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Effect of resonance mixing of states of a three-level system on the emergence of multiphoton transitions from them

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Abstract. **It is established experimentally for the first time that multiphoton transitions, which are forbidden by the parity selection rules in the dipole approximation, occur from resonantly mixed states in a three-level system with probabilities approximately equal to the probabilities of multiphoton transitions that are allowed by such selection rules.**

Keywords: *resonance mixing of states, three-level system, multiphoton transitions.*

We studied experimentally atomic transitions in the Ba atom from a system of resonantly mixed *n, m, k* states: transitions from the ground ($6s^2\,{}^1S_0$) state, the first resonant ($6s6p\,{}^1P_1^{\rm o}$) state, and the metastable ($6s5d\,{}^3D_2$) state.

We used radiation from two pulsed lasers: a dye laser (DL) and a colour-centre laser (CCL). The radiation frequency ω_1 of the DL was chosen equal to the frequency ω_{mn} corresponding to a single-photon transition between the ground state and the first excited state, i.e., to the $6s^2\,{}^1S_0 \rightarrow 6s6p\,{}^1P_1^{\rm o}$ transition ($\omega_{nm} = 18060$ cm^{-1}). The radiation frequency ω_2 of the CCL varied in the vicinity of the frequency ω_{mk} corresponding to a single-photon transition between the first excited state and the metastable state, i.e., to the $6s6p\,{}^1P_1^{\rm o} \rightarrow 6s5d\,{}^3D_2$ transition ($\omega_{mk} = 8845$ cm^{-1}). The radiation from both lasers was linearly polarised with a parallel orientation of the light vectors. Both radiations were synchronised in time and focused at a beam of Ba atoms. The maximum strengths ε_1 and ε_2 of the field produced by the DL and CCL in the interaction region were 6×10^4 and 2×10^6 V cm^{-1}, respectively.

Estimates obtained by using the data on the probabilities of the $6s^2\,{}^1S_0 \rightarrow 6s6p\,{}^1P_1^{\rm o}$ and $6s6p\,{}^1P_1^{\rm o} \rightarrow 6s5d\,{}^3D_2$ transitions [1, 2] show that the Rabi frequencies Ω of these transitions ($\Omega = V/2$, where $V = d\varepsilon$ is the transition matrix element, and d is the transition dipole moment) are $\Omega_{nm} \approx 25$ cm^{-1} and $\Omega_{mk} \approx 100$ cm^{-1}, respectively, for the above values of ε_1 and ε_2. These frequencies considerably exceed the natural width of the atomic levels and laser linewidths used in our experiments ($\Delta\omega \approx 3$ cm^{-1}).

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It is known that the simultaneous action of two intense resonance radiations on a three-level system should result in resonance mixing of the three states [3]. In this case, all the three levels will be populated. The populations of these levels depend on the ratio of the matrix elements of single-photon transitions $n \rightarrow m$ and $m \rightarrow k$ (V_{nm} and V_{mk}) between the states of the three-level system and on the detunings $\Delta_1 = \omega_{nm} - \omega_1$ and $\Delta_2 = \omega_{mk} - \omega_2$. If the condition $|\omega_2 - \omega_{mk}| > \Omega_{mk}$ is satisfied, the three-level system is transformed into a two-level system. In this case, only the states $6s^2\,{}^1S_0$ and $6s6p\,{}^1P_1^{\rm o}$ with populations $N_n \approx N_m \approx 0.5$ and $N_k \approx 0$ will be mixed.

Upon the simultaneous action of DL and CCL radiation on Ba atoms according to the above scheme, the states $6s^2\,{}^1S_0$, $6s6p\,{}^1P_1^{\rm o}$ and $6s5d\,{}^3D_2$ are not only populated, but transitions from these states to higher-energy states can also occur. We studied such transitions by the method of ionisation spectroscopy. The yield N^+ of Ba$^+$ ions formed upon a simultaneous exposure of a beam of Ba atoms to DL and CCL radiation was measured as a function of the frequency ω_2 of the CCL radiation. We used the experimental setup applied for investigations of resonance processes in two laser radiation fields by ionisation spectroscopy [4].

