Influence of an electric field on Rydberg \( nD \) states of cold lithium atoms

S Ya Bronin\(^1\), A A Bobrov\(^1\), D E Fomichev\(^{1,2}\), S A Saakyan\(^1\), V A Sautenkov\(^{1,4}\), B B Zelener\(^{1,2,3}\) and B V Zelener\(^1\)

\(^1\) Joint Institute for High Temperatures of the Russian Academy of Sciences, Izhorskaya 13 Bldg 2, Moscow 125412, Russia
\(^2\) National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), Kashirskoe Shosse 31, Moscow 115409, Russia
\(^3\) National Research University Moscow Power Engineering Institute, Krasnokazarmennaya 14, Moscow 111250, Russia
\(^4\) Lebedev Physical Institute of the Russian Academy of Sciences, Leninsky Avenue 53, Moscow 119991, Russia

E-mail: bobozel@mail.ru

Abstract. Influence of a weak electric field on the coherent Rydberg resonances was studied on two-photon transitions \( 2S-nD \) by recording the resonance fluorescence of lithium atoms in a magneto-optical trap. It was observed that an isolated spectral line \( 2P-nD \) transferred to the spectral band width growing with a principal number \( n \) (or an electric field \( E \)). It was shown that the frequency tuning of a resonant radiation on the left (right) edge of the band allows to create a gas of Rydberg atoms with fixed and large dipole moments \( d \propto e a_0 n^2 \) (\( e \) is the elementary charge, \( a_0 \) is the Bohr radius), oriented against (along) the electric field. The radiation frequency was tuned on the center of the band would produced atomic states with zero dipole moments, but with a big contribution of states with maximum angular momentum quantum number \( l \sim n \).

1. Introduction

Rydberg atoms are very sensitive to external electric and magnetic fields. In most experiments, an important contribution to the width of the observed Rydberg transitions is provided by uncompensated stray electric fields. The sensitivity of Rydberg atoms to the field grows very fast with increasing the principal quantum number.

In [1], the field was decreased to 45 \( \mu \)V/cm, which allowed observing Rydberg states of barium with \( n = 520 \). In [2], Rydberg states were used to measure weak electric fields and the accuracy of order of 20 \( \mu \)V/cm was achieved. In real experimental conditions, it is practically impossible to completely compensate the electric fields. Measuring equipment and charged particle counting systems are potential sources of an electric field. The investigated atoms can be deposited as films on the elements of the recording system and create a contact potential difference [3]. At the same time, the residual electric field may lead to the excitation of Rydberg states with a high orbital quantum number \( l \). States with a high \( L \) (hydrogenic states) [4] have a high dipole moment.

In this work, the influence of weak residual electric field on the width of Rydberg resonances \( 49D, 59D, 70D \) and \( 82D \) was investigated experimentally by the reduction of the resonance...
Figure 1. The diagram of energy levels in $^7$Li (a) and the principal scheme of our experiment (b).

fluorescence of neutral atoms of $^7$Li in magneto-optical trap (MOT) [5,6]. Quantum mechanical calculations for these Rydberg states were performed using perturbation approach taking into account the Stark effect. Calculations results agree well with the experiment if field value $E = 12$ mV/cm is substituted into the equations. Earlier [7] we made estimation for residual field in our MOT using a different technique. The estimation result is close to the field value obtained here. It was also shown that in the presence of even a weak electric field the resonant radiation tuned to the left or right edge of the band makes it possible to obtain a gas of Rydberg atoms with a permanent high dipole moment oriented respectively along or against the field.

2. Experimental setup

All the experiments described in this work were carried out on MOT for lithium atoms, a detailed description of which can be found in works [5–7].

Two lasers with a wavelength of 671 nm are used for laser cooling and trapping lithium atoms into MOT. The first cooling laser with intensity $I_c$ is stabilized at the transition frequency $2S_{1/2}|F=2\rangle\rightarrow2P_{3/2}|F'=3\rangle$. The second cooling laser $I_{op}$ is stabilized at the transition frequency $2S_{1/2}|F=1\rangle\rightarrow2P_{3/2}|F'=3\rangle$. In each of six beams forming MOT, there was a constant relation of $I_c/I_{tot}=0.7$ and $I_{op}/I_{tot}=0.3$ ($I_{tot}=I_c+I_{op}$). The total intensity of MOT beams is $I_{tot}=72$ mW/cm$^2$.

