{\text{min}} = \hbar \Gamma / 4$ when $\Delta = -\Gamma/2$. The corresponding temperature is $T_D = 2E/k_B = \hbar \Gamma / (2k_B)$, where k_B is the Boltzmann constant. This temperature T_D is called the Doppler limit, which is the lowest temperature that can be achieved by the Doppler cooling.

2.4.2 Atoms in a harmonic trap

Next, we consider the center-of-mass motion of the atom in a harmonic trap with a trap frequency of ω_ρ . In a harmonic trap, the motion of the atom is quantized according to following Hamiltonian as

$$\mathcal{H}_{\text{trap}} = \frac{\hat{p}^2}{2m} + \frac{1}{2}m\omega_\rho^2\hat{x}^2 = \hbar\omega_\rho\left(\hat{a}^\dagger\hat{a} + \frac{1}{2}\right), \quad (2.91)$$

where $\hat{x}$, $\hat{p}$ is the position and momentum operator, and related to the annihilation and creation operators as

$$\begin{aligned} \hat{x} &= \sqrt{\frac{\hbar}{2m\omega_\rho}}(\hat{a} + \hat{a}^\dagger) = x_0(\hat{a} + \hat{a}^\dagger) \\ \hat{p} &= i\sqrt{\frac{\hbar m\omega_\rho}{2}}(\hat{a}^\dagger - \hat{a}) = p_0(\hat{a}^\dagger - \hat{a}). \end{aligned} \quad (2.92)$$

Assuming that the atom has only two internal states $|g\rangle$ and $|e\rangle$, the atomic states are labeled as $|g, n_g\rangle$ and $|e, n_e\rangle$, where n_g, n_e is the vibrational quantum number for each internal state. Then the transition matrix element can be written as

$$\langle g, n_g | \mathcal{H}_{\text{int}} | e, n_e \rangle = \hbar\Omega_0 \langle n_g | e^{ik\hat{x}} | n_e \rangle = \hbar\Omega_0 \langle n_g | e^{i\eta(\hat{a} + \hat{a}^\dagger)} | n_e \rangle, \quad (2.93)$$

where Ω_0 is the Rabi frequency for the carrier transition ($n_g = n_e$), and $\eta = kx_0 = \sqrt{E_R/(\hbar\omega_\rho)}$ is the Lamb-Dicke factor. When the trap frequency is much larger than the recoil frequency ($\omega_\rho \gg E_R/\hbar$), which means that the atoms are tightly confined in the trap, the Lamb-Dicke factor is much smaller than 1, and the matrix element is approximated as

$$\Omega_0 \langle n_g | e^{i\eta(\hat{a} + \hat{a}^\dagger)} | n_e \rangle \simeq \Omega_0 \langle n_g | (1 + i\eta(\hat{a} + \hat{a}^\dagger)) | n_e \rangle = \begin{cases} \Omega_0 & (n_g = n_e) \\ i\eta\sqrt{n_e}\Omega_0 & (n_e = n_g + 1) \\ i\eta\sqrt{n_g}\Omega_0 & (n_e = n_g - 1) \\ 0 & (\text{otherwise}) \end{cases} \quad (2.94)$$

The expressions means that the transition with a motional state change, which are called sidebands, is suppressed by the factor $\eta \ll 1$. This regime is called the Lamb-Dicke regime. If we calculate the matrix element without the approximation, we obtain [1]

$$\Omega_0 \langle n_g | e^{i\eta(\hat{a} + \hat{a}^\dagger)} | n_e \rangle = \Omega_0 e^{-\eta^2/2} \eta^{\Delta n} \sqrt{\frac{n_{<}!}{n_{>}!}} L_{n_{<}}^{\Delta n}(\eta^2), \quad (2.95)$$

where $n_{<} = \min(n_g, n_e)$, $n_{>} = \max(n_g, n_e)$, $\Delta n = |n_g - n_e|$, and $L_{n_{<}}^{\Delta n}(\eta^2)$ is the Laguerre polynomial as

$$L_n^\alpha(x) = \sum_{k=0}^n \binom{n+\alpha}{n-k} \frac{(-x)^k}{k!}. \quad (2.96)$$

It is important to note that the above calculation assumes the trap satisfies the magic condition, meaning the trap frequency is the same for each internal state. Under this condition, the wavefunctions of motional states with the same vibrational quantum number in different internal states are identical. Specifically, this implies that $\langle n_g | n_e \rangle = 1$ when $n_g = n_e$. However, in reality, this assumption does not always hold, and the wavefunctions of the motional states differ between internal states. In such cases, $\langle n_g | n_e \rangle \neq 1$ for $n_g = n_e$ and $\langle n_g | n_e \rangle \neq 0$ for $n_g \neq n_e$, explicitly written as [136]

$$\langle 0_g | 0_e \rangle = \left[\sum_{m=0}^{\infty} \frac{(2m-1)!!}{(2m)!!} \left(\frac{\omega_e - \omega_g}{\omega_e + \omega_g} \right)^{2m} \right]^{-1/2} \quad (2.97)$$

$$\langle n_g | 0_e \rangle = (-1)^{n_g/2} \sqrt{\frac{(n_g-1)!!}{n_g!!}} \left(\frac{\omega_e - \omega_g}{\omega_e + \omega_g} \right)^{n_g/2} \langle 0_g | 0_e \rangle \quad (\text{for even } n_g) \quad (2.98)$$

$$\langle n_g | 0_e \rangle = 0 \quad (\text{for odd } n_g) \quad (2.99)$$

$$\langle n_g | n_e \rangle = \frac{1}{2\sqrt{n_e}} \left[\alpha_+ \sqrt{n_g} \langle n_g - 1 | n_e - 1 \rangle + \alpha_- \sqrt{n_g + 1} \langle n_g + 1 | n_e - 1 \rangle \right], \quad (2.100)$$

where ω_g, ω_e is the trap frequency for the ground and excited states, respectively, and

$$\alpha_{\pm} = \sqrt{\frac{\omega_e}{\omega_g}} \pm \sqrt{\frac{\omega_g}{\omega_e}}. \quad (2.101)$$

This results in unintended spreading of the vibrational quantum number during the dynamics when the sidebands are not well-resolved for the transition. This leads to the heating of the atoms in the trap.

Chapter 3

Ytterbium atoms and Rydberg states

This chapter reviews the fundamental properties of ytterbium atoms and Rydberg states. The first part summarizes the Yb energy levels and transitions utilized in this study. The second part discusses the physical characteristics of Rydberg states, the multi-channel quantum defect theory, the unique properties of two-electron Rydberg atoms, and the interactions between Rydberg atoms.

3.1 Ytterbium

3.1.1 Atomic structure

Ytterbium (Yb) is a lanthanoid atom with an atomic number of 70. It has seven stable isotopes, five bosons (^{168}Yb , ^{170}Yb , ^{172}Yb , ^{174}Yb , ^{176}Yb) and two fermions (^{171}Yb , ^{173}Yb). The natural abundance of each isotope is summarized in table 3.1. The electron configuration is written as $[\text{Xe}]4f^{14}6s^2$, with two valence electrons, similar to alkaline-earth atoms. This configuration gives Yb unique properties compared to alkali atoms, offering advantages for quantum computing in neutral atom arrays.

Table 3.1 Isotopes of ytterbium

	Natural abundance (%)	Nuclear spin ($\hbar$)	μ_I/μ_N	Statistics
^{168}Yb	0.13	0		Boson
^{170}Yb	3.05	0		Boson
^{171}Yb	14.3	1/2	0.4919	Fermion
^{172}Yb	21.9	0		Boson
^{173}Yb	16.1	5/2	-0.6776	Fermion
^{174}Yb	31.8	0		Boson
^{176}Yb	12.7	0		Boson

3.1.2 Energy levels and transitions

With two valence electrons forming spin singlet and triplet pairs, Yb atoms exhibit a greater variety of energy levels than alkali atoms. The energy levels are primarily described by the LS coupling scheme, where the total electronic orbital angular momentum L and the total electronic spin angular momentum S are coupled to form the total electronic angular momentum J , and each electronic level is denoted by $^{2S+1}L_J$. Notably, because Yb is a heavy atom, spin-orbit coupling cannot

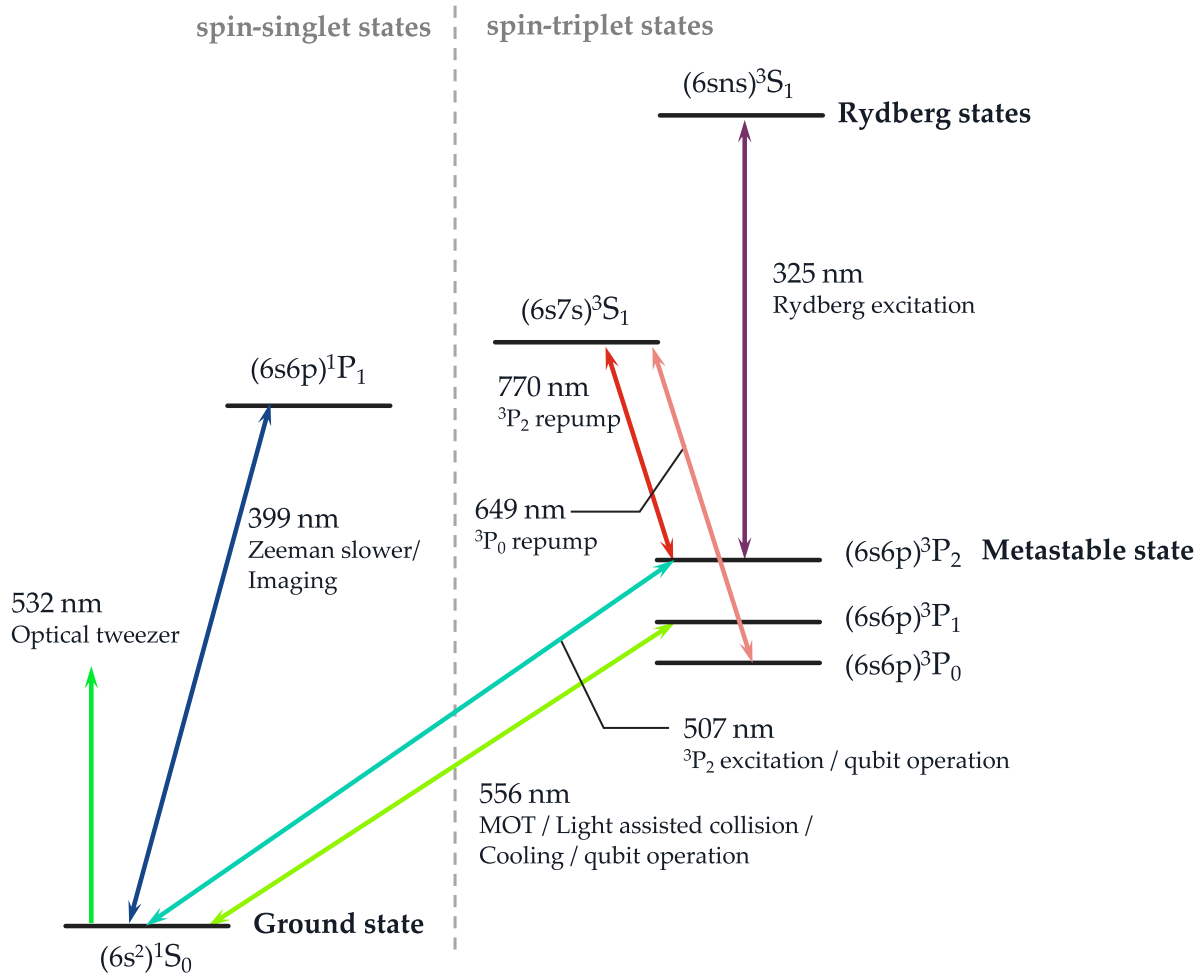


Figure 3.1 Yb energy levels relevant to this work.

be ignored, leading to slight perturbations in the LS coupling scheme. As a result, the intercombination transitions $\Delta S = 1$, which is prohibited in the selection rule of the E1 transtion, is allowed. The optical transitions and energy levels used in this study are shown in figure 3.1. The characteristics of each transition are summarized below.

$(6s^2)^1S_0 - (6s6p)^1P_1$ transition

This transition is an E1 transition with a wavelength $\lambda = 398.9$ nm and a natural width $\Gamma = 29$ MHz [137], making it a very strong transition. Since the 1P_1 state slightly decays to $(5d6s)^3D_1$ and $(5d6s)^3D_2$ [138], this transition is not completely closed. In this study, this transition is used for the Zeeman slowing and fluorescence imaging of single atoms.

$(6s^2)^1S_0 - (6s6p)^3P_1$ transition

This transition is an intercombination transition connecting the singlet and the triplet states ($\Delta S = 1$). The transition wavelength is $\lambda = 555.8$ nm, and the natural width is $\Gamma = 182.4$ kHz [139].

Although this transition is forbidden in the E1 transition selection rule, the heavy Yb atom has a pure LS coupling level $|^1P_1^0\rangle, |^3P_1^0\rangle$ due to the spin-orbit interaction as

$$\begin{aligned} |^1P_1\rangle &= \alpha |^1P_1^0\rangle + \beta |^3P_1^0\rangle \\ |^3P_1\rangle &= \alpha |^3P_1^0\rangle - \beta |^1P_1^0\rangle, \end{aligned} \quad (3.1)$$

and the transition becomes allowed. The values of α and β are obtained from the lifetime τ and energy E of each state as

$$\frac{|\alpha|^2}{|\beta|^2} = \frac{\tau(^3P_1)E(^3P_1)^3}{\tau(^1P_1)E(^1P_1)^3}. \quad (3.2)$$

From this equation, we obtain $|\alpha|^2 = 0.9834$ and $|\beta|^2 = 0.0166$. This transition is perfectly closed transition and has a much lower Doppler cooling limit temperature of $T_D = \hbar\Gamma/(2k_B) = 4.4 \mu\text{K}$ than alkali atoms due to its narrow natural linewidth. This linewidth is narrow enough for atom cooling and simultaneously broad enough for making magneto-optical trap (MOT) without the need for other transitions [140]. Furthermore, this transition can be employed for fluorescence imaging of single atoms with enough scattering rate and low atom temperature [111, 112]. In this study we use this transition for the magneto-optical trap (MOT), Doppler cooling, light-assisted collision (LAC), Raman sideband cooling (RSC), optical pumping (OP), and single qubit gates.

Basic properties and isotope shifts of $^1S_0 \leftrightarrow ^1P_1$ and $^1S_0 \leftrightarrow ^3P_1$ transitions are summarized in table 3.2 and table 3.3, respectively.

Table 3.2 Basic properties of $^1S_0 \leftrightarrow ^1P_1$ and $^1S_0 \leftrightarrow ^3P_1$ transitions

		$^1S_0 \leftrightarrow ^1P_1$	$^1S_0 \leftrightarrow ^3P_1$	Unit
Wavelength	λ	398.9	555.8	nm
Lifetime	τ	5.5	873	ns
Natural linewidth	$\Gamma/2\pi = 1/(2\pi\tau)$	29	0.182	MHz
Saturation intensity	I_{sat}	59	0.139	mW/cm ²
Recoil energy	E_R/k_B	344	178	nK
Doppler cooling limit	T_D	690	4.37	μK

(6s²)¹S₀ – (6s6p)³P₂ transition

This transition is M2 transition with a wavelength $\lambda = 507.35 \text{ nm}$. As we discussed in section 2.1.3, this transition is very weak and thus ultranarrow. In the case of fermionic isotope, the 3P_2 state is perturbed by hyperfine mixing as [141]

$$|^3P_2, F, m_F\rangle = \sum_{\alpha} \frac{\langle ^3P_2^0; F, m_F | \mathcal{H}_{\text{HF}} | \alpha \rangle}{E(\alpha) - E(^3P_2^0)} | \alpha \rangle, \quad (3.3)$$

Table 3.3 Isotope shifts of $^1S_0 \leftrightarrow ^1P_1$ [137] and $^1S_0 \leftrightarrow ^3P_1$ [139] transitions

$^1S_0 \leftrightarrow ^1P_1$	Isotope shift (MHz)	$^1S_0 \leftrightarrow ^3P_1$	Isotope shift (MHz)
^{168}Yb	1887.4	$^{173}\text{Yb}, (F = 3/2)$	3807.3
^{170}Yb	1192.4	$^{171}\text{Yb}, (F = 3/2)$	3804.6
$^{171}\text{Yb}, (F = 1/2)$	1153.7	^{168}Yb	3655.1
$^{171}\text{Yb}, (F = 3/2)$	832.4	$^{173}\text{Yb}, (F = 5/2)$	2311.4
$^{173}\text{Yb}, (F = 7/2)$	588.0	^{170}Yb	2286.4
^{172}Yb	533.3	^{172}Yb	1000.0
$^{173}\text{Yb}, (F = 3/2)$	516.0	^{174}Yb	0
^{174}Yb	0	^{176}Yb	-954.8
$^{173}\text{Yb}, (F = 5/2)$	-253.4	$^{171}\text{Yb}, (F = 1/2)$	-2132.1
^{176}Yb	-509.3	$^{173}\text{Yb}, (F = 7/2)$	-2386.7

where α is the intermediate states, and the hyperfine interaction Hamiltonian $\mathcal{H}_{HF}$ is given as [127]

$$\mathcal{H}_{HF} = A \frac{\mathbf{I} \cdot \mathbf{J}}{\hbar^2} + B \frac{3(\mathbf{I} \cdot \mathbf{J})^2 / \hbar^2 + 3(\mathbf{I} \cdot \mathbf{J}) / (2\hbar) - I(I+1)J(J+1)}{2I(2I-1)J(2J-1)}, \quad (3.4)$$

with the hyperfine constants A and B . As a result, the electronic dipole coupling with the ground state occurs as

$$\sum_{\alpha} \frac{\langle ^3P_2^0; F, m_F | \mathcal{H}_{HF} | \alpha \rangle \langle \alpha | \mathbf{d} \cdot \mathbf{E} | ^1S_0; F', m_{F'} \rangle}{E(\alpha) - E(^3P_2^0)}. \quad (3.5)$$

This has non-zero value for $F = 3/2$ of ^{171}Yb and $F = 3/2, 5/2$ of ^{173}Yb , and the transition becomes hyperfine mixing induced E1 (HFM-E1) transition, with which we can obtain one order magnitude stronger Rabi frequency than the M2 transition. Table 3.4 summarizes the basic properties of the $^1S_0 \leftrightarrow ^3P_2$ transition.

Table 3.4 Basic properties of $^1S_0 \leftrightarrow ^3P_2$ transition [141, 142]

		Boson	^{171}Yb	^{173}Yb	Unit
Wavelength	λ		507.35		nm
Lifetime	τ	15	6.3	7.2	s
Natural linewidth	$\Gamma/2\pi = 1/(2\pi\tau)$	10.6	25	22	mHz
Saturation intensity	I_{sat}	1.1×10^{-8}	2.5×10^{-8}	2.2×10^{-8}	mW/cm ²
Recoil energy	E_R/k_B		214		nK

(6s6p) 3P_2 – (6s7s) 3S_1 transition

This transition is E1 transition with a wavelength $\lambda = 770.2$ nm and used for 3P_2 state repumping. The (6s7s) 3S_1 state decays to not only 3P_2 but also 3P_0 and 3P_1 states. Therefore, we need

additional repumping of the 3P_0 state to perfectly repump the 3P_2 state.

(6s6p) 3P_0 – (6s7s) 3S_1 transition

This transition is E1 transition with a wavelength $\lambda = 649.1$ nm and used for 3P_0 state repumping. The branching ratio of the (6s7s) 3S_1 state is summarized in table 3.5.

Table 3.5 Basic properties of $^3S_1 \leftrightarrow ^3P_J$ transitions. The Radiative decay rate is given by $\Gamma_J = \omega_J^3 |\langle ^3P_J || \mathbf{d} || ^3S_1 \rangle|^2 / (9\pi\epsilon_0\hbar c^3)$ with the transition angular frequency ω_J and the reduced matrix element $\langle ^3P_J || \mathbf{d} || ^3S_1 \rangle$ [142].

		$^3S_1 \rightarrow ^3P_0$	$^3S_1 \rightarrow ^3P_1$	$^3S_1 \rightarrow ^3P_2$	Unit
Wavelength	λ	649.1	680.1	770.2	nm
Radiative decay rate	$\Gamma_J / (2\pi)$	1.54	4.26	6.00	MHz
Branching ratio		0.13	0.36	0.51	

(6s6p) 3P_2 – (6snl) Rydberg state transition

This transition is E1 transition with a wavelength $\lambda \simeq 325.5$ nm. The properties of the Rydberg state highly depends on the principal quantum number as we will discuss later.

3.2 Rydberg states

3.2.1 General properties of Rydberg atoms

Rydberg states are highly energetic states of atoms, where one of the valence electrons is excited to an orbital with a large principal quantum number n . Since the excited electron feels approximately $+e$ charge from the core, the properties of Rydberg states are in good analogy to the hydrogen atom. As the energy levels of the hydrogen atom is given by

$$E_n = -hc \frac{R_\infty}{n^2}, \quad (3.6)$$

with the Rydberg constant $R_\infty = m_e e^4 / (8\epsilon_0^2 \hbar^3 c)$ and electron mass m_e , the energy levels of the Rydberg atom is given by [143]

$$E_{n,l,J} = -hc \frac{R^*}{\nu^2}, \quad (3.7)$$

with the mass scaled Rydberg constant $R^* = R_\infty / (1 + m_e/m_c)$, core mass m_c , and the effective principal quantum number $\nu = n - \delta_{n,l,J}$. The quantum defect $\delta_{n,l,J}$ is introduced to take into account the deviation from the hydrogen atom due to the core potential, and depends on the principal quantum number n , the orbital angular momentum l , and the total angular momentum J of the Rydberg state, which is determined by the spectroscopy of the Rydberg states.

The large orbit of the Rydberg electron makes the properties of Rydberg atoms quite different from those of low-lying states. From the scaling law of the orbital radius $\langle r \rangle \sim n^2$, we can obtain the scaling law of various quantities as shown in table 3.6.

Table 3.6 Scaling laws of the properties of Rydberg states

Property		n scaling
Binding energy	E_n	n^{-2}
Energy interval	ΔE_n	n^{-3}
Orbital radius	$\langle r \rangle$	n^2
Dipole moment	$\langle g -er nl \rangle$	$n^{-3/2}$
	$\langle nl -er nl' \rangle$	n^2
Radiative lifetime	τ	n^3
Dipole interaction	C_3	n^4
Polarizability	α	n^7
van der Waals interaction	C_6	n^{11}

3.2.2 Quantum defect theory

The quantum defect theory (QDT) is used to describe the deviation of the Rydberg wavefunction from the hydrogen atom due to the core potential. While the Rydberg electron feels approximately $+e$ charge from the core when it is far from the core, the interaction between the Rydberg electron and the core is not negligible when the distance between them gets shorter. We can set this characteristic distance as r_0 , and the space is divided into two regions: the outer region $r > r_0$ where the system can be treated as hydrogen-like, and the inner region $r < r_0$ where the core potential is taken into account. The QDT extract the quantities associated with the Rydberg states from the boundary condition at $r = r_0$ and $r \rightarrow \infty$. In the following, we review the (single channel) QDT for one-electron atoms and multichannel QDT (MQDT) for two-electron atoms, based on Refs. [78,143–149]. Note that we use atomic units in the following discussion.

Single channel

We start the discussion with the schrödinger equation for hydrogen atom as

$$\left(-\frac{1}{2} \nabla^2 - \frac{1}{r} \right) \Psi(r, \theta, \phi) = E \Psi(r, \theta, \phi), \quad (3.8)$$

where E is the energy, r is the distance between the electron and the proton, and θ, ϕ are the angles of the spherical coordinate. The solution of the schrödinger equation is given by the spherical harmonics $Y_l^m(\theta, \phi)$ and the radial wavefunction $R(r)$ as

$$\Psi(r, \theta, \phi) = R(r) Y_l^m(\theta, \phi), \quad (3.9)$$

with the quantum number l and m . The radial wavefunction $R(r)$ satisfies the radial schrödinger equation as

$$\frac{d^2 R(r)}{dr^2} + \frac{2}{r} \frac{dR(r)}{dr} - \frac{l(l+1)}{r^2} R(r) + \frac{2R(r)}{r} + 2ER(r) = 0. \quad (3.10)$$

This equation has two physically interesting solutions as

$$R(r) = \frac{f(l, E, r)}{r}, \quad \frac{g(l, E, r)}{r}, \quad (3.11)$$

where f and g are the regular and irregular Coulomb functions, respectively, which have the asymptotic form at $r \rightarrow 0$ as

$$f(l, E, r) \propto r^{l+1}, \quad g(l, E, r) \propto r^{-l}. \quad (3.12)$$

As $r \rightarrow \infty$ and $E > 0$, f and g behave as sine and cosine functions, and therefore they have phase difference of $\pi/2$. For $E < 0$, f and g can be written as

$$\begin{aligned} f &= u(l, E, r) \sin(\pi\nu) - v(l, E, r) e^{i\pi\nu}, \\ g &= -u(l, E, r) \cos(\pi\nu) + v(l, E, r) e^{i\pi\nu+1/2}, \end{aligned} \quad (3.13)$$

where $\nu = 1/\sqrt{-2E}$ and u and v are exponentially increasing and decreasing functions of r . Considering that only f is allowed as $r \rightarrow 0$ and the boundary condition at $r \rightarrow \infty$, $\sin(\pi\nu)$ in equation (3.13) should be zero, meaning that ν should be an integer n . Therefore, we obtain the energy levels of the hydrogen atom as $E_n = -1/(2n^2)$.

In the case of Rydberg state of alkali atoms, the core potential is not negligible. When the Rydberg electron penetrates the charge cloud of the core electrons, it feels deeper potential than $-1/r$ due to decrease of the nuclear charge shielding. As a result, the wavefunction of the Rydberg electron is pulled towards the core. Then, the radial wavefunction of the Rydberg electron $\phi(r)$ at $r > r_0$ can be written as the linear combination of f and g

$$\phi(r) \propto [f(l, E, r) \cos(\pi\delta_l) - g(l, E, r) \sin(\pi\delta_l)]/r, \quad (3.14)$$

with the radial phase shift $\pi\delta_l$. Considering the boundary condition $\phi(r) \rightarrow 0$ at $r \rightarrow \infty$, we obtain the constraint as

$$\cos(\pi\delta_l) \sin(\pi\nu) + \sin(\pi\delta_l) \cos(\pi\nu) = 0, \quad (3.15)$$

or equivalently $\sin(\pi\nu + \pi\delta_l) = 0$. This condition means that $\nu = n - \delta_l$ with an integer n , and the energy levels of the Rydberg states are given by $E_{n,l} = -1/(2\nu^2) = -1/(2(n - \delta_l)^2)$. Here, δ_l is the quantum defect. Once the quantum defects are obtained from the spectroscopy of the Rydberg states, we can calculate the wavefunction of the Rydberg electron and various quantities associated with the Rydberg states.

Multi-channel

In contrast to the one-electron atoms, the two-electron atoms have much more complicated structure in Rydberg states due to multiple configurations of the core, which we refer as “channels”.

For $r > r_0$, the radial wavefunction $\phi_i(r)$ of long-range (or collision) channel i is written as

$$\phi_i(r) = [f(l_i, E_i, r) \cos(\pi\delta_i) - g(l_i, E_i, r) \sin(\pi\delta_i)]/r, \quad (3.16)$$

where $E_i := -1/(2\nu_i^2) = E - I_i$ is the asymptotic kinetic energy of the Rydberg electron with the ionization limit I_i for i -th channel, l_i is the orbital angular momentum of the Rydberg electron, and $\pi\delta_i$ is the phase shift. This equation is the counterpart of equation (3.14) for the one-electron atom. From the boundary condition at $r \rightarrow \infty$ as equation (3.15), we can replace $\delta_i = n - \nu_i$ and rewrite equation (3.16) as

$$\phi_i(r) = [f(l_i, E_i, r) \cos(\pi\nu_i) + g(l_i, E_i, r) \sin(\pi\nu_i)]/r. \quad (3.17)$$

The atomic wavefunction Ψ can then be written as

$$\Psi = \mathcal{A} \sum_{i=1}^N A_i \phi_i \chi_i, \quad (3.18)$$

where N is number of channels, A_i is the channel coefficients, χ_i is angular and spin components, and $\mathcal{A}$ is anti-symmetrized operator.

On the other hand, for $r < r_0$, the radial wavefunction $\{\phi_i\}$ is no longer good basis for the system because the singly charged core picture is invalid in this region. Instead, the wavefunction is characterized by the doubly charged core and the coupling of two valence electrons. We introduce new basis $\{\psi_\alpha\}$, where α is the short-range (or close-coupling) channel index, and the wavefunction Ψ is written as

$$\Psi = \sum_{\alpha=1}^N B_\alpha \psi_\alpha, \quad (3.19)$$

with the channel coefficients B_α . The basis $\{\psi_\alpha\}$ is converted to the basis $\{\phi_i\}$ for the long-range channels by a unitary transformation U . Therefore, ψ_α can be written as

$$\psi_\alpha = \frac{1}{r} \left[\mathcal{A} \sum_{i=1}^N U_{i\alpha} f(l_i, E_i, r) \chi_i \cos(\pi\mu_\alpha) - \mathcal{A} \sum_{i=1}^N U_{i\alpha} g(l_i, E_i, r) \chi_i \sin(\pi\mu_\alpha) \right], \quad (3.20)$$

where $U_{i\alpha}$ is the matrix element of U , and μ_α is eigenchannel quantum defect. From equations (3.17) to (3.20), we can obtain the relation between the channel coefficients A_i and B_α as

$$\begin{aligned} A_i \cos(\pi\nu_i) &= \sum_{\alpha=1}^N U_{i\alpha} B_\alpha \cos(\pi\mu_\alpha), \\ A_i \sin(\pi\nu_i) &= - \sum_{\alpha=1}^N U_{i\alpha} B_\alpha \sin(\pi\mu_\alpha). \end{aligned} \quad (3.21)$$

Because U is unitary, we can rewrite the above equations by multiplying $U_{\alpha i}$ and taking summation over i as

$$\begin{cases} \sum_i U_{\alpha i} A_i \cos(\pi v_i) = B_\alpha \cos(\pi \mu_\alpha) \\ \sum_i U_{\alpha i} A_i \sin(\pi v_i) = -B_\alpha \sin(\pi \mu_\alpha) \end{cases} \iff \begin{cases} \sum_i U_{\alpha i} \cos(\pi(v_i + \mu_\alpha)) A_i = B_\alpha \\ \sum_i U_{\alpha i} \sin(\pi(v_i + \mu_\alpha)) A_i = 0 \end{cases} \quad (3.22)$$

for $\alpha = 1, 2, \dots, N$. These equations have non-trivial solutions only when

$$\det [U_{\alpha i} \sin(\pi(v_i + \mu_\alpha))] = 0 \quad (3.23)$$

is satisfied. This equation gives a constraint on the effective principal quantum number v_i for given values of μ_α and $U_{\alpha i}$. As mentioned above, the boundary condition that $\Psi \rightarrow 0$ as $r \rightarrow 0$ leads to the additional constraint on v_i as

$$E = I_i + E_i = I_i - \frac{1}{2v_i^2}. \quad (3.24)$$

By optimizing the parameters of $U_{\alpha i}$, μ_α , I_i , and v_i in equations (3.23) and (3.24) to match the spectroscopy data of the Rydberg states, we can obtain an empirical MQDT model for the Rydberg series.

When the spin-orbit interaction is considerable, a frame transformation by introducing a purely LS-coupled channel $\bar{\alpha}$ is convenient to express the collision channel i in terms of the jj-couple channel. In this case, the matrix U is decomposed as

$$U_{i\alpha} = U_{i\bar{\alpha}} V_{\bar{\alpha}\alpha}. \quad (3.25)$$

The matrix element $U_{i\bar{\alpha}}$ corresponds to the LS-jj frame transformation and is given as

$$\begin{aligned} U_{i\bar{\alpha}} &= \langle ((S_c L_c) J_c (s l) j) J | ((S_c s) S (L_c l) L) J \rangle \\ &= \sqrt{(2S+1)(2L+1)(2J_c+1)(2j+1)} \begin{Bmatrix} S_c & L_c & J_c \\ s & l & j \\ S & L & J \end{Bmatrix}, \end{aligned} \quad (3.26)$$

where S_c , L_c , J_c are the total spin, orbital angular momentum, and total angular momentum of the core, s , l , j are the spin, orbital angular momentum, and total angular momentum of the Rydberg electron, and S , L , J are the total spin, orbital angular momentum, and total angular momentum of all electrons. For channel i (left-hand-side of the bracket), $S_c + L_c = J_c$, $s + l = j$, and $J_c + j = J$ while for channel $\bar{\alpha}$ (right-hand-side of the bracket), $S_c + s = S$, $L_c + l = L$, and $S + L = J$. The curly bracket $\{\cdot\}$ in equation (3.26) is the Wigner 9-j symbol. The matrix element $V_{\bar{\alpha}\alpha}$ is expressed with multiple rotation matrices $R(\theta)$ as

$$V_{\bar{\alpha}\alpha} = \prod_{k=1}^{N(N-1)/2} R(\theta_k), \quad (3.27)$$

where k runs over all combinations of the jj-coupled channels $(i, j) = (1, 2), (1, 3), \dots, (N-1, N)$, and θ_k is the Euler angle around channels i and j .

When the atom has a nuclear spin I , the hyperfine interaction should be taken into account. In this case, equation (3.23) is modified as

$$\det [U_{\alpha_F i_F} \sin(\pi(\nu_{i_F} + \mu_{\alpha_F}))] = 0, \quad (3.28)$$

where F represents the total angular momentum $F = J + I$. Then the matrix U is decomposed as

$$U_{i_F \alpha_F} = U_{i_F i} U_{i \alpha_F}, \quad (3.29)$$

with the transformation matrix of

$$U_{i_F i} = \langle ((J_c I) F_c j) F | ((J_c j) J I) F \rangle = (-1)^{I+j+F_c+J} \sqrt{(2J+1)(2F_c+1)} \begin{Bmatrix} I & J_c & F_c \\ j & F & J \end{Bmatrix}, \quad (3.30)$$

where $\{\cdot\}$ is the Wigner 6-j symbol. Since the hyperfine interaction is smaller than the non-Colombic interaction in close-coupling region, it can be treated as a perturbation. Thus, we can start to construct a MQDT model with the parameters obtained for the isotope with zero nuclear spin and equation (3.29).

