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Highly sensitive detection of Rydberg atoms with fluorescence loss spectrum in cold atoms*

Xuerong Shi(师雪荣)[1,2], Hao Zhang(张好)[1,2,†], Mingyong Jing(景明勇)[1,2], Linjie Zhang(张临杰)[1,2], Liantuan Xiao(肖连团)[1,2], and Suotang Jia(贾锁堂)[1,2]

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Fluorescence loss spectrum for detecting cold Rydberg atoms with high sensitivity has been obtained based on lock-in detection of fluorescence of $6P_{3/2}$ state when cooling lasers of the magneto-optical trap are modulated. The experiment results show that the signal to noise ratio has been improved by 32.64 dB when the modulation depth (converted to laser frequency) and frequency are optimized to 4 MHz and 6 kHz, respectively. This technique enables us to perform a highly sensitive non-destructive detection of Rydberg atoms.

Keywords: fluorescence loss spectrum, Rydberg atoms, signal to noise ratio

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1. Introduction

In recent years, significant progress in the research field of ultra-cold atoms has taken place, such as coupling ultra-cold atoms with ions systems.[1–3] Besides, ultra-cold atomic ensembles also provide an ideal platform for investigating the behavior of interacting many-body quantum systems due to their large degree of control and tunability.[4–6] Rydberg atoms excited in cold atoms have attracted lots of interests of scientists, and much of the recent breakthrough in the field of quantum information has been made by taking advantage of strong interactions between Rydberg atoms,[7–9] quantum nonlinear optics,[10–13] quantum information processing,[14–16] as well as Rydberg-dressed atoms based on laser cooling and trapping.[17,18]

The decay rates of Rydberg states are quite small according to their long lifetimes which are proportional to n^3,[19] and multiple decay channels and non-radiative decay make the radiation of Rydberg states a much complicated process. Therefore, the fluorescence of a high lying Rydberg state is too weak to be observed directly in this case. There are several methods to detect Rydberg atoms, such as ions detection, in which the state selective field ionization (SFI) techniques are generally used to detect specific Rydberg states, however, the Rydberg atoms are ionized in this case and this detection method is destructive.[20–24] Electromagnetically induced transparency (EIT) has been successfully applied to provide a non-destructive method for Rydberg states detection.[25–28] Besides absorption spectroscopy and imaging,[29] the fluorescence loss spectrum is also used for recording Rydberg excitation spectra in a magneto-optic trap spanning a series of principal quantum numbers.[30,31] However, in those techniques, the signal-to-noise ratio (SNR) shows large room to improve. In this study, we use a method to directly measure the fluorescence of the excited states of the cooled atoms in the magneto-optic trap, when the coupling laser is tuned at the resonance with a certain Rydberg level we could observe a sudden drop of the magneto-optical trap (MOT) fluorescence signal, which can be utilized for the detection of the Rydberg atoms, and in order to improve the detection sensitivity, a frequency modulation is applied on the cooling lasers. The precision laser spectroscopy measurements of Rydberg states are made effectively using pure optical detection with a vapor cell sample based on frequency modulation as described in Ref. [32]. High-resolution photoassociation spectroscopy by using the modulation technique is also carried out in a cesium atomic magneto-optical trap.[33,34]

In this paper, we demonstrate a highly sensitive fluorescence loss spectroscopy based on ultra-cold cesium atoms in a magneto-optical trap by adding a frequency modulation to the cooling lasers. The modulation frequencies and depth have been optimized that lead to the improvement of SNR at least 30 dB compared to the unmodulated spectrum. The modulated fluorescence loss detection technique provides a simpler, versatile, and non-destructive method for the detection of Rydberg atoms.

