Two-photon coherent spectroscopy of ultracold Li atoms

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Abstract. Our work is devoted to theoretical study of the two-photon coherent spectroscopy of ⁷Li atoms continuously cooled in a magneto-optical trap (MOT) on the 2S–2P transition. The ultracold atoms are transferred to highly excited Rydberg states in a two-step coherent excitation process by red and UV lasers. The red laser is detuned by -600 MHz from 2S–2P transition frequency and UV laser frequency detuning is scanned in the vicinity of +600 MHz from 2P–nS(D) transition where n=40-100 is principal quantum number. The fluorescence signal on the 2P–2S cooling transition makes it possible to obtain a two-photon absorption spectrum. Atom-field interaction is considered in the simple three-level approximation involving a density matrix formalism. It is shown that the effect of the MOT beams on the shape of the two-photon absorption line can be taken into account by an appropriate change in the 2S–nS(D) coherence decay rate.

1. Introduction

Investigation of the properties of systems consisting of highly excited Rydberg atoms is important for various applications, such as, for example, quantum computations [1, 2]. It is convenient to use two-photon or three-photon laser excitation to transfer atoms to Rydberg states. The standard method for diagnosing Rydberg states uses ionization of excited atoms by an electric field [3]. For a simple and reliable diagnostics of Rydberg states in the two-photon excitation process, we proposed a new method based on recording the resonance fluorescence of atoms in the MOT [4, 5, 6]. The advantages of this method include simplicity and high sensitivity. High sensitivity is due to the low rate of escape of atoms from the MOT and the low atoms capture rate in the MOT. Small rates determine small nonlinearities that ensure saturation of the signal.

In this paper we propose a simple theoretical model that describes well the shape of the spectral line of the two-photon absorption, and also makes it possible to estimate the densities of atoms in the ground and Rydberg states.
2. Experiment

A schematic diagram of the experiment on two-photon excitation of ultracold $^7$Li atoms and the involved atomic levels are shown in figure 1. The Li atoms are continuously cooled in the MOT at the $2S(F=1)\rightarrow 2P$ transition, optical pumping is performed at the $2S(F=2)\rightarrow 2P$ transition. The temperature of the atoms is in the range $T \approx 300 – 500 \mu K$ (depending on the power of the MOT beams). The experimental cooling technique is described in more detail in [7]. The cloud of ultracold atoms is irradiated by counter propagating beams of red and UV lasers. The red laser is detuned from the transition frequency $2S(F=1)\rightarrow 2P$ to $\delta_1 \approx 600$ MHz. The detuning $\delta_2$ of the UV laser is scanned in the vicinity of $+600$ MHz from the $2P\rightarrow nS(D)$ transition, where $n$ is the principal quantum number of the corresponding Rydberg level of Li. By tuning the frequency of the UV laser, $n$ can be set within wide limits $n \approx 40–100$.

At close absolute values of the detunings of the red and UV lasers, a two-photon coherent absorption process occurs, leading to the formation of Rydberg Li atoms with definite values of the principal $n$ and orbital $l$ quantum numbers. This, in turn, leads to an effective decrease in the density of atoms in the ground state and an experimentally observed drop in fluorescence at the $2P\rightarrow 2S$ transition (see figure 2).

Figure 1. Schematic diagram of the experiment and involved $^7$Li levels.

Figure 2. The fluorescence signal of $2P\rightarrow 2S$ transition as a function of the detuning of the UV laser from the $2P\rightarrow 58S$ transition (shifted so that the center of the line is approximately at zero). The solid line is the experimental result, the dashed line is the solution of equations (1). The line FWHM is $3.5$ MHz, the total intensity of the MOL beams is $8.4$ mW/cm$^2$. 
In the next section, we propose a simple model that describes well the experimental results obtained.

3. Theoretical model and comparison with the experiment

For a theoretical description of the experimental results, we will use the density matrix formalism.

Since the relative population of the intermediate level 2P is low [4] and large absolute values of detunings of the red and the UV lasers are used, the Rydberg states are populated during a purely coherent two-photon absorption. This allows us to consider a three-level system consisting of 2S(F=1), 2P and nS(D) states, on which only the fields of the red and the UV lasers operate. In this case, the direct action of the MOT beams on the kinetics of the population of highly excited levels can be neglected.