Two-photon resonances in Rydberg states are excited by two counter propagating laser beams $\vec{k}_{671}$ and $\vec{k}_{350}$ (figure 1) to reduce the Doppler broadening. The laser beam $\vec{k}_{671}$ is formed by the cooling laser that is stabilized at the transition $2S_{1/2}|F=2\rangle\rightarrow2P_{3/2}|F'=3\rangle$ and tuned from the transition $2P_{3/2}|F'=3\rangle$ at the frequency $\delta = 593$ MHz toward the red side.

Beam $\vec{k}_{350}$ is the result of doubling the frequency of the titanium–sapphire laser, which is locked on a stable Fabry–Perot cavity by the Pound–Drever–Hall method [8].

The intensity of uv-beam $\vec{k}_{350}$ in all the experiment did not exceed 3 mW/cm$^2$. Resonances to Rydberg states were registered by the reduction in the resonant fluorescence of neutral lithium atoms in MOT [6]. The shape of the resonant spectral lines for all $n$ is determined by the combined action of the Doppler broadening and the Lorentz broadening, which is due mainly the
Figure 2. The resonance on the two-photon coherent transition $2S-2P-49D$. The black solid curve presents experimental data. The red dashed curve presents result of the calculations.

optical field broadening. In all experiments, the temperature of the atoms was about 0.6 mK, at this temperature the Doppler width is 1.8 MHz. The total intensity of cooling beams is 72 mW/cm$^2$, which corresponds to the Lorentz width of the order of 2.8 MHz. A detailed study of the effect of the intensity of MOT beams on the width of two-photon Rydberg resonances can be found in the work [9]. The total width of lines of the red and uv lasers (in figure 1(b) $\vec{k}_{671}$ and $\vec{k}_{350}$ respectively) was about 0.2 MHz [10].

In figure 2, a spectral profile of the two-photon coherent transition $2S-2P-49D$ in cold lithium atoms is shown. Here, the effect of the weak electrical field induced a shift of transition $2P-49D$ and splitting, which are small to compare with the width of the observed resonance. The calculated resonance is the Voigt convolution of Lorentz and Gaussian functions. The ratio of the optical field broadening and the Doppler width equals to 1.5. The width of the observed resonances was increased with growing of the principal quantum number $n$. In order to interpret such broadening of the resonances the Stark splitting of $nD$ states has to be taken into account. In section 3 the Stark effect is considered and in section 4 the comparison of the experimental data with calculations is performed.

3. Calculation of Stark splitting
In the absence of an electric field, the excitation spectrum in the interval between two adjacent hydrogen levels is represented by two spectral lines $2P_{3/2}-nD$ and $2P_{3/2}-nS$. A general approach to the modification of spectral lines by the Stark effect is discussed in [11,12]. When the electric field is non zero, the excitation process, due to the Stark effect, involves $5(n-3)$ levels with orbital number values $l > 2$ and magnetic quantum numbers $m = 0, \pm 1, \pm 2$. In the factorization of each Stark state with respect to the unperturbed eigenstates, there is an $nD$ component. Therefore each such a state is related to the $2P_{3/2}$ level by a dipole matrix element making possible a resonant transition to this state. As a result, with increasing field strength, a single spectral line
turns into a band consisting of $5(n-2)$ lines. The levels forming these bands are connected to each other in pairs by matrix elements $V_{n,l,m} = \langle n, l, m | E \Delta | n, l + 1, m \rangle = E_{er} \xi_{l,m}$ ($E$ is electric field, $d$ is dipole moment, atomic radius $r_{nl}$ is calculated using quasiclassical approximation, $\xi_{l,m} = \xi_{l,-m}$, $\xi_{l,m} = \int_{-\frac{1}{2}}^{\frac{1}{2}} \Theta_{l,m}(t) \Theta_{l+1,m}(t) dt$, where $Y_{l,m}(\theta, \phi) = \Theta_{l,m} \cos \theta \exp(i m \phi) / \sqrt{2\pi}$, $\Theta_{l,m}(\cos \theta)$ is a factor in the composition of the spherical function $Y_{l,m}(\theta, \phi)$, $\xi_{l,m} \to 1/2$ when $l \to \infty$).