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2. Experimental setup

Cold cesium atoms are prepared in a magneto-optical trap with the temperature about 200 μK. A cuboid shaped quartz vacuum chamber with a background pressure of about 1.5×10^{-10} Torr is connected with a cesium reservoir via a metal valve. The quadrupole magnetic field for MOT is generated by a pair of anti-Helmholtz coils together with magnetic fields compensations, and the magnetic field gradient of the quadrupole field along the symmetry axis of the coils is about 10 G/cm. The cooling beams are provided by a commercial diode laser (DL pro, Toptica) of which the linewidth is smaller than 1 MHz and the frequency is stabilized to transition $6S_{1/2}(F=4)\rightarrow 6P_{3/2}$(crossover of $F'=3$ and 5) using a saturated absorption spectroscopy, and then shifted with an acoustic optical modulator (AOM) to be red detuned from the $6S_{1/2}(F=4)\rightarrow 6P_{3/2}(F'=5)$ atomic transition, and the detunning is about twice of the natural linewidth of state $6P_{3/2}$. The power of the cooling beams is about 1.3 mW each and the diameter is about 5 mm. To prevent the accumulation of atoms on state $6S_{1/2}(F=3)$ decaying from $6P_{3/2}(F'=4)$ level, another repumping laser is necessary which is provided by an external-cavity diode laser (New Focus, model 6305), and the frequency is locked to the transition of $6S_{1/2}(F=3)\rightarrow 6P_{3/2}(F'=4)$ by using a polarized absorption spectrum. In our experiments, the first excitation laser which drives the atoms from ground state $6S_{1/2}$ to excited state $6P_{3/2}$ is performed by the cooling lasers, and another 510 nm laser provided by a frequency-doubled laser (TA-SHG pro, Toptica) couples the transition from the excited state to $47D_{5/2}$ Rydberg state. The frequency of the upper transition laser is scanned around certain Rydberg state to record the fluorescence spectrum, and the diameter is about 0.4 mm, which is a little bit larger than the size of the atoms cloud (0.3 mm in diameter). In this case, all the atoms can be illuminated by both excitation lasers. By using the absorption method, the number of the trapped atoms is measured to be about 4×10^{7}, yielding a peak density of 3.5×10^{11} cm^{-3}. The fluorescence loss spectra are investigated based on the three-level systems, and the related energy level diagrams and experimental setup are shown in Fig. 1.

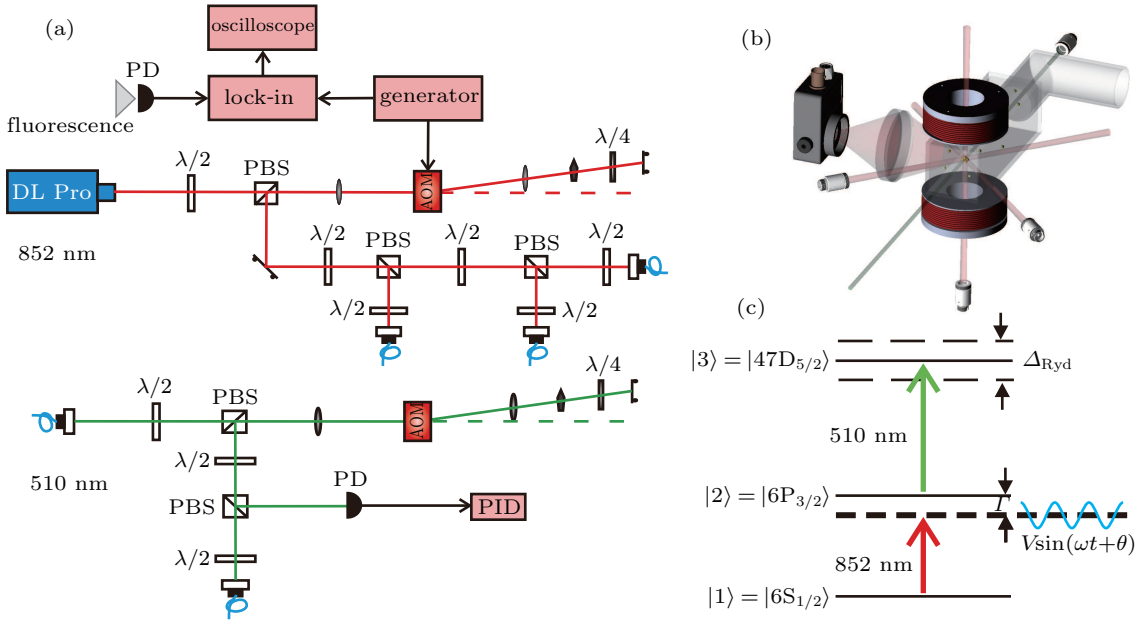

Fig. 1. Experiment setup and levels diagram. (a) Lasers setup, where the 852 nm laser (upper left) is coupled into three PM fibers respectively as the cooling beams. The 510 nm coupling beam (left lower) is split into two parts, one of which is used for power stabilization via PID, and the other one is coupled into a PM fiber for Rydberg excitation. (b) Zoom in of the trapping area, including cooling and excitation lasers, MOT coils, and photodiode for fluorescence collection. (c) Cascade three levels of Rydberg excitation. Cesium atoms are excited from $6S_{1/2}$ ground state to $6P_{3/2}$ excited state with 852 nm cooling lasers and then from the excited state to $47D_{5/2}$ Rydberg state by 510 nm coupling laser. The detuning of the first excitation laser is periodically varied due to the modulation of the 852 nm cooling lasers.