We denote the levels 2S(F=1), 2P, nS(D) as 1, 2 and 3 respectively. We denote the transitions 2S(F=1)→2P and 2P→nS(D) as 1 and 2 and introduce for them the Rabi frequencies $\Omega_i = d_i E_i / \hbar$, where $d_i$ and $E_i$ are the dipole matrix elements of the corresponding transitions and amplitudes of the radiation fields of red and UV lasers.

We will consider an open system, in this case the diagonal terms of the density matrix $\rho$ correspond to the relative populations of the levels. Following [8], we write the equations for the elements of the density matrix in the three-level approximation in the following form:

\[
\begin{align*}
\dot{\rho}_{11} &= 0.5i\Omega_1(\rho_{21} - \rho_{12}) + 2\gamma_2\rho_{22} + \gamma_{\text{trap}}(1 - \rho_{11}) \\
\dot{\rho}_{22} &= 0.5i\Omega_2(\rho_{22} - \rho_{12}) - 2\gamma_2\rho_{22} + 0.5i\Omega_2(\rho_{32} - \rho_{23}) \\
\dot{\rho}_{33} &= 0.5i\Omega_2(\rho_{33} - \rho_{32}) - 2\gamma_3\rho_{33} - \gamma_R\rho_{33} \\
\dot{\rho}_{12} &= 0.5i\Omega_1(\rho_{21} - \rho_{12}) - \rho_{12}(\gamma_2 + i(\delta_1 - k_1 v)) - 0.5i\Omega_2\rho_{13} \\
\dot{\rho}_{23} &= 0.5i\Omega_2(\rho_{23} - \rho_{22}) - \rho_{23}(\gamma_3 + \gamma_2 + i(\delta_2 + k_2 v)) + 0.5i\Omega_1\rho_{13} \\
\dot{\rho}_{13} &= 0.5i\Omega_1\rho_{23} - 0.5i(\delta_1 + \delta_2 - (k_1 - k_2) v)\rho_{13} - 0.5i\Omega_2\rho_{12} - \rho_{13}\gamma_{13} \\
\dot{\rho}_{ij} &= \rho_{ji} 
\end{align*}
\]

Relaxation terms containing the corresponding rates are added to the equations: $2\gamma_2$ is the rate of radiative decay of the 2P level, $\gamma_{\text{trap}}$ is the rate of escape of the 2S atoms from the trap, $2\gamma_3$ is the rate of radiative decay of the Rydberg state, $\gamma_R$ is the rate of escape of Rydberg atoms from the trap, and $\gamma_{13}$ is the rate of decay of the coherence between the ground and Rydberg states.

Unlike experiments with heavier elements, in the analysis of experiments with Li we cannot neglect the Doppler broadening. The Doppler shifts in (1) for red and UV lasers are denoted by $k_1 v$ and $-k_2 v$ respectively.

Equations (1) are written for the case when the radiative decay of a Rydberg atom removes it from the system. This approximation is made, since in the described experiments there is no radiation recorded that indicates that during the radiation decay the atom could return to the ground state and be again captured in the MOT. The characteristic rates of radiative relaxation processes can be estimated from the extrapolation of data of [9]: for the 40S state $2\gamma_3 \approx 2 \times 10^4 \text{ s}^{-1}$, for the 40D $2\gamma_3 \approx 3 \times 10^4 \text{ s}^{-1}$ state.

One of the features of the described experiments is that due to relatively low Li atomic mass, the capture rate of atoms in the trap is much less than the escape rate of Rydberg atoms (which are not held by the MOT forces), i.e. $\gamma_{\text{trap}} \ll \gamma_R$. The capture rate of atoms can be estimated from the rates of filling and emptying of the MOT as $\gamma_{\text{trap}} \approx 0.1 - 0.01 \text{ Hz}$, while the escape rate of Rydberg atoms, based on the temperature of the atomic cloud and the size of the trap, is $\gamma_R \approx 10^5 \text{ Hz}$ [5]. Such a ratio of the rates of the processes leads to a high contrast of the obtained two-photon absorption spectrum.