The defect of the $nD$ level is equal to $\delta_2 = 0.002$. The defects of the other levels can be neglected so that in the unperturbed system the energy levels are represented by two values:

$$E_0 = -R_y/(n - 0.002)^2, \quad l = 2,$$

and the energy of the $n$-hydrogen level

$$E_1 = -R_y/n^2, \quad l = 3, 4, \ldots, n - 1,$$

the distance between which is equal

$$\varepsilon_0 = E_1 - E_0 \approx 2R_y\delta_2/n^3. \quad (3)$$

At not very high values of the field $E$, when the effect of the $nP$ level can be neglected, perturbed energy levels and corresponding states are eigenvalues and eigenfunctions of the matrix operator $H = H_0 + V$ with dimension $(n-2) \times (n-2)$. The values of matrix elements $H_{i,j}$ are equal to: $H_{1,1} = -\varepsilon_0$, $H_{i,i} = 0$, $H_{i,i+1} = V_{n,i+1,m}$, $H_{i+1,i} = V_{n,i,m}^*$, $i = 2, 3, \ldots, n - 2$. At low values of the field $E$, the second order Stark effect takes place, while at large ones it is linear. The boundary value of the field $E_{St}$ is its magnitude, at which $E_{St} 3e a_0 n^2/2 = \varepsilon_0$ and $E_{St} = 2e\delta_2/3a_0^2 n^5$ ($e$ is the elementary charge, $a_0$ is the Bohr radius).

Eigenenergy values $\varepsilon_k = E_{n,m}^{(k)} - E_0$, $k = 1, 2, \ldots, n - 2$ are determined by a numerical solution of the equation $\det[H_{i,j} - \varepsilon \delta_{i,j}] = 0$. Based on the known values of $\varepsilon_k(n,m)$, coefficients of the expansion of eigenfunctions with respect to functions of the unperturbed system $|\psi(n,m)\rangle = \sum_{l=-2}^{l} C_l^{(k)}(n,m) |n,n,l,m\rangle$ are determined from the system of equations:

$$-(\varepsilon_k + \varepsilon_0) C_2^{(k)} + V_{n,2,m} C_3^{(k)} = 0; \quad \text{(4)}$$
$$V_{n,2,m} C_2^{(k)*} - \varepsilon_k C_3^{(k)*} + V_{n,3,m} C_4^{(k)*} = 0; \quad \text{(5)}$$
$$\ldots$$
$$V_{n,l-1,m} C_{l-1}^{(k)*} - \varepsilon_k C_l^{(k)*} + V_{n,l,m} C_{l+1}^{(k)*} = 0; \quad \text{(6)}$$
$$\ldots$$
$$V_{n,n-2,m} C_{n-2}^{(k)*} - \varepsilon_k C_{n-1}^{(k)*} = 0. \quad \text{(7)}$$

The bandwidth $\Delta \varepsilon = \varepsilon_{n-2} - \varepsilon_1$ at $E > E_{St}$, like for a hydrogen, is given by the equation $\Delta \varepsilon \approx 2\varepsilon_0 E/E_{St} = 3e a_0 n^2 E$. As in each of eigenstates $n - 2$ there is the state $|n,2,m\rangle$ with a certain coefficient $C_2^{(k)}(n,m)$, each of them is connected to the state $2P_{3/2}$ with an oscillator strength proportional to $|C_2^{(k)}|^2$, so that $\sum_k |C_2^{(k)}|^2 = 1$. For this reason, when the field is involved, an individual line with a fixed value of the magnetic number turns into a band consisting of $n - 2$ lines with the same total oscillator strength and width proportional to the magnitude of the electrical field at $E > E_{St}$.

Eigenstates $|\psi(n,m)\rangle$, unlike states $|n,l,m\rangle$, have other than 0 average values $d^{(k)}$, the projection of the dipole moment on the field direction:

$$d^{(k)} E = \langle \psi_k | dE | \psi_k \rangle = E \sum_{l=2}^{n-2} (C_l^{(k)})^* C_{l,l+1}^{(k)} d_{l,l+1} + C_{l+1}^{(k)*} C_l^{(k)} d_{l+1,l}), \quad \text{(8)}$$

where $d_{l,l+1} = \langle n,l,m | d_z | n,l+1,m \rangle = V_{n,l,m}/E$. 