As mentioned above, the frequency of the 510 nm coupling laser is scanned and at the same time the oscilloscope is used to record the fluorescence loss spectrum of Rydberg atoms excitation. The scanning speed of the 510 nm laser is slow and carefully chosen to be 0.8 MHz/s with which the signal intensity is almost twice larger than that of fast scan (e.g., 3.2 MHz/s) and the signal is symmetry for both scan directions. The fluorescence is collected by an achromatic lens whose focal length is 50 mm and detected with a photoelectric detector (PDA36A-EC, Thorlabs). The frequency of the cooling lasers is modulated by feeding a weak oscillation through the frequency MOD interface of the cooling laser AOM. The varying ranges of depth (converted to laser frequency) and frequencies of modulation are 0.2–5.7 MHz and 1–11 kHz in steps of about 0.4 MHz and 1 kHz, respectively. The fluorescence signal is then sent into and demodulated with a lock-in amplifier (SR830, Standford Research Systems), of which the time constant is 300 ms and the sensitivity is 200 mV.[35]

3. Results and discussion

The direct fluorescence loss spectra are shown in Fig. 2. The blue and black dots represent the signal with and without modulation, respectively. The red line is the theoretical calculation. All the data are normalized with respect to the peak of the modulated signal. Obviously, the signal increases a lot and the noise narrows down when the cooling lasers are modulated which leads to a significantly enhancement of SNR of the fluorescence loss spectrum.

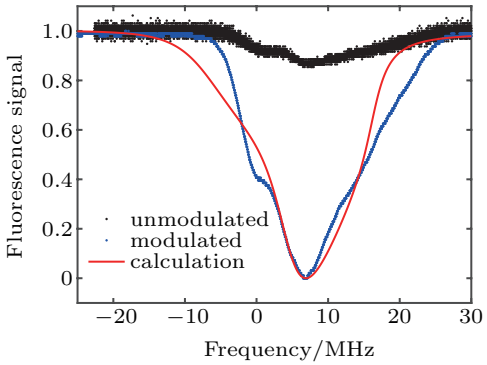

Fig. 2. Direct fluorescence loss spectra with (blue dots) and without (black dots) modulation. Red line is the result of theoretical calculation.

In order to study the fluorescence variation of excited states, we consider a three-level system composed of states $|1\rangle$, $|2\rangle$, and $|3\rangle$, corresponding to the ground $6S_{1/2}$, excited $6P_{3/2}$, and Rydberg $47D_{5/2}$ states, respectively, which is illustrated in Fig. 1(c). The atoms are excited to the Rydberg states through two-photon excitation $6S_{1/2}$–$6P_{3/2}$–$47D_{5/2}$. The system Hamiltonian is written in the form

$$H = \begin{pmatrix} 0 & \Omega_{852} & 0 \\ \Omega_{852} & -2\Delta_{510} & \Omega_{852} \\ 0 & \Omega_{852} & -2(\Delta_{510}+\Delta_{852}) \end{pmatrix},$$

in which Ω_{852} and Ω_{510} represent the Rabi frequencies of the 852 nm first excitation and 510 nm coupling lasers, respectively, Δ_{510} and Δ_{852} denote the detunings associated with the 510 nm laser and 852 nm laser, respectively. The Von Neumann equation is applied to investigate the time evolution of this system. The decay and decoherence of the system can be attributed to the finite lifetime of the atomic levels, interaction and collisions between atoms, which lead to a Liouville term in the equation. Then the time evolution of the density matrix becomes

$$\dot{\rho} = -\frac{\mathrm{i}}{\hbar}[H,\rho] - \frac{1}{2}\{\Gamma,\rho\}, \tag{1}$$

where Γ has $\langle n|\Gamma|m\rangle = \gamma_n\delta_{nm}$. Then we have the following optical Bloch equation:

