Single-Photon Rydberg Excitation and Trap-Loss Spectroscopy of Cold Cesium Atoms in a Magneto-Optical Trap by Using of a 319-nm Ultraviolet Laser System

Jiandong Bai, Shuo Liu, Jieying Wang, Jun He, and Junmin Wang

Abstract—We demonstrate the single-photon Rydberg excitation of cesium atoms in a magneto-optical trap (MOT). We excite atoms directly from $6S_{1/2}$ ground state to $nP_{3/2}$ ($n = 70–100$) Rydberg state with a narrow-linewidth 319-nm ultra-violet laser. The detection of Rydberg states is performed by monitoring the reduction of fluorescence signal of the MOT as partial population of $6S_{1/2}$ ($F = 4$) ground state are transferred to Rydberg state. We clearly observe Autler-Townes doublet in the trap-loss spectra due to the cooling lights. Utilizing the large electric polarizability of Rydberg atoms, we observe Stark splitting in the Autler-Townes doublet induced by background DC electric fields. We investigate the dependence of Stark shift on electric fields by theoretical analysis, and then infer the DC electric field from the measured Stark splitting. We find that there is a 44.8(4) mV/cm DC electric field in the vicinity of the cold ensemble. It indicates that high-lying Rydberg atoms can be used as sensors for DC electric fields.

Index Terms—Single-photon Rydberg excitation, magneto-optical trap, trap-loss spectra, Stark effect, electric field sensing.

I. INTRODUCTION

RYDBERG atoms with principal quantum numbers $n > 10$ have many exotic characters in terms of their unique structure. This includes large polarizability and microwave-transition dipole moments and lower field ionization threshold, which is proportional to $n^7$, $n^2$, and $n^{-4}$ [1], respectively, giving rise to an extreme sensitivity to the electric field environment. It makes Rydberg atoms promising for applications such as quantum information [2] and quantum metrology [3]–[6]. Due to the invariance of atomic properties, the atom-based field measurement has obvious advantages over other methods because it is calibration-free. Moreover, atom-based metrology has made significant progress in resolution, accuracy, and reproducibility, making it expanded into electric-field sensing. However, most experiments diagnosed DC electric field by the field ionization of high Rydberg states [7]–[11]. While this detection method has high efficiency and discrimination, the detected atoms after field ionization has been destroyed and cannot be reused. Moreover, this detection system is more complex, and it is difficult to be miniaturization due to larger space usage. So it is necessary to perform the nondestructive detection of Rydberg states for the involved quantum information applications.

For purely optical detection of Rydberg states in a cold atomic sample, commonly employed scheme is the cascaded two-photon excitation [12], [13]. Two-photon excitation can avoid ultraviolet (UV) wavelengths making it easier to implement technically. However, it has following drawbacks: atomic decoherence due to the photon scattering from the lower transition via intermediate state. If we utilize off-resonant cascaded two-photon excitation to reduce the decoherence effect, it will introduce the light shift of the involved ground and Rydberg states, leading to a lower Rydberg excitation efficiency. Alternatively, direct excitation from the ground state to the desired highly-excited Rydberg state can avoid these drawbacks, but it demands narrow-linewidth tunable UV laser. Thanks to the development of the efficient laser frequency conversion technology and the fiber lasers as well as fiber amplifiers, high-power narrow-linewidth continuous-wave UV laser has been implemented [14]–[17]. Recently, Hankin et al. performed the single-photon Rydberg excitation using a continuous-wave UV laser, and demonstrated the coherent control of two single atoms in the $84P_{3/2}$ states [15], [18]. However, compared with the cold atomic ensemble, the signal-to-noise ratio of the spectrum based on the single atoms is lower.

In this paper, we demonstrate the single-photon Rydberg excitation of cesium cold atomic ensemble with a purely optical detection method, instead of ionization detection. We excite atoms directly from $6S_{1/2}$ ground state to $nP_{3/2}$ ($n = 70–100$) Rydberg state with a narrow-linewidth 319 nm UV laser. The
diameter of transitions. 