Fig. 1 shows the results of investigations. One can see that the dependence $N^+(\omega_2)$ exhibits resonance structure. An analysis of the results shows that only peaks B and D can be assigned to normal multiphoton transitions in accordance with the selection rules for dipole transitions. In particular, the peak D can be assigned to several transitions that are induced only by the CCL radiation, or by a simultaneous exposure to the CCL and DL radiation and occur from all the three initial states $6s5d\,{}^3D_2 + \omega_1 + \omega_2 \rightarrow 5d6d\,{}^1F_3(\omega_2 = 8890$ cm$^{-1})$; $6s5d\,{}^3D_2 + 3\omega_2 \rightarrow 6s8p\,{}^1P_1^{\rm o}$ ($\omega_2 = 8892$ cm^{-1}); $6s6p\,{}^1P_1^{\rm o} + 2\omega_2 \rightarrow 6s8p\,{}^1P_1^{\rm o}(\omega_2 = 8916$ cm^{-1}); $6s^2\,{}^1S_0 + \omega_1 \rightarrow 6s6p\,{}^1P_1^{\rm o} + 2\omega_2 \rightarrow 6s8p\,{}^1P_1^{\rm o}$ ($\omega_2 = 8916$ cm^{-1}); $6s5d\,{}^3D_2 + \omega_1 + \omega_2 \rightarrow 5d6d\,{}^3D_2$ ($\omega_2 = 8925$ cm^{-1}) and $6s^2\,{}^1S_0 + 3\omega_2 \rightarrow 5d6p\,{}^1F_3^{\rm o} + \omega_2 \rightarrow 6s7d\,{}^3D_3$ ($\omega_2 = 8940$ cm^{-1}).

The occurrence of several resonance transitions in this case is indicated by a comparatively large width of peak D. The contribution of each of the above-mentioned transitions to the formation of this peak is determined by the population of the initial states and the probabilities of these transitions.

As for the peak B, it is located at the frequency $\omega_{1s} = 8800$ cm^{-1} corresponding to the single-photon transition between the states $5d6p\,{}^1F_3$ and $6p^2\,{}^3P_1^{\rm o}$. It follows from the results presented in Ref [5] that the emergence of a

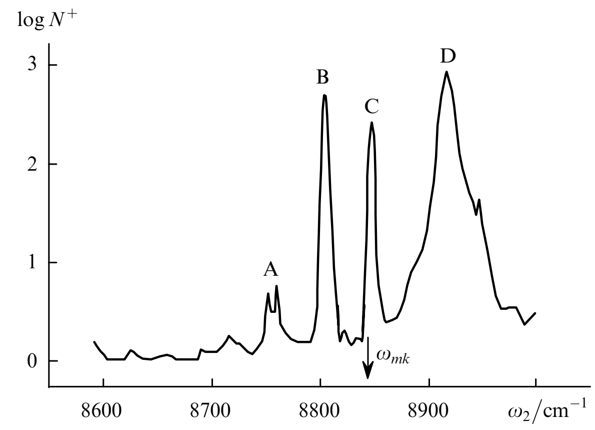

Figure 1. Yield of Ba^+ ions produced upon simultaneous irradiation of Ba atoms by the CCL and DL as a function of the CCL radiation frequency; the vertical arrow indicates the frequency $\omega_{mk} = 8845$ cm^{-1}.

resonance peak in this case is caused by multiphoton excitation and ionisation of strongly perturbed states, the perturbation of the states being due to the dynamic Stark effect under the conditions when dynamic polarisabilities have large absolute values and a strong frequency dependence. We shall not go into the details of the origin of peak B, but simply mention that a detailed description of the mechanism of the emergence of such resonance peaks in the yield of ions is presented in Refs [5, 6].

The remaining peaks A and C on the dependence $N^+(\omega_2)$ cannot be attributed to transitions in which the selection rules for dipole transitions are obeyed. However, the frequencies ω_2 corresponding to these peaks coincide within the CCL radiation linewidth with the frequencies corresponding to the transitions that are forbidden in the dipole approximation by the parity selection rules. The schemes of these transitions are presented in Fig. 2.