Despite the fact that the radiation field of the MOT beams does not directly affect the population of the Rydberg states, the constant excitation of atoms on the 2S→2P transition during cooling leads to the decay of the coherence between the ground and Rydberg states,
which in turn provide an additional line broadening. Calculations show that a good agreement with the experimental data is obtained when the coherence decay rate is qualitatively the same as the excitation rate at the 2S–2P transition by a cooling laser, calculated in the two-level approximation. At minimum total power of the MOT beams $\gamma_{13}$ is about hundreds of kHz in order of magnitude, which is much higher than the rates of other relaxation processes that can lead to a loss of coherence — $\gamma_3$ and $\gamma_R$. This suggests that the decay of coherence is determined mainly by the MOT influence. A more detailed investigation of the dependence of the line broadening on the intensity of cooling beams in the MOT is beyond the scope of this paper.

The frequency scanning rate of the UV laser in the experiments under discussion is sufficiently low to consider the system being in a stationary state for any value of detuning $\delta_2$. Therefore, the derivatives on the left-hand side of (1) can be set equal to zero. Since the low saturation of 2S–2P transition the fluorescence signal observed in the experiments is considered to be proportional to the population of the ground state $\rho_{11}$. Figure 2 shows the calculated shape of the two-photon absorption line in the stationary approximation for one of the experiments. In the experiment the excitation of the 58S level was carried out, the total intensity of the MOT beams was 8.4 mW/cm$^2$, the intensity of the red laser was 28 mW/cm$^2$, the intensity of the UV laser was 64 mW/cm$^2$. Calculation parameters are: $\Omega_1 = 10^8$ s$^{-1}$, $\Omega_2 = 4 \times 10^5$ s$^{-1}$, $2\gamma_2 = 37 \times 10^6$ s$^{-1}$, $2\gamma_3 = 6 \times 10^3$ s$^{-1}$, $\gamma_{\text{trap}} = 2\pi 0.13$ s$^{-1}$, $\gamma_R = 2\pi 10^3$ s$^{-1}$, $\gamma_{13} = 3.3 \times 10^6$ s$^{-1}$. The results of calculations (1) were averaged over the Maxwell distribution for atoms with $T=363$ $\mu$K.

4. Conclusion
The paper presents a model which in a simple three-level approximation makes it possible to describe the results of experiments on two-photon coherent spectroscopy of ultracold Li atoms. The proposed model allows estimating the relative densities of Li atoms in the ground and Rydberg states in these experiments. In this model, both the kinetics of the atoms in the MOT and the broadening of the two-photon absorption line induced by the action of laser cooling beams are taken into account. There is a good agreement of theoretical results with experimental data. As the next step we would like to study the temperature of atoms in MOT under different conditions by using the two-photon spectroscopy with co-propagating and contra-propagating laser beams [6].

References
[1] Ryabtsev I I, Beterov I I, Tret’yakov D B, Entin V M and Yashina E A 2016 Phys. Usp. 59 196
[2] Pillet P and Gallagher T F 2016 J. Phys. B: At. Mol. Opt. Phys. 49 174003
[3] Gallagher T F 2005 Rydberg Atoms (Cambridge: Cambridge University Press)
[4] Zelener B B, Saakyan S A, Sautenkov V A, Manykin E A, Zelener B V and Fortov V E 2014 Jetp Lett. 100 366
[5] Sautenkov V A, Saakyan S A, Vilshanskaya E V, Murashkin D A, Zelener B B and Zelener B V 2016 Laser Phys. 26 115701
[6] Sautenkov V A, Saakyan S A, Vilshanskaya E V, Zelener B B and Zelener B V 2017 Journal of Russian Laser Research 38 1
[7] Zelener B B, Saakyan S A, Sautenkov V A, Akulshin A M, Manykin E A, Zelener B V and Fortov V E 2013 Jetp Lett. 98 670
[8] Stenholm S 1984 Foundations of laser spectroscopy (New York: John Wiley & Sons, Inc.)
[9] Theodosiou S E 1984 Phys Rev E 30 2881