NONRESONANT MIXING OF METASTABLE LEVELS OF Ba ATOMS BY LASER RADIATION

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The results of experimental investigations of atoms perturbed by laser radiation under conditions when the energy of a Stark-shifted level becomes close to the energy of a neighboring level are presented.

Introduction

We present the results of experimental studies of perturbation of atoms perturbed by laser radiation under conditions when the change δE_n in the energy of some level n is comparable with the difference ΔE_{nk} between the energy of the considered level n and the neighboring level k (ΔE_{nk}), i.e., $\delta E_n \approx \Delta E_{nk}$. We studied in such condition the perturbation of $6s5d\ ^3D_1$ and $6s5d\ ^3D_2$ metastable states of *Ba* atom. To perturb these levels, we employed radiation of a color-center laser (CCL) with a frequency tunable within the range ω_1 = 8650 – 8900 cm^{-1}. The field strength of laser radiation in our experiments was equal to $\varepsilon_1 = 5 \times 10^6$ V/cm. This radiation strongly perturbs the $6s5d\ ^3D_2$ state of Ba atoms, since the frequency range covered by our laser includes the frequency ω_{nm} =8845 cm^{-1}, which corresponds to a one-photon transition from the $6s5d\ ^3D_2$ state to the $6s6p^1P_1^O$ state. Under these conditions, the dynamic polarizability α_n of the $6s5d^3D_2$ state should be large, and its frequency dependence can be represented as [1]

$$\alpha_n = \alpha_n(\omega) \approx \frac{(\omega_{nm} - \omega_1)d_{nm}^2}{(\omega_{nm} - \omega_1)^2 + \Gamma_m^2/4}, \quad (1)$$

where d_{nm} is the matrix element of the $6s5d\ ^3D_2 - 6s6p^1P_1^O$ transition and Γ_m is generally defined as the maximum among the width of the $6s6p^1P_1^O$ level and the line width of CCL radiation. As it follows from formula (1), the dynamic polarizability of the $6s5d^3D_2$ state is positive for frequencies $\omega_1 \le \omega_{nm}$. Due to the ac–Stark effect, the change in the energy of this state ($\delta E_n = -\alpha_n\varepsilon_1^2/4$) should be negative in the considered spectral range.

Note that, within the range of frequencies $\omega_1 \le \omega_{nm}$, the dynamic polarizability of the $6s5d\ ^3D_1$ level (α_k) is described by a formula similar to (1). This polarizability is also positive due to the fact that the condition $\omega_1 \le \omega_{km}$, where ω_{km} is the frequency of a one-photon transition from the $6s5d\ ^3D_1$ state to the $6s6p^1P_1^O$ state (ω_{km} = 9027cm^{-1}), is satisfied for frequencies from the above-specified range. However, the dynamic polarizability of the $6s5d^3D_2$ state in the above-specified spectral range is much higher than the dynamic polar1izability of the $6s5d^3D_1$ state[2]. Thus, as the CCL field strength grows within a laser pulse under our experimental conditions, the shift of the $6s5d^3D_2$ level exceeds the shift of the $6s5d\ ^3D_1$ level, and the energies of these two states may become approximately equal to each other with some CCL field strength.

The results of perturbation were studied in our experiments by means of ionization resonance spectroscopy. We determined the frequencies of maxima in the yield of Ba^+ ions emerging from the ionization of perturbed states. Perturbed states of Ba atoms in these experiments were excited by the joint action of CCL radiation and radiation of a dye laser (DL) with a fixed frequency (ω_2=17735 cm^{-1}). The field strength of DL radiation was equal to $\varepsilon_2 = 4 \times 10^5$ V/cm. The perturbation of *Ba* atoms due to the action of

DL radiation is much weaker than the perturbation of these atoms due to the action of CCL radiation.

The joint action of CCL and DL radiation on *Ba* atoms may give rise to a stimulated Raman scattering (SRS) process, when a Ba atom absorbs one photon of DL radiation and emits one photon at the frequency of CCL radiation. This process leads to the excitation of perturbed $6s5d\ {}^3D_1$ and $6s5d^3D_2$ states. These states were then ionized by CCL and DL radiation. We brought light beams of these lasers into a spatial coincidence and focused them into the center of Ba atoms. Both laser beams were linearly polarized, and the vectors of the light fields in these beams were parallel to each other.

Experimental results

The results of our measurements are shown in Fig.1. As can be seen from these data, three resonance maxima are observed in the yield of Ba^+ ions. The frequencies of CCL radiation corresponding to these maxima are equal to 8705, 8715, and 8725 cm^{-1}. All three maxima in Fig.1 are due to the excitation caused by the joint action of CCL and DL radiation on Ba atoms.