Up to this point, the discussion has primarily focused on the matrix U . However, it is less favorable for numerical algorithms due to the presence of the sine function in equation (3.23). Instead, equivalent to equation (3.23), we can transform equation (3.22) as

$$\begin{aligned} \sum_i U_{\alpha i} \sin(\pi(\nu_i + \mu_{\alpha})) A_i = 0 &\iff \sum_i [U_{\alpha i} \tan(\pi \nu_i) + \tan(\pi \mu_{\alpha}) U_{\alpha i}] \cos(\pi \nu_i) A_i = 0 \\ &\iff \sum_i \left[\delta_{ji} \tan(\pi \nu_i) + \sum_{\alpha} (U_{j\alpha} \tan(\pi \mu_{\alpha}) U_{\alpha i}) \right] \cos(\pi \nu_i) A_i = 0 \quad (3.31) \\ &\iff \sum_i [K_{ji} + \delta_{ji} \tan(\pi \nu_i)] a_i = 0, \end{aligned}$$

where we define $K := U^\dagger \text{diag}(\pi \mu_{\alpha}) U$ and $a_i = \cos(\pi \mu_i) A_i$. Note that δ_{ji} is the Kronecker delta. The above equation has non-trivial solutions only when

$$\det [K_{ji} + \delta_{ji} \tan(\pi \nu_i)] = 0. \quad (3.32)$$

The matrix K is real symmetric matrix and referred as the reactance (or reaction) matrix, which has the information of the coupling between the channels. From equations (3.24), (3.32) and the spectroscopy data, we can estimate K matrix and ν_i , and obtain an empirical MQDT model. The frame transformation can be applied to the reaction matrix so as done to the matrix U .

3.2.3 Two-electron Rydberg atoms

In contrast to the Rydberg states of alkali atoms, the Rydberg state of two-electron atoms have an optically active inner valence electron. This offers unique properties of the two-electron Rydberg

atoms: isolated core excitation (ICE) and trappable Rydberg states, which can be advantages for quantum simulation and computing. In the following, we take ytterbium atom as an example of two-electron Rydberg atoms.

Isolated core excitation

ICE is the transition from a $(6snl)$ Rydberg state to $(6p_{1/2}n'l')$ state [150]. This transition can be used to induce light shifts of the Rydberg states without perturbing the ground state. Combining with local addressing of the ICE laser in an optical tweezer array, we can control the Rydberg excitation locally even with global irradiation of the Rydberg excitation laser [121, 122]. The important point is that the ICE does not affect the ground state, and thus the scattering error of the qubits encoded in the ground state can be suppressed.

Another application of the ICE is auto-ionization of the Rydberg states [74]. Since the $(6p_{1/2}n'l')$ state has larger energy than the ionization limit of a single electron, the state can be connected to the continuum states $|e\rangle$. As a result, the Rydberg electron is transitioned into a continuum state by absorption of a photon, that is ionization. This process is very quick (typically, GHz order) and used for fast and high-fidelity detection of the Rydberg states.

Trappable Rydberg states

The Rydberg electron feels a potential from the oscillating electric fields of the trapping laser, which is called the ponderomotive potential and given as [151]

$$U_{\text{pond}} = \frac{1}{2\epsilon_0 c} \frac{e^2}{m\omega^2} \int d^3r |\psi_{nl}(\mathbf{r})|^2 I(\mathbf{r} + \mathbf{R}), \quad (3.33)$$

where ω is the angular frequency of the trapping light, $\mathbf{r}$ is the Rydberg electron position with respect to the nucleus $\mathbf{R}$, $I(\mathbf{r})$ is trapping light intensity, and $\psi_{nl}(\mathbf{r})$ is the Rydberg electron wavefunction. This potential is always positive, and therefore the Rydberg electron feels a repulsive force. This is the reason why the Rydberg atoms of alkali are not trappable with commonly used red-detuned dipole traps. For this, the traps are usually turned off during the Rydberg excitation, limiting the interaction time between the Rydberg atoms and leading a risk to heat and lose the atoms [58].

On the other hand, in the case of two-electron Rydberg atoms, the ionic core has a finite polarizability α_{ion} . In such a situation, the light shift is written as

$$U_{\text{total}} = -\frac{1}{2\epsilon_0 c} \alpha_{\text{ion}} I(\mathbf{R}) + \frac{1}{2\epsilon_0 c} \frac{e^2}{m\omega^2} \int d^3r |\psi_{nl}(\mathbf{r})|^2 I(\mathbf{r} + \mathbf{R}). \quad (3.34)$$

For large n , the overlap of the Rydberg electron wavefunction and the trapping laser is small, and the second term is smaller than the first term, resulting in the attractive potential [120]. Therefore one can fully utilize the Rydberg lifetime and suppress the heating and loss of the Rydberg atoms.

3.2.4 Rydberg interaction and entanglement generation

Due to the large dipole moment of the Rydberg states, the atoms in Rydberg states can interact with each other even at a distance of several micrometers. This interaction is used to generate entanglement between the atoms in quantum computation [152, 153]. The interaction is described by the Hamiltonian given as

$$\mathcal{V}_{dd} = \frac{1}{4\pi\epsilon_0} \frac{\mathbf{d}_1 \cdot \mathbf{d}_2 - 3(\mathbf{d}_1 \cdot \mathbf{R}/R)(\mathbf{d}_2 \cdot \mathbf{R}/R)}{R^3}, \quad (3.35)$$

where $\mathbf{d}_1$ and $\mathbf{d}_2$ are the dipole moments of the atoms, and $\mathbf{R}$ is the distance vector between the atoms. Let us consider the atoms in the same Rydberg state $|nj\rangle$, where n is the principal quantum number and j is the other quantum number of the Rydberg state. The energy shift of the pair state $|nj; nj\rangle$ can be calculated by the perturbation theory as

$$E_{dd}^{(2)} = \sum_{n_1 j_1 n_2 j_2} \frac{|\langle nj; nj | V_{dd} | n_1 j_1; n_2 j_2 \rangle|^2}{\Delta E}, \quad (3.36)$$

with $\Delta E = 2E_{nj} - (E_{n_1 j_1} + E_{n_2 j_2})$. Here n_1, j_1, n_2, j_2 run over all possible Rydberg states except for $|nj\rangle$, and E_{nl} represents the unperturbed energy of the Rydberg state. Note that the first order perturbation term $\langle nj; nj | V_{dd} | nj; nj \rangle$ vanishes because the interaction Hamiltonian of equation (3.35) has odd parity. From table 3.6, one can find the scaling law of the Rydberg interaction as

$$E_{dd}^{(2)} = \frac{C_6}{R^6} \sim \frac{1}{R^6} \frac{(n^2 \cdot n^2)^2}{n^{-3}} = \frac{n^{11}}{R^6}. \quad (3.37)$$

This interaction is proportional to R^{-6} and is called van der Waals interaction. In optical tweezer arrays, the matrix element $|\langle nj; nj | V_{dd} | n_1 j_1; n_2 j_2 \rangle|^2$ is typically smaller than the energy difference ΔE due to relatively large site intervals of several micrometers, and thus the Rydberg interaction becomes van der Waals type.

However, ΔE sometimes becomes close to zero coincidentally for a certain Rydberg state, which is called Förster resonance. In this situation, the first order perturbation does not vanish due to the energy degeneracy, and the Rydberg interaction becomes

$$E_{dd}^{(1)} = \frac{C^3}{R^3} \sim \frac{n^4}{R^3}. \quad (3.38)$$

This interaction is called resonant dipole-dipole interaction and stronger than the van der Waals interaction. The Förster resonance is found in $|59D_{3/2}59D_{3/2}\rangle \leftrightarrow |61P_{1/2}57F_{5/2}\rangle$ of Rubidium system [154], for example. Recently, Förster resonances due to the ionic core hyperfine splitting of the two-electron atoms are found in ytterbium and strontium system [78, 149]. Note that in addition to the intrinsic Förster resonance, the electric field can induce the resonance by tuning the energy levels of the Rydberg states, which is called Stark-tuned Förster resonance [143, 155, 156].

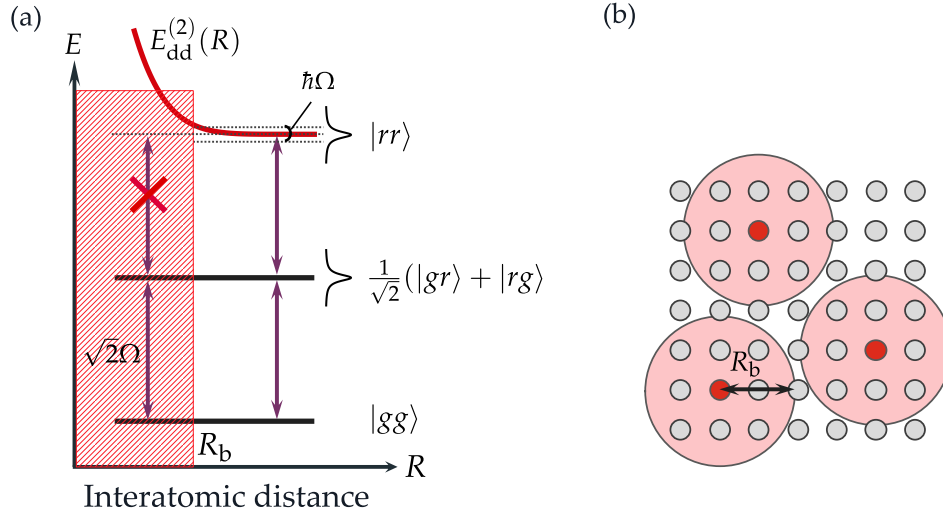


Figure 3.2 Rydberg blockade. (a) The Rydberg excitation of a second atom is suppressed due to the energy shift caused by the Rydberg interaction. (b) Rydberg blockade in an atom array. Only a single atom can be excited to the Rydberg state within the blockade radius.

The dipole-dipole interaction has angular dependence. The interaction Hamiltonian is decomposed into the spherical basis as [157]

$$\begin{aligned} \mathcal{V}_{dd} = \frac{1}{4\pi\epsilon_0 R^3} \big[& \mathcal{A}_1(\theta)(d_{1+}d_{2-} + d_{1-}d_{2+} + 2d_{1z}d_{2z}) \\ & + \mathcal{A}_2(\theta)(d_{1+}d_{2z} - d_{1-}d_{2z} + d_{1z}d_{2+} - d_{1z}d_{2-}) \\ & + \mathcal{A}_3(\theta)(d_{1+}d_{2+} - d_{1-}d_{2-}) \big], \end{aligned} \quad (3.39)$$

where $d_{k,\pm} = \mp(d_{k,x} \pm id_{k,y})$ are the spherical components of the dipole moment, and θ is the angle between the distance vector $\mathbf{R}$ and the quantization axis. The coefficients $\mathcal{A}_i(\theta)$ are given as

$$\mathcal{A}_1(\theta) = \frac{1 - 3\cos^2\theta}{2}, \quad \mathcal{A}_2(\theta) = \frac{3\sin\theta\cos\theta}{\sqrt{2}}, \quad \mathcal{A}_3(\theta) = \frac{3\sin^2\theta}{2}. \quad (3.40)$$

The index i of $\mathcal{A}_i(\theta)$ corresponds to the difference of the magnetic quantum number between the pair states ($\Delta m = 0, \pm 1, \pm 2$). Note that, in the case of van der Waals interaction, the angular dependence is almost averaged out in the summation of the pair states, and the interaction is nearly isotropic unless the pair states are close to the Förster resonance.

Rydberg blockade

When the Rydberg interaction strength V_{dd} is larger than the Rydberg excitation Rabi frequency Ω , the simultaneous Rydberg excitation is suppressed (figure 3.2). This phenomenon is called Rydberg blockade [57]. Since the interaction strength depends on the interatomic distance, we can define the blockade radius R_b as the distance where the interaction strength equals to the Rabi frequency.

quency as

$$R_b = \left(\frac{C_6}{\hbar\Omega} \right)^{1/6}. \quad (3.41)$$

More quantitatively, the Hamiltonian of two atoms under irradiation of the Rydberg excitation laser is given as

$$\begin{aligned} \mathcal{H} &= \frac{\hbar\Omega}{2} (|r\rangle\langle g| \otimes I + I \otimes |r\rangle\langle g| + \text{h.c.}) + V_{dd} |rr\rangle\langle rr| \\ &= \hbar \frac{\sqrt{2}\Omega}{2} \left(\frac{|gr\rangle + |rg\rangle}{\sqrt{2}} \langle gg| + |rr\rangle \frac{\langle rg| + \langle gr|}{\sqrt{2}} + \text{h.c.} \right) + V_{dd} |rr\rangle\langle rr|, \end{aligned} \quad (3.42)$$

where $|g\rangle$ and $|r\rangle$ are the ground and Rydberg states, respectively, and $I = |g\rangle\langle g| + |r\rangle\langle r|$ is the identity operator. While the first term and its hermite conjugate represent the Rabi oscillation between $|gg\rangle$ and $|\psi\rangle = (|gr\rangle + |rg\rangle)/\sqrt{2}$ with zero detuning, the second, third term, and their hermite conjugate represent the Rabi oscillation between $|\psi\rangle$ and $|rr\rangle$ with a detuning of V_{dd} . Within the blockade radius, where the Rabi frequency is smaller than the interaction strength, the simultaneous excitation is blocked due to large detuning. As a result, the state of the two atoms oscillates between the ground state $|gg\rangle$ and the entangle state $|\psi\rangle$ with a frequency of $\sqrt{2}\Omega$. Similarly, when N atoms are in the blockade radius, the state of the system oscillates as

$$|g_1 g_2 \cdots g_N\rangle \leftrightarrow \frac{1}{\sqrt{N}} \sum_{i=1}^N |g_1 g_2 \cdots r_i \cdots g_N\rangle, \quad (3.43)$$

with a frequency of $\sqrt{N}\Omega$.

Implementation of controlled-Z gate via Rydberg interaction

The most important application of the Rydberg interaction in the optical tweezer arrays is implementation of the controlled-Z (CZ) gates between the qubits, which are essential in universal quantum computation. In the following, we review the Rydberg interaction mediated CZ gates.

The first CZ gate is proposed in 2000 [152] and experimentally demonstrated in 2010 [22–24]. The gate scheme is based on local addressing of the Rydberg excitation laser and the Rydberg blockade. Let us consider two atoms, one of which is “control” qubit and the other is “target” qubit. The atoms have qubit subspace $|0\rangle$ and $|1\rangle$, and the Rydberg state $|r\rangle$. First, a π -pulse that transfers $|1\rangle$ to $|r\rangle$ is applied to the control qubit. Then, a 2π -pulse is applied to the target qubits. If the control qubit is initially in $|1\rangle$, the Rydberg excitation is suppressed due to the Rydberg blockade, and the state of the target qubit remains unchanged. On the other hand, if the control qubit is initially in $|0\rangle$, the Rydberg excitation of the target qubit occurs, and the state of the target qubit obtains a phase of -1 . Finally, a π -pulse is applied to the control qubit. This sequence of the pulses changes the state

as

$$\begin{aligned}
|0\rangle_c |0\rangle_t &\longrightarrow |0\rangle_c |0\rangle_t \longrightarrow |0\rangle_c |0\rangle_t \longrightarrow |0\rangle_c |0\rangle_t \\
|0\rangle_c |1\rangle_t &\longrightarrow |0\rangle_c |1\rangle_t \longrightarrow -|0\rangle_c |1\rangle_t \longrightarrow -|0\rangle_c |1\rangle_t \\
|1\rangle_c |0\rangle_t &\longrightarrow i|r\rangle_c |0\rangle_t \longrightarrow i|r\rangle_c |0\rangle_t \longrightarrow -|1\rangle_c |0\rangle_t \\
|1\rangle_c |1\rangle_t &\longrightarrow i|r\rangle_c |1\rangle_t \longrightarrow i|r\rangle_c |1\rangle_t \longrightarrow -|1\rangle_c |1\rangle_t
\end{aligned}$$

where the subscript c and t represent the control and target qubits, respectively. One can find that this state change is equivalent to the CZ gate by relabeling the states $|0\rangle$ and $|1\rangle$, up to global phase.

The above scheme relies on local control of the Rydberg excitation laser. However, this is technically demanding in the optical tweezer arrays. To overcome this difficulty, the CZ gate with global irradiation of Rydberg excitation laser is demonstrated in 2019 [73]. This scheme uses two Rydberg pulses with a certain detuning and relative phase jump between the pulses. While the dynamics of the state $|01\rangle$ and $|10\rangle$ are given by the single atom coupling of $|1\rangle \leftrightarrow |r\rangle$ with a Rabi frequency of Ω and detuning of Δ , the state $|11\rangle$ evolves with the collective coupling of $|11\rangle \leftrightarrow |\psi\rangle = (|1r\rangle + |r1\rangle)/\sqrt{2}$ with a Rabi frequency of $\sqrt{2}\Omega$ due to the Rydberg blockade. For a given detuning Δ , one can choose the pulse width τ such that the pulse becomes 2π -pulse for the state $|11\rangle$, and the phase jump of Rabi frequency $\Omega \rightarrow \Omega e^{i\zeta}$ such that the second pulse returns the state $|01\rangle$ and $|10\rangle$ to the initial state up to phase shift. This sequence changes the states as

$$\begin{aligned}
|00\rangle &\rightarrow |00\rangle, \\
|01\rangle &\rightarrow |01\rangle e^{i\phi}, \\
|10\rangle &\rightarrow |10\rangle e^{i\phi}, \\
|11\rangle &\rightarrow |11\rangle e^{i(2\phi-\pi)},
\end{aligned} \tag{3.44}$$

where ϕ is the phase obtained during the dynamics, which correspond to the geometric surface area of the Bloch sphere enclosed by the trajectory of the state. This state change is equivalent to the CZ gate up to global phase.

More recently, the CZ gate with the global irradiation of single pulse which has amplitude and phase modulation is proposed [158]. This gate scheme is called time-optimal CZ gate, and the modulation is optimized by the gradient ascent pulse engineering (GRAPE) technique. This gate is expected to be more robust against the finite Rydberg blockade strength, finite Rydberg lifetime, and experimental imperfection such as the fluctuation of the Rabi frequency and detuning. With this scheme, gate fidelity around 0.995 is achieved [76–79, 159]. The highest fidelity of the state of the art is 0.9972(3) demonstrated in ytterbium system [81].

Chapter 4

Experimental apparatus and techniques

This chapter reviews the apparatus and techniques used in the experiments. The chapter is divided into two sections. The first section describes the experimental apparatus, including the vacuum system, the magnetic field system, the laser system, and the optical setup. The second section describes the experimental techniques, including the optical tweezer array generation and trapping and imaging of single ytterbium atoms.

4.1 Experimental apparatus

4.1.1 Vacuum system

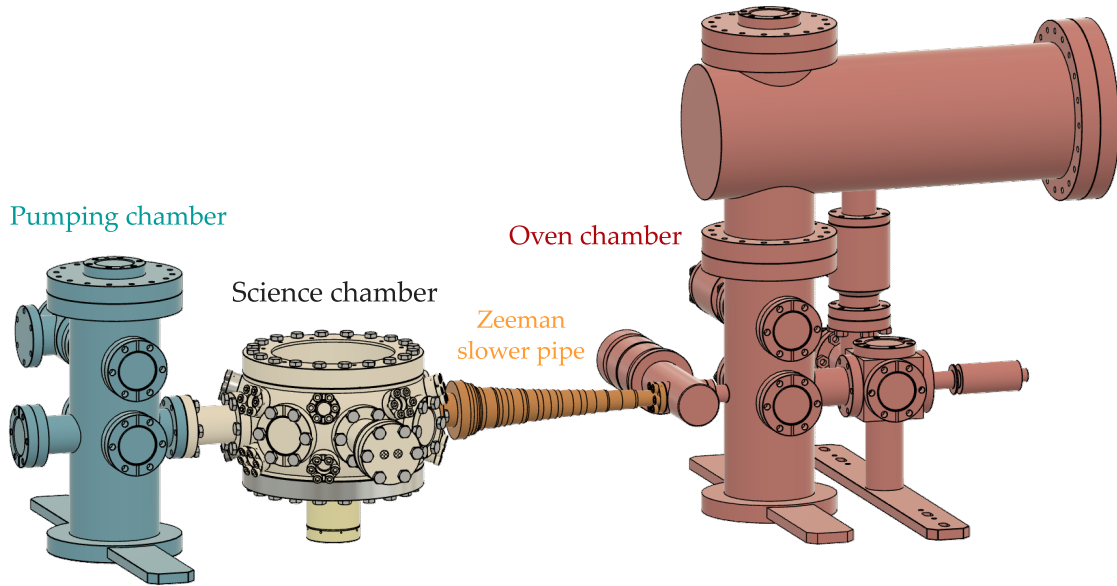


Figure 4.1 Overview of the vacuum chamber.

Our vacuum chamber consists of four parts: oven chamber, Zeeman slower pipe, science chamber, and pumping chamber (figure 4.1). The oven chamber is used to heat the ytterbium oven to produce ytterbium gas. The generated Yb gas is then cooled by Zeeman slowing technique in the Zeeman slower pipe. The pre-cooled Yb atoms are trapped and further cooled by magneto-optical trap (MOT) via $^1S_0 \rightarrow ^3P_1$ transition in the science chamber. The pumping chamber has an ion pump to maintain the vacuum in the science chamber. With the aid of additional ion pump and titanium sublimation pumps, the pressure at the science chamber is maintained at approximately 3×10^{-11} Torr.

The science chamber is specially designed for the optical tweezer array experiment and Rydberg spectroscopy (figure 4.2). The chamber is based on a commercial chamber (Kimball Physics, MCF800-ExtOct-G2C8A16), which has two ICF203, eight ICF70 and sixteen ICF34 flanges. Two reentrant viewports are installed on the top and bottom of the chamber (ICF203 flanges) for hosting the objective lenses, which are used for generating optical tweezers and imaging single atoms. Eight electrodes and a micro-channel plate (Hamamatsu Photonics, F4655-11) are installed inside the chamber for controlling the electric field and detecting the Rydberg atoms. The windows of reentrant viewports are indium-tin-oxide (ITO) coated to avoid charging up and to provide a good stability of the electric fields at the atom position. The details of the electrodes and the micro-channel plate are described in Ref. [160].

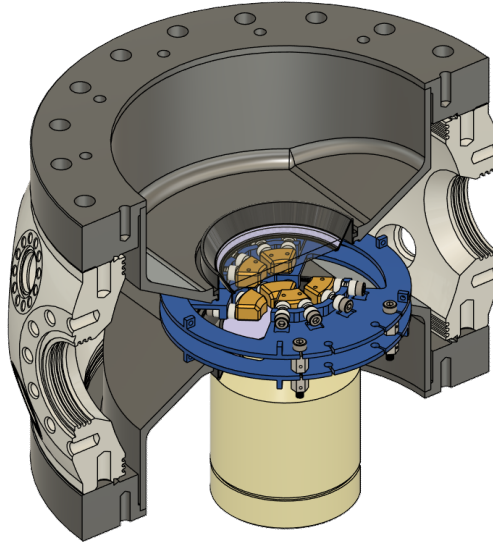


Figure 4.2 Schematic of the science chamber. The chamber has two reentrant (black) for the objective lenses, eight electrodes for the electric field control (yellow), and a micro-channel plate for the Rydberg atom detection.

4.1.2 Magnetic field system

There are four coils around the science chamber: the MOT coil and three compensation coils for x , y , and z direction. The MOT coil is used to generate the magnetic field gradient for the magneto-optical trap with anti-Helmholtz configuration, and strong bias field along z direction with Helmholtz configuration. The compensation coils are used to control the magnetic field at the atom position. Table 4.1 shows the specifications of the coils.

4.1.3 Laser system

Table 4.1 Specifications of the coils

	Number of turns	Configuration	Magnetic field
MOT coil	42×14	Anti-Helmholtz	11.6 (G/cm) / A
		Helmholtz	42.7 G/A
xy compensation	70	Helmholtz	0.5 G/A
z compensation	40	Helmholtz	3.4 G/A

399 nm lasers for probe

We have two 399 nm lasers for the probe of single atoms. The first laser is a commercial laser (Toptica, DL pro) with a power up to 70 mW. The second laser is a homemade interference-filter based external-cavity diode laser (IFDL) with a power of approximately 10 mW. The former is used for the probe of ^{171}Yb and the latter is for ^{174}Yb . These are frequency stabilized with a wavemeter (HighFinesse, WS8-10).

399 nm laser for Zeeman slowing

The Zeeman slower laser is generated by an intra-cavity second harmonic generation (SHG) from 798 nm light. The fundamental light with a power of 1.5 W from a commercial laser (Toptica, TA pro) is sent to a homemade SHG cavity with a lithium triborate (LBO) crystal. The cavity has a free spectral range (FSR) and finesse of 420 MHz and 380, respectively. The SHG output power is approximately 250 mW and delivered to the Zeeman slower through an end-cap fiber. The frequency of the fundamental light is stabilized by a wavemeter.

507 nm laser for metastable state excitation

The 507 nm laser is consisted of a commercial 1014 nm IFDL (Optoquest), a homemade tapered amplifier (TA), and a SHG with a periodically poled lithium niobate (PPLN) crystal (NTT electronics). Since the 1S_0 – 3P_2 transition is ultranarrow, the 1014 nm IFDL is frequency stabilized with an ultra-low expansion (ULE) cavity with a FSR of 1.5 GHz and a finesse of 5.8×10^5 , resulting in a laser linewidth of approximately 20 Hz, measured by a beat note with a stabilized optical frequency comb. The output power of the 507 nm laser is approximately 120 mW.

532 nm laser for optical tweezers

The optical tweezers are generated by a 532 nm diode-pumped solid-state laser (Coherent, Verdi V-10) with a power of approximately 10 W. The polarizabilities of the 1S_0 , 3P_1 , and 3P_2 states of ^{174}Yb for this wavelength are summarized in table 4.2. From these values, we can find that this wavelength can satisfy the near-magic conditions for the $|^1S_0\rangle \leftrightarrow |^3P_1, m_J = 0\rangle$ and $|^1S_0\rangle \leftrightarrow |^3P_2, m_J = 0\rangle$ transitions for ^{174}Yb as shown in figure 4.3, enabling efficient cooling and narrow linewidth excitation.

Table 4.2 Scalar and tensor polarizabilities of the 1S_0 [161], 3P_1 [161], and 3P_2 [162] states of ^{174}Yb at 532 nm. The polarizabilities are given in $\text{Hz}/(\text{W}/\text{cm}^2)$ and the light shift can be calculated by equation (2.69).

State	α_{nJ}^S	α_{nJ}^T
1S_0	37.9	—
3P_1	22.4(2)	−7.6(1)
3P_2	46.0(2)	7.5(3)

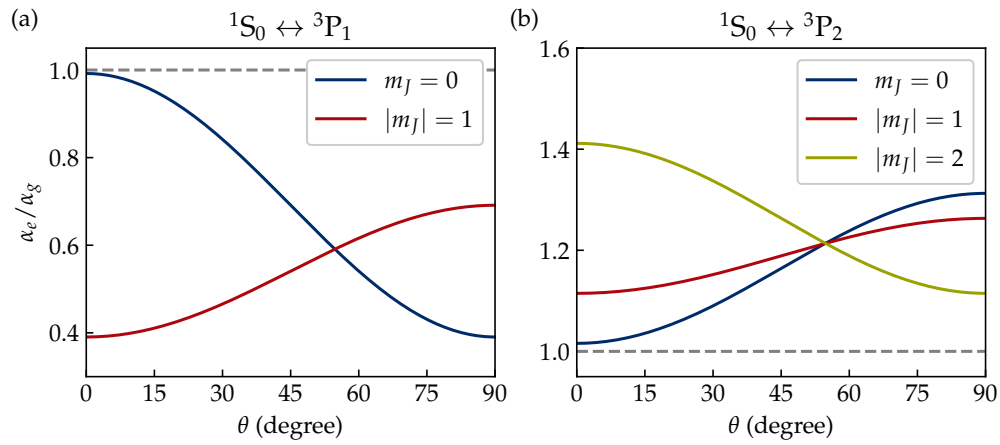


Figure 4.3 Polarizabilities of the 1S_0 , 3P_1 , and 3P_2 states of ^{174}Yb at 532 nm. (a) The ratio of the polarizabilities of the 1S_0 (α_g) and the 3P_1 (α_e) states. The 3P_1 atoms feel shallower trap than that for 1S_0 because $\alpha_e/\alpha_g < 1$. (b) The ratio of the polarizabilities of the 1S_0 (α_g) and the 3P_2 (α_e) states. The 3P_2 atoms feel deeper trap than that for 1S_0 because $\alpha_e/\alpha_g > 1$. The near-magic conditions for the $|^1S_0\rangle \leftrightarrow |^3P_1, m_J = 0\rangle$ and $|^1S_0\rangle \leftrightarrow |^3P_2, m_J = 0\rangle$ transitions are satisfied when the tweezer polarization is parallel to the quantization axis ($\theta = 0$).

556 nm lasers for MOT

We have two 556 nm laser systems for the MOT of ^{171}Yb and ^{174}Yb . The first laser is consisted of a commercial 1112 nm laser (Toptica, TA pro), an additional homemade TA, and two PPLN-based SHG modules (NTT electronics). The output power of the 556 nm is approximately 200 mW for both modules. The first module is used for the MOT of ^{171}Yb and the second is for Z gate operation on the nuclear spin qubits. The fundamental light is frequency stabilized with a ULE cavity, which is the same as for the 507 nm laser and has a finesse of 2×10^4 for this wavelength, resulting in a laser linewidth of approximately 50 kHz.

The second laser system is consisted of a commercial 1112 nm laser (Toptica, TA pro), and a PPLN-based SHG module (NTT electronics), and used for the MOT of ^{174}Yb . The output power of the 556 nm laser is approximately 200 mW. The frequency of the fundamental light is stabilized by

a offset lock to the first 1112 nm laser described above.

556 nm laser for nuclear spin manipulation

The 556 nm light for the nuclear spin manipulation is generated by a commercial 1112 nm fiber laser (Menlo systems, Orange-one) and two PPLN-based SHG modules (NTT electronics). The output power of the 556 nm laser is approximately 200 mW for both modules. The former is used for Raman sideband cooling and the latter is for X gate operation on the nuclear spin qubits. The frequency of the fundamental light is stabilized by a wavemeter.

649 nm and 770 nm lasers for repumping

The 649 nm and 770 nm lasers for repumping of the metastable 3P_2 state are IFDLs (Optoquest) with a power of approximately 10 mW and 20 mW, respectively. The frequency of the lasers are stabilized by a wavemeter.

325 nm laser for Rydberg excitation

The 325 nm laser for Rydberg excitation is consisted of a 1300 nm laser (Toptica, TA pro), a Raman fiber amplifier (MPB communications) and waveguide SHG module, and a homemade SHG cavity with a BBO crystal. The details of this laser system are described in chapter 7.

4.1.4 Objective lenses

We have two custom-made objective lens made by Special Optics. One is for the optical tweezer array generation and 556 nm fluorescence imaging of single atoms, and the other is for the 399 nm fluorescence imaging. Hereafter, the former is referred as the visible (VIS) objective and the latter is the ultraviolet (UV) objective. The specifications of the objective lenses are summarized in table 4.3. While the VIS objective is designed to have diffraction limited performance for 532 nm, 556 nm, and 759 nm light, the UV objective is designed for 399 nm and 325 nm.

Table 4.3 Specifications of the objective lenses

	NA	Working distance	Focal length	Field of View
VIS objective	0.6	20.25 mm	20.0 mm	200 μm $\times$ 200 μm
UV objective	0.6	23.38 mm	28.7 mm	200 μm $\times$ 200 μm

The VIS objective is mounted on a assembled stage of 3-axis translation stage (Suruga, B71-80-C) and 5-axis stage (Newport, 9082-V-M), as shown in figure 4.4 (a). This stage allows us to adjust the position of the objective lens with a high precision. The UV objective is mounted on a piezo linear Z stage (PI, P-528.ZCD), which is fixed to a hexapod (PI, H811.I2), as shown in figure 4.4 (b). The hexapod is used for fine adjustment of the UV objective position and the piezo stage is for dynamical adjustment of the focus during three-dimensional optical tweezer array experiment.

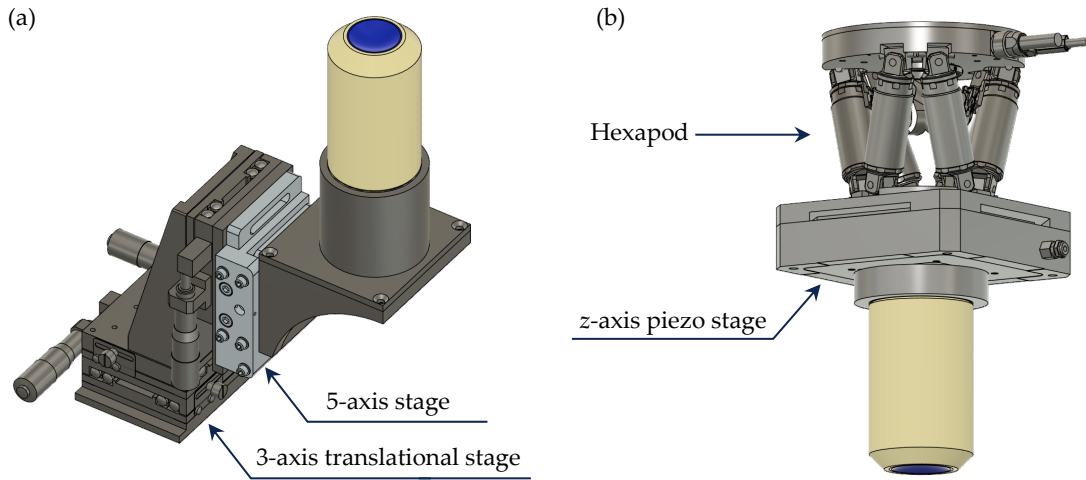


Figure 4.4 Assembly of the objective and stages. (a) VIS objective mounted on the 3-axis and 5-axis stages. (b) UV objective mounted on the piezo Z stage and the hexapod.

4.1.5 Optical setup

The optical path around the science chamber is shown in figure 4.5 and 4.6. On the first floor of the optical table, a diode-pumped solid-state laser (Coherent, Verdi V-10) is placed for optical tweezer array generation. The laser output is firstly divided into two paths: one is for static tweezer array, and the other for the moving tweezers. Each beam is then passes through an acousto-optic modulator (AOM) and coupled to a single mode fiber to clean the beam profile and stabilize the beam pointing. The AOMs are used for switching and power stabilizing of the tweezer beams. The fiber output for static tweezers is delivered to a spatial light modulator (SLM) (Hamamatsu Photonics, X15213-16L). Another fiber output for moving tweezer is sent to an acousto-optic deflector (AOD) (AA Opto-Electronic, DTSX-400-532) for horizontal scanning and then to another AOD for vertical scanning via a $4f$ -system. The beams coming from the SLM and AODs are combined by a polarizing beam splitter (PBS) and sent to the second floor of the optical table.

The fiber outputs for MOT Z beam of ^{171}Yb and ^{174}Yb are also placed on the first floor. The beams are combined by a PBS and sent to the second floor. On the second floor, the MOT beams are focused onto the mini mirror, which is a two inches fused silica substrate with a 10 mm mirror coating at the center. This mini mirror configuration allows us to separate the retroreflected MOT Z beam and 556 nm atom fluorescence [113], and thus enables 556 nm fluorescence imaging of the single atoms. The MOT beams reflected by the mini mirror are expanded by a telescope and combined with the optical tweezer beams by a dichroic mirror. Then the combined beams are sent to the science chamber through the VIS objective on the second floor. Note that the MOT beams are focused around the back focal plane of the objective to avoid focusing at the atom position.

