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FEATURES OF ATOM SPECTRA Ca, Ba, Sr

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Abstract. *This paper deals with the issues of atomic spectroscopy of multiphoton ionization of alkaline earth atoms. The cycle of researches, which substantially complements the modern understanding of the physical essence of processes, is described. The analysis and generalization of the obtained experimental and available theoretical materials are carried out, and studies on the influence of the correlation of valence electrons on the process of multiphoton ionization of alkaline earth atoms are presented in detail. The appearance of intense intercombination transitions coincides in n with a sharp change in the magnitude of the quantum defect. Since an abrupt change in the magnitude of the quantum defect of a state is caused by the perturbing action of an extraneous level from another configuration, therefore, the appearance of intercombination transitions is due to the effect of the interaction of configurations. The results of experiments and their interpretation are given, namely:*

- the efficiency of excitation of forbidden intercombination transitions and allowed transitions;

- the behavior of alkaline earth atoms within the L-S bond and the classification of the energy levels of these atoms;

-collective effects in atoms and molecules and detection of the phenomenon of violation of the dipole approximation.

The results of an experimental study of the intermediate region of the spectrum of the Ca atom provide information on the relative probability of spin-forbidden transitions and its dependence on the principal quantum number of the excited state.

Keywords: *spectral dependence, three-photon ionization, multi-quantum transitions, two-photon processes, intermediate resonance level, cascade process, spectral width of radiation, transition oscillator, intercombination transitions.*

INTRODUCTION. In the experiments, the results of which are presented above, the process of three-photon ionization of alkaline earth atoms was investigated. When the frequency of the laser radiation was changed, intermediate resonances with bound states were observed, which manifested themselves in a sharp increase in the amplitude of the ion signal.

In this case, the ionization process was of a resonant nature. Since in the future we are going to investigate the probability of excitation of certain states, it is necessary to discuss to what extent information on the population of the excited state can be obtained from such an integral characteristic as the ion yield.

So, if excitation and ionization occur in different fields, moreover, separated in time, then it is obvious that the number of formed ions is proportional, in particular, to the probability of excitation of atoms in the first field. In the case of resonant ionization in one field, this is not so

obvious, since ionization can occur due to the simultaneous absorption of three quanta of radiation.

In other words, this is the question of the relationship between multiquantum and step (or cascade) transitions [1, 2]. Figure 1 shows a diagram of a three-photon transition from the ground state g to continuum 1 with an intermediate two-photon resonance with the r state.

MATERIAL AND METHODS. In the case when the resonance detuning Δ = wrg-2w (where w = wrg = Er– Eg) is much greater than the width of the level r, i.e. at Δ »G, the multiquantum and cascade ionization processes are well distinguishable and the probability of ionization is the sum of the probabilities of these two processes Wn = Wm-Wk.

Strictly speaking, for the same finite electron energy, these two processes should occur at different radiation frequencies. And, conversely, at the same frequency, multi-quantum and cascade processes at A » G will lead to different final energies of electrons (Fig. 1), which in the language of quasi-energies represent an optical repetition of the quasi-energy levels Eg and Er in an external field with a frequency wl - [1,2]. In the case when the detuning is comparable with the width of the resonance level (A ≤ G), the multiquantum and cascade processes lead to the same final electron energy and one should add not the transition probabilities, but the amplitudes

$A_{gi} = A_{gr} A_{ri} + A'_{gi}$,

where $A_{gi}= V^{(3)}_{gi}$, $A_{gr}=V^{(2)}_{gr}$, $V^{(k)}_{m1}$ - composite matrix element K- th order. In this case, the transition probability will be equal not to the sum of the squares of the amplitudes, but to the square of the sum.

