Two-photon excitation of ultracold atoms to Rydberg states

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Abstract. In this work, we discuss two-photon excitation and diagnostic of ultracold Rydberg atoms in a magneto-optical trap. Lithium atoms were excited by using ultraviolet cw laser. For identification of Rydberg transitions, we recorded resonance fluorescence of ultracold atoms. Spectra of transitions $2P-nS$, $2p-nD$ were measured. Our results are in good agreement with calculations and experimental data available in literature. Presented work is a part of our project focused on preparation and study of Rydberg matter and ultracold plasma.

Our research project is focused on study of a Rydberg matter and non-ideal plasma [1, 2]. Results of the research can be useful for fundamental physics and for some applications as quantum computers. Atoms can be optically transferred to definite Rydberg states by two-photon or multi-photon excitation processes [3, 4]. The problem is to diagnose highly-excited states. In addition to the traditional approach, induced ionization of Rydberg atoms by applied electric field [2], recently EIT [5] and FWM [6] techniques have been used for identification of the Rydberg transitions. As the first step in our research project we have performed cw two-step excitation of ultracold lithium atoms from the ground state to a highly-excited state. In the current paper we describe our experimental setup and observation of the Rydberg transitions in lithium atoms in magneto-optical trap (MOT). A basic description of the MOT for lithium atoms is presented in [7]. Our vacuum system is similar to an experimental apparatus which used in laboratory of A. Turlapov for investigation of the Fermi gas of $^6$Li atoms [8]. The atomic transitions, which participated in excitation of Rydberg states, are shown in figure 1.

Optical transitions in $D_2$ line are used for optical cooling and trapping of lithium atoms. Optical transitions in $D_1$ line are used for absorption measurements of an atomic cloud in MOT by using a weak probe laser. In figure 1 a blue arrow shows an ultraviolet transition (350 nm) from the excited state $2P_{3/2}$ to highly excited states.

In our laboratory an experimental setup for trapping and ultraviolet excitation of $^7$Li atoms is assembled. Our setup is described in [9–11]. It consists of vacuum and optical systems. The vacuum system includes an oven (the source of a thermal atomic beam), a Zeeman slower, and a main vacuum chamber (residual air pressure $<10^{-9}$ mbar), where $^7$Li atoms are trapped.

In the optical part of the setup we use two amplified external cavity diode lasers (ECDL) for the cooling and trapping of $^7$Li atoms. One of them is the cooling laser, which is slightly detuned from the cycling transition $2S_{1/2}(F = 2) - 2P_{3/2}(F' = 3)$. This laser is locked to the thermally stabilized reference cavity with tunable transmission resonances. The other laser is repump laser...
Figure 1. Diagram of energy levels in $^7$Li atom.

which is locked to the Doppler-free saturation resonance in a hot lithium vapor ($\sim 700$ K). The laser frequency is detuned by 20 MHz from the transition $2S_{1/2}(F = 1)\rightarrow 2P_{3/2}(F' = 2)$. The repump laser needs to return atoms from sub-level $F = 1$ to the $F = 2$ sub-level in the ground state. The emitted light from these two lasers are recombined by polarized-beam-splitters (PBS) and wave-plates to several beams with circular polarization. An optical beam with power near 100 mW is used in the Zeeman slower. Other optical beams with total power of 150 mW are sent to the vacuum chamber to cool and trap $^7$Li atoms. The diameters of the optical beams in the vacuum chamber are equal to 2.4 cm. For characterization of the cloud of ultracold atoms we use a weak probe ECDL. Also the setup includes an ultraviolet laser system (350 nm) to create Rydberg atoms.

Using the CCD camera, we have measured the intensity profile of fluorescence from the atomic cloud in the MOT. The results of these measurements are presented in figure 2, which correspond to a magnetic field gradient of 21 G/cm. The profiles of the atomic cloud are very close to the Gaussian distribution with diameters (FWHM) $d_x = 0.2$ cm and $d_y = 0.14$ cm [10].

Complementary high-resolution spectroscopy of $^7$Li atoms in the MOT is performed by the probe laser. The diameter of the probe beam is much less than the size of the cloud. The intensity of the probe beam is reduced to ensure the regime of linear absorption. The results are presented in figure 3. As one can see, the hyperfine splitting of the D$_1$ line is resolved, whereas the hyperfine splitting of the D$_2$ line is not resolved. In general, both spectra can be used to estimate the number density of atoms in the MOT. In order to improve the accuracy we estimated population of ground state sublevels by using the transmission spectra on D$_1$ line. In this case the absorption is relatively small and excited state $2P_{1/2}$ is unpopulated. The concentration of the trapped atoms on the ground state $S_{1/2}(F_g)$ can be calculated from
transmittance $T$ by using the relation

$$T = \exp(-\sigma(F_g, F_e)nd_x).$$

According to [12] the absorption cross section for the hyperfine components of the D$_1$ line can
Figure 4. Density of trapped atoms on sub-levels \( F = 2 \) and \( F = 1 \) of the ground state \( 2S_{1/2} \). The “squares” and “circles” are experimental data for densities \( n_{g1} \) on sub-level \( F = 1 \) and \( n_{g2} \) on sub-level \( F = 2 \). The curves are guides for eye.