Figure 3. (a) Values $|C_2^{(k)}(n, 0)|^2$ in the expansion of the $k$-th Stark component into spherical functions depending on the energy of the Stark level $\varepsilon_k/|\varepsilon_0|$ (the energy being counted from the $n$-th hydrogen level, $\varepsilon_0$ is the energy of the level $|n, 2, m\rangle$) for $n = 49, 59, 70, 82$, and $E = 12$ mV/cm. (b) Dipole moment of Rydberg atoms divided on its maximum value $d_0$ ($d_0 = 3e\alpha_0n^2/2$) as a function of resonance excitation frequency, $n = 70$, $E = 12$ mV/cm.

Multiplying each equation (8) by $C_2^{(k)}$ and summing up give

$$
d^{(k)}E = (\varepsilon_k + \varepsilon_0)|C_2^{(k)}|^2 + \varepsilon_k \sum_{l=3}^{n-1} |C_l^{(k)}|^2 = (\varepsilon_k + \varepsilon_0)|C_2^{(k)}|^2 + \varepsilon_k(1 - |C_2^{(k)}|^2)
$$

At low $E/E_{St}$ the right-hand side of equation (9) is proportional to $E^2$ and $d^{(k)} \propto E$, at high $E/E_{St}$ it is proportional to $E$ and the average value of the dipole moment does not depend on $E$. The sum on $k$ of all values $d^{(k)}$ is always equal to 0, as can be verified by summing up (9):

$$
\sum_{k=1}^{n-2} d^{(k)}E = \sum_{k=1}^{n-2} \varepsilon_k + \varepsilon_0 \sum_{k=1}^{n-2} |C_2^{(k)}|^2.
$$

The first sum on the right-hand side of (10) is equal to the spur of the operator $H$ (SpH = $-\varepsilon_0$), the second sum is equal to one because values $C_1^{(k)}$ are coefficients of the expansion of states $|n, l, m\rangle$ based on the normalized system of functions $|\Psi^k\rangle$. Finally, we have $\sum_{k=1}^{n-2} d^{(k)} = 0$.

Figure 3(a) shows values $|C_2^{(k)}(n, 0)|^2$ depending on the energy of Stark levels counted from $n$-th hydrogen level for $n = 49, 59, 70, 82$, $m = 0$, and $E = 12$ mV/cm. The values of ratio $E/E_{St}$ are respectively equal to: 0.49, 1.24, 2.92, and 6.45. At $n = 49$ and 59 with low values of ratio $E/E_{St}$, prevailing are coefficients $C_2^{(1)}(n, 0)$, which represent slightly shifted states $|n, 2, 0\rangle$ (second order Stark effect). At $n = 70$ and 82 all $n - 2$ Stark levels takes part in the formation of the band. With an increasing $n$, the band becomes more and more symmetrical in the relation to the hydrogen level (a linear Stark effect), coming closer in the limit to the result realized for the hydrogen atom. As $E/E_{St} \propto En^5$ the same evolution takes place with increasing $E$ at a fixed $n$. Figure 3(b) shows the values of dipole moments of Stark levels depending on their energy counted from the hydrogen level for $n = 70$, $m = 0$ and $E = 12$ mV/cm. Practically an ideal proportional dependence of these magnitudes means the linearity of the Stark effect, at which the values of the dipole moments cease to depend on the magnitude of the field.
Figure 4. The weight at which unperturbed functions with a given orbital angular momentum \( l \) are part of the state arising from resonant excitation by radiation of a given frequency for \( n = 70, E = 12 \) mV/cm.

Thus, in the presence of even a weak electric field, the resonance radiation tuned to the edge of the Stark band makes it possible to obtain a gas of Rydberg atoms with a high dipole moment \( d_0 \propto n^2 \) oriented either along the field (the right-hand side of the band \( k = n - 2, d = d_0 \)) or against the field (the left-hand side \( k = 1, d = -d_0 \)).