Consequently, interference terms will appear, which, in fact, do not allow separating ionization into multiquantum and cascade, but require consideration of a single process of transition of an atom to their ground state

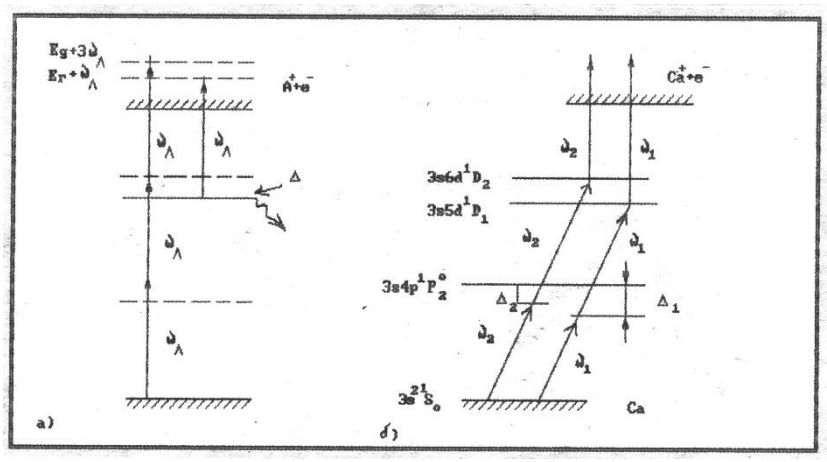

Fig. 1.

a) Scheme of three-photon ionization of a Ca atom with an intermediate resonance

b) Scheme of three-photon ionization of a Ca atom with an intermediate resonance with the levels 3s5d1D2 and 3s6d1D2 at frequencies w1 and w2, respectively g into final I with the absorption of three quanta [1,2].

However, even in this case, one can try to introduce a quantitative criterion that will make it possible to single out one of the processes under consideration, similar to how it was done in

Ref. [13], for resonant Raman scattering, comparing the lifetimes of the excited state caused by stimulated and spontaneous processes.

1. Identification and analysis of dispersion dependences of atoms

The lifetime in an excited state can be characterized by three quantities: the spontaneous relaxation time τδ or the natural width of the Gδ level, the time of the forced transition to the ground state τf or the field width Gf, and the ionization time τi by the ionization width G_i.

If the width due to spontaneous relaxation is greater than the field and ionization widths Gδ> Gf, Gi, then the idea of the excitation of an atom to a real state is justified, and in the first approximation we can speak of a cascade process consisting in the excitation of a level and its subsequent ionizationzations.

This representation, however, is valid only when the characteristic time τ of the ionization process is much larger than the reciprocal width of the level: τ »G-1 = τδ. In the case of τ ~ G, even if the condition Gδ »Gf, Gi is satisfied, the separation into multiquantum and cascade processes is impossible and they must be considered simultaneously. At τ «G, the multiquantum process of resonant ionization dominates.

This can be shown more clearly as follows. The probability of a cascade ionization process is equal to the product of the probability of excitation of the r - Wgr state by the probability of ionization from this state Wri (Fig. 1): Wk - Wgr - Wri. These probabilities are equal [8]:

$$w_{gr} = 2\pi \frac{G_\delta}{\Delta^2+1/4G_\delta^2}\left|v_{gr}^{(2)}\right|2\tau; w_{ri} = 2\pi|v_{ri}|2\tau \qquad (1)$$

Therefore, the total probability of a cascade transition is:

$$w_{\text{kack.}} = 4\pi^2 \frac{\left|v_{gr}^{(2)}\right|2|v_{ri}|2\, G_\delta \tau^2}{\Delta^2+1/4\, G_\delta} \qquad (2)$$

(here in after, it is taken into account that Gδ »Gi, Gf)

The probability of resonant ionization in a weak monochromatic field can be represented as [9]

$$w_{м} = 2\pi \frac{\left|v_{gr}^{(2)}\right|2|v_{ri}|2\, G_\delta \tau}{\Delta^2+1/4\, G_\delta^2} \qquad (3)$$

The ratio of the probabilities of cascade and direct ionization in the case of exact resonance Δ = 0 is:

$$w_{\text{kack}} / w_{\text{mn}} = 2\text{п}\ G\tau, \qquad (4)$$

It is clearly seen from (4) that if the characteristic ionization time is much greater than the reciprocal width of the level, that is, G δτ »1, then the process of resonant ionization is of a cascade nature; with the inverse ratio Gδτ« 1, the resonant multiphoton process prevails.