$$\begin{aligned}
\dot{\tilde{\rho}}_{11} &= \mathrm{i}\frac{\Omega_{852}}{2}(\rho_{21}-\rho_{12}),\\
\dot{\tilde{\rho}}_{22} &= -\gamma_2\rho_{22} - \mathrm{i}\frac{\Omega_{852}}{2}(\rho_{21}-\rho_{12}) - \mathrm{i}\frac{\Omega_{510}}{2}(\rho_{23}-\rho_{32}),\\
\dot{\tilde{\rho}}_{33} &= -\gamma_2\rho_{33} + \mathrm{i}\frac{\Omega_{510}}{2}(\rho_{23}-\rho_{32}),\\
\dot{\tilde{\rho}}^*_{21} &= \dot{\tilde{\rho}}_{12} = (-\gamma_{12} + \mathrm{i}(\Delta_{510}-\Delta_{852}))\tilde{\rho}_{12}\\
&\quad - \mathrm{i}\frac{\Omega_{510}}{2}\tilde{\rho}_{13} + \mathrm{i}\frac{\Omega_{852}}{2}\tilde{\rho}_{32},\\
\dot{\tilde{\rho}}^*_{31} &= \dot{\tilde{\rho}}_{13} = (-\gamma_{13} - \mathrm{i}\Delta_{852})\tilde{\rho}_{13}\\
&\quad + \mathrm{i}\frac{\Omega_{852}}{2}(\tilde{\rho}_{33}-\tilde{\rho}_{11}) - \mathrm{i}\frac{\Omega_{510}}{2}\tilde{\rho}_{12},\\
\dot{\tilde{\rho}}^*_{32} &= \dot{\tilde{\rho}}_{23} = (-\gamma_{23} - \mathrm{i}\Delta_{510})\tilde{\rho}_{23}\\
&\quad + \mathrm{i}\frac{\Omega_{510}}{2}(\tilde{\rho}_{33}-\tilde{\rho}_{22}) - \mathrm{i}\frac{\Omega_{852}}{2}\tilde{\rho}_{21}.
\end{aligned} \tag{2}$$

Theoretically, the absolute fluorescence radiance, B_F, in the absence of self-absorption is obtained through the relation[36]

$$B_\mathrm{F} = n_0\rho_{22}h\nu_{12}A_{21}l/4\pi, \tag{3}$$

where n_0 is the total population (number of atoms per cm^3), $h\nu_{12}$ is the energy difference between the ground and excited states, A_{21} (in s^{-1}) represents the Einstein spontaneous emission probability, and l (in cm) denotes the depth of the (homogeneous) fluorescing volume as seen in the direction of observation. The ρ_{22} is the probability density distribution on the excited state and it is determined by the frequency detuning as well as the Rabi frequency of the cooling laser beams and the 510 nm laser beam, and the steady state solution is

$$\rho_{22} = \frac{4(\Delta_{852}+\Delta_{510})^2\Omega_{852}^2}{4\gamma_2^2(\Delta_{852}+\Delta_{510})^2 + (-4\Delta_{852}(\Delta_{852}+\Delta_{510}) + \Omega_{510}^2)^2 + 2(4(\Delta_{852}+\Delta_{510})^2 + \Omega_{510}^2)\Omega_{852}^2 + \Omega_{852}^4}, \tag{4}$$

where γ_2 is the decay rate of the excited state and the decay rate of Rydberg state γ_3 is ignored throughout the calculation as it is far smaller than Δ_{852}. When we add the frequency modulation to the cooling lasers, in the expression of ρ_{22}, the Δ_{852} can be substituted with $(-3/2\gamma_2 + A\cos(2\pi t\delta))$ and Ω_{852} can be substituted with $(\Omega_{852(0)}^2 - Ak\cos(2\pi t\delta))^{1/2}$, where A represents the modulation depth, δ represents the modulation frequency, and k represents a coefficient of the modulation amplitude impact on the laser intensity.

Besides, the relationship between ρ_{22} and ρ_{33} can be written as

$$\frac{\rho_{33}}{\rho_{22}} = \frac{\Omega_{510}^2 + \Omega_{852}^2}{4(\Delta_{852}+\Delta_{510})^2}. \tag{5}$$

Thus, in this paper, we mainly use ρ_{22} to investigate the vari-

ation of the fluorescence loss spectrum. Since the cooling and repumping lasers are applied continuously on the atoms, the atomic populations on the ground and excited states are in dynamical equilibrium. Then the rapid decrease of the atomic population on the excited state is mainly due to the excitation to Rydberg state rather than decay. Since the cooling lasers also serve as the first excitation laser, the number of cooled atoms in the MOT can be evaluated by observing the fluorescence of the atom cloud, in this way we measure the dependency of fluorescence of the excited state on the detuning of the cooling laser beams without both modulation and Rydberg excitation as shown in inset of Fig. 3, which was also illustrated in Refs. [37,38]. The fluorescence signal peaks around the position when the cooling lasers are red-detuned by 7.5 MHz. Then modulation with the frequency of 6 kHz and depth of 0.2–5.7 MHz is applied on the cooling lasers. The fluorescence signal of the excited state decreases as expected, as shown in Fig. 3.