The main optical paths, such as MOT XY beams, probe beam, optical pumping (OP) beam, Raman sideband cooling (RSC) beams, metastable excitation beam, repumping beam, and Rydberg

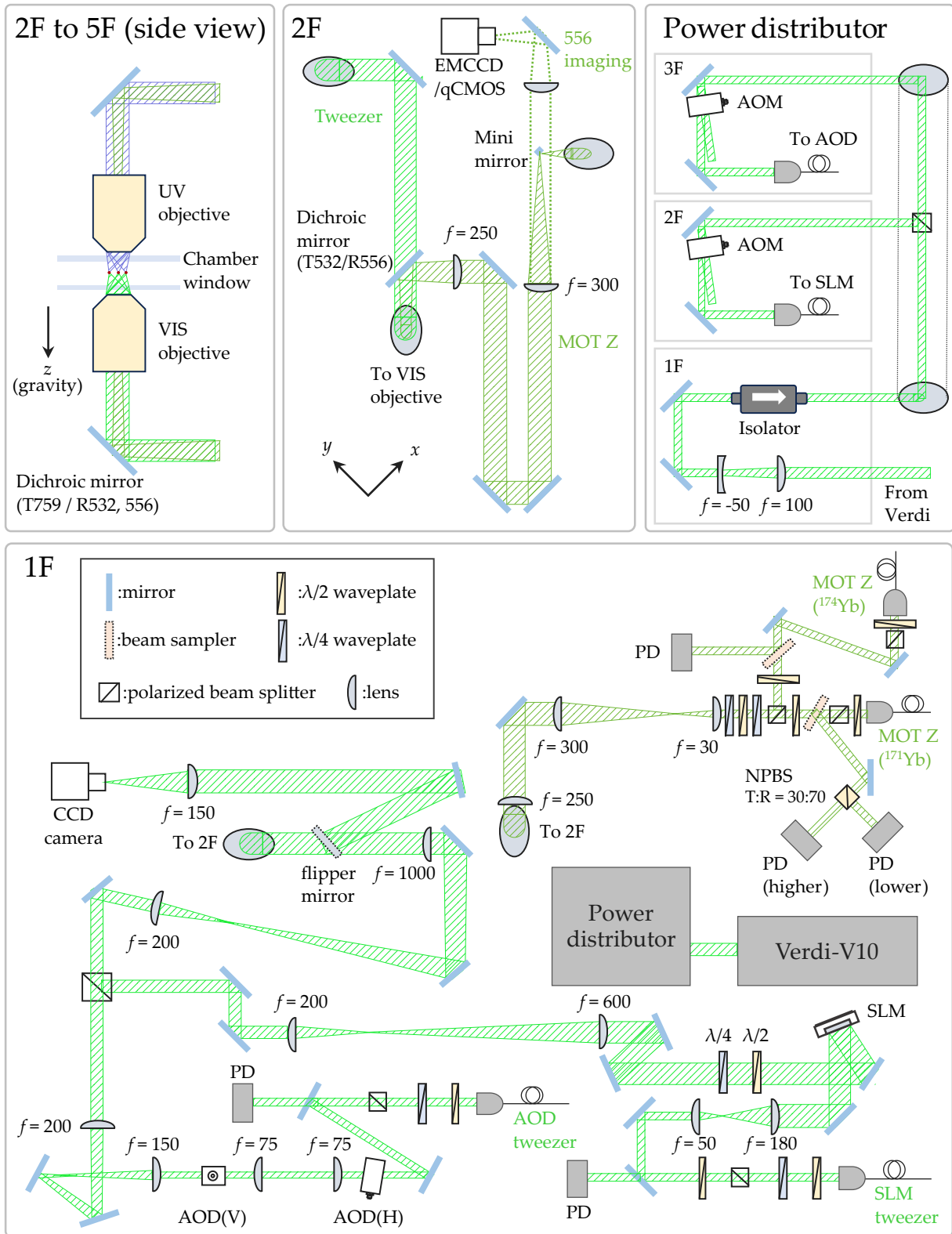


Figure 4.5 Optical setup of the first and second floor.

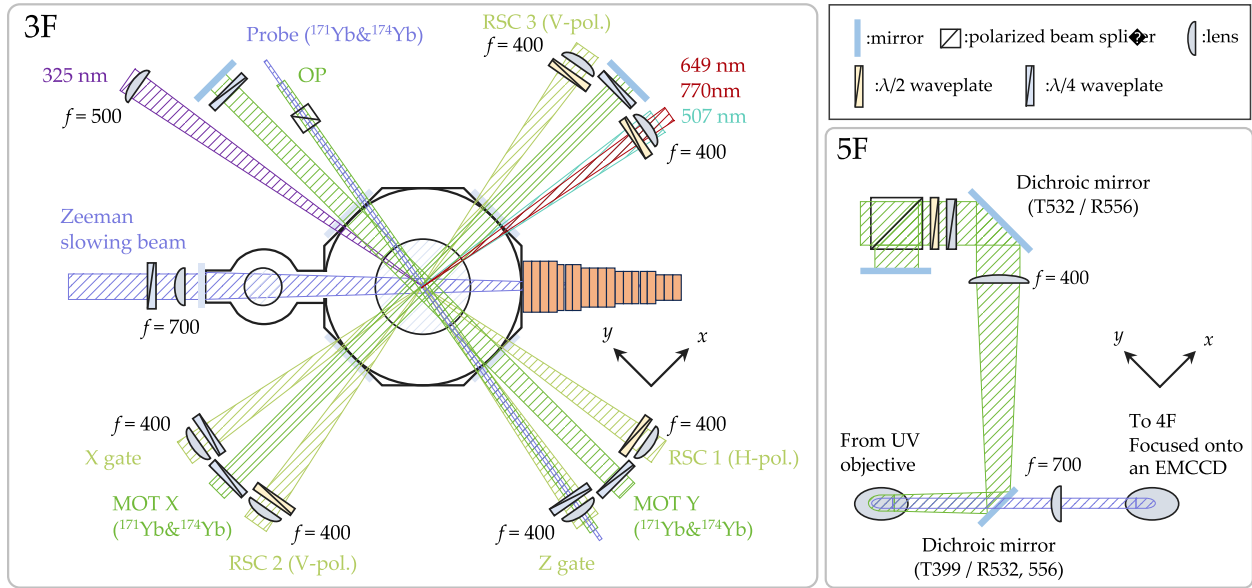


Figure 4.6 Optical setup of the third and fifth floor.

excitation beam, are placed on the third floor as shown in 4.6.

On the fourth and fifth floor, the optical paths for MOT Z beams and atom imaging are placed. The hexapod, which has the piezo Z stage and the UV objective, is inversely fixed to the fifth floor. The MOT Z beams, tweezer beams, and 399 nm atom fluorescence passing through the UV objective are separated by dichroic mirrors on the fifth floor. The MOT Z beams then pass a lens and are retroreflected by a mirror. The atom fluorescence is focused onto an electron-multiplying charge-coupled device (EMCCD) camera (Andor, iXon Ultra 897 EXF) on the fourth floor.

4.2 Experimental techniques

4.2.1 Optical tweezer array generation

The optical tweezer array is the essential technique for this study. In the following, we overview the generation of the optical tweezer array by SLM and AOD.

Static tweezer array by SLM

The static tweezer array is generated by an SLM. The SLM displays a hologram, which can modulate the wavefront of the incident beam arbitrarily, resulting in arbitrary intensity pattern at the focal plane [95]. We use the weighted Gerchberg-Saxton algorithm [163, 164] to design the hologram for the optical tweezer array.

Let us consider that the incident beam of a wavelength λ has an electric field amplitude A_0 , and the target intensity pattern at the focus of a lens with a focal length of f is given as I_t (figure 4.7). We

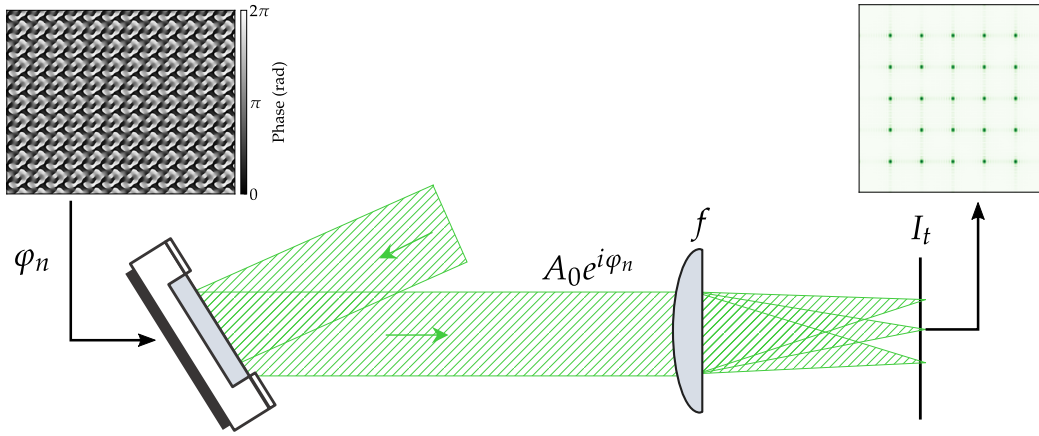


Figure 4.7 Optical tweezer array generation by SLM.

iteratively update the phase hologram φ_n on the SLM from a random phase φ_0 to minimize the error between the target intensity pattern and the actual intensity pattern I_n at the focus. We introduce the complex amplitude of the electric field of the k -th trap $V_n^{(k)}$ at the focal plane in the n -th iteration as [164]

$$V_n^{(k)} = \iint A_0 \exp \left[i \left(\varphi_{n-1}(x_s, y_s) - \Delta^{(k)}(x_s, y_s) \right) \right] dx_s dy_s, \quad (4.1)$$

with the pixel coordinates of the SLM (x_s, y_s) and the phase $\Delta^{(k)}$ given as

$$\Delta^{(k)} = \frac{2\pi M}{f\lambda} \left(x^{(k)} x_s + y^{(k)} y_s \right), \quad (4.2)$$

where M is the magnification of the optical system, $(x^{(k)}, y^{(k)})$ are the coordinates of the k -th trap. The intensity of the k -th trap $I_n^{(k)}$ is calculated as $I_n^{(k)} = |V_n^{(k)}|^2$. If the calculated intensity is converged to the target intensity, the iteration is stopped. If not, the phase hologram is updated by the following equations:

$$w_n^{(k)} = w_{n-1}^{(k)} \sqrt{\frac{\langle I_n \rangle}{I_n^{(k)}}}, \quad (4.3)$$

$$\theta_n^{(k)} = \arg \left[V_n^{(k)} \right], \quad (4.4)$$

$$\varphi_n = \arg \left[\sum_k w_n^{(k)} \exp \left[i \left(\Delta^{(k)}(x_s, y_s) + \theta_n^{(k)} \right) \right] \right], \quad (4.5)$$

where $w_n^{(k)}$ is the weight for the site k in the n -th iteration, $\langle I_n \rangle$ is the average of each site intensity $I_n^{(k)}$. This algorithm ensures that the efficiency of the diffraction $e = \sum_k I^{(k)} / \langle I \rangle$ is maximized, and the inhomogeneity of the intensity pattern $\sigma = \sqrt{\langle I^2 \rangle - \langle I \rangle^2} / \langle I \rangle$ is minimized (figure 4.8 (a)).

Although the algorithm works efficiently and predicts the intensity inhomogeneity of 1%, the actual intensity inhomogeneity at the atom position is around 10% due to the imperfection of the

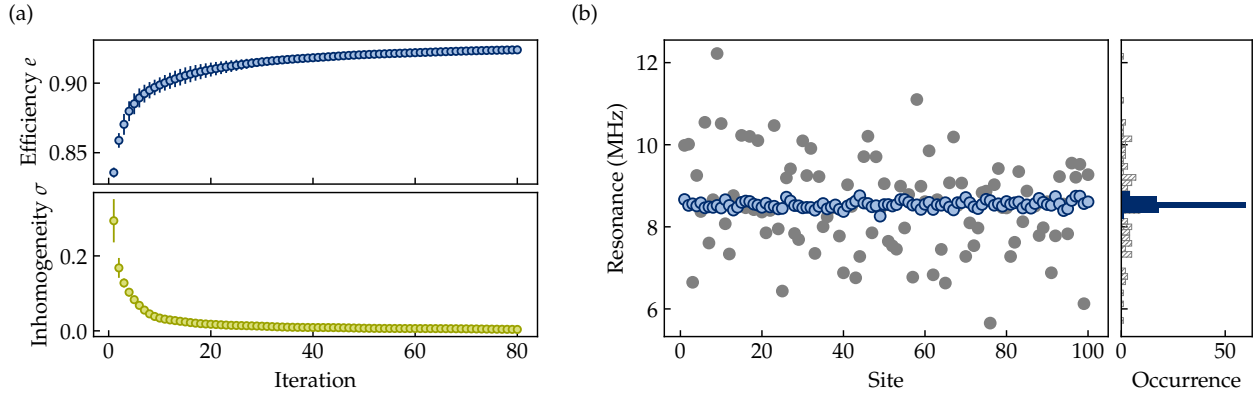


Figure 4.8 Performance of the WGS algorithm. (a) The calculated diffraction efficiency and inhomogeneity of the tweezer array as a function of the iteration of the WGS algorithm. After 80 iterations, the algorithm converges to the maximum efficiency of 92 % and minimum inhomogeneity of 0.3 %. (b) The actual inhomogeneity of the optical tweezer array before and after the equalization via spectroscopy of the $|^1S_0\rangle \leftrightarrow |^3P_1, m_J = \pm 1\rangle$ transition of ^{174}Yb . The intensity inhomogeneity is reduced from 13.3 % (gray) to 1.03 % (blue).

optical system. Therefore, we need to equalize the intensity of the optical tweezers by feedback method. For this, we measure the differential light shift (DLS) of the 1S_0 and the 3P_1 states in each site using $|^1S_0\rangle \leftrightarrow |^3P_1, m_J = \pm 1\rangle$ transition for ^{174}Yb or, $|^1S_0\rangle \leftrightarrow |^3P_1, F = 3/2, m_F = \pm 1/2\rangle$ transition for ^{171}Yb . Based on the measured DLS, which is proportional to the intensity of the optical tweezer, we adjust the intensity of the optical tweezer according to equation (4.3). After this feedback, we typically achieve the intensity inhomogeneity of approximately 1 % as shown in figure 4.8 (b).

The attractive feature of the SLM is that we can compensate the aberration by adding the Zernike polynomial phases to the hologram. The Zernike polynomial Z_n^m is given as

$$Z_n^m(\rho, \theta) = \begin{cases} R_n^m(\rho) \cos(m\theta) & (m > 0) \\ R_n^{|m|}(\rho) \sin(|m|\theta) & (m < 0), \end{cases} \quad (4.6)$$

with

$$R_n^m(\rho) = \sum_{k=0}^{\frac{n-m}{2}} \frac{(-1)^k (n-k)!}{k! (\frac{n+m}{2} - k)! (\frac{n-m}{2} - k)!} \rho^{n-2k}. \quad (4.7)$$

Each Zernike polynomial corresponds to the specific aberration, such as astigmatism, coma, trefoil, and so on. We optimize the weights a_n^m of the Zernike polynomials to minimize the trap waist, which can be estimated from the trap frequencies measured by Raman sideband spectroscopy of ^{171}Yb , as

$$\varphi_{\text{aber}} = \sum_{m,n} a_n^m Z_n^m(\rho, \theta). \quad (4.8)$$

Figure 4.9 presents the optimized φ_{aber} in our setup.

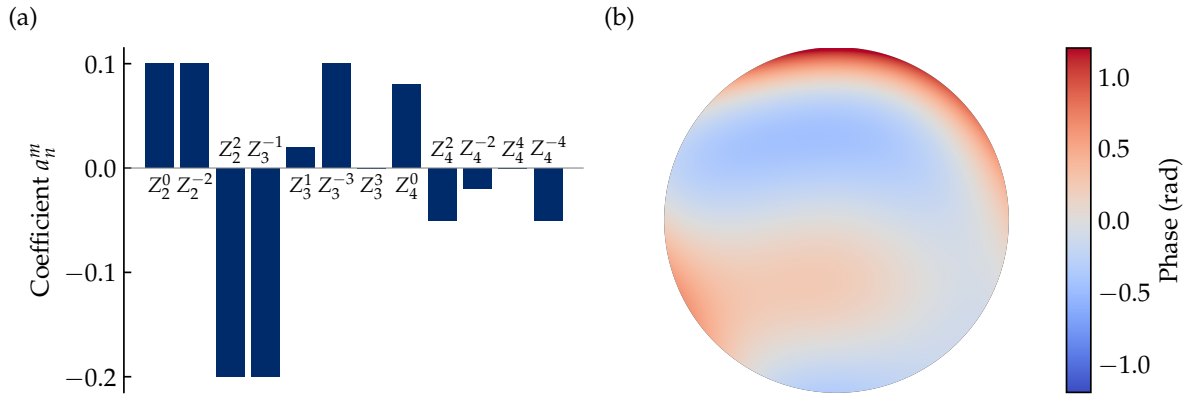


Figure 4.9 Optimized Zernike polynomial phase for the optical tweezer array in our setup. (a) The coefficients a_n^m for the optimized φ_{aber} . (b) Calculated Zernike polynomial phase.

Moving tweezers by AOD

Moving tweezers play a pivotal role in neutral atom based quantum computer. They are used for making a defect-free atom array [28,29,165] and for implementing zoned-architecture [88,89].

The AOD has a crystal inside, which can deflect the incident beam by the Bragg diffraction when radio frequency (RF) signal is applied. The diffraction angle θ_D and the diffracted light frequency f_D are given as

$$\theta_D = \sin^{-1} \left(\frac{cf_{\text{RF}}}{vf_{\text{photon}}} \right) \quad (4.9)$$

$$f_D = f_{\text{photon}} + f_{\text{RF}},$$

where c is the speed of light, v is the acoustic velocity in the crystal, f_{RF} is the RF frequency, and f_{photon} is the photon frequency. Since the diffraction angle is proportional to the RF frequency, when

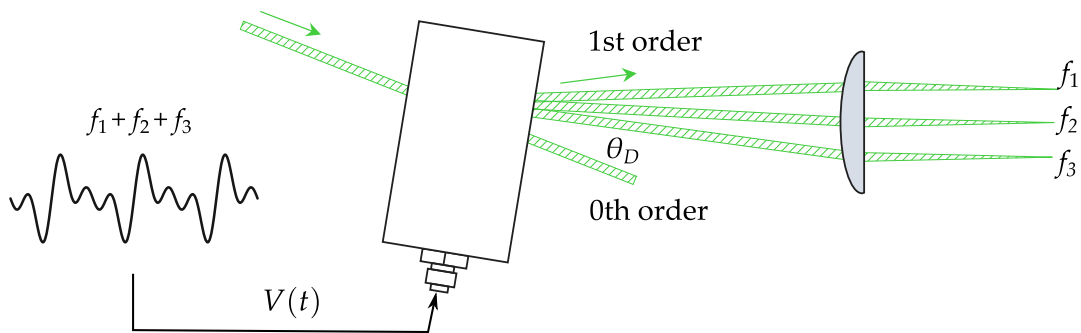


Figure 4.10 Optical tweezer generation by AOD.

a multitone RF signal given as

$$V(t) = \sum_{i=1}^N A_i \sin(2\pi f_i t + \phi_i), \quad (4.10)$$

is applied to the AOD, N diffracted beams are generated (figure 4.10). Here, A_i , f_i , and ϕ_i are the amplitude, frequency, and phase of the i -th RF signal, respectively.

Although the phases $\{\phi_i\}$ seems to be arbitrary, they are important to maintain the diffraction efficiency when the number of the frequency components is large. The limited RF voltage range of the AOD requires low peak-to-average power ratio (PAPR) defined as $V_{\max}^2 / \langle V^2 \rangle$. If the maximum voltage $V_{\max}$ is linearly proportional to the number of frequency components, the amplitudes $\{A_i\}$ have to be reduced not to exceed the applicable voltage range, resulting in low diffraction efficiency. To avoid this, it is necessary to choose the phases $\{\phi_i\}$ to achieve the PAPR. For this, we employ $\{\phi_i\}$ by Kitayoshi algorithm [166] as

$$\phi_k = \frac{\pi k(k+1)}{N}. \quad (4.11)$$

Figure 4.11 shows the multitone signals with and without employing the Kitayoshi algorithm. The PAPR is reduced from 33.4 to 3.62 for the 16-tone signal.

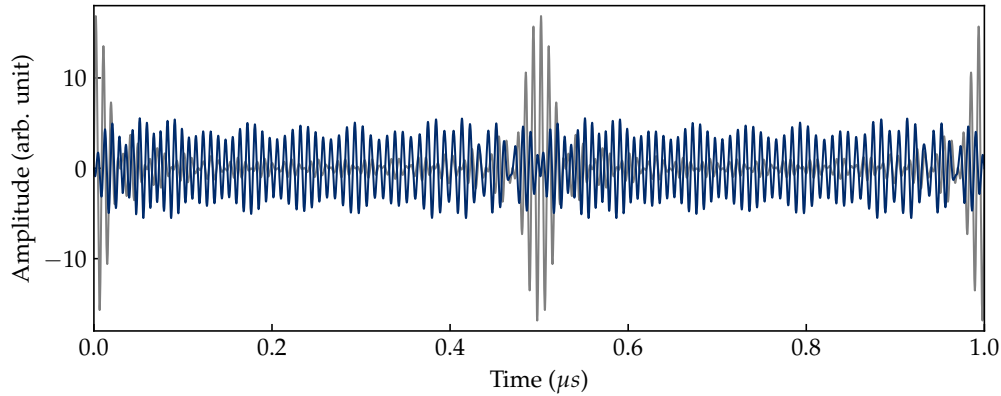


Figure 4.11 Multitone signal with the Kitayoshi phase, respectively. Gray and blue lines represent 16-tone signals with and without the Kitayoshi phase. The frequencies are equally distributed from 100 MHz to 132 MHz. The PAPR is reduced from 33.4 to 3.62 for this case.

Frequency sweep of the RF signal can be used for the fast scanning of the optical tweezer, that is moving tweezer. The sweep schedule have to be carefully designed to avoid the atom heating during the transport. We use the following trajectory for the moving tweezer:

$$f(t) = f_s + (f_e - f_s) \sin^2 \left(\frac{\pi t}{2T} \right), \quad (4.12)$$

where f_s, f_e are the start and end frequency, and T is the sweep time. This trajectory ensures that the acceleration of the tweezer is zero at the start and end of the sweep. Other groups use different

trajectory called minimal jerk trajectory given as

$$f(t) = f_s + (f_e - f_s) \left[6 \left(\frac{t}{T} \right)^5 - 15 \left(\frac{t}{T} \right)^4 + 10 \left(\frac{t}{T} \right)^3 \right], \quad (4.13)$$

which make the jerk minimal during the transport [88, 167]. Recently, the shortcut to adiabaticity theory is applied to the moving tweezer to minimize the transport time and the atom heating [168].

4.2.2 Light-assisted collision

When atoms are loaded into optical tweezers from the MOT, each site initially contains multiple atoms. To prepare single atoms trapped in the tweezers, we utilize light-assisted collisions (LAC). This process induces two-body loss, leading to single-atom occupancy in tweezers with an odd number of atoms. Consequently, the probability of achieving single-atom occupancy is typically 50 %.

The LAC process is illustrated in figure 4.12. Consider two atoms separated by an interatomic distance R . When both atoms are in the ground state $|S\rangle$, the interatomic potential is described by $V_{|S+S\rangle}(R) = -C_6/R^6$, where C_6 represents the van der Waals interaction coefficient. In contrast, if one atom is in the excited state $|P\rangle$, the potential is given by $V_{|S+P\rangle}(R) = \hbar\omega_0 \pm C_3/R^3$, where ω_0 is the resonant frequency of the $|S\rangle \leftrightarrow |P\rangle$ transition, and C_3 is the dipole-dipole interaction

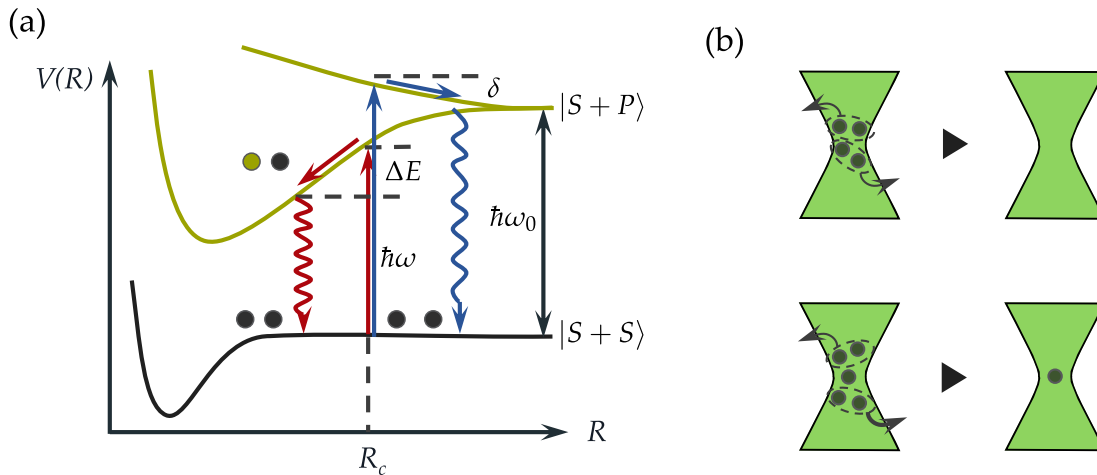


Figure 4.12 Light-assisted collision process. (a) When atoms are excited by a red-detuned LAC beam, they experience an attractive potential, resulting in the acquisition of kinetic energy ΔE as the interatomic distance decreases. Conversely, excitation with a blue-detuned LAC beam allows control of the kinetic energy up to the detuning δ , thereby enhancing the loading probability. (b) After the LAC process, no atoms remain in the tweezers initially containing an even number of atoms, whereas a single atom persists in the tweezers with an odd number of atoms. For red-detuned LAC, the single-atom loading probability is approximately 50 %.

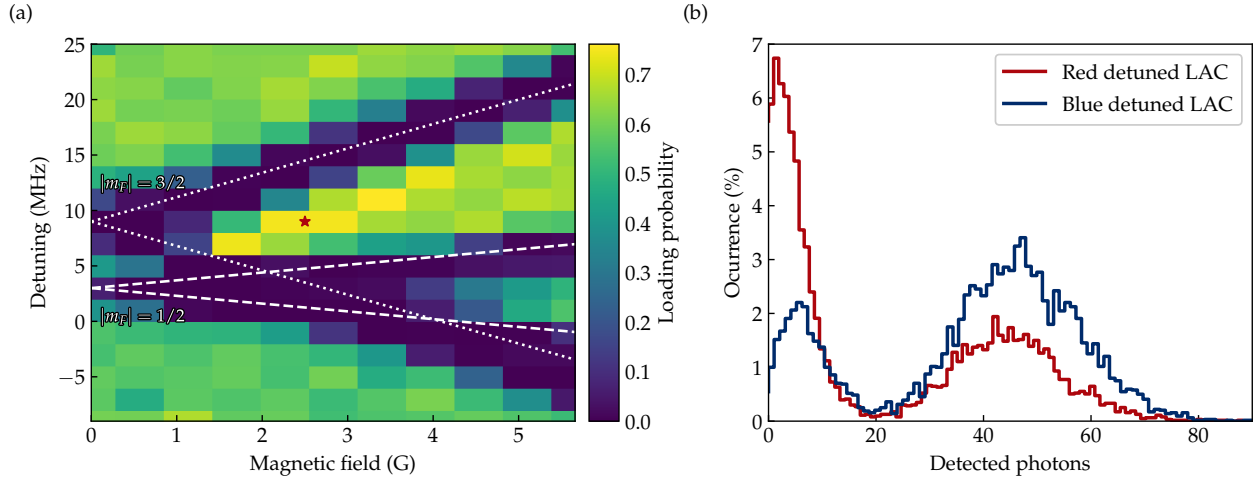


Figure 4.13 Blue-detuned LAC. (a) Loading probability as a function of the magnetic field and LAC beam detuning. When the magnetic field and the LAC detuning are set to around 2.5 G and 9.5 MHz from the free space resonance, respectively, the loading probability is enhanced. The white dashed and dotted line represent the resonances of $|m_F| = 1/2$ and $|m_F| = 3/2$ states, respectively. (b) Histograms of detected photons. When we set the LAC parameters to the optimal point, which is indicated by a star in (a), the loading probability is increased from 45 % to 78 %.

coefficient. When a red-detuned light with frequency ω is applied and the interatomic distance R_c satisfies the condition $V_{|S+P\rangle}(R_c) - V_{|S+S\rangle}(R_c) = \hbar\omega$, one atom is excited to the $|P\rangle$ state. Due to the attractive nature of the interatomic potential in this configuration, the atoms acquire kinetic energy as they move closer together. After the excited atom decays back to the ground state, the atoms gain an energy ΔE , which is sufficient for them to escape the optical tweezer potential. This results in two-body loss.

If the trap volume is so small that the loading rate of atoms is much smaller than the two-body loss rate, the number of atoms in the tweezer after LAC is restricted to zero or one. This phenomenon is called collisional blockade [169].

In the above discussion of LAC, we assumed the red-detuned LAC beam. However, the blue-detuned LAC beam can also be used to induce LAC. Notably, this process enhance the single atom loading probability up to 90 % [97, 113, 165, 170–172]. In this case, the blue-detuned LAC beam with a frequency of $\omega_0 + \delta$ excite one of the atoms to the $|P\rangle$ state. The interatomic potential is then repulsive, and the atoms acquire kinetic energy up to δ . When the detuning δ is larger than the trap depth, but smaller than twice of the trap depth, only one atoms can escape from the trap. Consequently, the single atom loading probability is enhanced.

In our setup, we observed the enhanced loading by the blue-detuned LAC transition in the ^{171}Yb tweezer array. We use the $|^1S_0\rangle \leftrightarrow |^3P_1, F = 3/2\rangle$ transition for this purpose. Figure 4.13 shows the loading probability as a function of the magnetic field and LAC beam detuning. When the magnetic

field along the tweezer polarization is set to 2.5 G, the loading probability is increased up to 78 %. Further enhancement can be achieved by increasing the atomic density around the tweezer during the loading process.

4.2.3 Single atom imaging

The single atom imaging is performed by the fluorescence imaging technique. In this study, we use the 399 nm beam with an intensity of $1 \times 10^{-3} I_{\text{sat},399}$ as a probe, and the 556 nm beams, which are the same as the MOT beams, with a total intensity of $10 I_{\text{sat},556}$ as a simultaneous cooling light. Note that the 556 nm fluorescence imaging is also possible in our setup [112], but the 532 nm tweezer beam induces auto-fluorescence around 556 nm at the objective housing [111], preventing high fidelity imaging in large scale array.

The photon emission pattern from the atoms obeys the dipole radiation pattern $\Phi(\theta, \phi)$, which is given as [127]

$$\Phi(\theta, \phi) = \begin{cases} \frac{3}{8\pi} \sin^2 \theta & (\Delta m = 0) \\ \frac{3}{16\pi} (1 + \cos^2 \theta) & (\Delta m = \pm 1), \end{cases} \quad (4.14)$$

where θ is the angle from the quantization axis and Δm is the change of the magnetic sublevels in the transition. To maximize the photon collection efficiency, we set the probe beam polarization to be linear and in xy -plane, that is orthogonal to the objective axis (z -axis). When the quantization

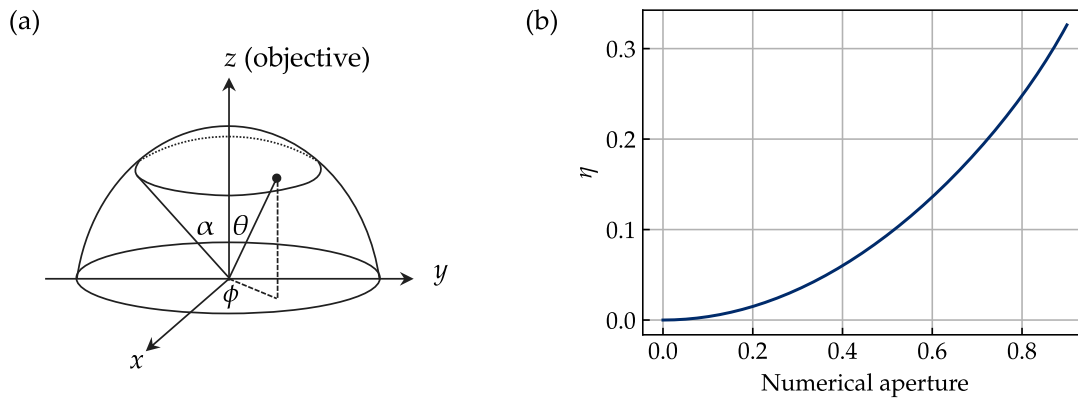


Figure 4.14 Calculation of the photon collection efficiency. (a) Coordinates for the calculation. (b) Photon collection efficiency as a function of the numerical aperture of the objective.