be written

\[
\sigma(F_g, F_e) = (\lambda^2/4\pi)(A/\Gamma)g(F_g, F_e)(2J_e + 1)/(2J_g + 1).
\]

Here \( A \) is the Einstein coefficient and \( \Gamma \) is the spectral width of the atomic transition. In our case, \( A = \Gamma = 2\pi\gamma \). Natural spectral width of the transition \( \gamma \) is of 6 MHz. The factor \( g(F_g, F_e) \) can be expressed by using 6J symbols [13]. The calculated concentrations of the ground state atoms v.s. detuning of the cooling laser are shown in figure 4.

Note that the populations of the ground-state \( 2S_{1/2} \) sub-levels are quite close by magnitude. It can be explained by the fact that the upper state \( 2P_{3/2} \) sublevels are not resolved (hf-splitting is the same order as the natural width \( \gamma \)). Therefore it is impossible to select the circling transition for the effective optical cooling. Atoms are continuously transferred from \( F = 2 \) to \( F = 1 \) and back. In the maximum (detuning range from \( 3\gamma \) to \( 6\gamma \)) the estimated population of the excited state \( 2P_{3/2} \) is order \( 10^{-2} \) of the total population of the ground state. Therefore the total density of the trapped atoms \( n \) in the maximum is near \( 5 \times 10^{10} \text{ cm}^{-3} \). An estimated total number \( N \) of the trapped atoms is of \( 4 \times 10^8 \). Estimated temperature of the trapped atoms is order of \( 3 \times 10^{-4} \text{ K} \). The density \( n \) and total number \( N \) of atoms can be increased by increasing the magnetic field gradient. The linear dependence of \( n \) and \( N \) on the magnetic field gradient in the range 20–35 Gauss/cm has been observed [10].

For excitation of Rydberg atoms we use an ultraviolet laser system (Newport Spectra Physics). This cw laser system works at a wavelength of 350 nm with an output power up to 100 mW. The ultraviolet laser beam (diameter \( \approx 1 \text{ cm} \)) is sent onto the atomic cloud. The laser frequency can be scanned. When the laser frequency passes through the Rydberg transition, we observe the reduction of the resonance fluorescence of the trapped atoms. It is gone down to its partial or complete vanishing depending on the ultraviolet laser beam power and on the quantum number of upper state.
Figure 5. Reduction of resonance fluorescence of the trapped atoms on the transition $2P_{3/2} - 100D$.

For the more reliable determination of the transition frequency, in addition to the CCD camera, we install a photodetector for recording the fluorescence intensity. The frequency of ultraviolet laser is controlled by a high precision wavemeter WSU (High Finesse Angstrom). In our experiment, the wavemeter is calibrated by using a stabilized ECDL, whose frequency is locked to Doppler-free absorption resonance in rubidium-85 atomic vapor. The diagram depicting the working energy levels is shown in figure 1. According to the selection rules, the transitions from the 2P-state are possible only to the nS and nD-states. In the experiment we have measured frequencies $\nu_{nl}$ of Rydberg transitions $2P_{3/2} - nl$. To identify Rydberg states we calculated the quantum numbers $nl$ taking into account the quantum defect $\Delta n$ and using the expression [4, 11, 14]:

$$n^* = \sqrt{\frac{\text{Ry}}{\nu_i - \nu_{SP} - \nu_{nl}}}.$$  

Here $n^*$ is quantum number with quantum defect $\Delta n$, Ry is the Rydberg constant, $\nu_i$ is the ionization threshold frequency and $\nu_{SP}$ is a frequency interval between the centers of gravity of the $2S_{1/2}$ and $2P_{3/2}$ states.

The curve in figure 5 is an example of our observations. The dips in the fluorescence, which correspond to transition $2P_{3/2} - 100D$, are shown. The frequency of the spectral component $2P_{3/2} - 100D$ is of $856.5727(3)$ THz. We have recorded also other $2P - nS$ and $2P - nD$ transitions and measured the frequencies of transitions. The observed resonances are quite broad.

The spectral broadening and shape of the components of the Rydberg transitions will be studied more carefully. We plan to vary powers of the laser beams, magnetic field gradient and other experimental parameters. It will be exciting to measure an influence of interactions between Rydberg atoms on the spectral width.

In conclusion we note that the cw two-step excitation and diagnostic of Rydberg atoms is demonstrated. Lithium atoms were excited by using ultraviolet laser system. For identification of Rydberg transitions we recorded the resonance fluorescence of ultracold lithium atoms in MOT. The challenge is to investigate many-body interactions in a gas of ultracold Rydberg atoms or non-ideal plasma [15–18].

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