III. RESULTS AND DISCUSSIONS

A. Trap-Loss Spectra of Cesium MOT for Single-Photon Rydberg Transition

To study the Rydberg atomic characters and the interaction between them, it is of great importance to perform high-resolution spectroscopy. Generally, the continuous-scan and step-scan techniques are used to perform the spectroscopic measurement. The continuous-scan technique has some limitations: the unstable frequency will broaden the observed spectra, moreover, the continuous loading of the MOT during Rydberg excitation will result in complex population dynamics. This distorts the shape of the spectra. Compared to this technique, the step-scan technique has better signal-to-noise ratio and much narrower linewidth. The spectral lineshapes are highly symmetrical. Consequently, we employ the step-scan technique to perform Rydberg spectroscopy in the following way. In the process, the 319 nm UV beam is stabilized to a high-finesse ultra-low-expansion (ULE) optical cavity inside an ultra-high vacuum chamber using the electronic sideband locking method [22]. The relative frequency deviation of the UV beam is estimated to be less than 15 kHz. The high-stability narrow-linewidth UV beam can be continuously tuned over a range of 4 GHz while maintaining the lock.

Employing the frequency-stabilized tunable UV laser system, we perform the trap-loss spectrum of Rydberg state 71P3/2, as shown in Fig. 2(a). Here, the UV beam’s power is fixed at 1 mW. Because the cooling beams’ Rabi frequency is greater than natural linewidth of the 6P3/2 state, the existence of the strong cooling beams results in Autler-Townes (AT) splitting in the ground state [6S1/2(F = 4)] and the excited state [6P3/2(F′ = 5)]. While a weak 319 nm UV beam is scanned across the transition of 6S1/2(F = 4) → 71P3/2, the spectrum exhibits two loss peaks. To verify this, we measure the separation of AT doublet for 71P3/2 state as a function of the UV coupling intensity.
of 6S₁/₂ state in the center of the MOT along the z direction (Δf₂ ≈ \mu n g₁(S₁/₂) \frac{\partial B_z}{\partial z} \Delta z \approx 4.6 \text{ MHz} with g₁(S₁/₂) = 2), and Stark broadening of 2.8 MHz from the background electric fields which will be discussed below. In the experiment, because the intensity of the 319-nm UV laser is weak, only some of the ground-state atoms in the MOT are excited to Rydberg state, resulting in a relatively low number density of Rydberg atoms. So the van der Waals interactions between Rydberg atoms have little effect on the spectral linewidth. Therefore, the other broadening of \sim 1.7 \text{ MHz} may contribute from the collisional broadening among the atoms.

B. The Influence of Background DC Electric Field

The orbital radius of Rydberg atoms scale to \( n^2 \) [1], and the spacings between adjacent states decrease sharply with increasing \( n \). This implies exaggerated sensitivity to DC electric field due to its large electric polarizability. We calculate that the electric polarizability of 71P₃/2 state is on the order of 10^{11} times larger than that of 6S₁/₂ ground state. We exploit the large electric polarizability to assess the influence of the electric field environment in the vicinity of the MOT on the Rydberg spectra of nP₃/2 states.

We first model Stark splitting of Rydberg state by including the perturbing effects of electric field. The total Hamiltonian for an atom in a static electric field is written as:

\[
H_{\text{total}} = H_{\text{atom}} + H_{\text{Stark}}
\]

Here, \( H_{\text{atom}} \) is the Hamiltonian of bare atom and its matrix elements can be calculated using quantum defect theory [25]. The final term is the Stark interaction which is given by

\[
H_{\text{Stark}} = -\mu_d E.
\]

Here, \( \mu_d \) and \( E \) are the electric-dipole moment and a DC electric field, respectively. The eigenstates \( |\chi\rangle \) of \( H_{\text{total}} \) in a static electric field \( E \) are a linear superposition of the unperturbed atomic \( |n, s, l, j\rangle \) states:

\[
|\chi(E)\rangle = \sum_{n,l,j} c_{n,l,j}(E) |n, s, l, j\rangle
\]