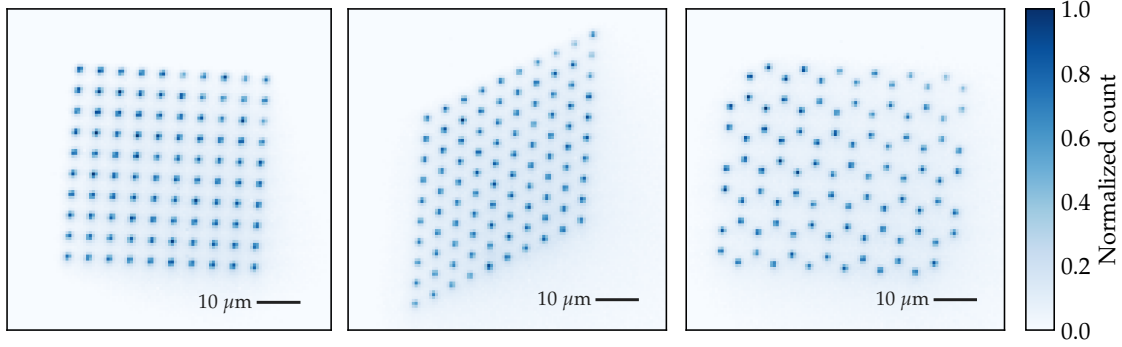


Figure 4.15 Averaged fluorescence images of single ^{174}Yb atoms trapped in various tweezer arrays. (left) Square array of 600 shots average. (middle) Triangle array of 300 shots average. (right) Honeycomb array of 300 shots average.

axis is set to be the z -axis, the photon collection efficiency η is given as

$$\begin{aligned}\eta &= \int_0^{2\pi} d\phi \int_0^\alpha d\theta \sin \theta \Phi(\theta, \phi) = \frac{3}{16\pi} \int_0^{2\pi} d\phi \int_0^\alpha d\theta \sin \theta (1 + \cos^2 \theta) \\ &= \frac{1}{8} [4 - \cos \alpha (3 + \cos^2 \alpha)] \\ &= \frac{1}{8} [4 - (4 - \text{NA}^2) \sqrt{1 - \text{NA}^2}],\end{aligned}\tag{4.15}$$

where $\text{NA} = \sin \alpha$ is the numerical aperture of the objective. Similarly, when the quantization axis is set to be in the xy -plane, the photon collection efficiency is given as

$$\begin{aligned}\eta &= \int_0^{2\pi} d\tilde{\phi} \int_0^\alpha d\tilde{\theta} \sin \tilde{\theta} \Phi(\tilde{\theta}, \tilde{\phi}) = \frac{3}{8\pi} \int_0^{2\pi} d\tilde{\phi} \int_0^\alpha d\tilde{\theta} \sin \tilde{\theta} (\cos^2 \tilde{\theta} + \sin^2 \tilde{\theta} \cos^2 \tilde{\phi}) \\ &= \frac{1}{8} [4 - \cos \alpha (3 + \cos^2 \alpha)] \\ &= \frac{1}{8} [4 - (4 - \text{NA}^2) \sqrt{1 - \text{NA}^2}].\end{aligned}\tag{4.16}$$

Here we take a rotated frame $(\tilde{\theta}, \tilde{\phi})$ so that the objective axis corresponds to $\tilde{\theta} = 0$ axis. In this frame, $\sin^2 \theta = \cos^2 \tilde{\theta} + \sin^2 \tilde{\theta} \cos^2 \tilde{\phi}$. From the above calculations, we find that the above two cases have the same photon collection efficiency. Figure 4.14 shows the photon collection efficiency as a function of the numerical aperture of the objective. In our setup, the NA of the objective is 0.6, corresponding to the theoretical photon collection efficiency of 13.6 %.

Figure 4.15 presents the 399 nm fluorescence image of single ^{174}Yb atoms trapped in various tweezer arrays. The imaging performance will be discussed in section 6.3.1.

4.2.4 Release and recapture of single atoms

The temperature of the single atoms in the optical tweezer array can be estimated by release and recapture method [173]. In this method, the atoms are firstly released from the optical tweezer

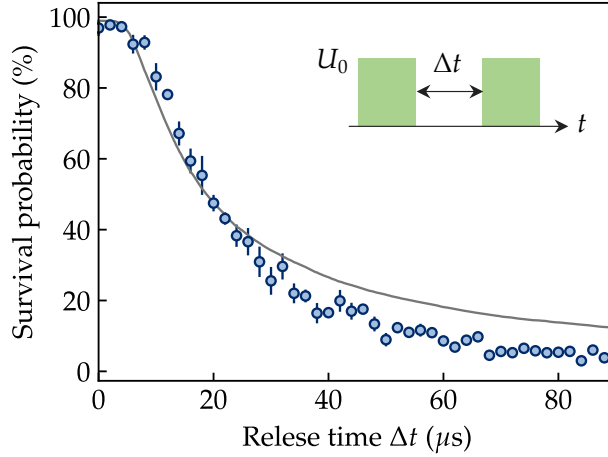


Figure 4.16 Release and recapture experiment of ^{174}Yb . The recapture probability as a function of the time of flight Δt . The gray line is the fitting result from 100 trials of the Monte Carlo simulation. The temperature is estimated to be $35(3) \mu\text{K}$.

array by turning off the optical tweezer beams. After certain time of flight Δt , the tweezer beams are turned on again to recapture the atoms. If the temperature of the atoms is high, the atoms are spread out and the recapture probability is rapidly reduced. By measuring the recapture probability as a function of Δt , we can estimate the temperature of the atoms by comparing the experimental data with the Monte Carlo simulation [174].

The procedure of the Monte Carlo simulation is as follows: (1) Generate the initial position (x_i, y_i, z_i) and velocity (v_{xi}, v_{yi}, v_{zi}) of the atoms. The initial position is normally distributed with a standard deviation of $\sigma_{x,y,z} = \sqrt{k_B T / (m \omega_{x,y,z}^2)}$ as

$$n(x, y, z) = \frac{1}{(2\pi)^{3/2} \sigma_x \sigma_y \sigma_z} \exp \left[- \left(\frac{x^2}{2\sigma_x^2} + \frac{y^2}{2\sigma_y^2} + \frac{z^2}{2\sigma_z^2} \right) \right], \quad (4.17)$$

and the initial velocity obeys the Maxwell-Boltzmann distribution given as

$$f(v) = 4\pi v^2 \left(\frac{m}{2\pi k_B T} \right)^{3/2} \exp \left(- \frac{mv^2}{2k_B T} \right). \quad (4.18)$$

Here, m is the mass of the atom, T is the atom temperature, k_B is the Boltzmann constant, and $\omega_{x,y,z}$ are the trap frequencies along each direction. Once the velocity is determined, the direction of the velocity is chosen from the uniform distribution in the solid angle. (2) Calculate the atom position $\mathbf{r}$ and velocity $\mathbf{v}$ after time of flight Δt as $\mathbf{r} = (x_i + v_{xi}\Delta t, y_i + v_{yi}\Delta t, z_i + v_{zi}\Delta t - g\Delta t^2/2)$ and $\mathbf{v} = (v_{xi}, v_{yi}, v_{zi} - gt)$, where g is the gravitational acceleration. (3) Check if the kinetic energy of the atom $mv^2/2$ is larger than the effective trapping potential at $\mathbf{r}$, resulting in the atom loss. Note that the trapping potential includes the gravitational potential. (4) Repeat the above steps for N samples and calculate the recapture probability. (5) Fit the experimental data with the Monte

Calro simulation by least square method. The fit parameters are the temperature of the atoms and the survival probability at $\Delta t = 0$.

Figure 4.16 shows the release and recapture experiment of ^{174}Yb . The temperature of the atoms is estimated to be $35(3) \mu\text{K}$, which is higher than the Doppler cooling limit of $4.4 \mu\text{K}$, and can be reduced by optimizing the intensity balance of the cooling beams. The mismatch of the experimental data and the Monte Carlo simulation at longer release time might be attributed to the irregularity of the tweezer beam shape due to the abberations, which is not considered in the simulation.

Chapter 5

Coherent manipulations of nuclear spin and optical clock qubits in ytterbium atoms

This chapter outlines the coherent manipulation of qubits in ytterbium atoms. First, we provide an overview of the fundamental properties of nuclear spin qubits in ^{171}Yb and present experimental results, including initialization, readout, and Raman sideband cooling. Subsequently, we focus on optical clock qubits in ^{174}Yb , highlighting the observation of coherent Rabi oscillations and the Ramsey interference experiment on the optical qubits.

5.1 Nuclear spin qubits in ^{171}Yb

5.1.1 Basic properties

The nuclear spin states in the $^1\text{S}_0$ and $^3\text{P}_0$ states of ^{171}Yb are ideal candidates for qubits [75, 81, 113, 115, 116, 119]. This suitability arises from the fact that the total angular momentum of the valence electrons in these states is zero, eliminating hyperfine coupling between the nuclear spin and the electron. Furthermore, the nuclear spin states exhibit remarkable insensitivity to magnetic fields, with a Zeeman splitting of only 751 Hz/G. As a result, nuclear spin qubits demonstrate exceptionally long coherence times [113, 116, 119], typically three orders of magnitude longer than those of hyperfine clock qubits in alkali atoms [88]. In this study, we specifically encode the qubit states using the ground-state nuclear spin states as $|0\rangle = |^1\text{S}_0, m_F = 1/2\rangle$ and $|1\rangle = |^1\text{S}_0, m_F = -1/2\rangle$.

A particularly compelling feature of ^{171}Yb is its nuclear spin of 1/2, which stands in contrast to the nuclear spin of 9/2 in strontium-87 atoms. This minimal nuclear spin structure enables simplified quantum state control and significantly facilitates fast and high-fidelity manipulations of the nuclear spin states via Raman transitions [113], as will be discussed in later sections. These attributes position ^{171}Yb as a promising platform for scalable quantum information processing.

5.1.2 State initialization and readout

The state initialization is achieved through optical pumping (OP) via the $|^1\text{S}_0, m_F = -1/2\rangle \rightarrow |^3\text{P}_1, F = 3/2, m_F = 1/2\rangle$ transition (figure 5.1). This process is implemented by irradiating a 556 nm laser beam in the horizontal plane under a 48 G magnetic field applied along the z -axis. The OP beam has a radius of 1 mm, an intensity of $3I_{\text{sat}}$, and is linearly polarized within the horizontal plane.

The state readout is performed by selectively blasting out the $|0\rangle$ state using the $|^1\text{S}_0, m_F = 1/2\rangle$

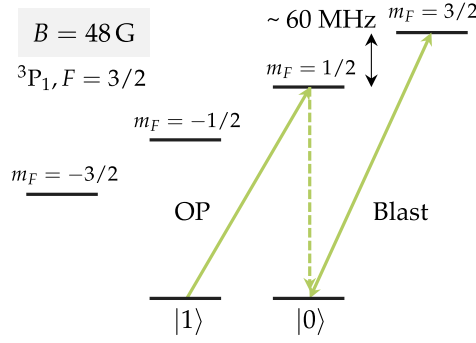


Figure 5.1 Relevant energy levels for OP and blast.

$\leftrightarrow |^3\text{P}_1, F = 3/2, m_F = 3/2\rangle$ transition (figure 5.1), followed by fluorescence imaging as described in section 4.2.3. The blast beam is identical to the OP beam, except that its intensity is increased to $9I_{\text{sat}}$.

The population of the qubit states during the blast obeys following rate equations:

$$\begin{cases} \frac{dN_0}{dt} = -LN_0 + PN_1, \\ \frac{dN_1}{dt} = -PN_1, \end{cases} \quad (5.1)$$

where N_0 and N_1 is the population of $|0\rangle$ and $|1\rangle$ states, respectively, L is the loss rate of the $|0\rangle$ state, and P is the unintended pumping rate from the $|1\rangle$ state to the $|0\rangle$ state induced by the blast beam. The solution of these equations is given by

$$\begin{cases} N_0(t) = \frac{PN_1(0)}{P-L}(e^{-Lt} - e^{-Pt}) + N_0(0)e^{-Lt}, \\ N_1(t) = N_1(0)e^{-Pt}. \end{cases} \quad (5.2)$$

By measuring the populations of the qubit states without applying OP, we can evaluate the performance of the blasting operation. Figure 5.2 (a) shows the population of the $|0\rangle$ state as a function of blast time. Fitting the data using equation (5.2) yields a loss rate of $L = 3.5(3)$ kHz, with the unintended pumping rate suppressed to $P < 5$ Hz. Under these conditions, blasting for 5 ms removes more than 99.9 % of the $|0\rangle$ state while preserving 98(3) % of the $|1\rangle$ state. Further improvement in the blasting performance can be achieved by increasing the strength of the magnetic field during the blasting process. A stronger magnetic field enhances the Zeeman splitting of the $^3\text{P}_1$ state, effectively suppressing unintended pumping transitions and increasing the selectivity of the operation.

The performance of the OP process is evaluated by measuring the population of the $|0\rangle$ state as a function of OP time. Figure 5.2 (b) illustrates the population of the $|0\rangle$ state after the blasting operation. Assuming that 99.9 % of the atoms in the $|0\rangle$ state are removed after the blasting process,

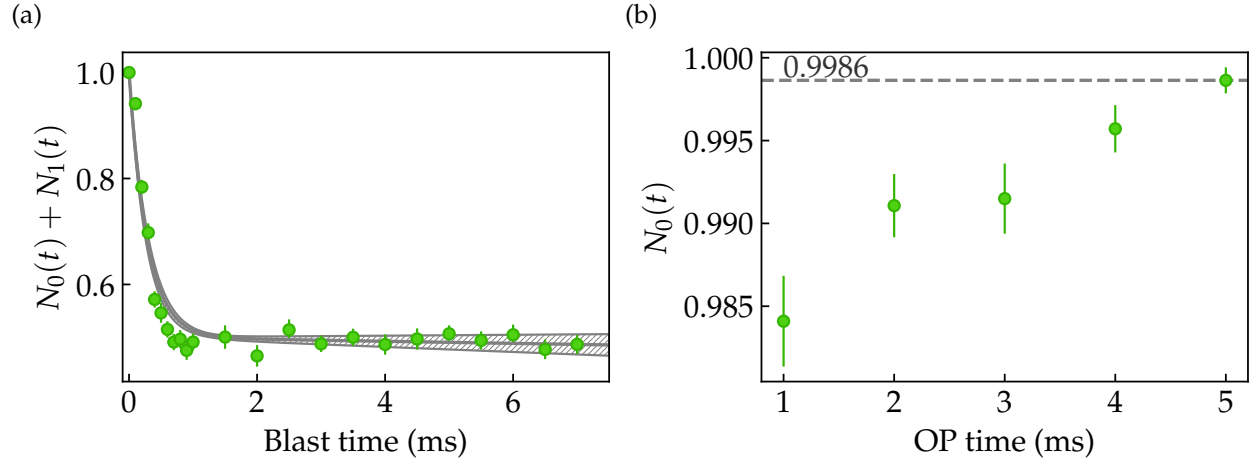


Figure 5.2 Evaluation of the blast and OP performance. (a) Population dynamics as a function of blast time. The solid line represents the fit to the data using equation (5.2). The shaded area represents the 1σ confidence intervals of the fit. From the fitting, the blast rate and unintended pumping rate is estimated to be $L = 3.5(3)$ kHz and $P < 5$ Hz, respectively. Note that $N_0(0) = N_1(0) = 1/2$ is assumed in this calculation. (b) Population of the $|0\rangle$ state as a function of OP time. After applying the OP pulse of 5 ms, 99.86(8) % of the atoms in the $|0\rangle$ state. The error bars in this figure represent the standard error of the mean.

the data indicate that 99.86(8) % of the atoms are pumped into the $|0\rangle$ state after an OP time of 5 ms.

5.1.3 Raman transitions between the qubit states

The Raman transitions are essential for the coherent manipulation of nuclear spin qubits and for Raman sideband cooling. Before presenting the experimental results, we provide an overview of the Raman transitions between nuclear spin states.

Formulation of the Raman transition

The strength of the transition, characterized by the Rabi frequency between the hyperfine states $|F_g, m_g\rangle$ and $|F_e, m_e\rangle$, is proportional to the matrix element $\langle F_g, m_g | \hat{d}_q | F_e, m_e \rangle$. Here, F_g and F_e represent the total angular momentum of the ground and excited states, respectively, while m_g and m_e are their projections onto the quantization axis. The operator $\hat{d}_q$ is the q -component of the electric dipole operator in the spherical basis. According to the Wigner-Eckart theorem, the matrix element can be expressed as

$$\langle F_g, m_g | \hat{d}_q | F_e, m_e \rangle = \frac{\langle F_g, m_g | F_e, m_e; 1, q \rangle}{\sqrt{2F_g + 1}} \langle F_g || \mathbf{d} || F_e \rangle, \quad (5.3)$$

where $\mathbf{d}$ is the electric dipole operator. The reduced matrix element is further decomposed as

$$\langle F_g || \mathbf{d} || F_e \rangle = (-1)^{F_e + J_g + 1 + I} \sqrt{(2F_g + 1)(2F_e + 1)} \begin{Bmatrix} J_g & J_e & 1 \\ F_e & F_g & I \end{Bmatrix} \langle J_g || \mathbf{d} || J_e \rangle. \quad (5.4)$$

Here, J_g and J_e denote the total electronic angular momentum of the ground and excited states, respectively, and I is the nuclear spin. The atom-light interaction Hamiltonian for the transition between the hyperfine states can be written as

$$\hat{\mathcal{H}}_{\text{int}} = \frac{\hbar}{2} \sum_q \left(\Omega_q \hat{\Sigma}_q + \Omega_q^* \hat{\Sigma}_q^\dagger \right), \quad (5.5)$$

with the Rabi frequency Ω_q given by

$$\Omega_q = - \frac{\langle J_g || \mathbf{d} || J_e \rangle E_q}{\hbar}, \quad (5.6)$$

where E_q is the electric field amplitude of the light, and

$$\hat{\Sigma}_q = \sum_{F_e} \sum_{m_g, m_e} S_{m_g m_e}^{(q)}(F_e) |F_g, m_g\rangle \langle F_e, m_e|. \quad (5.7)$$

Here, we define the coefficients $S_{m_g m_e}^{(q)}(F_e)$ as

$$S_{m_g m_e}^{(q)}(F_e) := (-1)^{F_e + J_g + 1 + I} \sqrt{2F_e + 1} \langle F_g, m_g | F_e, m_e; 1, q \rangle \begin{Bmatrix} J_g & J_e & 1 \\ F_e & F_g & I \end{Bmatrix}. \quad (5.8)$$

Let us consider a three-level system consisting of nearly-degenerated ground states of $|g\rangle = |F_g, m_g\rangle$ and $|g'\rangle = |F_g, m'_g\rangle$, and an excited state of $|e\rangle = |F_e, m_e\rangle$. The Raman transition between the $|g\rangle$ and $|g'\rangle$ states is driven by two laser beams with a frequency difference of δ and a detuning of Δ_{F_e} from the $|g\rangle \rightarrow |e\rangle$ and $|g'\rangle \rightarrow |e\rangle$ transitions. When the frequency difference δ matches the energy difference between the $|g\rangle$ and $|g'\rangle$ states, and the angular momentum change of the photons $q' - q$ satisfies the conservation law of the angular momentum $q' - q = m_g - m'_g$, the Raman transition occurs. Under these conditions, the effective Rabi frequency of the Raman transition is given by:

$$\Omega_{m_g m'_g}^{qq'} = \Omega_q \Omega_{q'}^* \sum_{F_e} \sum_{m_e} \frac{S_{m_g m_e}^{(q)}(F_e) S_{m'_g m_e}^{(q')}(F_e)}{2\Delta_{F_e}} = \frac{3\pi c^2}{\hbar \omega_0^3} (2J_e + 1) \Gamma \sum_{F_e} \sum_{m_e} \frac{S_{m_g m_e}^{(q)}(F_e) S_{m'_g m_e}^{(q')}(F_e)}{\Delta_{F_e}} \sqrt{I_q I_{q'}}. \quad (5.9)$$

Here, ω_0 is the resonance frequency of the $|g, g'\rangle \rightarrow |e\rangle$ transition, $I_q = \epsilon_0 c E_q^2 / 2$ is the intensities of the laser beams, and Γ is the natural linewidth of the excited state, which is given by

$$\Gamma = \frac{\omega_0^3}{3\pi \epsilon_0 \hbar c^3} \frac{1}{2J_e + 1} |\langle J_g || \mathbf{d} || J_e \rangle|^2. \quad (5.10)$$

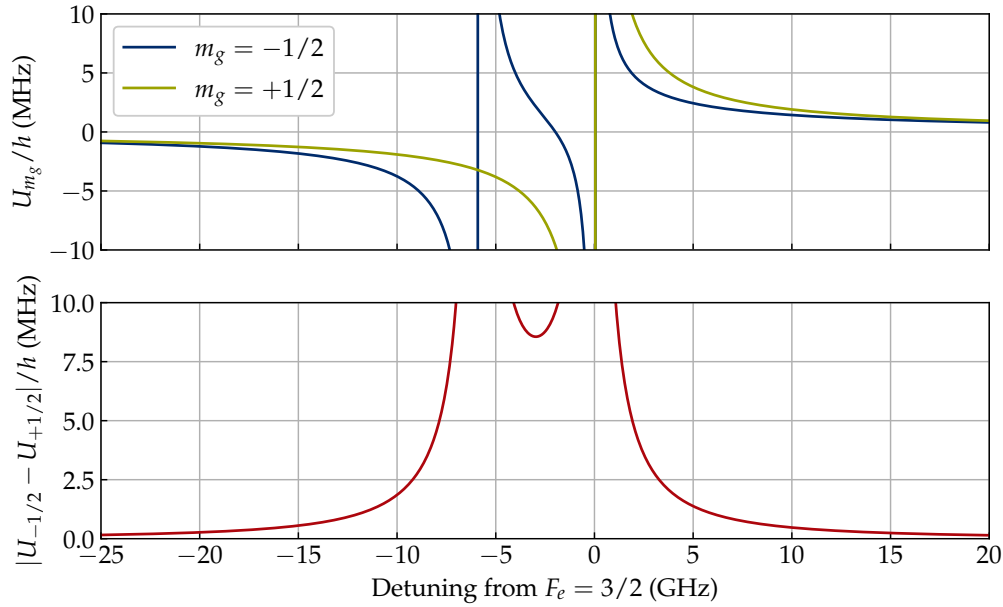


Figure 5.3 Light shift of the nuclear spin states. (Top) Light shift of each nuclear spin state as a function of the detuning from the $^3\text{P}_1, F_e = 3/2$ manifolds. (Bottom) Nuclear spin splitting induced by the light shift. In this calculation, we assume that the beam has a power of 100 mW, a beam radius of 100 μm , and circular polarization ($q = 1$).

The Raman beams also induce the light shifts and scattering of the ground states. Similar to equations (2.59) and (2.60), the light shift and scattering rate of the $|g\rangle$ state are given by

$$U_{m_g} = \frac{3\pi c^2}{2\omega_0^3} (2J_e + 1) \Gamma \sum_q \sum_{F_e} \sum_{m_e} \frac{|S_{m_g m_e}^{(q)}(F_e)|^2}{\Delta_{F_e, m_e}} I_q, \quad (5.11)$$

$$\gamma_{\text{sc}} = \frac{3\pi c^2}{2\hbar\omega_0^3} (2J_e + 1)^2 \Gamma^2 \sum_q \sum_{F_e} \sum_{m_e} \frac{|S_{m_g m_e}^{(q)}(F_e)|^2}{\Delta_{F_e, m_e}^2} I_q. \quad (5.12)$$

Figure 5.3 shows the light shift of the nuclear spin states as a function of the detuning of the Raman beams.

Here, it is worth noting that the above formula for the scattering rate is, strictly speaking, NOT entirely accurate. This is because the formula represents an independent summation of the scattering rates for each transition path and does not account for the detailed scattering processes, such as Raman and Rayleigh scattering. Despite this limitation, the formula provides a practical and convenient approximation for estimating the scattering rate in experimental settings. A more detailed and rigorous discussion on the scattering process, which includes all relevant contributions, will be discussed later.

Conventional Raman transitions

We first demonstrate the coherent manipulation of nuclear spin states using conventional Raman transitions driven by two laser beams. For this purpose, the $^3\text{P}_1$ state is chosen as the intermediate state for the Raman transitions. The beam configuration is illustrated in figure 5.4 (a) and figure 4.6.

Three beams, referred to as RSC1, RSC2, and RSC3, are employed. The RSC1 beam is linearly polarized in the horizontal plane, whereas the RSC2 and RSC3 beams are linearly polarized in the vertical plane. Each beam has an elliptical intensity profile with a beam radius of approximately $35\text{ }\mu\text{m}$ in the vertical direction and $350\text{ }\mu\text{m}$ in the horizontal direction at the atom position, ensuring uniform intensity across the atom array. The tweezer potential is approximately 1.6 mK deep, and trap frequencies are $2\pi \times (153, 173)\text{ kHz}$ in the radial directions, respectively¹.

Under a magnetic field of 48 G applied along the z -axis, the pairs of RSC1 and RSC2, as well as RSC1 and RSC3, are capable of driving Raman transitions between the $|0\rangle$ and $|1\rangle$ states when the frequency difference between the beams matches the Zeeman splitting of the qubit states of 33.8 kHz . Figure 5.4 (b) shows a typical Rabi oscillation between the $|0\rangle$ and $|1\rangle$ states driven by the RSC1 and RSC2 beams with a beam power of 2.5 mW with a detuning of -3 GHz from the $^3\text{P}_1, F = 3/2$ manifolds. The Rabi frequency is estimated to be $2\pi \times 14.5(1)\text{ kHz}$. We also perform

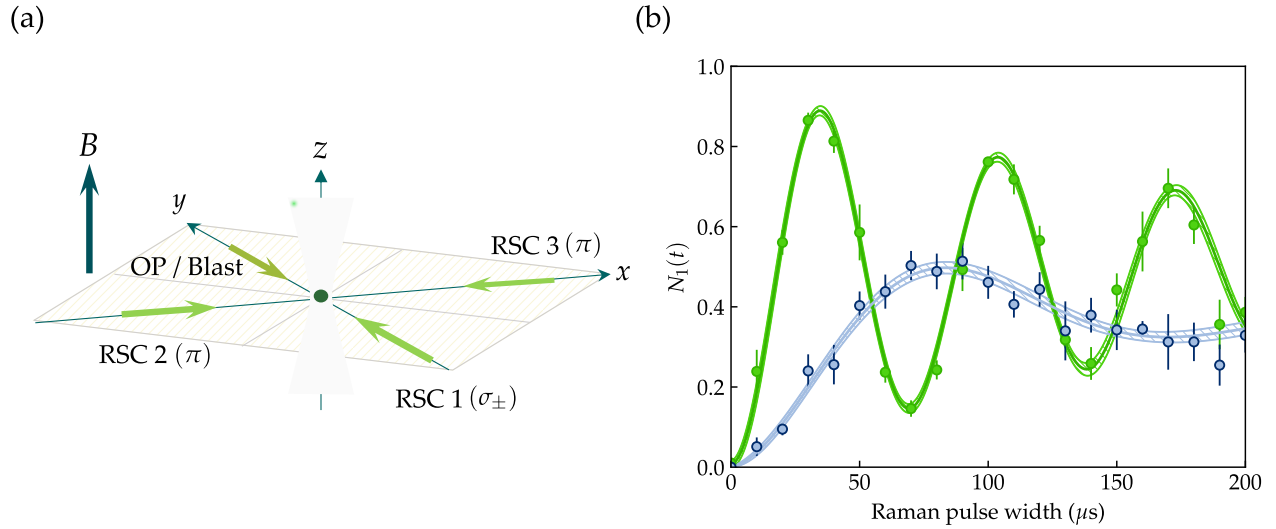


Figure 5.4 Raman transitions for nuclear spin manipulations. (a) Schematic of the beam configuration for the Raman transitions. (b) Rabi oscillations between the $|0\rangle$ and $|1\rangle$ states, driven by the RSC1 and RSC3 beams. The green circles correspond to the Rabi oscillations on the carrier transition, while the blue circles represent the oscillations on the blue sideband transition. The solid lines are the fits to the data using a damped sinusoidal function. The error bars represent the standard error of the mean.

¹The tweezer beam has an elliptical shape with beam radii of 500 nm and 560 nm along the two principal axes, primarily due to optical aberrations.

coherent manipulation on the blue-sideband Raman transition as shown in blue curve in figure 5.4 (b). The Rabi frequency of the blue-sideband transition is estimated to be $2\pi \times 5.8(3)$ kHz. The low contrast and fast decay of the carrier Rabi oscillation can be attributed to the high atom temperature.

Raman sideband cooling

As seen in the previous subsection, the atom temperature in the tweezer influences the coherent dynamics of nuclear spin states. In addition to affecting the Raman transitions, the clock transition—critical for achieving high-fidelity quantum operations—is also impacted by the atom temperature.

To mitigate these effects, we implement Raman sideband cooling (RSC) to cool the atoms to the motional ground state. This section begins with a theoretical discussion of motional dephasing, particularly its impact on the clock transition, to illustrate the necessity of RSC. We then describe the method and present the experimental results of RSC.

The Rabi frequency of a transition between two internal levels of an atom is given by [175]

$$\Omega_n = \Omega_{\text{el}} \prod_{j=x,y,z} |\langle n_j | \exp(ik_j j) | n_j \rangle|, \quad (5.13)$$

where Ω_{el} is the Rabi frequency of the electronic transition, n_j is the vibrational quantum number along the j -axis, and k_j is the wave number of the laser beam along the j -axis. When the laser propagates along a specific axis, such as the x -axis, the Rabi frequency simplifies to

$$\Omega_n = \Omega_{\text{el}} \exp(-\eta^2/2) L_n(\eta^2). \quad (5.14)$$

Here, $n = n_x$ is the vibrational quantum number along x -axis, $\eta = k_x \sqrt{\hbar/(2m\omega_x)}$ is the Lamb-Dicke parameter, ω_x is the trap frequency along x -axis, $L_n(\cdot)$ is the n -th order Laguerre polynomial, and we use equation (2.95) in this calculation. Consequently, the population dynamics of the excited state becomes

$$\rho_{ee}(t) = \frac{1}{1 + \bar{n}} \sum_{n=0}^{\infty} \left(\frac{\bar{n}}{1 + \bar{n}} \right)^n \sin^2 \left(\frac{1}{2} \Omega_n t \right), \quad (5.15)$$

where $\bar{n}$ is the mean motional level of the atom. The above equation indicates that the Rabi oscillation can quickly decay due to the distribution in the motional states.

This phenomenon can be simulated by solving the Schrödinger equation with the Hamiltonian

$$\mathcal{H} = -\hbar\delta |e\rangle\langle e| \otimes I_{\text{mo}} + \frac{\hbar\Omega_{\text{el}}}{2} \left(|g\rangle\langle e| e^{-i\eta(\hat{a} + \hat{a}^\dagger)} + |e\rangle\langle g| e^{i\eta(\hat{a} + \hat{a}^\dagger)} \right) + I_{\text{el}} \otimes \hbar\omega \sum_{n=0}^{N'} \left(n + \frac{1}{2} \right) |n\rangle\langle n|, \quad (5.16)$$

where δ is the detuning of the laser beam, ω is the trap frequency, N' is the truncation number of the motional states, and I_{el} and I_{mo} are the identity operators for the electronic and motional states, respectively. The operator $\hat{a}$ and $\hat{a}^\dagger$ are the annihilation and creation operator for the motional states, respectively. The simulation results are shown in figure 5.5. This simulation support that it is necessary to cool the atoms to motional ground state to achieve high-fidelity clock transitions.

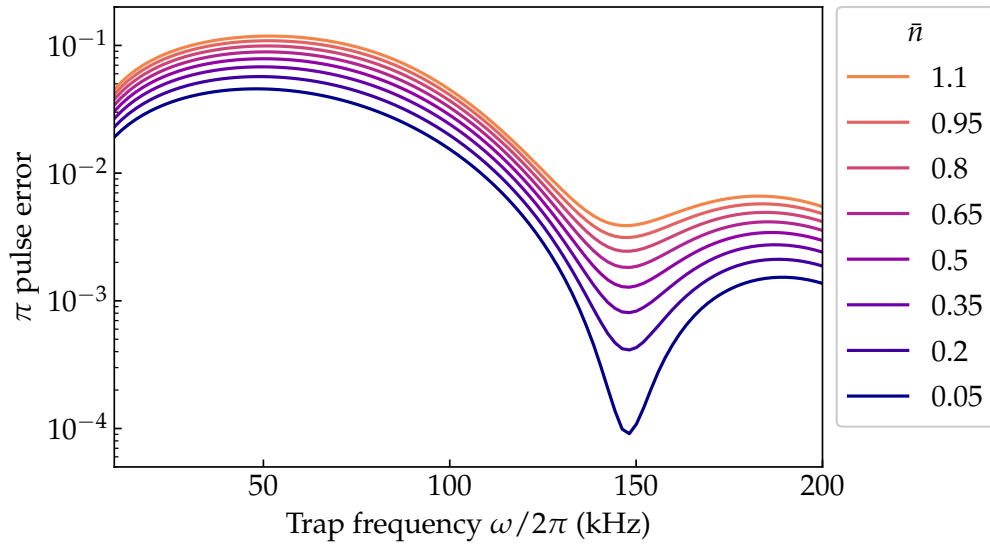


Figure 5.5 Simulation of clock excitation error. We set the Rabi frequency Ω_{el} to $2\pi \times 50$ kHz in this simulation. The results show that a lower mean motional level significantly enhances the fidelity of clock excitation. The simulation is performed using the QuTiP package [176, 177].

We now focus on the experimental implementation of Raman Sideband Cooling (RSC). The RSC process involves driving the red-sideband $|0\rangle \rightarrow |1\rangle$ transition, followed by optical pumping (OP) to return the atoms to the $|0\rangle$ state [16, 17, 113] (figure 5.6 (a)). The beam configuration for this process is shown in figure 5.4 (a). The RSC1 and RSC2 beams cool the atoms along the $x - y$ direction (axis 1), while the RSC1 and RSC3 beams cool along the $x + y$ direction (axis 2).

The pulse sequence used for RSC is depicted in figure 5.6 (b). It consists of 16 RSC beam pulses, each with a duration of 6 ms, separated by 2 ms intervals. During this sequence, the OP beam is simultaneously applied, detuned by -2 MHz from the optimal frequency for OP. After completing the cooling process along axis 1 using the RSC1 and RSC2 beam pair, the RSC sequence is repeated to cool the atoms along axis 2, utilizing the RSC1 and RSC3 beam pair.

To improve the efficiency of RSC, we utilize the HS1 pulse [178, 179], which is defined as

$$\begin{cases} \Omega(t) = \Omega_0 \operatorname{sech} \left[\beta \left(\frac{2t}{T} - 1 \right) \right], \\ \Delta(t) = \frac{\Delta_0}{2} \tanh \left[\beta \left(\frac{2t}{T} - 1 \right) \right], \end{cases} \quad (5.17)$$

where T is the pulse width, Ω_0 is the peak Rabi frequency, Δ_0 is the frequency sweep range, and β is the pulse shape parameter. This pulse is robust to the inhomogeneity of the trap frequency and enables efficient sideband cooling [180]. In this experiment, we set the pulse shape parameter $\beta = 5$, and the frequency sweep range $\Delta_0 = 2\pi \times 20$ kHz.