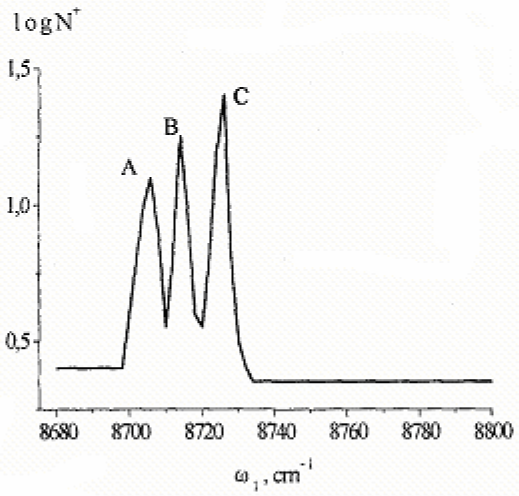

Fig. 1. The yield of Ba^+ ions produced under the joint action of CCL and DL beams on Ba atoms as a function of the frequency of CCL radiation.

Only one of these maxima can be attributed to the excitation of an unperturbed state: the maximum at the frequency ω_1=8705cm^{-1} (maximum *A* in Fig. 1) is related to the excitation of the unperturbed $6s5d\ {}^3D_1$ state through the abovedescribed SRS process (the relevant excitation scheme is shown in Fig. 2). The frequency of CCL radiation corresponding to this process is equal to ω_1 = 8702 cm^{-1}.

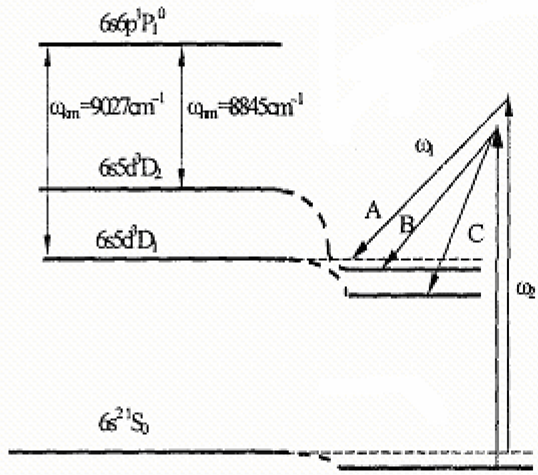

Fig. 2. Diagram of perturbation and excitation of *Ba* atoms. The bold dashed line shows the way the energies of *Ba* atom states change with the increase in ε.

In accordance with selection rules for transitions in the presence of two radiation fields [3], the excitation of the $6s5d^3D_1$ state is forbidden in the case of parallel light field vectors. The appearance of a resonance maximum in this case can be explained by deviations from the parallel orientation of light field vectors and a large interaction volume, where the unperturbed $6s5d^3D_1$ state is populated in the presence of CCL radiation with a moderate field strength.

Now, let us consider the resonance maxima *B* and *C* in Fig.1. The analysis shows that these maxima are related to the excitation of perturbed $6s5d^3D_2$ and $6s5^3D_1$ states in a situation when the shift of the $6s5d^3D_2$ state meets the condition $\delta En \approx \Delta E_{nk}$. The corresponding excitation schemes are labeled as *B* and *C* in Fig. 2.

In accordance with the theory presented in [4], the specific features of the perturbation of two levels n and k with a difference of total angular momenta ΔJ not exceeding 2 are determined in the case when $\delta E_n \approx \Delta E_{nk}$ by the interaction of these two states with the field of laser radiation. In particular, the shifts of each of these levels in such a situation depend on the dynamic polarizabilities of both levels (α_n and α_k) and the matrix element of the two-photon transition between these states:

$$\delta E_n^{/} = \delta E_n + \frac{1}{2}(E_k - E_n) - \sqrt{\frac{1}{4}(E_k - E_n)^2 + V_{nk}^{(2)} V_{kn}^{(2)}}\ ;$$
$$\delta E_k^{/} = \delta E_k + \frac{1}{2}(E_n - E_k) + \sqrt{\frac{1}{4}(E_n - E_k)^2 + V_{kn}^{(2)} V_{nk}^{(2)}}\ . \qquad (2)$$

In formulae (2): $E_{n,k} = E^{(0)}_{n,k} + \delta E_{n,k}$; $\delta E_{n,k} = -\alpha_{n,k}\varepsilon^2/4$: $E^{(0)}_{n,k}$ – the energy of unperturbed n and k states ($E^{(0)}_n > E^{(0)}_k$); $V^{(2)}_{nk} = -[V^{(2)}_{nk}(\omega) + V^{(2)}_{kn}(-\omega)]$ and $V^{(2)}_{kn} = -[V^{(2)}_{kn}(\omega) + V^{(2)}_{nk}(-\omega)]$.