Where \( c_{n,l,j} \) are the mixing coefficients. The Stark interaction does not mix states of different total angular momentum projection number \( |m_j\rangle \) and different total spin \( |m_j\rangle \), so we can calculate a separate Stark map for different \( |m_j\rangle \). The energy levels of different Rydberg states are much closer at higher \( n \) (energy spacings scale as \( n^{-3} \)), so the Stark map is fairly complex in high electric fields. However, in low electric fields, Stark shift of the Rydberg state \( \Delta \varepsilon \) can be given by second order perturbation theory as [27]:

\[
\Delta \varepsilon = -\frac{1}{2} \alpha E^2
\]

The total polarizability \( \alpha \) is given by:

\[
\alpha = \alpha_0 + \alpha_2 \frac{3m_j^2 - j(j+1)}{j(2j-1)}
\]

Here, the energy shift is decided by a scalar polarizability \( \alpha_0 \), and the splitting of the energy level is given by a tensor polarizability \( \alpha_2 \) for different \( m_j \) values. For nP₃/2 states, the total
polarizabilities are expressed as $\alpha = \alpha_0 - \alpha_2$ for $|m_j| = 1/2$ and $\alpha = \alpha_0 + \alpha_2$ for $|m_j| = 3/2$, respectively.

To accurately calculate the total polarizabilities, the scalar $\alpha_0$ and tensor $\alpha_2$ polarizabilities are evaluated as the sum over intermediate $|n', s', l', j'\rangle$ states allowed by the electric-dipole selection rules:

$$\alpha_0 = \frac{2}{3(2j + 1)} \sum_{n', l', j'} \left\langle n', s', l', j' \parallel D \parallel n, s, l, j \right\rangle^2$$

$$\alpha_2 = \sqrt{\frac{4(2j - 1)}{3(j + 1)(2j + 1)(2j + 3)}} \sum_{n', l', j'} (-1)^{j + j'} \left\langle n', s', l', j' \parallel D \parallel n, s, l, j \right\rangle^2 \times \{j \ 1 \ j' \} \left\langle n', s', l', j' \parallel E \parallel n, l, j \right\rangle^2 - E_{n, l, j}$$

Where $\left\langle n', s', l', j' \parallel D \parallel n, s, l, j \right\rangle$ is the electric-dipole matrix elements. $\{...\}$ represents the Wigner six-j symbol. $E_{n, l, j}$ and $E_{n', l', j'}$ are zero-field energies of the $|n, s, l, j\rangle$ state and the dipole allowed intermediate $|n', s', l', j'\rangle$ state, respectively.

For the calculation of the energy shifts in the range of the given electric fields, we diagonalize (1) for a large enough set of states to ensure convergence of the eigenvectors and eigenvalues of $H_{\text{total}}$. We calculate states up to $l = 10$, and a range of $n = \pm 5$. The simulated energy shifts for the $|m_j| = 1/2, 3/2$ components of cesium $nP_{3/2}$ state are shown in Fig. 3(a), which are consistent with the calculation results in the ref. [15], [28], [29]. Considering the contribution of tensor polarizability to the total polarizability, the $nP_{3/2}$ state is split into two components in an external electric field, corresponding to $|m_j| = 1/2$ and $|m_j| = 3/2$. Because the tensor polarizability is calculated to be negative, the $|m_j| = 3/2$ state has smaller polarizability than $|m_j| = 1/2$ from (4). That is, the Stark shift of $|m_j| = 1/2$ is larger than that of $|m_j| = 3/2$ in the same electric field. And the Stark shift becomes more severe with increasing $n$. Moreover, the separation between the $|m_j| = 1/2$ and $|m_j| = 3/2$ components also increases with principal quantum number and DC electric fields [Fig. 3(b)].