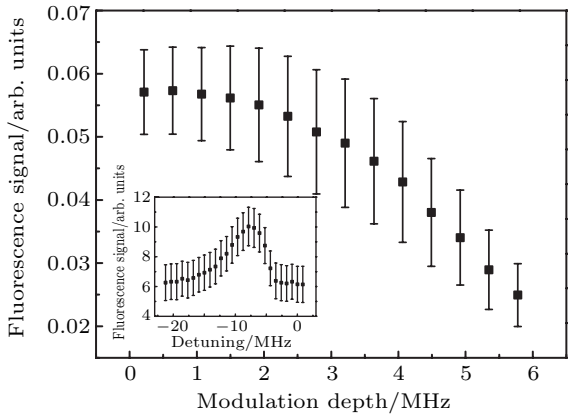

Fig. 3. The dependency of the excited state fluorescence on the depth of modulation of the cooling lasers. Inset: the dependency of fluorescence of cooling atoms on detuning of the cooling lasers. The error bars represent the fluorescence jitter.

Thus, we can simulate the dependence of Rydberg excitation on the modulation parameters of the 852 nm laser considering the population decrease in the cooling process caused by the modulation on the cooling lasers, and the optimal modulation parameters can be obtained from the theoretical simulation as shown by the red solid line in Fig. 4. In the experiment, we systematically investigate the effects of the modulation on the fluorescence loss spectrum by varying the depth and frequencies of the modulation. SNR data are extracted from each measurement as shown in Fig. 4. The dependencies of SNR on the modulation depth for each frequency are almost arch shapes of which the peaks are around 2.5–4.5 MHz, which agrees with the theoretical calculation, while it remains nearly the same value when the modulation frequencies are varied since the detunings and density of the first excitation are only affected by the depth of modulation rather than the frequencies. In the best result of measurements, the SNR of the modulated fluorescence loss spectrum is 52.48 dB, which improves by about 33 dB with respect to the mean value of 19.84 dB of the original fluorescence loss signals.

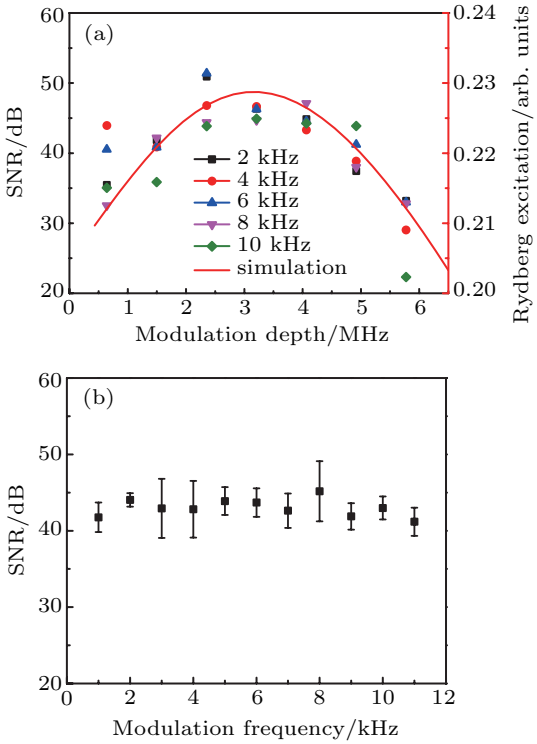

Fig. 4. (a) The SNR versus modulation depth with different modulation frequencies of 2 kHz, 4 kHz, 6 kHz, 8 kHz, and 10 kHz. (b) When the modulation depth is fixed at 4 MHz, we measure the SNR versus different modulation frequencies. The error bars represent the standard deviation uncertainty from fitting data.

We choose the D state of Rydberg atoms for detection in the experiment for its larger transition dipole moment compared to that of S state. However, the D state exhibits DC stark splitting due to the stray electric field which is not well compensated in the experiment, shown as a small bump in the left wing of signal in the fluorescence loss spectrum in Fig. 2. Fluctuations of atoms number due to power instability of cooling lasers and collisions between atoms and environment particles contribute to the errors of the experiment.

4. Conclusion

We obtained a highly sensitive fluorescence loss spectrum in the MOT of cesium atoms by optimizing the modulation depth and frequencies of the cooling lasers, which leads to a significant improvement of the signal to noise ratio of the spectrum, and the best result is 32.64 dB. This technique provides a robust and sensitive way for detections of Rydberg atoms non-destructively.

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