Figure 5.6 (c) and (d) present the results of RSC along axes 1 and 2, respectively. The Raman sideband spectra are taken with a $160 \mu\text{s}$ pulse of each RSC beam pair, corresponding to the π -pulse

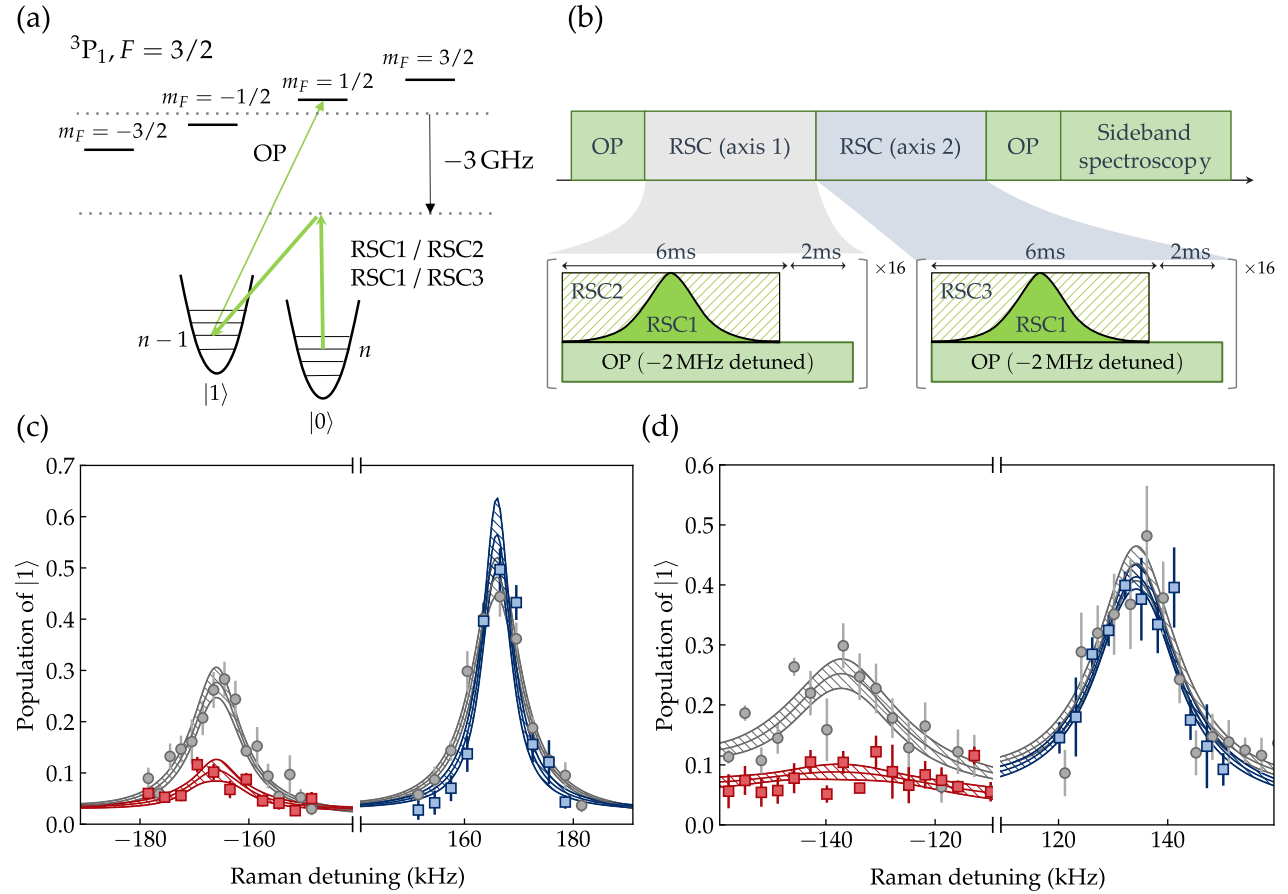


Figure 5.6 Raman sideband cooling. (a) Relevant energy levels for RSC. (b) Pulse sequence for RSC. (c) and (d) are the Raman sideband spectra along axis 1 and axis 2, respectively. The gray circles represent the data taken without RSC, while the blue and red squares corresponds to that with RSC. The solid lines are the fits to the data using a Lorentzian function, and the shaded areas are 1σ confidence intervals of the fit. The mean motional quantum number is reduced from $\bar{n} = 1.6(4)$ to $\bar{n} = 0.17(4)$ along axis 1 and from $\bar{n} = 1.7(6)$ to $\bar{n} = 0.18(4)$ along axis 2. The error bars represent the standard error of the mean.

for the blue sideband. The mean motional level of the atoms can be estimated from the relation as

$$\frac{A_R}{A_B} = \frac{\bar{n}}{1 + \bar{n}} \iff \bar{n} = \frac{A_R}{A_B - A_R}, \quad (5.18)$$

where A_R and A_B are the amplitudes of the red and blue sidebands, respectively. The mean motional quantum number after the RSC is estimated to be $\bar{n} = 0.17(4)$ along axis 1 and $\bar{n} = 0.18(4)$ along axis 2. These results correspond to a motional ground-state probability of $P_0 = (1 + \bar{n})^{-1} = 83\%$. To achieve more efficient cooling, a lower initial atom temperature is required, which can be realized using a magic tweezer array designed for the $^1S_0 \rightarrow ^3P_1$ transition [115, 119].

5.1.4 Fast gate operations on the nuclear spin qubits

The gate operation on the nuclear spin qubits can be significantly accelerated by employing single-beam Raman transitions [113]. In the following, we first provide a theoretical overview of the single-beam Raman transitions. We then present the experimental results of the fast nuclear spin control using single-beam Raman transitions.

The relevant energy levels for the single-beam Raman transitions, which are labeled as $|0\rangle$, $|1\rangle$, $|2\rangle = |^3\text{P}_1, F = 3/2, m_F = -1/2\rangle$, and $|3\rangle = |^3\text{P}_1, F = 3/2, m_F = 1/2\rangle$, are shown in figure 5.7 (a). Note that this labeling implicitly assumes that the detuning of the laser beam is not so far from the $^3\text{P}_1, F = 3/2$ manifolds, and thus the $^3\text{P}_1, F = 1/2$ manifolds are not involved in the Raman transitions. The Hamiltonian for the single-beam Raman transitions is given by

$$\mathcal{H}/\hbar = \begin{pmatrix} \Delta_N & 0 & \Omega_{02}/2 & \Omega_{03}/2 \\ 0 & 0 & \Omega_{12}/2 & \Omega_{13}/2 \\ \Omega_{02}^*/2 & \Omega_{12}^*/2 & \Delta_X & 0 \\ \Omega_{03}^*/2 & \Omega_{13}^*/2 & 0 & \Delta_X + \Delta_e \end{pmatrix}, \quad (5.19)$$

where Δ_N is the splitting between the $|0\rangle$ and $|1\rangle$ states, Δ_e is the splitting between the $|2\rangle$ and $|3\rangle$ states, Δ_X is the detuning of the laser beam from the $|1\rangle \leftrightarrow |2\rangle$ transition, and Ω_{ij} is the Rabi frequency between the $|i\rangle$ and $|j\rangle$ states. If we take a quantization axis along the z -axis and the laser beam direction along the x -axis, the electric field of the laser beam can be written as $\mathbf{E} = E_y \cos(\omega t + \phi_{H/V})\hat{\mathbf{y}} + E_z \cos(\omega t)\hat{\mathbf{z}}$ with the relative phase between the horizontal and vertical polarization $\phi_{H/V}$. The Hamiltonian is then rewritten as

$$\mathcal{H}/\hbar = \begin{pmatrix} \Delta_N & 0 & ie^{i\phi_{H/V}}\Omega_\sigma/2 & \Omega_\pi/2 \\ 0 & 0 & \Omega_\pi/2 & ie^{i\phi_{H/V}}\Omega_\sigma/2 \\ -ie^{-i\phi_{H/V}}\Omega_\sigma/2 & \Omega_\pi^*/2 & \Delta_X & 0 \\ \Omega_\pi^*/2 & -ie^{-i\phi_{H/V}}\Omega_\sigma^*/2 & 0 & \Delta_X + \Delta_e \end{pmatrix}, \quad (5.20)$$

with π -transition strength of Ω_π and σ -transition strength of Ω_σ . By adiabatically eliminating the excited states and assuming $\Delta_X \gg \Delta_e$, we obtain the effective Hamiltonian for the nuclear spin states as

$$\mathcal{H}_{\text{eff}}/\hbar = -\frac{1}{4\Delta_X} \begin{pmatrix} -4\Delta_N\Delta_X + \Omega_\pi^2 + \Omega_\sigma^2 & 2\sin(\phi_{H/V})\Omega_\pi\Omega_\sigma \\ 2\sin(\phi_{H/V})\Omega_\pi\Omega_\sigma & \Omega_\pi^2 + \Omega_\sigma^2 \end{pmatrix}. \quad (5.21)$$

Therefore, the Rabi frequency of the single-beam Raman transition is given by

$$\Omega_R = |\sin(\phi_{H/V})|\Omega_\pi\Omega_\sigma/\Delta_X. \quad (5.22)$$

This quantity is maximized when $\phi_{H/V} = \pm\pi/2$, that is, when the laser beam is circularly polarized.

Here, we provide an intuitive interpretation of the single-beam Raman transitions. Consider a nuclear spin polarized along the z -axis. In this scenario, the quantization axis (hereafter referred to

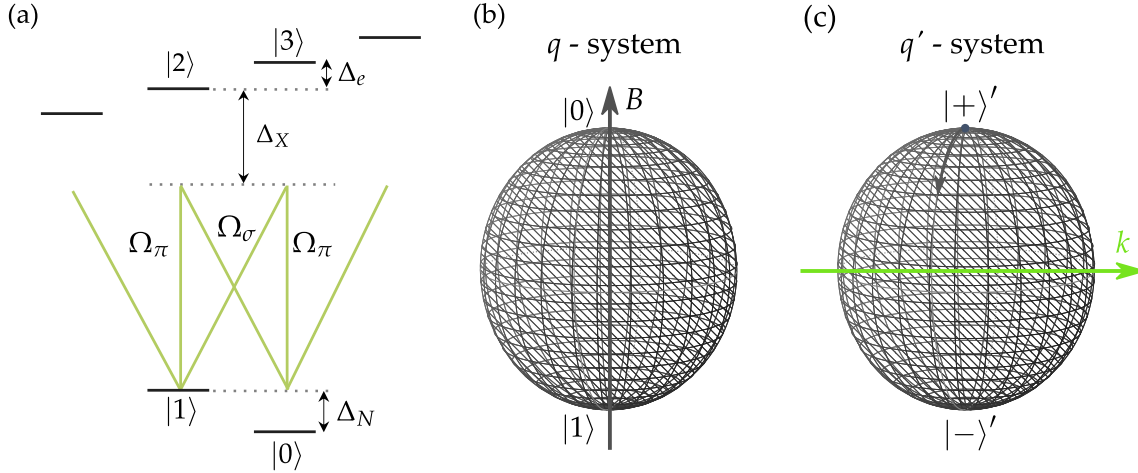


Figure 5.7 Single-beam Raman transition. (a) Relevant energy levels for the single-beam Raman transitions. (b) Bloch sphere representation of the nuclear spin states. The quantization axis is determined by the magnetic field. (c) Bloch sphere representation of the nuclear spin states in the q' -system. The strong gate beam defines the quantization axis. Consequently, the $|0\rangle$ state in the q -system is represented as the $|+\rangle'$ state in the q' -system and rotates around the x -axis with an angular frequency of Ω_R .

as the q -system) is defined by the magnetic field. The Bloch sphere representation of the nuclear spin states is shown in figure 5.7 (b). When a circularly polarized single-beam Raman laser is applied, the nuclear spin states experience state-dependent light shifts as given by equation (5.11), resulting in a nuclear spin splitting larger than the Zeeman splitting.

In this situation, the quantization axis is effectively determined by the direction of the laser beam (referred to as the q' -system). As a result, the $|0\rangle$ state in the q -system is represented as the $|+\rangle'$ state in the q' -system (figure 5.7 (c)). The state then rotates around the laser beam direction (x -axis) with an angular frequency of $\Omega_R = |U_{1/2} - U_{-1/2}|/\hbar$. This rotation can be interpreted as the Raman transition between the $|0\rangle$ and $|1\rangle$ states, corresponding to the implementation of the X gate.

We now present experiment results of the fast nuclear spin control of X and Z gates. The former is performed by employing single beam Raman transition described above, while the latter is by applying a circularly polarized laser beam along the quantization axis. The beam configuration is shown in figure 5.8 (a). The beams are focused onto the atom arrays with a beam radii of $45\text{ }\mu\text{m}$ in the vertical direction and $600\text{ }\mu\text{m}$ in the horizontal direction. The quantization axis is set along the Z gate beam direction (y -axis) by applying a magnetic field of 2.5 G. The gate beams are orthogonal to each other.

Figure 5.8 (b) present the experimental result of the X gate. The beam power is 105 mW and have a detuning of -3 GHz from the $^3\text{P}_{1/2}, F = 3/2$ manifolds. We successfully observe the Rabi oscillation between the $|0\rangle$ and $|1\rangle$ states with a Rabi frequency of $2\pi \times 1.27\text{ MHz}$. The fidelity of

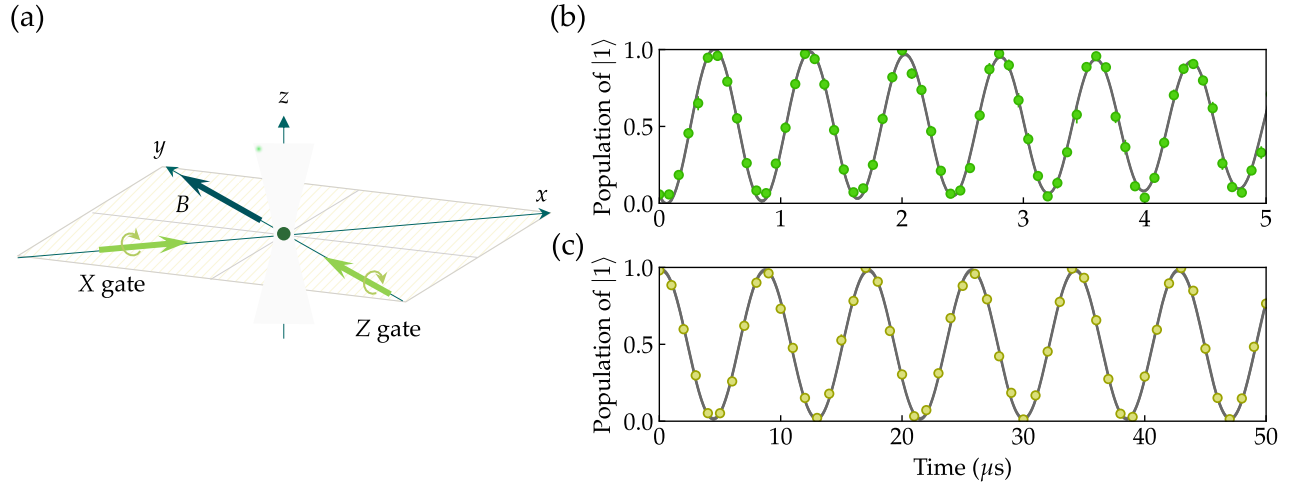


Figure 5.8 Gate operations on the nuclear spin qubits. (a) Schematic of the beam configuration for the X and Z gates. (b) Rabi oscillations between the $|0\rangle$ and $|1\rangle$ states driven by the X gate beam, with a Rabi frequency of $2\pi \times 1.27\text{ MHz}$. The solid line is the fit to the data using a damped sinusoidal function. (c) Phase rotation by the Z gate beam with a oscillation frequency of $2\pi \times 117\text{ kHz}$. The solid line is the fit to the data using a damped sinusoidal function. The error bars represent the standard error of the mean.

π -pulse $X(\pi)$ is found to be approximately 0.995 by comparing the survival probabilities with and without blasting out the atoms in $|0\rangle$ after applying a π -pulse. This fidelity might be limited by the inhomogeneity of the beam intensity over the atom array and the shot-to-shot fluctuation of the pulse area.

Figure 5.8 (c) shows the experimental result of the Z gate. We first apply $X(\pi/2)$ gate to prepare the superposition state of $|0\rangle$ and $|1\rangle$. We then irradiate the Z gate beam, followed by the additional $X(\pi/2)$ pulse to measure the population of the $|0\rangle$ state. The oscillation with a frequency of $2\pi \times 117\text{ kHz}$ does not exhibit any damping behavior within the measured range, indicating that the Z gate is implemented with high fidelity. The fidelity of the Z gate is estimated to be 0.999 by comparing the survival probabilities with and without the Z gate beam irradiation and compensating the infidelity of the $X(\pi/2)$ gates. More regorous evaluation of the gate fidelity will be performed with randomized benchmarking technique [181].

Optimal detuning for high-fidelity gate operations

Since photon scattering during gate operation can lead to infidelity, it is crucial to carefully select the detuning of the single-qubit gate beam to optimize the fidelity. One straightforward way to estimate the gate operation fidelity is to evaluate the ratio between the Rabi frequency and the scattering rate at a given detuning. The Rabi frequency of the single-beam Raman transition corresponds to

the difference in light shifts between the $|0\rangle$ and $|1\rangle$ states, which is derived from equation (5.11) as

$$\Omega_R = |U_{1/2} - U_{-1/2}|/\hbar \propto \frac{1}{\Delta^2} \quad (\text{for } \Delta \gg \Delta_{\text{HF}}), \quad (5.23)$$

where Δ_{HF} represents the hyperfine splitting of the excited state.

On the other hand, the scattering rate is given by equation (5.12) as

$$\gamma_{\text{sc}} \propto \frac{1}{\Delta^2}. \quad (5.24)$$

At first glance, this suggests that the ratio between the Rabi frequency and the scattering rate is independent of detuning. However, this conclusion is incorrect because the scattering rate in equation (5.12) is not entirely accurate, as previously noted. The scattering rate must account for both Raman and Rayleigh scattering. The former is described by the Kramers-Heisenberg formula [182] as

$$\gamma_{ij} = g^2 \Gamma \sum_q \left(\sum_{F', m_{F'}} A_{F', m_{F'}, q}^{ij} \right)^2, \quad (5.25)$$

while the latter is expressed as

$$\gamma_{\text{el}} = g^2 \Gamma \sum_q \left[\sum_{F', m_{F'}} \left(A_{F', m_{F'}, q}^{+1/2, -1/2} - A_{F', m_{F'}, q}^{-1/2, +1/2} \right) \right]^2. \quad (5.26)$$

Here, $g = dE_0/(2\hbar)$ and $d = (3\pi\epsilon_0\hbar c^3\Gamma/\omega_0^3)^{1/2}$, where ω_0 is the transition frequency, and the electric field of the light is $E_0 = \sum_q b_q \hat{\epsilon}_q$. The scattering amplitude for transitions between nuclear spin states $|i\rangle$ and $|j\rangle$ is given by

$$A_{F', m_{F'}, q}^{ij} = \frac{b_q \langle j | \hat{d}_q \cdot \hat{\epsilon}_{q+(i-j)}^* | F', m_{F'} \rangle \langle F', m_{F'} | \hat{d}_q \cdot \hat{\epsilon}_q | i \rangle}{\Delta_{F', m_{F'}} d^2}, \quad (5.27)$$

where $\Delta_{F', m_{F'}}$ is the detuning from the intermediate state $|F', m_{F'}\rangle$.

For the equal superposition state $|+\rangle = (|0\rangle + |1\rangle)/\sqrt{2}$, the total scattering rate becomes $\gamma_{\text{sc}} = (\gamma_{+1/2, -1/2} + \gamma_{-1/2, +1/2} + \gamma_{\text{el}})/2$. The ratio of the scattering rate to the Rabi frequency is then expressed as [113]

$$\frac{\gamma_{\text{sc}}}{\Omega_R} \simeq \frac{\Gamma}{\sqrt{6}} \left| \frac{\Delta_{\text{HF}}}{\Delta(\Delta - \Delta_{\text{HF}})} \right|, \quad (5.28)$$

where Δ is the detuning from the $F_e = 3/2$ manifolds of the $^3\text{P}_1$ state. Figure 5.9 shows the ratio of the scattering rate to the Rabi frequency as a function of the detuning. When the detuning lies between the hyperfine splitting, the ratio is locally minimized. Considering the Rabi frequency at a given detuning (figure 5.3), this detuning range is preferable. Further suppression of scattering can be achieved by increasing the detuning, albeit at the cost of a reduced Rabi frequency. In Yb system, a single qubit gate fidelity of 0.999 68(3) is achieved with a detuning of -5.8GHz from the $^3\text{P}_1, F' = 1/2$ manifolds.

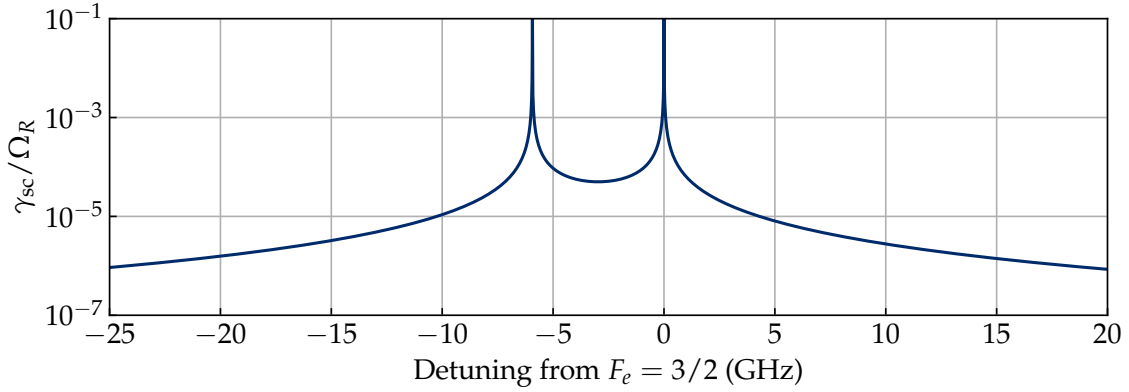


Figure 5.9 Detuning dependency of the ratio of the scattering rate to the Rabi frequency.

5.1.5 Coherence time evaluation of the nuclear spin qubits

The coherence time of the nuclear spin qubits is evaluated by using the Ramsey sequence. The sequence consists of two $X(\pi/2)$ pulses separated by a variable free evolution time T . The Ramsey signal is obtained by measuring the population of the $|1\rangle$ state after the second $X(\pi/2)$ pulse. The coherence time is then extracted by fitting the Ramsey signal to a damped sinusoidal function. Figure 5.10 shows the experimental results of the Ramsey sequence. In this measurement, we set the magnetic field to 2.5 G along y -axis and the trap potential to 35 μK . The coherence time is estimated to be $T_2^* = 3.8(4)$ s. The coherence time is limited by the magnetic field noise. The coherence time can be improved by using dynamical decoupling technique [183].

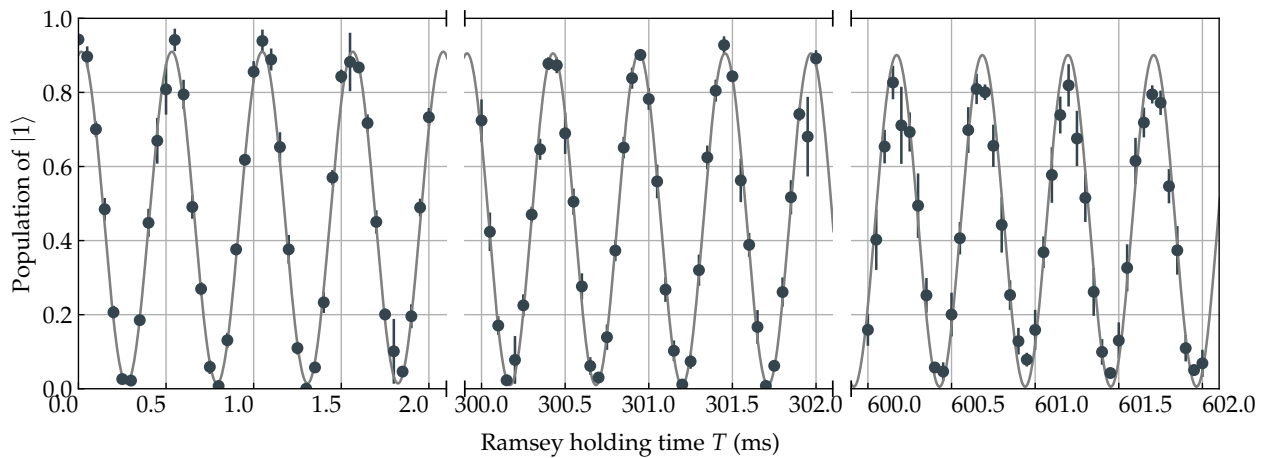


Figure 5.10 Coherence time evaluation of the nuclear spin qubit. The Ramsey signal is obtained by measuring the population of the $|1\rangle$ state after the second $X(\pi/2)$ pulse. The solid line is the fit to the data using a damped sinusoidal function. The coherence time is found to be $T_2^* = 3.8(4)$ s. The error bars represent the standard error of the mean.

5.1.6 Coherent $^3\text{P}_2$ excitation of ^{171}Yb

As a first step towards Rydberg excitation, we demonstrate the coherent excitation of the $^3\text{P}_2$ state in ^{171}Yb . We begin by cooling the motional states of the atoms using the RSC technique described in the previous section. Subsequently, we adiabatically reduce the magnetic field from 48 G to 1 G and lower the trapping depth to 35 μK to minimize the differential light shift between the ground $^1\text{S}_0$ and the metastable $^3\text{P}_2$ states. The excitation is then performed using a 507 nm beam, with beam radii of 35 μm in the vertical direction and 500 μm in the horizontal direction, propagating along the x -axis. The polarization of the excitation beam is set to horizontal, enabling the HFM-E1 $|0\rangle \leftrightarrow |^3\text{P}_2, F = 3/2, m_F = 3/2\rangle$ transition under the z -axis magnetic field.

Figure 5.11 (a) shows the spectroscopy result for the $|^3\text{P}_2, F = 3/2, m_F = 3/2\rangle$ state. The beam power is 1.5 mW, and the spectroscopy pulse duration is set to 400 μs . The observed linewidth is 2.6(4) kHz, primarily limited by magnetic field fluctuations during spectroscopy. Increasing the beam power to 35 mW, we perform coherent excitation. Figure 5.11 (b) displays the Rabi oscillation between the $|0\rangle$ and $|^3\text{P}_2, F = 3/2, m_F = 3/2\rangle$ states, with a measured Rabi frequency of $2\pi \times 9.1(2)$ kHz.

The low excitation efficiency to the $^3\text{P}_2$ state is attributed to the differential light shift between the $^1\text{S}_0$ and $^3\text{P}_2$ states. This efficiency can be enhanced by employing an adiabatic rapid passage technique or a magic tweezer array optimized for the $^1\text{S}_0 \leftrightarrow ^3\text{P}_2$ transition.

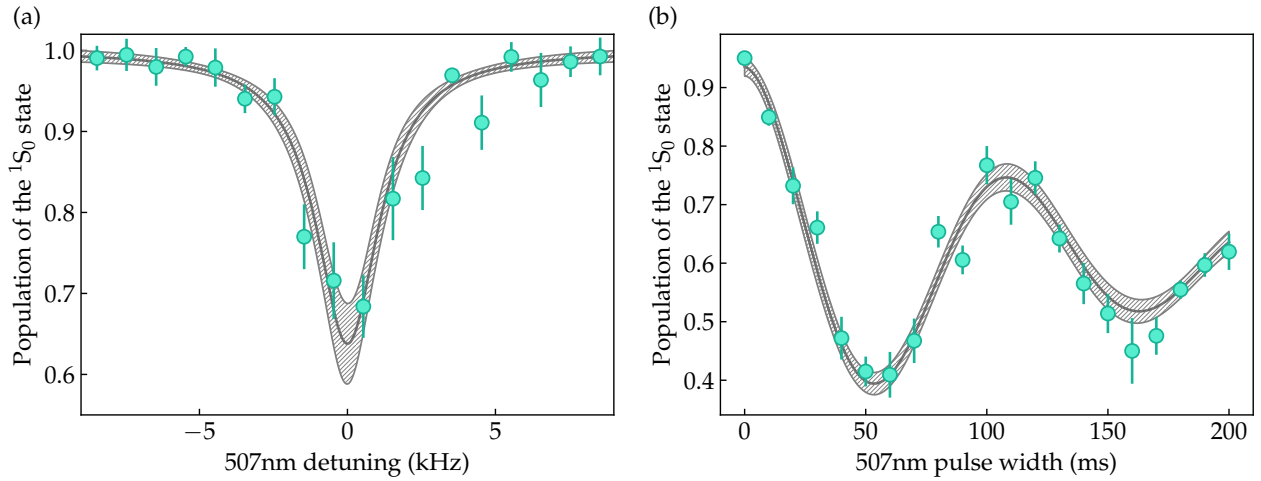


Figure 5.11 Spectroscopy and coherent excitation of the $^3\text{P}_2$ state of ^{171}Yb . (a) Spectrum of the $|^3\text{P}_2, F = 3/2, m_F = 3/2\rangle$ state with a linewidth of 2.6(4) kHz. The solid line is the fit using a Lorentzian function. The shaded area represents 1σ confidence interval of the fit. (b) Rabi oscillation between the $|0\rangle$ and $|^3\text{P}_2, F = 3/2, m_F = 3/2\rangle$ states with a Rabi frequency of $2\pi \times 9.1(2)$ kHz. The solid line is the fit using a damped sine function. The error bars represent the standard error of the mean.

5.2 Optical clock qubits in ^{174}Yb

5.2.1 Basic properties

The optical clock qubits encoded in the ground state and a metastable state of bosonic alkaline-earth(-like) atoms are promising candidates for ancilla qubits due to several advantageous properties. First, the qubit states with $m_J = 0$ are highly insensitive to magnetic field fluctuations, which is crucial for preserving qubit coherence. Indeed, second-order coherence times have been reported in previous studies [51, 52]. Second, state-selective qubit readout without necessity of state-selective atom loss or requiring specialized techniques such as shelving is naturally feasible, as the metastable state serves as a dark state for the probe transition. This unique feature makes these qubits particularly appealing as ancilla qubits.

In this study, we utilize an optical clock qubit encoded as $|0\rangle = |^1S_0\rangle$ and $|1\rangle = |^3P_2, m_J = 0\rangle$. This choice is motivated by the fact that our tweezer wavelength of 532 nm is near the magic wavelength for these qubit states, minimizing the differential light shift. Furthermore, the transition strength of the $^1S_0 \leftrightarrow ^3P_2$ transition is significantly larger than that of the $^1S_0 \leftrightarrow ^3P_1$ transition, enabling faster gate operations. This is because the former is an M2 transition, while the latter relies on magnetic-field-induced state mixing [184]. Additionally, the 3P_2 state is less sensitive to magnetic field fluctuations compared to the 3P_1 state [185], which is advantageous for maintaining qubit coherence.

5.2.2 Narrow-line spectroscopy of the 3P_2 state

We first perform spectroscopy of the 3P_2 state to evaluate the transition linewidth in our setup. The excitation beam at 507 nm is applied from the horizontal direction with vertical polarization. The magnetic field is oriented at an angle of 45° relative to the excitation laser beam, and the tweezer polarization is aligned to the magnetic field. This configuration maximizes the transition strength of the $^1S_0 \leftrightarrow ^3P_2, m_J = 0$ transition (see figure 2.2). The beam power is set to 3 mW, and the spectroscopy pulse duration is 10 ms.

We repeat the spectroscopy at various trap depths ranging from 35.4 μK to 212.4 μK to investigate the ratio of polarizabilities between the qubit states. The results are shown in figure 5.12. The slope of the resonance shift as a function of trap depth is expressed as $A = 1 - \alpha_e/\alpha_g$, where α_e and α_g are the polarizabilities of the 3P_2 and 1S_0 states, respectively. From the linear fit, we obtain $A = -0.142(1) \text{ kHz}/\mu\text{K}$, corresponding to $\alpha_e/\alpha_g = 1.0068(1)$. This result is consistent with the theoretical prediction of $\alpha_e/\alpha_g = 1.016$ (figure 4.3). Note that we use the theoretical ground-state polarizability of $\alpha_g = 37.9 \text{ Hz}/(\text{W}/\text{cm}^2)$ in this calculation. Without any theoretical input, we estimate the differential polarizability as $\Delta\alpha = \alpha_e - \alpha_g = -6.44(5) \times 10^{-2} \text{ Hz}/(\text{W}/\text{cm}^2)$ using the tweezer beam radii and the beam power.

The spectrum of the 3P_2 state in the 35.4 μK trap is shown in figure 5.13. The observed linewidth

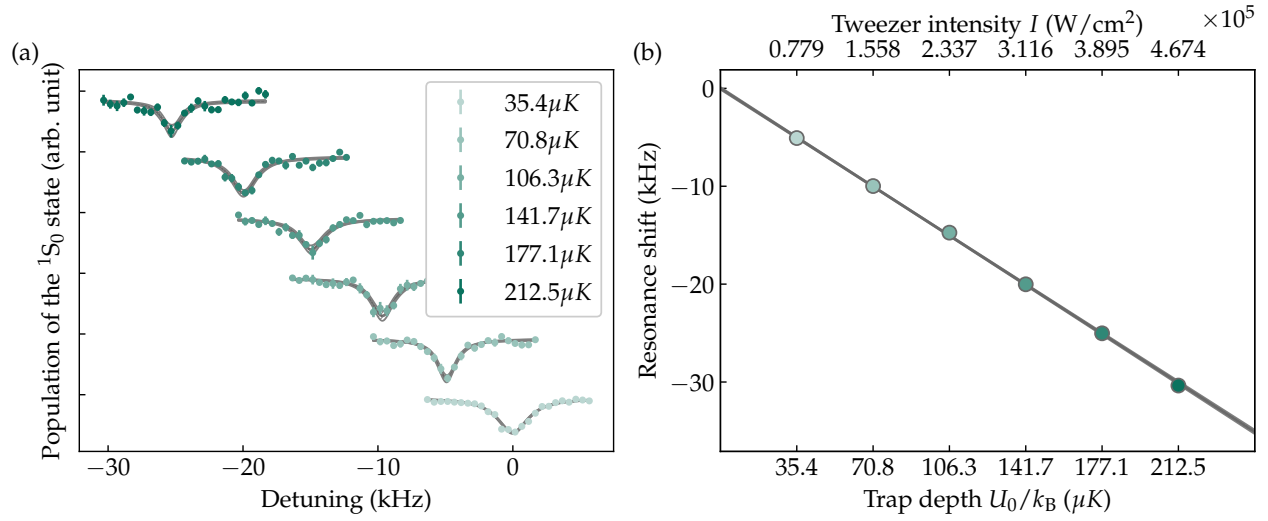


Figure 5.12 Measurement of the differential light shift between the 1S_0 and the 3P_2 states of ^{174}Yb . (a) The spectra taken at various trap depths. The solid lines are the fits to the data using a Lorentzian function. (b) The resonance shift as a function of the trap depth is shown. The solid line is the fit to the data using a linear function. The differential polarizability is estimated to be $\Delta\alpha = -6.44(5) \times 10^{-2} \text{ Hz}/(\text{W}/\text{cm}^2)$. The error bars represent the standard error of the mean.

is 1.3(2) kHz, which is narrower than the linewidth of the 3P_2 state in ^{171}Yb (2.6(4) kHz), owing to its insensitivity to the magnetic field fluctuations and near-magic condition. However, the linewidth is still too broad considering the differential polarizability of $\Delta\alpha = -6.44(5) \times 10^{-2} \text{ Hz}/(\text{W}/\text{cm}^2)$, described above. This indicates that there can be additional sources of the linewidth broadening. We leave the detailed investigation of the broadening to future work.

5.2.3 Coherent control and coherence time measurement of the optical clock qubit

We next perform coherent control of the optical clock qubit. For this, we narrow the excitation beam radii to 45 μm in vertical direction and 105 μm in horizontal direction, and increase the beam power to 210 mW to obtain enough intensity. Figure 5.14 (a) shows the Rabi oscillation between the $|0\rangle$ and $|1\rangle$ states in a 35.4 μK tweezer array. The Rabi frequency is measured to be $2\pi \times 9.5(2)$ kHz. It should be noted that the Rabi oscillation is primarily damped by the thermal distribution of the motional states in the tweezer potential [175]. In this experiment, the mean vibrational quantum number $\bar{n}$ is approximately $\bar{n} = 1$, resulting in variations in the Rabi frequency due to motional coupling. This can be addressed through sideband cooling to the motional ground state via the clock transition [186].