To determine the size of background DC electric field, we characterize the spectra of Rydberg states with different principal quantum numbers and compare the results with the theoretical prediction. The measured Rydberg spectra for $nP_{3/2}$ ($n = 84, 90, 95$ and 100) states are shown in Fig. 4. In order to determine the position of these peaks more accurately, a multi-peak Voigt function (red solid line) is exploited to fit the experimental data (black squares). We use a continuous-wave UV laser in a range of 100-MHz laser frequency to implement the Rydberg excitation experiment. Although we expect a single peak without any external perturbations, the observed spectrum is constituted of two nondegenerate sub-peaks (marked as 1, 2 or 3, 4) in any one of AT doublet. We use three sets of Helmholtz coils to compensate the geomagnetic field in the vicinity of the atomic cloud in three dimensions. Therefore, the degeneracy breaking of Rydberg spectra in Fig. 4 is due to the background DC electric field shifts the resonances by Stark effect.

Fig. 4 shows that the spectrum shifts red and Stark splitting increases with $n$. Because the separation of AT doublet is relatively small, the peak-3 gets closer to peak-2 with increasing $n$, so that the two peaks gradually overlap in higher $n$. Compared the measured Stark splitting (the spacings between the peak-1 and peak-2) with the theoretical value in Fig. 3(b), we can estimate the size of background DC electric field. The DC electric field in the vicinity of the atoms is 44.8 mV/cm with an uncertainty of $\pm 0.4$ mV/cm, which is an average for five Rydberg states, as shown in Fig. 5. The error is mainly caused by the fluctuation of background DC electric field and the intensity fluctuation of the UV beam. The experimental data are in excellent agreement with the theory.

To characterize the sensitivity of the whole system to DC electric field, Rayleigh criterion is commonly used to define the minimum resolution of the spectrum. The criterion states that when the center of one peak falls on the edge of the other, the two peaks can just be distinguished. For $100P_{3/2}$ Rydberg state, the spectral linewidth is about 10.7 MHz for $|m_j| = 1/2$ or 3/2, leading to a minimum detectable field strength of $\sim 43$ mV/cm. However, when the two peaks of Stark map overlap, the spectral linewidth will be squeezed with the reduced electric field. Thus, the measurement sensitivity of electric field is mainly limited to the frequency and intensity noises of the UV beam. We estimate
Fig. 4. The trap-loss spectra of cesium \(nP_{3/2}\) \((n = 84, 90, 95\) and 100\) Rydberg states. The spectrum shifts red and Stark splitting increases with \(n\). Because the separation of AT doublet is relatively small, the peak-3 is much closer to peak-2 with increasing \(n\), so that the two peaks gradually overlap in higher principal quantum number \((n = 95\) and 100\).

Fig. 5. Sensing the background DC electric field using highly-excited cesium \(nP_{3/2}\) \((n = 71, 84, 90, 95\) and 100\) Rydberg states. The red line represents the mean value for experimental data (blue diamond cube). From the measured Stark splitting, the background DC electric field is deduced to be 44.8 mV/cm, with an uncertainty of ±0.4 mV/cm. The error is mainly due to the fluctuation of background DC electric field and the intensity fluctuation of the UV beam.

that the sensitivity of DC electric field can be up to \(\sim 1\) mV/cm for 100\(P_{3/2}\) Rydberg state. For the smaller DC electric field measurement, Schramm et al. [10] reduced electric field to \(\sim 0.1\) mV/cm by exciting argon atoms to \(n = 400\) Rydberg state. For \(n = 400\), its polarizability is on the order of 10\(^4\) times larger than that of \(n = 100\) Rydberg state. Therefore, we can also excite the atoms to higher Rydberg states to enhance the atomic sensitivity to DC electric fields. The measurement sensitivity of DC electric field will be better than \(\sim 7\) \(\mu\) V/cm for Cs Rydberg state with \(n = 400\). It indicates that Rydberg atoms have promising for applications in sensing unknown electric fields and can provide atom-based traceable standard for the electrometry.