As follows from these formulae, even states with low dynamic polarizabilities may be subject to considerable shifts. In addition, the interaction of such states with the field of laser radiation cannot lead to the intersection of these states. The difference in the energies of such states under these conditions is determined by the relevant two-photon matrix element $V^{(2)}_{nk}$ and $V^{(2)}_{kn}$.

The experimentally observed dependences corresponding to the processes B and C are consistent with the above-described scenario of perturbation. Specifically, the frequencies of both maxima differ from the frequencies corresponding to the excitation of unperturbed $6s5d\,^3D_1$ and $6s5d\,^3D_2$ states. A small frequency difference characteristic of the considered maxima B and C ($\Delta\omega \approx 10\text{cm}^{-1}$) is due to the smallness of the relevant two-photon matrix element.

The fulfillment of the condition $\delta E_n \approx \Delta E_{nk}$ also implies that the states n and k are mixed. The wave function of each of these states in such a situation can be represented as a superposition of the wave functions of both unperturbed states. The fact that the amplitudes of the considered maxima are approximately equal to each other indicates that the effect described above plays a noticeable role in our experiments. If the levels under study were not mixed, the amplitudes of the relevant maxima would considerably differ from each other, since, in accordance with selection rules for parallel polarizations of light field vectors, excitation is allowed for the $6s5d\,^3D_2$ state and forbidden for the $6s5d\,^3D_2$ state [3].

Results of calculation

Using the obtained experimental data and formulae (2) we performed the calculation of dependence of $6s5d\,^3D_1$ and $6s5d\,^3D_2$ states energy ($E^{(0)}_{k,n} + \delta E'_{k,n}$) on radiation field strength ε. The results of calculation are given in Fig.3.

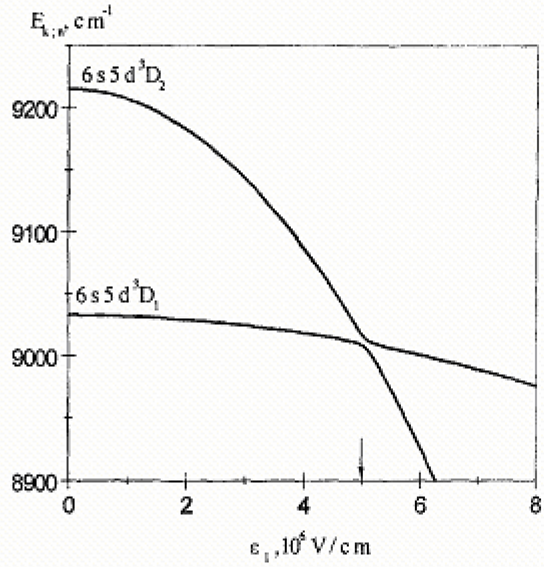

Fig.3.The result of calculation of $6s5d\,^3D_1$ and $6s5d\,^3D_2$ states as function field strength at frequency $\omega \approx 8720\ \text{cm}^{-1}$. The vertical arrow the value ε_1 at which the experimental investigations were performed

In these calculations the matrix elements $V^{(2)}_{nk}$ and $V^{(2)}_{kn}$ were supposed to be not essentiallyy dependent on the frequency. Therefore the product of these matrics elements could be presented as $V^{(2)}_{nk}V^{(2)}_{kn} \approx Z^2\varepsilon^4$. The values of Z and polarisabilities of $6s5d\ ^3D_1$ and $6s5d\ ^3D_2$ states were measured from the position of B and C maxima on the frequency scale and the difference between relevant to these maxima frequensies. These values in atomic units were folowing: $Z \approx 23$; $\alpha_k \approx 4.1\times10^2$ and $\alpha_n \approx 3.6\times10^3$.

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НЕРЕЗОНАНСНЕ ПЕРЕМІШУВАННЯ МЕТАСТАБІЛЬНИХ СТАНІВ АТОМІВ Ва ПІД ДІЄЮ ЛАЗЕРНОГО ВИПРОМІНЮВАННЯ

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Представлено результати експериментальних досліджень збурення атомів під дією лазерного випромінювання за умов, коли енергія рівня, зміщеного внаслідок ефекту Штарка, наближається до енергії сусіднього рівня.