Furthermore, we evaluate the coherence of the optical clock qubits using a Ramsey sequence. Figure 5.14 (b) shows the Ramsey signal obtained by scanning the holding time between the two $\pi/2$ pulses on the $^1S_0 \leftrightarrow ^3P_2$ transition. Fitting the data with a damped sine function yields a

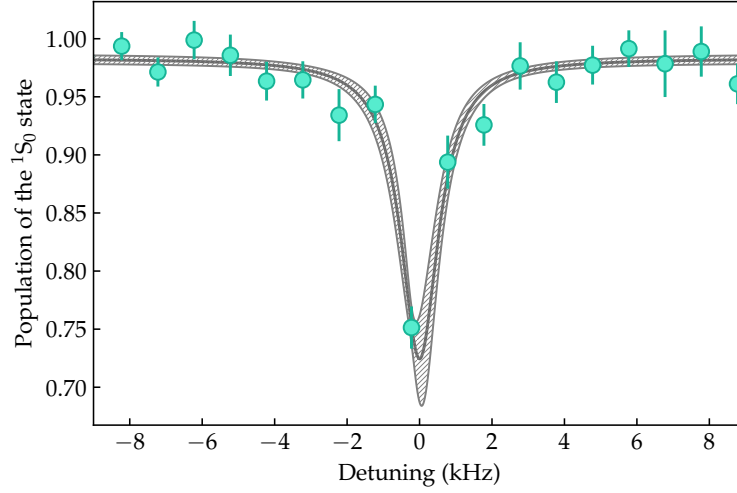


Figure 5.13 Narrow-line spectroscopy of the $^3\text{P}_2$ state of ^{174}Yb . The spectrum is taken at a trap depth of $35.4\ \mu\text{K}$. The solid line is the fit to the data using a Lorentzian function, yielding the linewidth of $1.3(2)\ \text{kHz}$. The shaded area represents 1σ confidence interval of the fit. The error bars represent the standard error of the mean.

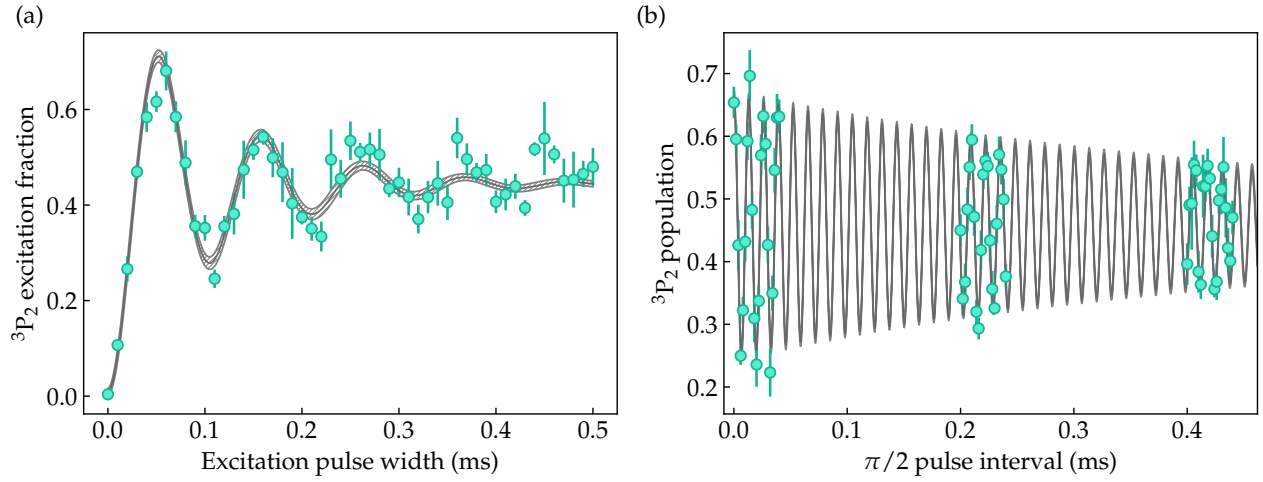


Figure 5.14 Coherent manipulation of the optical clock qubits. (a) Rabi oscillation between the ground and the $^3\text{P}_2$ states. The gray curve is a fitting function with a damping sine function, yielding a Rabi frequency of $2\pi \times 9.5(2)\ \text{kHz}$. The shaded area is 1σ -confidence intervals of the fitting. The error bars represent the standard error of the mean. (b) The coherence of the optical clock qubit measured by the Ramsey sequence. From the fitting with a damping sine function, the coherence time is estimated to be $0.55(7)\ \text{ms}$. The error bars represent the standard error of the mean.

coherence time of 0.55(7) ms. The oscillation frequency of 76.55(4) kHz corresponds to the probe light shift induced by the 507 nm beam. The coherence time might be currently shortened due to the spatial distribution of the differential light shift between the ground and $^3\text{P}_2$ states in each tweezer potential [109], as well as the aforementioned thermal distribution, which induces slight shot-to-shot variations in the Ramsey frequency. Meanwhile, we anticipate achieving a coherence time on the order of several milliseconds for the $^3\text{P}_2$ optical clock qubits in magic-wavelength traps, as recently demonstrated in a strontium system [133].

Chapter 6

Hybrid atom tweezer array of nuclear spin and optical clock qubits

This chapter reports on the experimental demonstration of a hybrid atom tweezer array of nuclear spin and optical clock qubits based on the published paper [187]. The results show that this system enables low crosstalk measurement of the optical clock qubits while maintaining the nuclear spin qubits, which is a key step towards mid-circuit measurements for quantum error correction.

6.1 Challenges with mid-circuit measurements in neutral atom arrays

Ancilla qubits play a central role in quantum error correction (QEC), which is essential for realizing fault-tolerant quantum computing [188]. The efficacy of ancilla-qubit-based QEC protocols has been recently demonstrated in quantum computing architectures with superconducting qubits [84–86] and trapped ion qubits [82,83], supporting their importance and utility in advancing the field.

Concurrently, significant progress has been made in developing the optical tweezer array system, enabling the creation of large-scale qubit arrays [63,65,67–70] and high-fidelity gate operations [54,76–80]. The attractive features of this system, such as dynamical qubit connection aided by moving tweezers [88,89] and error-detecting methods utilizing the rich internal state of atoms [75], open up the possibilities for realizing QEC protocols [90,123,189–191].

One of the recent main focuses in the optical tweezer array systems is efficient control and readout of the ancilla qubits while minimizing qubit loss and crosstalk with neighboring data qubits. The difficulties in achieving this arise primarily from the reliance on globally irradiating light for these operations, which can induce heating and decoherence of the atoms [58].

One approach to addressing those challenges is to hide the data qubits spatially or energetically from the controlling light for the ancilla qubits. The former takes advantage of the moving tweezers that allow qubit transport between spatially separated zones designated for a specific operation [89,192]. The latter utilizes a local addressing beam, which induces light shifts, sometimes followed by advanced state manipulation, to make data qubit atoms in a dark state [115,119,193].

Another approach is to prepare two distinct qubits in a single optical tweezer array, with each qubit acting as a data qubit and an ancilla qubit, respectively. Dual-species [194–196] and dual-isotopes [197] atom arrays of alkali atoms have successfully demonstrated the capability of qubit readout with negligible crosstalk, owing to their isolated resonant frequencies.

A simple system consisting of data qubits with inherently long coherence times and ancilla

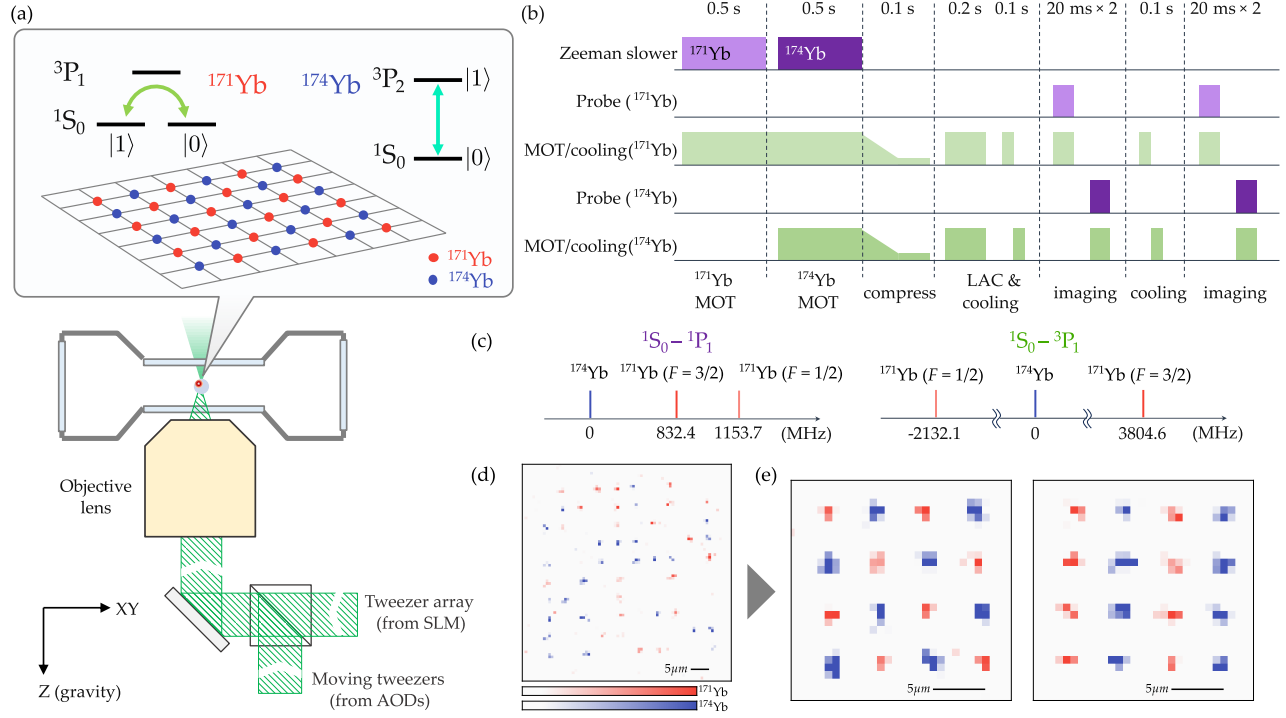


Figure 6.1 Overview of dual-Ytterbium atom array. (a) The vacuum chamber and the tweezer beam path. The tweezer array beam and moving tweezers are combined with a polarized beam splitter and focused at the center of the chamber through an objective lens with $\text{NA} = 0.6$. The fermionic isotope ^{171}Yb offers a nuclear spin qubit with a long coherence time and fast qubit control. The qubit states are encoded in the ground state. The bosonic isotope ^{174}Yb provides an optical clock qubit encoded in the ground state and a metastable excited state, which has the capability of non-destructive qubit readout. (b) Experimental sequence. First, the dual MOT of ^{171}Yb and ^{174}Yb is prepared by switching the frequency of the Zeeman slower. Then, atoms are loaded into the tweezers by compressing the dual-MOT. After LAC and cooling for each isotope, the atoms are sequentially imaged by a 399 nm probe and 556 nm cooling beams. (c) Isotope shifts of the $^1S_0 \leftrightarrow ^1P_1$ ($\lambda = 399 \text{ nm}$, $\Gamma/(2\pi) = 29 \text{ MHz}$) [137] and $^1S_0 \leftrightarrow ^3P_1$ ($\lambda = 556 \text{ nm}$, $\Gamma/(2\pi) = 182 \text{ kHz}$) [139] transitions. We use the $F = 3/2$ state of the 1P_1 and 3P_1 states for imaging and cooling for ^{171}Yb . (d) Combined single-shot image of dual-Yb array. The optical tweezer array is 10×10 with $5 \mu\text{m}$ intervals. The red and blue dots represent single ^{171}Yb and ^{174}Yb atoms, respectively. (e) Rearranged dual-Yb atom array in checkerboard (left) and stripe (right) patterns. The atoms are moved between adjacent rows or columns to avoid the filled sites.

qubits with non-destructive measurement capability, along with negligible crosstalk between them and no need for additional techniques, would be ideal for an ancilla-based QEC protocol [198]. Here, we develop a hybrid system consisting of dual-isotope alkali-earth-like ytterbium (Yb) atom array, where the fermionic isotope ^{171}Yb provides a nuclear spin qubit as a data qubit with long coherence and fast qubit control [113, 116], and the bosonic isotope ^{174}Yb offers an optical clock qubit as an ancilla qubit with the capacity of non-destructive state-selective qubit readout [117] (figure 6.1 (a)). In the following sections, we describe the experimental setup and the results of the hybrid atom tweezer array of nuclear spin and optical clock qubits.

6.2 Preparation of a dual-Yb atom array

Our experimental sequence starts by simultaneously trapping both isotopes in a MOT (figure 6.1 (b)). Subsequently, the atoms are loaded into the 10×10 tweezer array of $k_B \times 0.9$ mK depth with a beam radius of approximately 550 nm. The tweezer wavelength is 532 nm, corresponding to a near-magic wavelength for both the $^1S_0 - ^3P_1$ transition [111, 161] and the $^1S_0 - ^3P_2$ transition [112, 199] of ^{174}Yb . After applying red-detuned LAC beams of 556 nm for each isotope to prepare single atoms in the tweezers, we image them successively for 20 ms exposure time by irradiating a single resonant 399 nm beam and simultaneously cooling with MOT beams (figure 4.6). The intensities of the probe and cooling beams are $1 \times 10^{-3} I_{s,399}$ and $10 I_{s,556}$, respectively. Here $I_{s,399} = 59 \text{ mW/cm}^2$ and $I_{s,556} = 0.14 \text{ mW/cm}^2$ are the saturation intensities of the $^1S_0 - ^1P_1$ and $^1S_0 - ^3P_1$ transition, respectively. The atom temperatures are 38(2) μK for ^{171}Yb and 25(1) μK for ^{174}Yb , which are relatively higher than the previous work [111, 113] due to intensity imbalance of the cooling beams. The atom survival probabilities after imaging are 98.7 % for ^{171}Yb and 98.8 % for ^{174}Yb . Since the isotope shifts of the relevant transitions between ^{171}Yb and ^{174}Yb are on the order of 1 GHz and much larger than the natural linewidth of the $^1S_0 - ^1P_1$ ($^1S_0 - ^3P_1$) transition of 29 MHz (182 kHz) [137, 139] (Figure 6.1 (c)), the probe and cooling beams of one isotope do not cause atom loss of the other. Thus, the survival probabilities are not degraded compared to the single-isotope case.

6.2.1 Dual atom rearrangement

We use 532 nm moving tweezers generated with a pair of acousto-optic deflectors (AODs) to rearrange the atoms into an arbitrary pattern, as shown in figure 6.1 (e). Since the 532 nm tweezers can trap both isotopes and the atoms are randomly loaded in the optical tweezer array, there can be misfilled sites where the ^{174}Yb atoms fill the site designated for ^{171}Yb atoms and vice versa. Additionally, we must prevent the atoms from passing through the correctly-filled sites during the transport of the atoms. These difficulties complicate the rearrangement procedure compared with the single-isotope case. Previous research on a dual-isotope atom array has developed protocols that move the atoms along trajectories on the array grid after removing misplaced atoms that may block the paths [200, 201]. Instead, we adopt trajectories that pass between the rows and columns

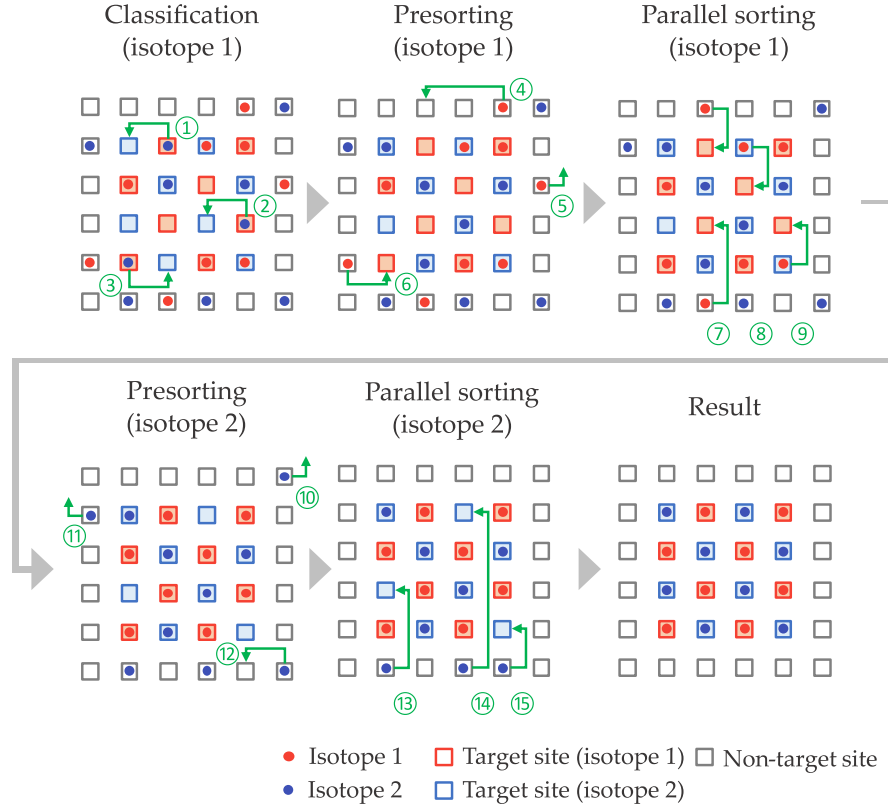


Figure 6.2 Rearrangement protocol of a dual-isotope atom array. The protocol consists of classification, presorting, and main sorting steps for each isotope. In the classification step, the atoms that occupy incorrect target sites are moved to non-target sites or correct target sites. After pre-sorting to prepare a sufficient number of atoms for each column, the atoms are transported to their target sites. In all steps, the moving tweezers pass between the rows or columns to avoid the occupied sites.

of the array and extend the parallel sort-and-compression algorithm [165], originally designed for single-isotope cases, to apply to a dual-isotope atom array.

Our protocol consists of three steps for each isotope: “classification”, “presorting”, and “main sorting” (figure 6.2). In the classification step, the isotope 2 atoms that occupy the sites designated for the isotope 1 are moved to correct or non-target sites. Next, to make the number of the isotope 1 atoms in each column equal to or larger than the number of sites designated for the isotope 1, we re-distribute the isotope 1 atoms between columns in the presorting step. The excess atoms are removed from the optical tweezer array during this step if necessary. Finally, the isotope 1 atoms are transported one by one, or simultaneously if possible, to the designated sites column by column in the main sorting step. After rearrangement of the isotope 1, we repeat the same procedure for the isotope 2. Note that, if the number of the isotope 1 atoms is equal to the number of the designated sites for the isotope 1 in each column, all isotope 1 atoms are successfully rearranged

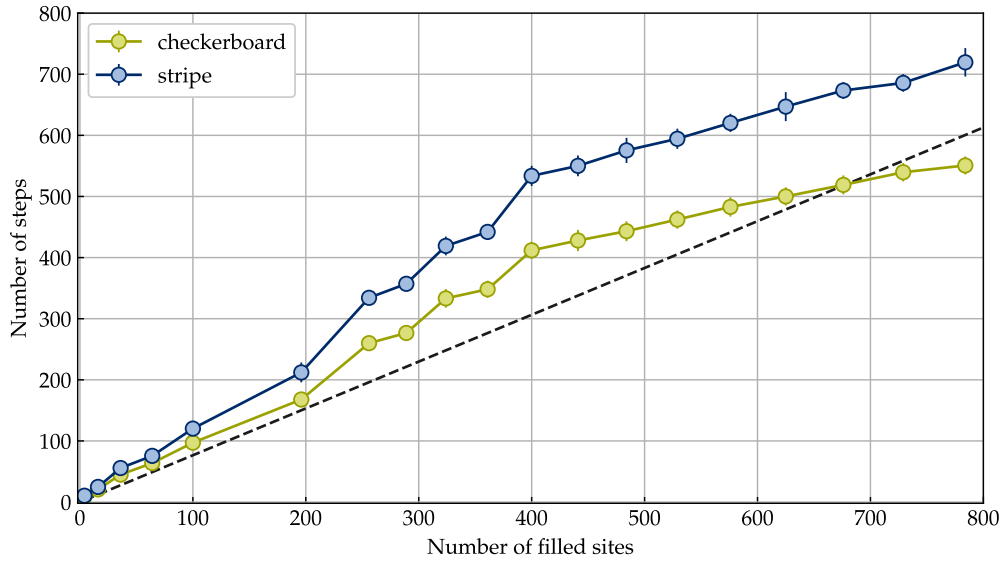


Figure 6.3 The scaling of the rearrangement protocol. The efficiency is comparable to that in the pioneering work [200] represented as a dashed black line. The scaling slope becomes more gradual as the target array size increases, indicating the efficiency may improve for larger arrays due to parallel sorting. The error bars represent the standard deviations of ten trials.

to the designated sites, and then the classification step for the isotope 1, scheduled before the pre-sorting step for the isotope 2, will be skipped. Importantly, the moving tweezers used in all steps pass between the adjacent rows or columns to avoid the filled sites.

The scalability of the rearrangement is primarily determined by the efficiency of the rearrangement algorithm. As shown in figure 6.3, the scaling of our algorithm is comparable to the pioneering work on dual-species rearrangement [200] for mid-sized arrays. Notably, the milder scaling slope in larger array sizes suggests that our algorithm may perform even more efficiently for large arrays, likely due to the parallel sorting technique demonstrated in [165].

It should be noted that the size of the defect-free array shown in figure 6.1 (e) is currently much smaller than the full 10×10 tweezer array. This is primarily due to our unoptimal transport trajectories, where atoms follow parabolic paths as they move between adjacent rows and columns. In this trajectory, the moving tweezers may pass close to other filling sites during transportation, leading to atom loss [68]. We have observed that atom loss occurs when the moving tweezer is activated at a distance of approximately $1 \mu\text{m}$ from the filling sites. In our setup, the success probability of making a 4×4 defect-free array from 10×10 array with a 20% loading of each isotope is only $\sim 1\%$. However, this could be improved by iterative rearrangement, as demonstrated in [165]. Once we use a proper sweep trajectory that does not pass close to the filling site such as the one used in [89], the size of the defect-free array will then be limited primarily by the loading probability of each isotope.

6.2.2 Controlling the dual occupancy probability

The probability of dual occupancy strongly depends on the frequency of the LAC beam for ^{174}Yb . Through frequency scanning, we determine the optimal detuning to minimize dual occupancy at $-0.50(2)$ MHz from the resonance of the $^3\text{P}_1$ state in the tweezers (figure 6.4). At this detuning, the probability of the simultaneous loading of both isotopes decreases, indicating that heteronuclear single-photon photoassociation (PA) occurs. Consequently, each of the loading probabilities of ^{171}Yb and ^{174}Yb becomes $21(5)\%$, with a dual occupancy probability of only $0.2(7)\%$. Regarding the system size, the sequential loading with shelving atoms to a metastable state [202] and blue-detuned LAC [97, 113, 170] could enhance the loading probabilities, thereby facilitating the creation of larger dual-Yb arrays.

The tunability of the dual occupancy is unique property in the dual-Yb array while this behavior

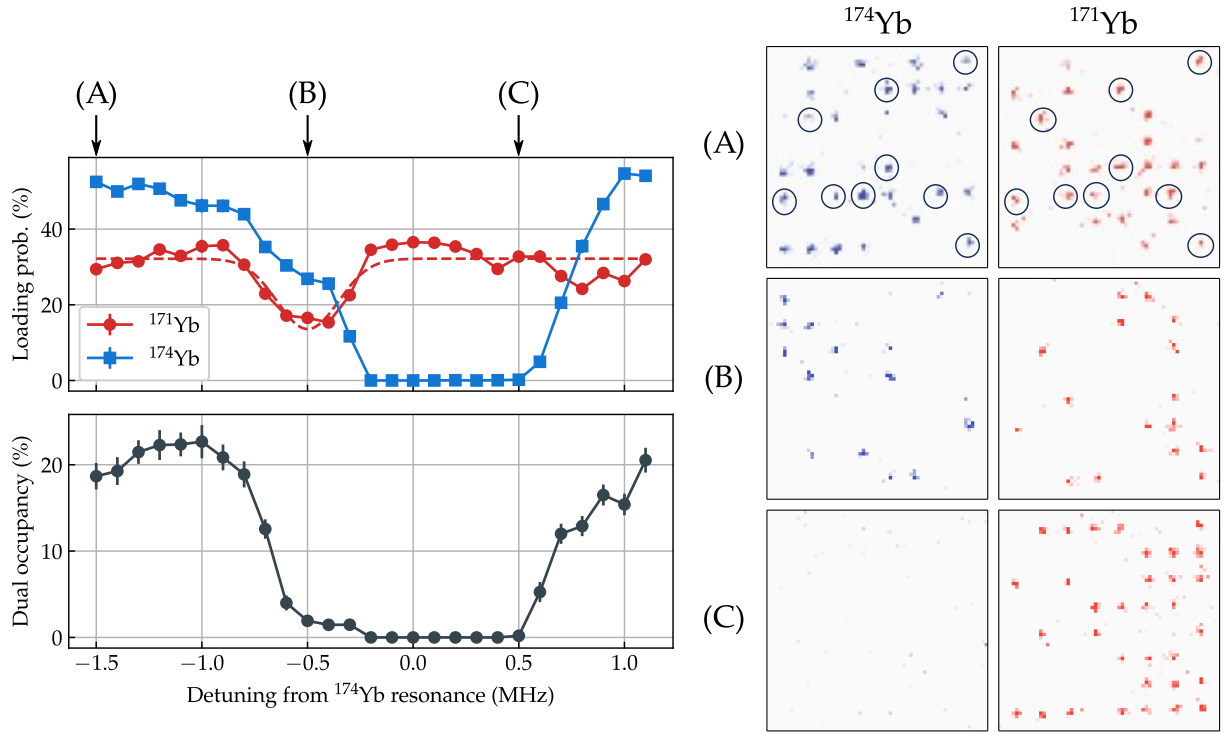


Figure 6.4 Frequency dependence of dual occupancy probability. Loading probability of each isotope (left top) and dual occupancy (left bottom) as a function of the LAC frequency around the $^3\text{P}_1$ resonance of ^{174}Yb . The dip of ^{171}Yb loading probability around -0.5 MHz indicates one-photon heteronuclear PA. The dashed line is a fitting curve by a Gaussian function. The right panels show typical atom array images at the LAC detuning of -1.5 MHz (A), -0.5 MHz (B), and 0.5 MHz (C), respectively. The black circle represents dual occupancy. Note that the loading probability of each isotope without applying heteronuclear PA fluctuates in the range of $40 \pm 10\%$, depending on the experimental conditions. The error bars represent the standard error of the mean.

is not observed in the dual-isotope alkali system [200]. The difference between the dual-isotope ytterbium and alkali systems arises from the difference in the natural linewidth Γ of the cooling transition.

The interatomic potential of the heteronuclear molecule bound near the dissociation limit is governed by the van der Waals interaction as $V(R) \propto -C_6/R^6$. From the LeRoy-Bernstein formula [203], the derivative of the binding energy $E(v)$ to the vibrational level v for the heteronuclear molecule is analytically calculated as

$$\frac{dE(v)}{dv} \propto C_6^{-1/6} (D - E(v))^{2/3}, \quad (6.1)$$

where D is the dissociation limit energy. Since the detuning of the cooling beams, which induces light-assisted collisions or photo-association, is on the order of several Γ , $D - E(v)$ in the above equation can be replaced with Γ . Furthermore, considering that the C_6 coefficient is derived from the second-order perturbation of the dipole-dipole interaction with a strength d^2 and thus, $C_6 \propto |d^2|^2 = \Gamma^2$, we get the scaling of the binding energy as

$$\frac{dE(v)}{dv} \propto (\Gamma^2)^{-1/6} \Gamma^{2/3} = \Gamma^{1/3}. \quad (6.2)$$

From the above equation, we can estimate the number of the weakly-bound states per Γ as $\Gamma / (dE/dv) = \Gamma^{2/3}$.

Since the ratio of the linewidth between the 1S_0 - 3P_1 transition of ytterbium and the D_2 transition of rubidium is around 1 : 30, the number of the bound states per Γ around the cooling detuning for alkali atoms is approximately 10 times as large as ytterbium. As a result, the cooling beams of alkali atoms are likely to overlap the photo-association resonance and naturally induce heteronuclear light-assisted collisions while the photo-association resonances of ytterbium are resolved compared to the linewidth of 1S_0 - 3P_1 transition as detailed later. We note that dual occupancy is naturally avoided in the dual-species alkali system by working with the two-color tweezer system [194, 204].

6.2.3 Observation of heteronuclear photoassociation resonances

Additionally, we explore other photoassociation (PA) resonances and identify four resonances at $-268(2)$ MHz, $-281(1)$ MHz, $-698.8(8)$ MHz, and $-716.4(5)$ MHz (figure 6.5). To confirm that these lines correspond to heteronuclear PA resonances, we perform a theoretical calculation of the PA resonance positions.

To calculate the PA resonances of the excited $^{171}\text{Yb}^{174}\text{Yb}$ molecule, which asymptotically connects to the 1S_0 - 3P_1 state, we start with the interaction Hamiltonian under a Born-Oppenheimer approximation [205], given as

$$\mathcal{H}_{\text{int}} = \frac{\vec{d}_{171} \cdot \vec{d}_{174} - 3d_{z,171}d_{z,174}}{4\pi\epsilon_0 R^3} + A\hbar \vec{I}_{171} \cdot \vec{J}_{171}. \quad (6.3)$$

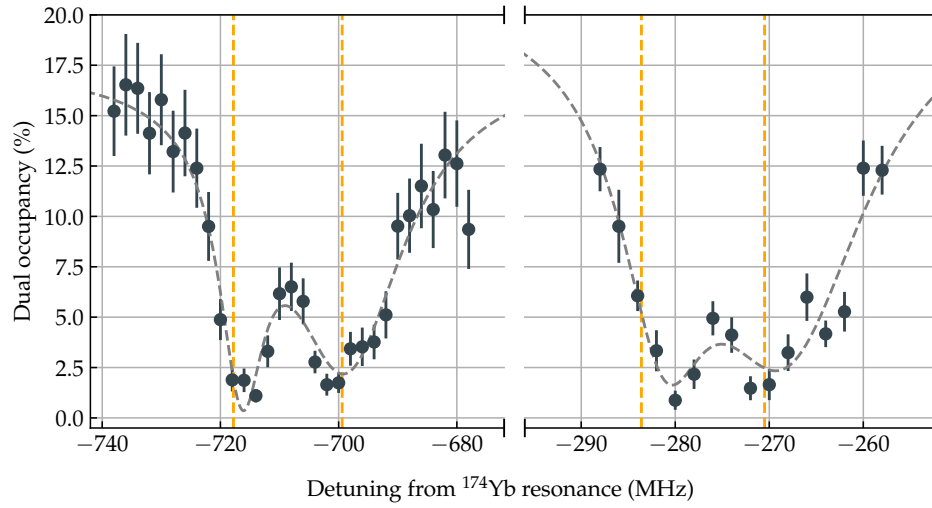


Figure 6.5 Heteronuclear PA resonances of deeper bound states around -710 MHz and -280 MHz. The data are fitted by a double Lorentzian function. The yellow dashed lines represent the theoretical PA resonances. The error bars represent the standard error of the mean.

Here, $\vec{d}$ and d_z are the dipole operator and its projection onto the interatomic axis for each atom, ϵ_0 is the permittivity of vacuum, R is the interatomic distance, h is the Planck constant, A is the hyperfine coupling constant of ^{171}Yb , and $\vec{I}$ and $\vec{J}$ are the nuclear spin and electronic angular momentum operators of ^{171}Yb , respectively. We take the basis of $|G_i\rangle_{171} \otimes |E_j\rangle_{174}$ and $|E_i\rangle_{171} \otimes |G\rangle_{174}$ for the diagonalization, where $|G_i\rangle_{171}$ ($i = 1, 2$) and $|E_i\rangle_{171}$ ($i = 1-6$) are the ground states and excited 3P_1 states of ^{171}Yb , and $|G\rangle_{174}$ and $|E_j\rangle_{174}$ ($j = 1-3$) are the ground state and excited 3P_1 states of ^{174}Yb , respectively. By diagonalization of the Hamiltonian (equation (6.3)), we obtain six interaction potentials, as shown in figure 6.6. Note that the retardation effect on the resonant dipole interaction [206] is involved in this calculation. The potential No. 1–3 are attractive, thus they can support weakly-bound heteronuclear molecules. Then, we add a centrifugal potential, van der Waals potential, and a short-range repulsive potential to the obtained potential $\langle \mathcal{H}_{\text{int}} \rangle$ and get a model potential [207] as

$$V(R) = \langle \mathcal{H}_{\text{int}} \rangle + \hbar^2 \frac{T_e(T_e + 1) + \langle \vec{F}^2 \rangle - 2\Omega^2}{2\mu R^2} - \frac{C_6}{R^6} + \frac{C_{12}}{R^{12}}, \quad (6.4)$$

where $\hbar = h/(2\pi)$ is reduced Planck constant, μ is the reduced mass of ^{171}Yb and ^{174}Yb , T_e and Ω are the total electronic angular momentum and its projection onto the molecular axis, C_6 and C_{12} are the coefficients for the van der Waals potential and the short-range repulsive potential, respectively. $\vec{F} = \vec{I}_{171} + \vec{J}_{171} + \vec{J}_{174}$ is the sum of electronic angular momenta of ^{171}Yb and ^{174}Yb and nuclear spin of ^{171}Yb . Finally, we solve the Schrödinger equation with the model potential (equation (6.4)) by Numerov method [208] and obtain the eigenenergies. The parameters for this calculation are summarized in table 6.1.