The absorption coefficient of the band \( f_2(\nu) \) is the sum of all contributions of the eigenstates of the perturbed system and is described by the following equation:

\[
f_l(\nu) = \frac{1}{5} \sum_{m=-2}^{2} \sum_{k=1}^{n-2} |c_l^{(k)}(n,m)|^2 f_0(\nu - \nu_k(n,m)),
\]

(11)

where \( f_0(\nu) \) is the form of the spectral line common for all Stark states, \( \nu_k \) are energy levels for given values \( m \) and \( n \) expressed in the units of frequency and \( \int f_l(\nu)d\nu = 1 \).

As follows from the calculations made in [9], the experimental conditions correspond to the Voigt profile of the spectral line \( f_0(\nu) \) with a ratio of the Lorentz width to the Doppler width equal to 1.51. The calculated spectra are presented as exp\( [-\lambda f_2(\nu)] \), where \( \lambda \) is an adjustable parameter characterizing the optical depth of the atomic cloud.

In figure 4, curves show functions \( f_l(\nu) \). Equation (11) determines the statistical weights of unperturbed functions with a given orbital angular momentum \( l \). Because of the broadening of the lines described by the Voigt profile \( f_0(\nu) \), several neighboring Stark levels are mixed in the excited state. It can be seen (figure 4) that maximum orbital momentum states give the main contribution to the Stark states near the energy corresponding to the hydrogen level (i.e., when the quantum defect is zero).
Figure 5. Two-photon coherent resonances in Rydberg states (a) 59D, (b) 70D, (c) 82D. The black curve is the experiment, the red dashed line is the theoretical curve at a residual field in the vacuum chamber of 12 mV/cm. Zero along the X axis corresponds to the frequency of the hydrogen level with the corresponding principal quantum number.

4. Results and discussion

Figure 2 shows the resonance to the state 49D. Here, the effect of the electrical field is reduced to a small shift of line 2P_{3/2}–49D and its splitting into three components, which corresponds to Zeeman sublevels and is insignificant in comparison with the width of the line itself.

Figure 5(a–c) shows coherent two-photon resonances to 59, 70, and 82 states. The width of the observed resonances is in good agreement with the theoretical calculation. At n = 59, the splitting into Zeeman sublevels becomes noticeable: the shift of the sublevels corresponding to |m| = 2 exceeds the width of the lines. In addition, the contribution of the residual Stark levels forming the band to the right of the main line becomes significant. At n = 70 and 82, single spectral lines are replaced by spectral bands in which all Stark levels participate, including levels with energies higher than those of the n-th hydrogen level.

The calculation presented in figure 4, as well as the experimental and theoretical curves in figure 5(c), shows the possibility of excitation of atoms into states with any orbital angular momentum for a given principal quantum number with a probability that depends on the excitation energy.

The Rydberg atoms with the maximum values of the orbital angular momenta can be prepared with a probability of about 14% in the presence of a small field, when the laser frequency will be tuned to the level with zero quantum defect.

The analysis of the obtained experimental results have shown that, even in the presence of a weak electric field, an individual line of the Rydberg energy level turns into a band and the resonance radiation tuned to the left or right edge of the band can produce a gas of Rydberg atoms with a permanent high dipole moment. Under the experimental conditions, the temperature of this gas is \( \sim 10^{-3} \) K and the density can reach \( 10^9 \) cm\(^{-3}\).

In [13–17], methods of the spatial visualization of Rydberg atoms were proposed and tested. There is an indication that in [13], a chain of atoms in a dense gas was observed. The use of these techniques will make it possible to study the properties of the dense phase of a resonantly excited gas. The study of the self-organization and anisotropy of the properties of the Rydberg gas due to the presence of a selected direction of the electrical field opens up new directions in the physics of ultra-cold gases. It also makes it possible to take these effects into account in resonantly excited gases, for example, at room temperatures.
5. Conclusion
In the presented work, the influence of a weak electric field on the coherent Rydberg resonances was studied on two-photon transitions $2S$–$nD$ by recording the resonance fluorescence of lithium atoms in a magneto-optical trap. It was observed that due to the Stark effect the narrow transition is transferred to the band. The spectral width of the band increased with growing principal quantum number $n$. In order to describe the spectral shapes of the bands a perturbation calculation technique was applied. Calculations show that the linear Stark effect can lead to the formation of Rydberg atoms with permanent dipole moments. The recorded energy spectra of Rydberg $nD$ states of cold atoms are in a good agreement with results of our calculations.

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