To further enhance the sensitivity of electric field measurements, we will try our best to improve the signal-to-noise ratio of the spectra and suppress spectral broadening of Rydberg excitation signal which is essential for precision spectroscopy. The following measures can be taken to improve measurement accuracy: firstly, the ground-state atoms are excited by using of the short-pulsed UV light after turning off the gradient magnetic field and the trapping beams. In this way, it can not only eliminate the dressed splitting caused by the strong cooling lights, but also suppress the effect of Zeeman broadening on the spectra. Secondly, a fast feedback loop can be constructed to improve the UV frequency stability and narrow the laser linewidth [30]. In addition, we can use three pairs of electrode plates to compensate the background stray electric field in the vicinity of the atoms for future work. Then we apply a DC electric field to one pair of electrodes to accurately assess the minimum measurable value of electric field.

IV. CONCLUSION

In summary, we have demonstrated the purely all-optical detection of cesium Rydberg atoms in a MOT via the single-photon excitation, instead of ionization detection. Utilizing a home-made narrow-linewidth 319 nm UV laser system, we excite cesium atoms directly from \(6S_{1/2} (F = 4)\) ground state to \(nP_{3/2}\) \((n = 70–100)\) Rydberg state. We clearly observe Autler-Townes splitting in trap-loss spectra due to the cooling beams. Moreover, we see the distinct Stark splitting of Rydberg spectra in high principal quantum number due to the influence of background DC electric field. We investigate the dependence of Stark shift on DC electric fields, and then infer the background DC electric field from the measured Stark splitting. We find that there is a 44.8(4) mV/cm DC electric field at the location of cold ensemble. Rydberg atoms have very large electric polarizibility, therefore DC electric field induced Stark splitting of Rydberg spectra can be employed to sense the unknown DC electric field, while cold Rydberg Cs atoms serve as a sensor. Consequently, the precision spectroscopy of high-lying Rydberg atoms plays an important role in quantum metrology. Moderate improvements in the system and excitation of higher Rydberg states are expected to enable more precise DC electric field measurement.

REFERENCES

Jiandong Bai received the B.S. degree in physics from Shanxi University, Taiyuan, China, in 2014. He is currently working toward the Ph.D. degree in optics with the Institute of Opto-Electronics, Shanxi University. His main research interests include cold atomic physics, all-solid-state laser technology, and nonlinear optics.

Shuo Liu received the B.S. degree in physics from Luliang University, Shanxi, China, in 2017. He is currently working toward the M.S. degree in optics with the Institute of Opto-Electronics, Shanxi University, Taiyuan, China. His main research interests include cold atomic physics and nonlinear optics.

Jiaying Wang received the B.S. degree in physics from Shanxi University, Taiyuan, China, in 2012. She received the Ph.D. degree in optics from Shanxi University, China, in 2018. Her main research interests include cold atomic physics, all-solid-state laser technology, and nonlinear optics.

Jun He was born in 1982. He received the Ph.D. degree in physics from Shanxi University, Taiyuan, China, in 2011. He is currently an Associate Professor with the Institute of Opto-Electronics, Shanxi University. His research interests include cold atomic physics, quantum optics, and nonlinear optics.

Junnin Wang was born in 1967. He received the Ph.D. degree in physics from Shanxi University, Taiyuan, China, in 1999. From December 1999 to July 2001, he was a Visiting Scholar in the ultra-cold atom research group of Kastler-Brossel Laboratory, Department of Physics, Ecole Normale Superieure (ENS), Paris, France. From September 2005 to January 2006, he was a Visiting Scholar in the ultra-cold atom research group of Kastler-Brossel Laboratory, Department of Physics, Ecole Normale Superieure (ENS), Paris, France.

Jiadong Bai was born in 1982. He received the Ph.D. degree in physics from Shanxi University, Taiyuan, China, in 2014. He is currently working toward the Ph.D. degree in optics with the Institute of Opto-Electronics, Shanxi University. His main research interests include cold atomic physics, all-solid-state laser technology, and nonlinear optics.