Table 6.2 shows the resonance frequencies of the PA below 1 GHz for the potential No.2, in

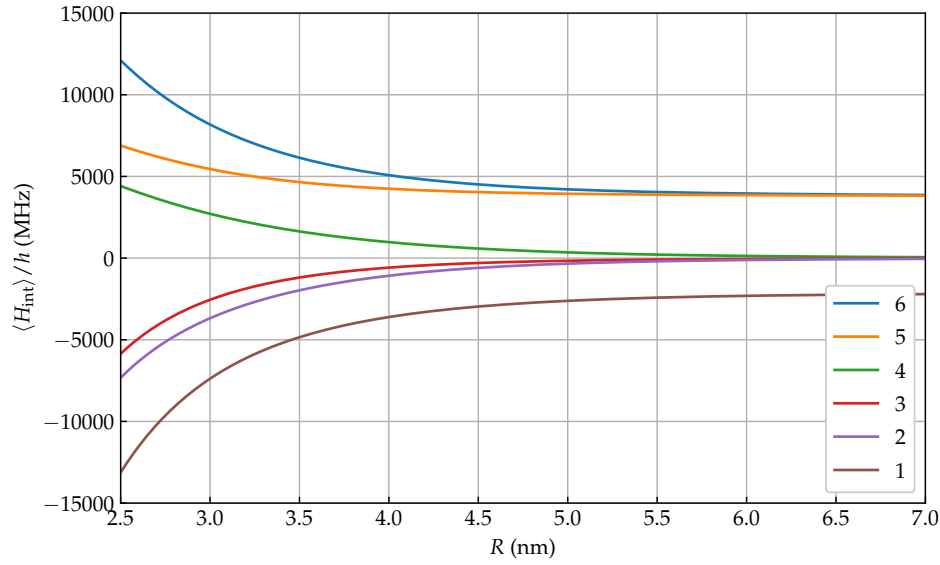


Figure 6.6 Interaction potentials between ^{171}Yb and ^{174}Yb obtained from equation (6.3). The potentials are plotted with respect to the $^3\text{P}_1$ state of ^{174}Yb . Potential No.2 and 3 asymptotically connect to the $|^1\text{S}_0\rangle_{171} |^3\text{P}_1\rangle_{174}$ atomic state in the dissociation limit.

Table 6.1 Parameters for PA resonance calculation. E_h is Hartree energy and a_0 is Bohr radius.

Parameter	Value
A	3957.781 MHz [139]
$d^2/(4\pi\epsilon_0)$	$0.08744313 E_h a_0^3$ [209]
C_6	$2405.364747 E_h a_0^6$ [209]
C_{12}	$9.319 \times 10^8 E_h a_0^{12}$ (fitting)

which the molecular states have $T_e = 1/2, 3/2$ and $|\Omega| = 1/2$. The calculated PA resonance frequencies agree with the experimental values with a few MHz precision.

These observations offer an important possibility of utilizing these PA lines in stimulated Raman adiabatic passage [210,211] or two-photon PA [212,213] to prepare ground-state heteronuclear molecules in the optical tweezers [213–219]. Such molecules possess multiple degrees of freedom, including rotational and vibrational levels, as well as nuclear spin states decoupled from hyperfine interactions. The presence of nuclear spin states with long coherence times [220,221] makes it feasible to realize proposals that leverage molecular degrees of freedom for quantum computation, thereby potentially enhancing the scalability of the system [222–224]. We note that while polar molecules consisting of alkali atoms do possess nuclear spin states in the $^1\Sigma$ electronic ground states, preparing such molecules is generally technically demanding [225]. In contrast, the dual-Yb system offers a relatively straightforward method [226–228] for producing heteronuclear molecules with

Table 6.2 PA resonances below 1 GHz for the potential No. 2.

T_e	Calculation (MHz)	Experiment (MHz)
1/2	-0.05	-0.50(2)
3/2	-55.3	—
1/2	-62.9	—
3/2	-270.5	-268(2)
1/2	-283.6	-281(1)
3/2	-699.4	-698.8(8)
1/2	-717.8	-716.4(5)

nuclear spin states.

It is also worth noting that the cooling beams during imaging do not induce the heteronuclear PA because they are detuned by approximately -3Γ from the PA resonance, as shown in the panel (A) of figure 6.4. This allows us to implement various operations while maintaining the dual occupancy sites, such as spin initialization of ^{171}Yb , cooling of the motional degrees of freedom via Raman sideband cooling [17, 113] for ^{171}Yb and a clock transition [186] for ^{174}Yb , as long as the operation lasers are detuned from the PA resonances. Furthermore, it is feasible to image atoms dissociated from a molecule after performing specific operations, allowing the internal state of the molecule to be detected without losing the atoms. These capabilities not only enhance the fidelity of molecule creation, but also simplify the overall process of conducting molecular experiments, making them more efficient and accessible.

6.3 Crosstalk measurement

In the dual-Yb atom array, we can utilize two distinct types of qubits: nuclear spin qubits and optical qubits. The nuclear spin qubit is encoded in the ground state of ^{171}Yb as $|0\rangle = |^1\text{S}_0, m_F = 1/2\rangle$ and $|1\rangle = |^1\text{S}_0, m_F = -1/2\rangle$, which exhibit a second-order coherence time and the capability of MHz-order fast control ([113, 116], and see section 5.1). The optical qubit, encoded in the ground state and the $m_J = 0$ state of an excited metastable state of ^{174}Yb , such as $^3\text{P}_0$, $^3\text{P}_2$, and $4f^{13}5d6s^2$ ($J = 2$) [229, 230], can be controlled by M2-transition [231] or magnetic-field induced E1 transition [184, 232] and imaged non-destructively, as the metastable state naturally serves as a dark state for the probe transition, successfully demonstrated in a recent work using the $^3\text{P}_0$ state [119].

We investigate the crosstalk between them, specifically the ability to image ^{174}Yb without losing the coherence of the nuclear spin qubits. Since the resonant frequencies of the probe and cooling light for ^{174}Yb are well-isolated from those for ^{171}Yb in GHz order (figure 6.1 (c)), the photon scattering rates of ^{171}Yb during the ^{174}Yb imaging are calculated to be approximately 30 Hz for 399 nm light and below 0.1 Hz for 556 nm light, respectively, which are small enough for our imaging con-

ditions of 20 ms exposure time. Note that ^{174}Yb is the best isotope for ^{171}Yb in terms of isotope shifts to suppress the crosstalk [137, 139].

To quantify this, we characterize the coherence property of the nuclear spin qubit using the Ramsey and Hahn-echo sequences under irradiation of imaging light for ^{174}Yb , where we consider two imaging methods: “399 nm imaging” with a 399 nm probe beam and 556 nm cooling beams, and “556 nm imaging” with a single 556 nm probe beam. We first prepare the dual-Yb atom array and initialize the nuclear spin qubit via optical pumping by the $|1\rangle \rightarrow |^3P_1, F = 3/2, m_F = 1/2\rangle$ transition. We then irradiate a circularly-polarized single 556 nm beam in the horizontal plane to rotate the qubits by single-beam Raman transitions [113]. The qubit control beam has an intensity of $2.4 \times 10^5 I_{s,556}$ and a detuning of -1.0 GHz (-3.1 GHz) from the resonance of $|^3P_1, F = 1/2\rangle$ of ^{171}Yb ($|^3P_1\rangle$ of ^{174}Yb), yielding the Rabi frequency of $2\pi \times 208$ kHz. After a $\pi/2$ -pulse and a fixed waiting time of $T + \Delta T$, another $\pi/2$ -pulse is applied to map the relative phase of the qubit states to their relative populations. Here, the additional waiting time of ΔT is used for scanning the relative phase between the qubit states. In the Hahn-echo sequence, we insert an additional π -pulse after a waiting time of $T/2$ from the first $\pi/2$ -pulse. Finally, we blow away the atoms in the $|0\rangle$ state via the $|0\rangle \leftrightarrow |^3P_1, F = 3/2, m_F = 3/2\rangle$ transition and image the remaining atoms. By scanning ΔT , we obtain a Ramsey (Hahn-echo) signal and compare its contrast with and without irradiation of imaging light for ^{174}Yb during holding time T to evaluate its influence.

6.3.1 Hahn-echo sequence

We first present the results of the Hahn-echo experiment using our current 399 nm imaging method. During the holding time T , with a trap depth of 0.9 mK, we apply a resonant 399 nm probe beam with a radius of 1.3 mm at an intensity of $1 \times 10^{-3} I_{s,399}$, as well as 556 nm cooling beams with radii of 4 mm (vertical) and 12 mm (horizontal) at a total intensity of $10 I_{s,556}$, which are detuned by -5.5Γ from the resonance. Figure 6.7 shows the Hahn-echo signals for holding time of $T = 20$ ms, which is typical in our current setup, with and without 399 nm imaging. The oscillation frequency is determined by the Zeeman splitting of the nuclear spin states with a magnetic field of 2.5 G applied during the holding time. The contrast ratio between the two conditions is 0.991(18), indicating that the coherence is maintained within the uncertainty.

Imaging performance

The detection fidelity of $|^1S_0\rangle = |0\rangle$ of ^{174}Yb is also important quantity. To evaluate the imaging performance, especially the discrimination fidelity of the states, we employ two methods: the usual fitting method and the model-free method [69].

First method relies on fitting of the histogram of the number of detected photons during imaging as shown in figure 6.8. To fit the histogram, we chose fitting functions as

$$P(x) = (1 - F)P_D(x) + FP_B(x), \quad (6.5)$$

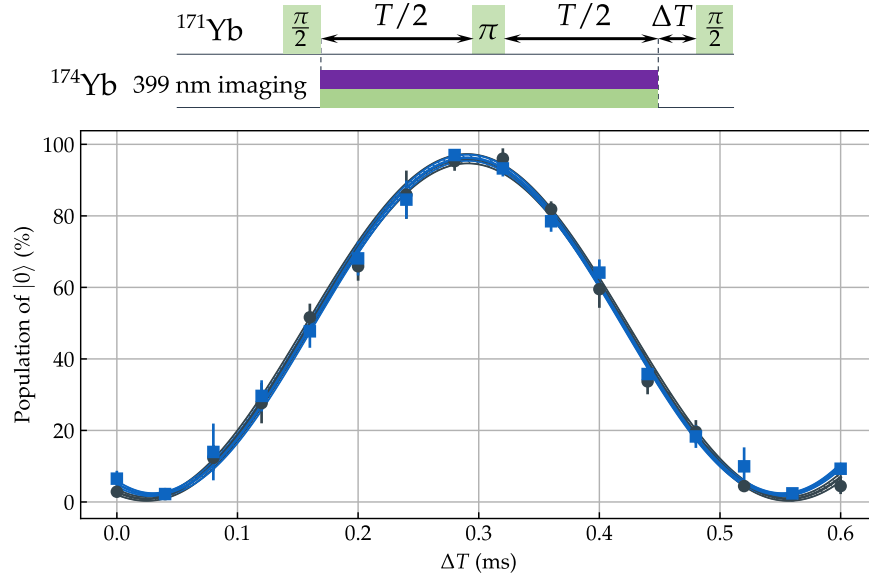


Figure 6.7 Influence on the coherence of the nuclear spin qubit from the imaging light for ^{174}Yb in the Hahn-echo sequence. Hahn-echo signal for holding time of $T = 20$ ms with (blue square) and without (black circle) 399 nm probe and 556 nm cooling beams. The solid lines are the fitting curves by a sinusoidal function. The contrast ratio is 0.991(18), highlighting the possibility of the 399 nm imaging without disturbing the coherence of the nuclear spin qubits. The error bars represent the standard error of the mean.

where F is the loading probability of atoms, and $P_D(x)$ and $P_B(x)$ are the distributions for dark and bright peaks of the histogram as

$$\begin{aligned} P_D(x) &= \frac{1}{\sqrt{2\pi}\sigma_D} \exp\left(-\frac{(x - \mu_D)^2}{2\sigma_D^2}\right) + a \exp(-bx) \\ P_B(x) &= \frac{1}{\sqrt{2\pi}\sigma_B} \exp\left(-\frac{(x - \mu_B)^2}{2\sigma_B^2}\right) \left(1 + \operatorname{erf}\left(c \frac{x - \mu_B}{\sqrt{2}\sigma_B}\right)\right) \end{aligned} \quad (6.6)$$

with the fitting parameters σ_D , μ_D , a , b , σ_B , μ_B , c . The $P_D(x)$ is composed of the Gaussian and exponential distributions, which account for the readout and amplification noise from the EMCCD camera. We note that strictly speaking, the amplification process of the camera count is explained by the Erlang distribution $P(x) = x^{n-1}e^{-x/g}/(g^n(n-1)!)$ with a photo-electron number n and a gain g [233]. However, for the dark distribution, where the number of photo-electrons is almost zero ($n \leq 1$), the Erlang distribution can be reduced to an exponential distribution. For the fitting to the bright peak, although the convolution of Poisson and Erlang distribution would be appropriate, we chose a skewed Gaussian distribution for computation simplicity and convenience, as used in

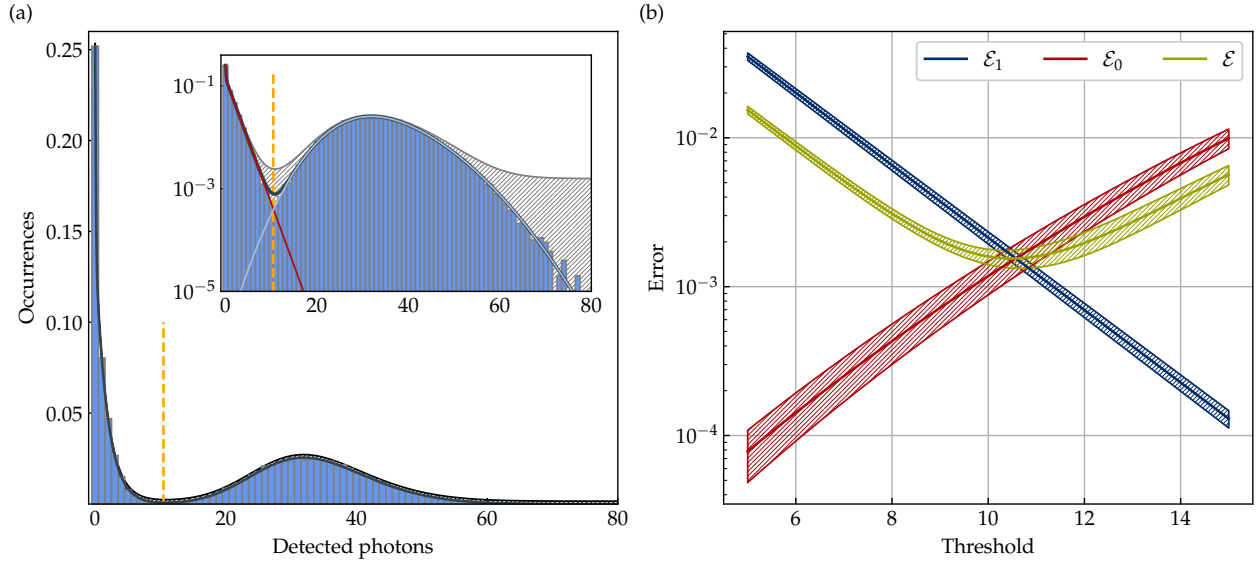


Figure 6.8 Fitting to the histogram of detected photons for imaging performance evaluation. (a) The histogram of the number of detected photons by 399 nm imaging with an exposure time of 20 ms. The black, red, and light blue curves represent the fitting function (Eq. 6.7), $P_D(x)$, and $P_B(x)$, respectively. The dashed orange is the threshold that maximizes the discrimination fidelity at 0.9985(2). (b) Imaging infidelities as a function of the threshold. The shaded areas in (a) and (b) are the 1σ confidence intervals.

previous work [80]. Figure 6.8 shows the fitting result and corresponding infidelities calculated as

$$\begin{aligned}\mathcal{E} &= (1 - F)\mathcal{E}_0 + F\mathcal{E}_1 \\ &= (1 - F) \int_{x_{\text{th}}}^{\infty} P_D(x) dx + F \int_0^{x_{\text{th}}} P_B(x) dx\end{aligned}\quad (6.7)$$

with the threshold for determining the presence of atoms and $\mathcal{E}_0$ and $\mathcal{E}_1$ are the false-negative and false-positive errors. The best fidelity is found to be 0.9985(2).

To precisely evaluate the fidelity without introducing any bias, we also employ a model-free method as detailed in [69]. In this approach, we successively capture three images under identical conditions during the cross-talk measurement using 399 nm imaging. When three images are taken in a single sequence, the probabilities of each event $\tilde{P}_{x_1 x_2 x_3}$, where $x_i \in \{0, 1\}$ represents whether a single atom is trapped (1) or not (0) in the i -th shot, are given by

$$\begin{aligned}\tilde{P}_{111} &= S^2 F, \\ \tilde{P}_{110} &= (1 - S) S F, \\ \tilde{P}_{100} &= (1 - S) F, \\ \tilde{P}_{000} &= 1 - F.\end{aligned}\quad (6.8)$$

Here, S represents the survival probability after imaging, and F denotes the loading (filling) prob-

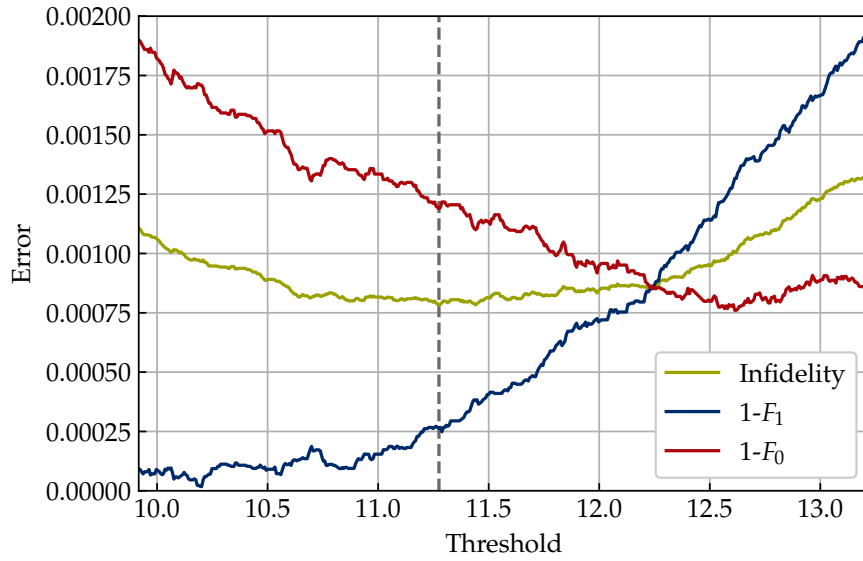


Figure 6.9 Imaging fidelity evaluation by model-free method. The best fidelity is found to be 0.9992 with a survival probability of 0.988 under a 20 ms exposure time. The loading probability is found to be 0.561 in this measurement. The dashed line indicates the threshold that minimize the discrimination infidelity.

ability. Given a threshold for detemining the presence of atoms, we can calculate the experimental value of the probability of each event as

$$P_{y_1 y_2 y_3} = \sum_{x_1 x_2 x_3} P(y_1|x_1)P(y_2|x_2)P(y_3|x_3)\tilde{P}_{x_1 x_2 x_3}, \quad (6.9)$$

where each conditional probability $P(y|x)$ is calculated from the discrimination fidelity $\mathcal{F}_0$ and $\mathcal{F}_1$ as $P(1|1) = \mathcal{F}_1$, $P(0|1) = 1 - \mathcal{F}_1$, $P(1|0) = 1 - \mathcal{F}_0$, and $P(0|0) = \mathcal{F}_0$. The above equations are explicitly written as

$$\begin{aligned} P_{111} &= (1 - \mathcal{F}_0)^3 \tilde{P}_{000} + \mathcal{F}_1(1 - \mathcal{F}_0)^2 \tilde{P}_{100} + \mathcal{F}_1^2(1 - \mathcal{F}_0) \tilde{P}_{110} + \mathcal{F}_1^3 \tilde{P}_{111} \\ P_{110} &= (1 - \mathcal{F}_0)^2 \mathcal{F}_0 \tilde{P}_{000} + \mathcal{F}_1(1 - \mathcal{F}_0) \mathcal{F}_0 \tilde{P}_{100} + \mathcal{F}_1^2 \mathcal{F}_0 \tilde{P}_{110} + \mathcal{F}_1^2(1 - \mathcal{F}_1) \tilde{P}_{111} \\ P_{100} &= (1 - \mathcal{F}_0) \mathcal{F}_0^2 \tilde{P}_{000} + \mathcal{F}_1 \mathcal{F}_0^2 \tilde{P}_{100} + \mathcal{F}_1(1 - \mathcal{F}_1) \mathcal{F}_0 \tilde{P}_{110} + \mathcal{F}_1(1 - \mathcal{F}_1)^2 \tilde{P}_{111} \\ P_{000} &= \mathcal{F}_0^3 \tilde{P}_{000} + (1 - \mathcal{F}_1) \mathcal{F}_0^2 \tilde{P}_{100} + (1 - \mathcal{F}_1)^2 \mathcal{F}_0 \tilde{P}_{110} + (1 - \mathcal{F}_1)^3 \tilde{P}_{111} \end{aligned} \quad (6.10)$$

By tuning the four parameters F , S , $\mathcal{F}_0$, and $\mathcal{F}_1$ for a given threshold to match the theoretical values with the experimental results, we obtain a discrimination fidelity expressed as $\mathcal{F} = (1 - F)\mathcal{F}_0 + F\mathcal{F}_1$. This procedure is iterated for various thresholds to determine the optimal fidelity that minimizes the infidelity. Using this method, we achieve a discrimination fidelity $\mathcal{F} = 0.9992$ with a survival probability $S = 0.988$ under a 20 ms exposure time and a loading probability $F = 0.561$, as shown in figure 6.9.

Long exposure time region

To assess the trend of the influence on the coherence in the longer exposure time region, we extend the holding time up to 1500 ms. Figure 6.10 illustrates the decay curve of the Hahn-echo contrast as a function of the exposure time. We estimate the coherence time T_2 in each case by fitting the data using the function $f(T) = A \exp(-(T/T_2)^n)$, where A , T_2 , and n are fitting parameters. The fitting results are summarized in table 6.3. In the 399 nm imaging, the coherence time is 0.67(5) s, which is shortened due to the scattering of the probe and cooling beams. However, as shown figure 6.7, we can still achieve high-fidelity imaging while maintaining the coherence. Moreover, considering the NA of our objective lens, there is still potential to improve the photon collection efficiency by a factor of three. If such an optimal imaging system is realized, the exposure time could be reduced by one-third, leading to a data qubit coherence retention of 99.7%. We note that the coherence without imaging is limited by the scattering from the tweezer with a trapping depth of 0.9 mK. This issue can be addressed by making the trap shallower at the data qubit sites while deepening it at the ancilla qubit sites.

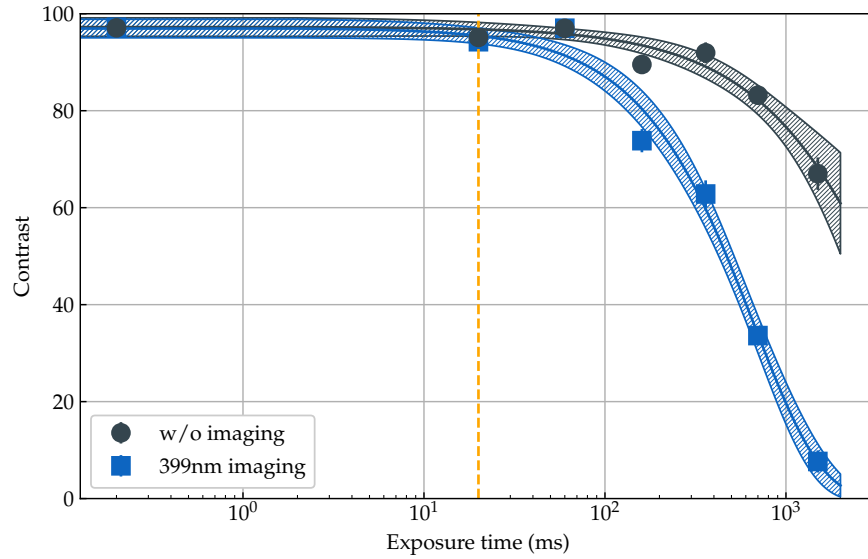


Figure 6.10 Decay curves of the Hahn-echo contrasts with and without exposure to imaging light for ^{174}Yb . The solid lines are fitting curves by $f(T) = A \exp(-(T/T_2)^n)$. The fitting results are summarized in Table 6.3. Although the coherence under the 399 nm imaging decreases faster than that of without imaging, the contrast of the Hahn-echo signal is almost the same for our typical exposure time of 20 ms, represented as an orange dashed line. The error bars and shaded regions represent the standard error of the mean and 1σ confidence intervals of the fit, respectively.

Table 6.3 Fitting results of the coherence curves in the Hahn-echo sequence.

	A	T_2 (s)	n
without imaging	0.97(2)	4.3(2.3)	1.0(3)
399 nm imaging	0.97(2)	0.67(5)	1.2(2)

6.3.2 Ramsey sequence

As a potential future improvement, we evaluate the influence on the coherence using the Ramsey sequence with the 556 nm imaging, which has proven highly effective for ^{174}Yb trapped in 532 nm tweezers [111]. The trap depth of 0.9 mK is the same as the Hahn-echo sequence. We apply a resonant single 556 nm beam for ^{174}Yb with a radius of 1.4 mm at an intensity of $3 I_{s,556}$, typical in the previous research [111], during the holding time for the 556 nm imaging. The probe beam diagonally propagates from the horizontal plane by 16 degrees and is horizontally polarized. Figure 6.11 presents the trend of the Ramsey contrast with a holding time up to 160 ms. While the contrast in the case of the 399 nm imaging rapidly decreases, the coherence under the 556 nm imaging remains almost the same as that without imaging. We again note that the observed coherence is reduced due to the tweezer potential of 0.9 mK, and does not exhibit a fundamental limitation. The fitting results, using the same function for the Hahn-echo experiment, are summarized in table 6.4. This result highlights the potential of the 556 nm imaging for low cross-talk measurements. Based on the experimental result in Ref. [111], the discrimination fidelity is expected to exceed 0.9999 with a 99 % survival probability for a 30 ms exposure time, with negligible impact on the data qubit coherence. Since our current optical setup does not support 556 nm imaging, we plan to verify its actual performance in the dual-Yb system in future work, which will be useful in fully evaluating its potential for improving low cross-talk measurements and enhancing overall system performance.

Table 6.4 Fitting results of the coherence curves in the Ramsey sequence.

	A	T_2^* (ms)	n
without imaging	0.98(2)	130(3)	2.0(2)
556 nm imaging	0.98(2)	122(5)	2.3(3)
399 nm imaging	0.99(2)	77(2)	1.8(1)

6.4 Discussion

The cross-talk measurement with our current 399 nm imaging, shown in figure 6.7, has revealed that the dual-Yb system has the capability of low cross-talk measurement with high-fidelity detection of the ancilla qubits. The current imaging performance is within reach of the requirement

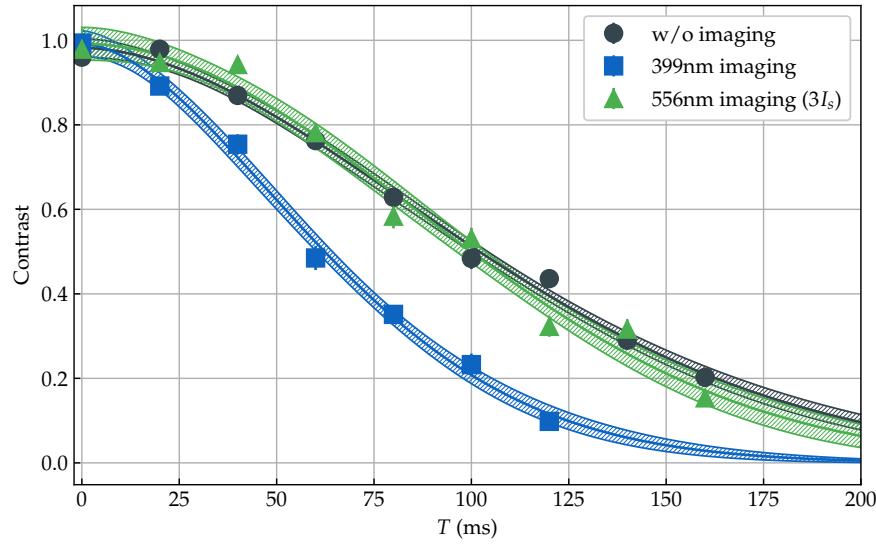


Figure 6.11 Influence on the coherence of the nuclear spin qubit from the imaging light for ^{174}Yb in the Ramsey sequence. Decay curves of the Ramsey contrast are shown as a function of exposure time. The solid lines are the fitting curves by $f(T) = A \exp(-(T/T_2^*)^n)$, where A , T_2^* , and n are fitting parameters. While the Ramsey contrast under 399 nm imaging decays faster than that without imaging, the contrast with 556 nm probe light with an intensity of $3 I_{s,556}$ (green triangle) remains the same as that of without irradiating a probe beam, suggesting that 556 nm imaging is a possible future improvement of low cross-talk measurements. The error bars and shaded regions represent the standard error of the mean and 1σ confidence intervals of the fit, respectively.

for the surface codes in neutral atom arrays [234]. Moreover, considering that the surface code is more resilient to readout errors than to coherent errors [235], our imaging performance, especially after the further optimization, is likely acceptable for surface code implementation. The coherence loss during imaging is less than 1 %, which is similarly qualified or even superior to the previous research using the omg-architecture [119], energy isolation by local light shift [115], and shelving technique [193] (table 6.5). The one important key aspect of the dual-Yb system lies in the simplicity of the operation for mid-circuit measurements while the other approaches including the spatial isolation methods [89, 192] require technically demanding local operations.

As is shown in figure 6.7, the crosstalk induced by the 399 nm imaging with our current parameters is only 0.9 %. However, when comparing the dual-Yb system with the dual-alkali system [195], the behavior of the contrast decay curves with and without imaging in the long exposure time region differs from that observed in the dual-alkali system. This is due to the limited frequency isolation, which is characterized by the ratio between the isotope shift $|\omega - \omega'|$ and the natural linewidth Γ of the transition, with $|\omega_{171\text{Yb}} - \omega_{174\text{Yb}}|/\Gamma_{399} \sim 30$ for the $^1S_0 \leftrightarrow ^1P_1$ transition of ytterbium, much smaller than $|\omega_{\text{Rb}} - \omega_{\text{Cs}}|/\Gamma \sim 10^6$ for the dual-alkali system. However, as shown in Fig. 5, 556 nm imaging holds the potential to achieve comparably low cross-talk measurements as the dual-alkali

Table 6.5 Comparison of low cross-talk measurement schemes

	Readout time	Imaging fidelity	Figure of merit
Shelving [193]	4 ms	94.9(8) % ($ 0\rangle$) 95.3(1.1) % ($ 1\rangle$)	$\mathcal{F} = 97.0(5) \%^a$
Local light shift [115]	5 ms	$\sim 99.9 \%$	97.5(1) % ($ 0\rangle$ imaging) ^b 98.7(8) % ($ 1\rangle$ imaging) ^b
omg-architecture [119]	3.5 ms	99.8 %	$F_{p,DQ} = 97.2(5) \%^c$
dual-alkali [195]	15 ms	98.9 %	$(A, \tau, n) = (0.97(2), 650(20)\text{ms}, 1.71(9))^d$ (w/o readout) $(A, \tau, n) = (0.935(2), 678(13)\text{ms}, 1.82(7))^d$ (w/ readout)
dual-Yb	20 ms	99.92 % (98.8 % survival)	99.1(1.8)% ^e

^a They estimated the process fidelity $\mathcal{F}$ of the shelving and echo by a Ramsey-type experiment with a measurement of ancilla qubits.

^b They compared the coherence contrast of the data qubits with and without imaging of the ancilla qubits during the holding time of the Ramsey sequence.

^c They estimated the process fidelity $F_{p,DQ}$ of mid-circuit measurement by quantum process tomography.

^d They fitted the contrast decay curves of the data qubits as a function of readout time using the function $f(t) = A \exp(-(t/\tau)^n) + B$ and compared the τ for cases with and without readout of ancilla qubits.

^e We compared the coherence contrast of the data qubits with and without imaging of the ancilla qubits during the hold time of the Hahn-echo sequence.

system, owing to its large frequency isolation of $|\omega_{171\text{Yb}} - \omega_{174\text{Yb}}|/\Gamma_{556} \sim 10^4$.

Here, we point out that the important difference in the alkali and Yb systems arises from the types of qubits. In the alkali system, the hyperfine state qubits with millisecond-order coherence, with no dynamical decoupling, are used for both data and ancilla qubits. Typically, the qubit detection in this system relies on the state-selective loss before imaging, which requires more frequent atom replenishment. Although the non-destructive imaging has been demonstrated in alkali systems using techniques such as shelving [193] and state transfer to a stretched state [80, 236], those operations may carry the risk of finite infidelity. In contrast, the dual-Yb system can offer a data

qubit (nuclear spin qubit) with inherently long second-order coherence time, and an ancilla qubit (optical qubit) that is intrinsically capable of non-destructive readout without state-selective loss. These advantages make the dual-Yb system preferable in implementing mid-circuit measurements for quantum error correction.

6.5 Conclusion

In summary, we have reported the realization of a dual-Yb atom array consisting of fermionic ^{171}Yb and bosonic ^{174}Yb . This hybrid system offers a nuclear spin qubit as a data qubit with a long coherence time and an optical clock qubit as an ancilla qubit, which provides the ability to read out the qubit states non-destructively. We found the tunability of the dual occupancy rate from almost zero to several tens of percent. Thus, this remarkable versatility could open the avenue to the experiment with heteronuclear molecules trapped in optical tweezers [237–241]. We evaluated the coherence of the nuclear spin qubits under exposure to the imaging beams for ^{174}Yb . In the case of 399 nm imaging with an exposure time of 20 ms, the coherence is maintained at 99.1(1.8) % level with a discrimination fidelity of 99.92 % and a survival probability after imaging of 98.8 % for ^{174}Yb . Although the current imaging performance is slightly worse than those in the state-of-the-art divalent atom arrays [75, 119], the coherence retention of the data qubits during ancilla readout is comparably as good as the previous work [115, 119, 193], even without technical local operations. Moreover, we confirm that the 556 nm imaging with $3 I_{s,556}$, typical in previous study [111], causes no detrimental effect on the coherence and could enable high-fidelity and low atom loss imaging, making it a promising candidate for mid-circuit measurements. Combined with further optimization of the imaging system, we anticipate that the dual-Yb system will be fully capable of mid-circuit operations.

As an outlook, we plan to integrate coherent controls on the optical qubits within this dual-Yb system. Leveraging the insensitivity to magnetic field fluctuations [185] and the magic-wavelength tweezer that compensates for tensor light shifts, the $^3\text{P}_2$ and $4f^{13}5d6s^2$ ($J = 2$) states, in addition to the $^3\text{P}_0$ state, present promising candidates for encoding optical qubits. Single-qubit operations on these optical qubits will employ π -pulses for X gates and phase modulation of the excitation laser for Z gates [54]. Furthermore, the fine-structure qubit recently demonstrated in strontium systems [109, 134] offers an intriguing direction, particularly due to its potential for fast and precise control. Regarding the generation of entanglement between nuclear spin qubits and clock transition qubits, the time-optimal two-qubit gates mediating the Rydberg interaction [158] can be used. For this, it is necessary to investigate the Rydberg interaction between the isotopes. Recent comprehensive Rydberg spectroscopy of Yb [78] and the development of sophisticated software for the Rydberg interaction calculation of divalent atoms [149] will aid in investigating the optimal conditions to achieve similarly high-fidelity two-qubit gates demonstrated in single-species atom arrays [54, 76–80]. Additionally, the inherent imbalance between the intra- and inter-isotope Rydberg interaction

has the potential to implement high-fidelity native multi-qubit gates [242], offering a pathway to efficient quantum computation. Those studies combining our dual-Yb system with some additional improvement and development will pave the way to the ancilla-based QEC protocols in the neutral atom architecture.