The peak A is assigned to the single-photon quadrupole $6s6p\,^1P_1^{\text{o}} + \omega_2 \to 5d6p\,^1F_3^{\text{o}}$ transition. Estimates using typical

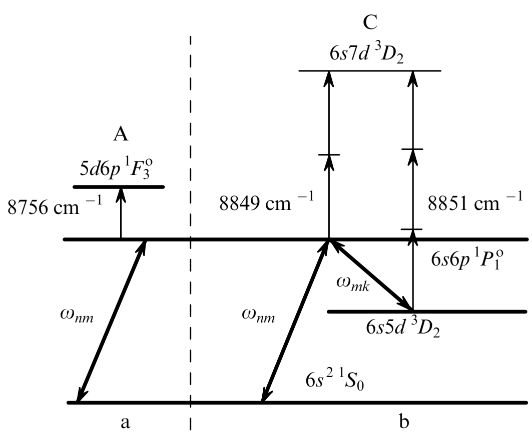

Figure 2. Schemes illustrating the resonance transitions with a violation of the parity selection rule for dipole transitions involving the mixing of states of (a) two-level ($6s^2\,^1S_0$ and $6s6p\,^1P_1^{\text{o}}$) and (b) three-level ($6s^2\,^1S_0$, $6s6p\,^1P_1^{\text{o}}$ and $6s5d\,^3D_2$) systems. Resonance transitions A and C correspond to the resonance peaks shown in Fig. 1; the numbers at the arrows are the CCL frequencies corresponding to these transitions. The thick double-ended arrows indicate the frequencies at which the mixing of states of the Ba atom takes place; the mixed states are shown by thick horizontal lines.

values of the probability of single-photon quadrupole transitions show that this is an unsaturated transition under our experimental conditions. One can see from Fig. 1 that this peak has a distinct doublet structure with the dip at the frequency $\omega_2 = 8755$ cm^{-1}, coinciding with the frequency of the above-mentioned quadrupole transition between unperturbed states.

This doublet structure is caused by the splitting of the initial $6s6p\,^1P_1^{\text{o}}$ state into two quasilevels due to its mixing with the ground $6s^2\,^1S_0$ state in the two-level system. In this case, the difference ($\delta\omega_2 \approx 10$ cm^{-1}) in frequencies ω_2 corresponding to the two components of the above doublet structure is equal to the matrix element $V'_{nm} = d_{nm}\varepsilon'_1$ of the $6s^2\,^1S_0 \to 6s6p\,^1P_1^{\text{o}}$ transition. Since the above-mentioned quadrupole transition is realised for maximum field strengths, the field strength ε'_1 of the DL radiation in this case corresponds to the region in which the highest field strength of the CCL radiation is attained. In the case investigated by us, $\varepsilon'_1 = 1.2 \times 10^4$ V cm^{-1}.

Consider now the resonance peak C. One can see from Fig. 2 that this peak is caused by multiphoton transitions, which are forbidden in the dipole approximation by the parity selection rules. In this connection, it is interesting to note that this peak has a comparatively large intensity. Indeed, one would expect that the intensity of peak C should be smaller than that of peak A because the probability of multiphoton transitions is lower than that of a single-photon transition.

Note that under our ewxperimental conditions, a tuning of the CCL can lead to several transitions (which are forbidden in the dipole approximation) from the same initial states upon absorption of the same number of photons to states with the same quantum numbers as for transitions corresponding to the resonance peak C. However, the dependence $N^+(\omega_2)$ in Fig. 1 does not show the peaks corresponding to these transitions.

The conditions for these transitions differ from those corresponding to peak C. One can see from Fig. 2 that peak C is caused by two transitions from two different states (a two-photon transition from the $6s6p\,^1P_1^{\text{o}}$ state and a three-photon transition from the $6s5d\,^3D_2$ state) to the same $6s7d\,^3D_2$ state. The frequencies ω_2 corresponding to these two multiphoton transitions ($\omega_2 = 8851$ cm^{-1} and $\omega_2 = 8849$ cm^{-1}) coincide within the linewidth of the CCL. At the same time, these frequencies are very close to the frequency $\omega_{mk} = 8845$ cm^{-1} at which resonance mixing of the initial states takes place. Therefore, interference phenomena may play a significant role in the absence of resonance transitions corresponding to peak C. As for the other transitions that are forbidden in the dipole approximation and for which no resonance peaks are observed, they are all caused by solitary multiphoton transitions and their resonance frequencies do not coincide with the frequency ω_{mk}.

Thus, the results of our experimental studies show that resonance mixing of states in a three-level system induces multiphoton resonance transitions from these states upon a violation of the parity selection rules for dipole transitions.

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