Chapter 7

Development of a high-power ultraviolet laser system and observation of fast coherent Rydberg excitation of ytterbium

Rydberg interactions in neutral atom arrays play crucial roles in quantum simulations of various physical models [32–34, 36, 37, 39, 42, 43, 50] and in generating entanglements for quantum information processing [22, 72, 73, 76, 152, 153, 158]. The Rydberg states, which are highly excited states close to the ionization limit of an atom, are typically accessed via a two-photon process in alkali atoms such as rubidium or cesium. However, this method can introduce unwanted scattering from the intermediate state, which can negatively impact the Rydberg excitation fidelity [58].

In contrast, alkali-earth or alkali-earth-like atoms (AEA) such as strontium [65, 105–110] and ytterbium [111–115] have recently attracted considerable interest. The optical tweezer systems employing the AEA enable Rydberg excitation via a single-photon process from a metastable excited state, offering a pathway to higher-fidelity operations [74, 77–79, 117, 159]. The key to further improving fidelity in these setups lies in the use of high-power lasers with low frequency noise [79, 243, 244]. However, this introduces a technical challenge: the wavelength required for Rydberg excitation in AEAs lies in the ultraviolet (UV) region, making it inherently difficult to construct a high-power laser with the necessary specifications for such systems.

This chapter describes the development of a high-power ultraviolet laser system and the observation of fast coherent Rydberg excitation of ytterbium, based on the published paper [245]

7.1 Development of a high-power ultraviolet laser system

7.1.1 Overview of the laser system

The transition wavelength from the metastable 3P_2 state to a Rydberg state in Yb is approximately 325 nm [112]. To achieve high-power output at this wavelength, we employ a two-stage frequency doubling scheme (figure 7.1). The laser system is composed of an external cavity diode laser and a tapered amplifier as a seed laser ($\lambda = 1300$ nm, Toptica, TA pro), followed by a Raman fiber amplifier coupled with a waveguide SHG module (MPB Communications, VRFA-P-3000-651.2-SF-Plus), which converts the wavelength from 1300 nm to 650 nm. The second stage of frequency doubling is achieved by developing an efficient SHG cavity system utilizing a beta barium borate (BBO) crystal, which further converts the wavelength from 650 nm to 325 nm.

The seed laser, with an output power of 100 mW, is first coupled to a single-mode fiber. It is

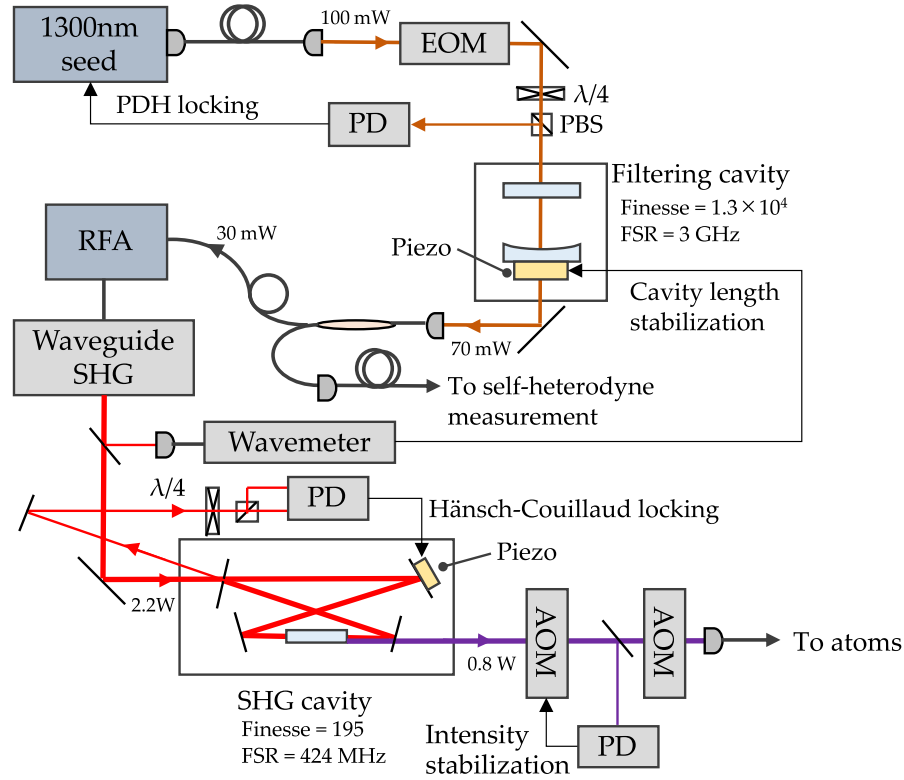


Figure 7.1 Overview of the UV laser system.

then passed through an electro-optic modulator (EOM) before being fed into a Fabry-Pérot cavity for frequency stabilization using the Pound-Drever-Hall (PDH) locking technique [246]. The cavity also serves as a filter to suppress the frequency noise to improve Rydberg excitation fidelity [72]. The cavity used in this setup has a finesse of 1.3×10^4 and a free spectral range (FSR) of 3 GHz, resulting in a 20 dB suppression in frequency noise of the transmitted light at an offset frequency of 1.5 MHz, as discussed later. The cavity length is tunable by piezoelectric transducers attached between the two mirror-holder tubes and locked to the desired frequency using a wavemeter (HighFinesse, WS8-10). This tunability enables us to lock the seed laser to various Rydberg resonances, optimizing the system for specific quantum operations. The transmission efficiency of the cavity is approximately 70 %, which is roughly consistent with the theoretical value based on the mirror specifications [247]. Notably, we have not observed any thermal drift of the cavity, which could potentially be caused by high intra-cavity power.

The transmitted light from the filtering cavity, with an output power of 70 mW, is coupled into a single-mode fiber and subsequently split using a 1×2 fiber splitter with a 50 : 50 ratio. One arm of the splitter, delivering approximately 30 mW of power, is directed to the RFA, while the other arm is employed in a self-heterodyne measurement for frequency noise evaluation. The 1300 nm light, amplified by the RFA, is then converted to 2.5 W of 650 nm light via the waveguide SHG module.

The converted 650 nm light is sent to the SHG cavity we developed, which consists of four

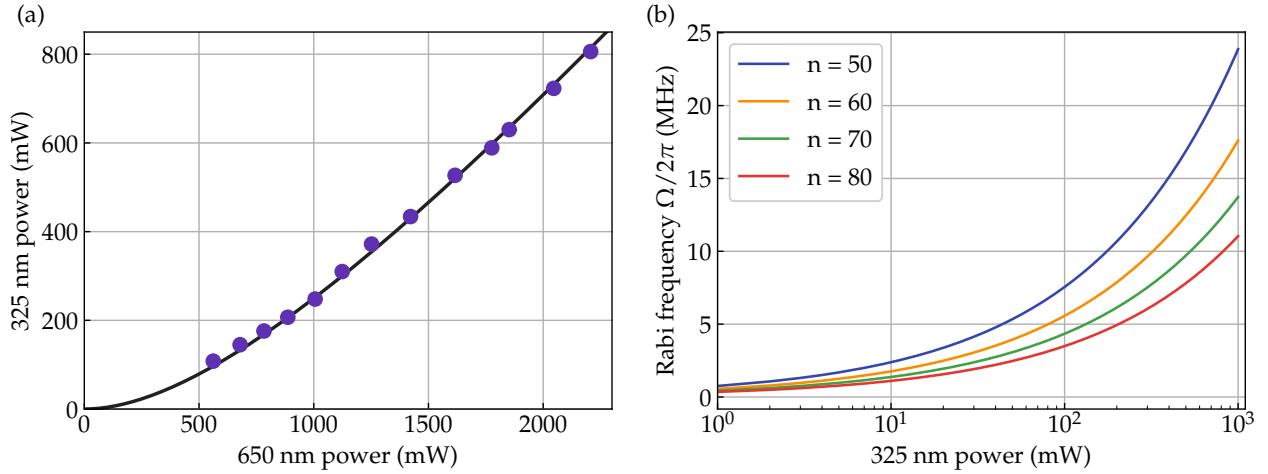


Figure 7.2 Output power of the UV laser system. (a) The 325 nm output power after the SHG cavity we developed. The maximum power exceeds 800 mW. The dashed line is the fitting curve with Eqs. (7.1) and (7.2) with the fitting parameter of l_{cav} . With the independently measured values of $E_{\text{nl}} = 9.4(1) \times 10^{-5} / \text{W}$, $T_1 = 2.1\%$, and $\eta = 82\%$, we estimate $l_{\text{cav}} = 1.08(8)\%$, consistent with the cavity finesse of 195. (c) Expected Rabi frequencies with various 325 nm beam powers. In the calculation, the reduced dipole matrix elements between $6p$ and ns states are taken from previous research [251], and a beam radius of $70 \mu\text{m}$ at the atom position is assumed.

mirrors (three mirrors with a reflectivity $R > 99.8\%$ and an input coupler with a transmittance $T_1 = 2.1\%$ at 650 nm, and all mirrors $R < 2\%$ at 325 nm) and a BBO crystal (3 mm (height) $\times$ 3 mm (width) $\times$ 10 mm (length), with cut angles of $\theta = 36.7^\circ$, $\phi = 90.0^\circ$). The nonlinear coefficient of the BBO crystal E_{nl} is $9.4(1) \times 10^{-5} / \text{W}$, measured by a single-pass SHG setup. The cavity finesse and FSR are 195 and 424 MHz, respectively. Figure 7.2 (a) shows the output power of the SHG cavity under Hänsch-Couillaud locking [248]. We obtain more than 800 mW output, which is sufficient for fast Rydberg excitation at MHz-order (figure 7.2 (b)). By fitting this result with the theoretical SHG output power P_{SHG} given as [249, 250]

$$P_{\text{SHG}} = E_{\text{nl}} P_{\text{ic}}^2, \quad (7.1)$$

with

$$P_{\text{ic}} = \frac{\eta T_1 P_{\text{in}}}{(1 - \sqrt{(1 - T_1)(1 - l_{\text{cav}})(1 - E_{\text{nl}} P_{\text{ic}})})^2}, \quad (7.2)$$

where P_{ic} is the intracavity power of 650 nm light, l_{cav} is the round-trip power loss in the cavity except for the input coupler, η is the coupling efficiency to the cavity, and P_{in} is the input power to the cavity, we determine the cavity loss as $l_{\text{cav}} = 1.08(8)\%$, with the independently measured value of $\eta = 82\%$. The estimated l_{cav} is consistent with the cavity finesse of 195.

The output from the SHG cavity passes through two acousto-optic modulators (AOMs), one for intensity stabilization [252] and the other for fast pulse shaping. Following this, the light is

delivered to the experimental chamber via a single-mode fiber and focused onto the atoms trapped in an optical tweezer array, with a beam radius of approximately $70\text{ }\mu\text{m}$. We note that the total power at the atom position is roughly 20 % of the SHG cavity output, which could be further increased by optimizing the spatial beam mode to better match the fiber mode.

7.1.2 Characteristics of the laser noises

It is crucial to characterize both frequency and intensity noises in the laser to estimate the fidelity of Rydberg excitation [79, 243]. Frequency noise around the Rydberg excitation Rabi frequency, preferably a few MHz and higher [79], negatively impacts the coherent dynamics during excitation [72] and should therefore be minimized. To achieve this, we employ a filtering cavity, as mentioned earlier. We measure the suppression factor for frequency noise in the MHz range by comparing the delayed self-heterodyne spectra of the seed laser with and without the filtering cavity. The spectra and suppression factor are presented in figure 7.3. We achieve more than 20 dB suppression in the MHz range, consistent with the filtering cavity linewidth (full width at half maximum) of 230 kHz.

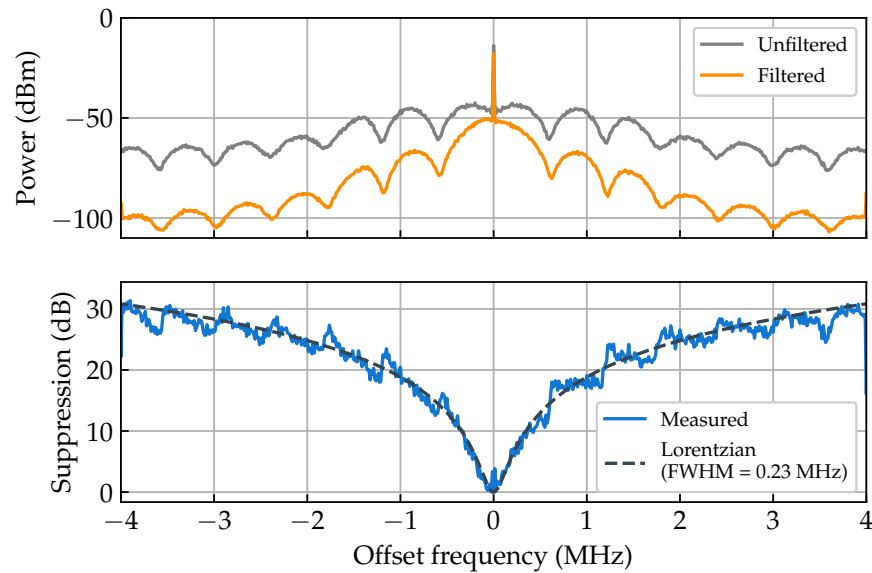


Figure 7.3 The performance of the filtering cavity. (Top) The spectra of the delayed self-heterodyne beat notes of the seed laser with and without the filtering cavity. The periodic dip structure corresponds to the delay line length of 350 m [243]. (Bottom) The suppression factor calculated from the ratio between the spectra. As a guide for the eye, a Lorentzian function with a full width at half maximum of 230 kHz, corresponding to the filtering cavity linewidth, is plotted with a dashed line.

Owing to the effective suppression of frequency noise in the megahertz range, the power spectral density (PSD) of the frequency noise at 1 MHz for the filtered seed laser is estimated to be $7 \times 10^2 \text{ Hz}^2/\text{Hz}$, as determined from the phase noise analysis of the delayed self-heterodyne signals

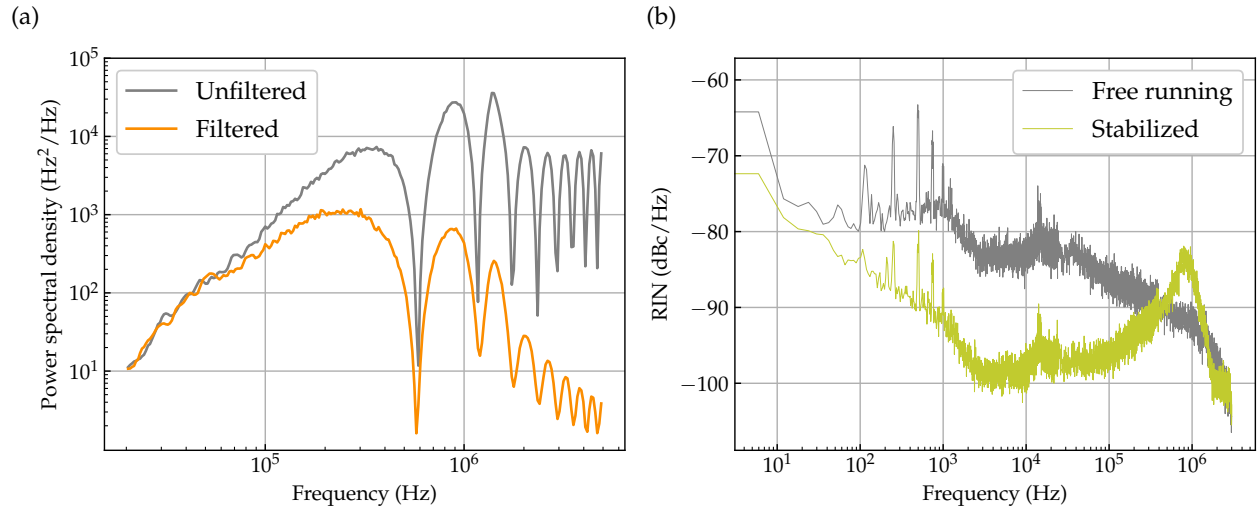


Figure 7.4 Characteristics of the laser. (a) The frequency noise PSDs of the unfiltered and filtered seed laser, evaluated from the delayed self-heterodyne signals. The frequency noise including the servo bump around 1 MHz of the unfiltered seed laser is effectively suppressed by the filtering cavity. The measurement sensitivity decreases at multiples of the Fourier frequency equivalent to the inverse of the delay time resulting in the periodic dip structure, as well as at low Fourier frequencies. Thus, the obtained PSD underestimates the actual frequency noise in these regions. (b) The relative intensity noise spectra of 325 nm light. The intensity noise below 100 kHz is effectively suppressed by the AOM-based stabilization. The increasing noise above 500 kHz is due to the limited bandwidth of the AOM, which can be addressed by installing an EOM-based feed-forward stabilization method [254].

using an oscilloscope (Keysight, MXR608A) (figure 7.4 (a)). According to the previous study [243], and assuming a Rabi frequency of 1 MHz, this corresponds to an error rate in Rydberg excitation of approximately 8×10^{-4} , which surpasses the threshold required for quantum error correction [253]. Further reduction in frequency noise could be achieved by replacing the seed laser with one that exhibits lower noise characteristics.

The relative intensity noise (RIN) spectra of 325 nm light, with and without AOM-based intensity stabilization, are shown in figure 7.4 (b). The RIN at the kHz range, which arises from the limited bandwidth of the SHG cavity locking and the frequency noise of the seed laser, is effectively suppressed by stabilization. However, stabilization becomes ineffective above 500 kHz due to the limited bandwidth of the AOM. As the Rabi frequency exceeds 1 MHz, the Rydberg excitation error is affected by the RIN integrated up to this range. As a result, the intensity stabilization increases the integrated RIN up to 3 MHz to approximately 5 %, slightly larger than that without intensity stabilization. This leads to a Rydberg excitation error of 0.15 % estimated from previous research [243]. To address this, enhancing the stabilization bandwidth by incorporating an EOM-based feed-forward

stabilization technique [254] would be effective.

7.2 Coherent excitation of Rydberg state in Ytterbium

We next employ the UV laser system for coherent Rydberg excitation of single ^{174}Yb atoms trapped in a 8×8 optical tweezer array. The site intervals are set to $10\mu\text{m}$ in this experiment. The time sequence of the Rydberg excitation is shown in figure 7.5 (a). The first step is the state transfer from the ground $^1\text{S}_0$ state to the $m_J = 0$ of the excited metastable $^3\text{P}_2$ state, as described above. For better signal-to-noise ratio in the Rydberg excitation experiments, the frequency of the 507 nm laser is chirped within a range of $\pm 6\text{kHz}$ around the resonance of the $^3\text{P}_2$ state for 8 ms as an adiabatic passage. Following this, we apply a 325 nm beam with vertical linear polarization for Rydberg excitation after irradiating a resonant 556 nm beam to blast out the remaining atoms in the ground state. Subsequently, the Rydberg atoms are quickly ionized by applying an electric field of 29 V/cm for $100\mu\text{s}$. We then repump back the remaining $^3\text{P}_2$ atoms to the $^1\text{S}_0$ state using 770 nm and 649 nm pulses for 5 ms. Finally, the atoms in the $^1\text{S}_0$ state are imaged to estimate the Rydberg excitation fraction.

We first perform the spectroscopy of the $(6s71s)^3\text{S}_1$ Rydberg state. In this measurement, the

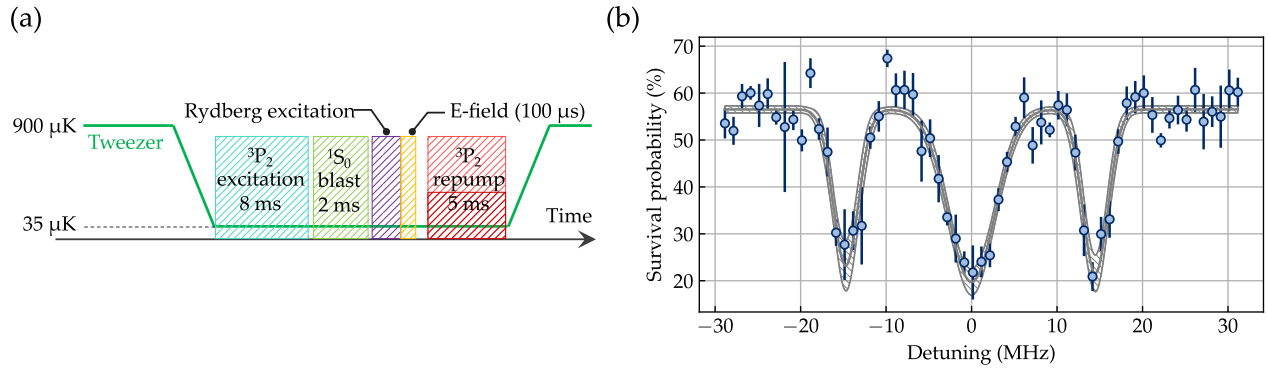


Figure 7.5 (a) Experimental sequence. After imaging the atoms, the trapping potential is reduced to $35\mu\text{K}$, and a 507 nm laser with a frequency chirp of $\pm 6\text{kHz}$ around the resonance is applied for 8 ms. Any remaining atoms in the ground state are then removed by a resonant 556 nm beam. Subsequently, a 325 nm beam is used for Rydberg excitation, followed by an application of an electric field pulse of 29 V/cm . Finally, the remaining atoms in the $^3\text{P}_2$ state are repumped back to the ground state and imaged after restoring the trapping potential to its original depth. The horizontal axis is not to scale. (b) Spectrum of the $(6s71s)^3\text{S}_1$ Rydberg state. The gray line is the fitting curve with a triple-peak Gaussian function. The shaded area represents 1σ confidence intervals. The three peaks correspond to the magnetic sublevels of the Rydberg state. The error bars represent the standard error of the mean.

325 nm beam power and pulse width are set to 15 mW and 1 ms, respectively. Note that the pulse width is much longer than the Rydberg state lifetime of several tens of microseconds, and the excitation fraction can be detected as atom loss even without applying the electric field. Figure 7.5 (b) shows a typical spectrum of the Rydberg state. The three dips represent the magnetic sublevels. The observed linewidth is broadened due to saturation, as the spectroscopy pulse duration is sufficiently long. Using the measured 1S_0 – 3P_2 frequency of 590.902 342 562(63) THz [255], the transition frequency of the 1S_0 to $(6s71s)^3S_1$ Rydberg state is found to be 1511.502 651 3(2) THz. The number in parenthesis represents the fitting uncertainty, and the accuracy is limited by the wavemeter, which has an accuracy of 20 MHz. The measured value is in good agreement with the value reported in the previous research [120].

Next, we perform coherent excitation of the $m_J = 0$ of $(6s71s)^3S_1$ Rydberg state. For this, we increase the 325 nm beam power to 70 mW and align the magnetic field direction to the beam polarization during excitation to maximize the Rabi frequency per excitation beam intensity. Figure 7.6 shows a Rabi oscillation between the 3P_2 and the Rydberg state obtained by scanning the pulse

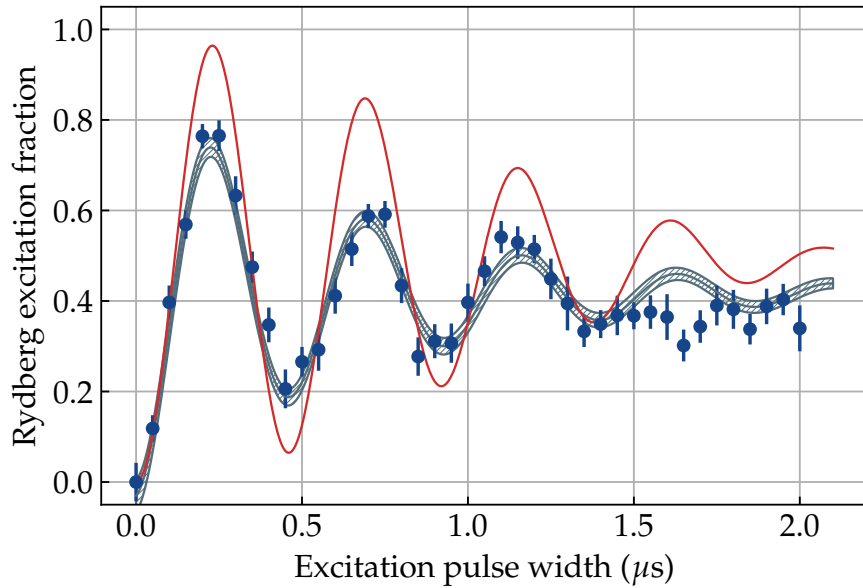


Figure 7.6 Coherent Rabi oscillation signal. The gray line represents the fit using a damped sine function, yielding a Rabi frequency of 2.13(3) MHz. The error bars represent the standard error of the mean. The red line shows a simulated dynamics model that includes measured shot-to-shot fluctuations in both intensity and frequency. The intensity fluctuation is characterized by $\sigma_1 = 8.7\%$, and the frequency fluctuation by $\sigma_2 = 0.3$ MHz. In the simulation, the relative intensity and detuning are sampled from normal distributions $\mathcal{N}(1, \sigma_1^2)$ and $\mathcal{N}(0, \sigma_2^2)$, respectively, where $\mathcal{N}(\mu, \sigma^2)$ is a normal distribution with a mean of μ and a standard deviation of σ . The averaged dynamics are computed over 10,000 iterations.

width from 0 μ s to 2 μ s. The Rabi frequency is $2\pi \times 2.13(3)$ MHz, which is as high as the previous demonstration of high-fidelity two-qubit gate for a neutral atom tweezer array [78]. We expect about a two-fold further improvement of the Rabi frequency by perfecting our beam shaping and alignments.

7.3 Discussion

The observed Rabi oscillation of the Rydberg excitation decays, primarily due to shot-to-shot fluctuations in both the intensity and frequency of the excitation laser. For the former, the intensity fluctuates by approximately 9 % around its mean value, as measured by monitoring the pulse area. This fluctuation may result from variations in pulse shape, as the RF frequency is applied to the AOM only during UV laser irradiation to protect the fiber from high-power UV light. As a result, the AOM does not fully thermalize before pulse generation, causing shot-to-shot fluctuations in the pulse area. To mitigate this issue, we can employ a sample-and-hold technique [72, 74] combined with an EOM-based intensity stabilization technique [254]. For the latter, we estimate that the frequency fluctuates by at least 0.3 MHz, even though the laser is stabilized using a filtering cavity. This estimation is based on the standard deviation of 0.3 MHz in the readings from the wavemeter, which we use for cavity stabilization. This limitation is due to the restricted frequency resolution of the wavemeter, which is used to lock the length of the filtering cavity. This issue can be addressed by locking the seed laser to a stable frequency reference; for example, an optical frequency comb stabilized by an ultranarrow laser [256]. We simulate the system dynamics while accounting for the aforementioned fluctuations, and the results are plotted as the red line in figure 7.6. Although the simulated excitation fraction does not perfectly match the experimental data – indicating the presence of other potential sources of decoherence such as the atom temperature, differential light shift between the 3P_2 and the Rydberg states, electric field noises, and so on – the decay of the oscillation qualitatively agrees with the observed experimental behavior.

7.4 Conclusion

In summary, we have developed a high-power UV laser system for Rydberg excitation from the 3P_2 state of Yb atoms. By employing two-stage frequency doubling with a RFA and a home-built SHG cavity, we successfully achieved an output power exceeding 800 mW at a wavelength of 325 nm. To mitigate frequency noise in the MHz range, which can adversely affect Rydberg excitation with an MHz-order Rabi frequency, we incorporated a filtering cavity with a linewidth of 230 kHz. The suppression factor for the seed laser’s frequency noise was measured to be 20 dB in the MHz range, suggesting the potential for high-fidelity Rydberg excitation. Additionally, we characterized the intensity noise at 325 nm and found that AOM-based intensity stabilization effectively suppresses up to 500 kHz. Moving forward, the stabilization bandwidth will be further enhanced

by integrating an EOM-based stabilization method [254].

As a first step to the Rydberg excitation, we demonstrated coherent manipulation on the 1S_0 – 3P_2 transition of ^{174}Yb , which also serves as the optical clock qubit with the capacity of non-destructive qubit readout. We observed Rabi oscillations between the ground state and the $m_J = 0$ state of the 3P_2 with a Rabi frequency of $2\pi \times 9.5(2)$ kHz. Furthermore, we evaluated the coherence time of the optical qubit to be 0.55(7) ms using the Ramsey sequence, providing the starting point for quantum computation utilizing this qubit.

Using the laser system we developed, we conducted spectroscopy and coherent excitation of the $(6s71s)^3S_1$ Rydberg state. The measured resonance frequency was in good agreement with the value reported in previous research [120]. By increasing the laser power, we successfully observed Rabi oscillations between the $m_J = 0$ states of the 3P_2 and the Rydberg state, with a frequency of $2\pi \times 2.13(3)$ MHz. The decay of the Rabi oscillation was qualitatively attributed to shot-to-shot fluctuations in both the frequency and intensity of the laser. These issues can be readily addressed using existing experimental techniques.

The high-power UV laser system with frequency noise suppression presented in this work serves as a foundational step toward fast and high-fidelity Rydberg excitation. By incorporating further laser noise suppression techniques, this system has the potential to significantly advance quantum simulation and computing in neutral atom arrays.

Chapter 8

Conclusion and outlook

8.1 Conclusion

This thesis has presented the construction of a hybrid atom tweezer array of nuclear spin and optical clock qubits based on dual-isotope ytterbium atoms, as well as the development of a high-power UV laser system for Rydberg excitation towards realizing quantum error correction. We briefly review the achievements of this thesis.

- *Coherent manipulations of nuclear spin and optical clock qubits in ytterbium atoms* (chapter 5)

We have demonstrated efficient initialization and coherent control of nuclear spin qubits in a ^{171}Yb atom array. The nuclear spin qubits exhibit second-order long coherence times and megahertz-order fast qubit controllability, highlighting their potential as promising data qubits for quantum computing.

Furthermore, we performed narrow-line spectroscopy and coherent manipulations of optical clock qubits in a ^{174}Yb atom array. Our results showed that the 532 nm tweezer satisfies the near-magic condition for the optical clock qubits, enabling high-fidelity state manipulations. Although the Rabi oscillations and the Ramsey signal show the limited coherence times of the optical clock qubits, these findings provide a solid foundation for the future utilization of optical clock qubits as ancilla qubits.

- *Hybrid atom tweezer array of nuclear spin and optical clock qubits* (chapter 6) [187]

We successfully constructed a hybrid atom tweezer array composed of ^{171}Yb and ^{174}Yb atoms. This system demonstrated atom rearrangement within the dual-Yb system and tunable loading probabilities for each isotope, highlighting its potential for molecule tweezer experiments, as well as neutral atom-based quantum computing.

Our investigation revealed that the optical clock qubit readout has a negligible impact on the coherence of nuclear spin qubits, representing a critical milestone for the implementation of mid-circuit measurements in quantum error correction protocols. Furthermore, we observed minimal coherence degradation from 556 nm imaging of the optical clock qubits, suggesting that further crosstalk suppression can be achieved through optimized 556 nm imaging techniques.

- *Development of a high-power ultraviolet laser system and observation of fast coherent Rydberg excitation of ytterbium* (chapter 7) [245]

We developed a high-power UV laser system tailored for Rydberg excitation of ytterbium

atoms. The system comprises a 1300 nm seed laser, a Raman fiber amplifier, and two frequency doubling systems. We achieved a 325 nm laser with a maximum output power of 0.8 W, enabling rapid Rydberg excitation. By incorporating a filtering cavity, the laser phase noise was suppressed by 20 dB at an offset frequency of 1.5 MHz, which is crucial for achieving high-fidelity Rydberg excitation. Using this system, we observed fast coherent Rydberg excitation of ytterbium atoms with a Rabi frequency of 2.13(3) MHz, establishing a key step towards high-fidelity two-qubit gate operations.

8.2 Outlook

Below, we outline several future directions for the hybrid atom tweezer array experiments.

- **Utilization of the 3P_0 state for the optical clock qubit**

The 3P_0 state in ^{174}Yb atoms is a metastable excited state and serves as a promising candidate for the optical clock qubit. Owing to the absence of electronic total angular momentum J , this state is immune to vector and tensor light shifts. By employing a magic wavelength laser for the optical tweezer array, the 3P_0 state is expected to exhibit extended coherence times [51, 52] and enable high-fidelity state manipulations [81, 119].

- **759 nm optical tweezer array**

This wavelength serves as a magic wavelength for the 3P_0 state [232, 257, 258], thereby enhancing the coherence and state manipulation fidelity of the 3P_0 state. Additionally, by tuning the angle between the tweezer polarization and the quantization axis, it satisfies a magic condition for the 3P_1 state in both ^{171}Yb [119] and ^{174}Yb [135] atoms, enabling efficient cooling and high fidelity imaging of single atoms in the tweezer array.

- **Exploring the inter-isotope Rydberg interactions and implementation of two-qubit gates**

Although the intra-isotope Rydberg interactions of ^{171}Yb and ^{174}Yb have been extensively studied and utilized for two-qubit gates, the inter-isotope Rydberg interactions remain unexplored. By investigating these interactions with the support of theoretical calculations informed by detailed Rydberg spectroscopy research of ytterbium [78], we aim to realize two-qubit gates between nuclear spin and optical clock qubits, which are essential for quantum error correction protocols.

Additionally, the potential existence of a Förster resonance in the inter-isotope Rydberg interactions could enhance the interaction range, offering a novel platform for quantum simulation. This would leverage the imbalance between inter- and intra-isotope Rydberg interactions in a dual-isotope atom array, enabling unique opportunities for exploring complex many-body quantum systems [259–262].

- **Single heteronuclear molecule creation in the tweezer array**

The tunability of dual occupancy of ^{171}Yb and ^{174}Yb atoms in the tweezer array offers a unique platform for the creation of single heteronuclear molecules. By employing techniques such as stimulated Raman adiabatic passage or two-photon photoassociation, we aim to produce single ground-state heteronuclear molecules within the tweezer array.

This development opens new avenues for encoding quantum information in the rich internal states of molecules, providing access to a diverse set of quantum states for computation [222]. Furthermore, it paves the way for novel quantum error correction protocols utilizing molecular qubits, leveraging their inherent rich internal structure [223, 224, 263].

- **Implementation of quantum error correction protocols**

Towards the ultimate goal of fault-tolerant quantum computing, we aim to implement quantum error correction protocols using the hybrid atom tweezer array. The demonstrated low-crosstalk readout of optical clock qubits, while preserving the coherence of nuclear spin qubits, positions us well to realize mid-circuit measurements essential for quantum error correction.

By integrating the high-power UV laser system for high-fidelity Rydberg excitation and leveraging insights from inter-isotope Rydberg interaction studies, we aim to achieve high-fidelity two-qubit gates between data and ancilla qubits. These advancements will enable robust error syndrome measurements and serve as a critical step towards the realization of fault-tolerant quantum computing.

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