In the intermediate case Gδτ ~ 1 it is impossible to distinguish a multiquantum or cascade process, since the interference terms in the expression for the total probability will be essential. In a situation that is far from saturation Witl «1, the characteristic time of the ionization process will be determined by the duration of the laser pulse τ ~ tl.

For an arbitrary ratio of the widths Gδ, Gf, Gi at large τ "G-1 and τ" G-1 times (G = max {Gδ, Gf, Gi}), the resonant ionization process was considered in detail in [13, 12], from the

results of which It follows that the use of simple resonance formulas of the Breit-Wigner type (4) with one or another resonance width to describe the linear (in time) ionization regime in the general case is impossible.

Each specific situation realized in the experiment requires a separate consideration.

Let us consider the experimentally realized process of ionization of Ca using the example of two resonances with levels 4s5d1D2 and 4s6d1D2 at frequencies w1 = 21459.5 cm-1 and w2 = 22494.9 cm-1. For the indicated levels, the probabilities of radiative decay and the oscillator strengths of transitions to the lower states are known [4], and therefore the most complete numerical estimates are possible.

The natural widths of these levels, determined by spontaneous relaxation, are equal: for 4s5d1D2 - Gδ = 4 10-3cm-1, for 4s6d1D2 - Gδ = 8 10-4cm-1. The nonresonant change in the energy of excited states in the field ε = 3-104 V cm-1, estimated by the asymptotic formula [13] δE = 1/4 ε2 / w2, is equal to δEδd ≈ 2.2 · 10-5 cm-1 and δEGd ≈ 1.3 · 10-5 cm-1.

The polarizability of the ground state of the Ca atom at frequencies w1 and w2 is approximately the same and is equal to αo = 195 AU. [8]. Hence, the change in the energy of the ground state is δE0 = -1/4 α E2 = - 2 · 10-10 a.e. = 4-10-5 cm-1. As can be seen, the nonresonant change in the energy of the levels due to the dynamic Stark effect is much smaller than the natural widths of the excited states and, therefore, they can be neglected.

The field width of levels 5d and 6d will be determined by the two-photon Rabi frequency [12]: $G_{fdsd} \sim \Omega^{(2)} = 1[2\sqrt{\Delta^2 + 4}\left|v^{(2)}\right|^2$, those. just like the nonresonant level shift, it quadratically depends on the field and at exact resonance *(*$\Delta = E_d - 2w = 0$*)* is equal to $G_f = |\, v_{\delta d}^{(2)}| = \frac{\left|D_{\delta p}\right|\left|D_{pd}\right|}{\Delta}\varepsilon^2$, where D is the dipole matrix element of the transition, Δ_p - detuning with intermediate state

$4s4p^1P_1^0$. The matrix elements of the transitions can be estimated from the known oscillator strengths $f_{ik} = -\frac{2w_{ik}}{2j+1}\left|<1\left|D\right|k>\right|^2$, where wik is the frequency of transition to → 1 J is the full moment of the upper level. To go $4s^2\ {}^1S_0$-$4s4p^1P_1^0$, $f_{\delta p}$ =1.75, for $4s4p^1p_1^0$ - $4s5d^1D_2$, f_{pd} = 0,27; and for $4s4p\ {}^1p_1^0$ -$4s6d^1D_2$, f_{pd} = 4,4-10^{-2} [49[. The estimates give the following field widths: for the level 5d – $G_f \approx 9\cdot10^{-4}$ sm^{-1}, for 6d – $G_f \approx$ 8,4-10^{-4} sm^{-1}. The fact that the field widths for the 5d and 6d levels are almost the same is explained by the fact that the difference in the strengths of the oscillators $f_{pd1} > f_{pd2}$ compensated by the difference in detunings $\Delta_{p1} > \Delta_{p2}$ with intermediate level $4s4p^1p_1^0$ at frequencies w1 and w2.

The ionization width of the levels is $G_i = 2\pi\left|v_{dE}^{(2)}\right|^2$. For non-hydrogen-like atoms, the VdE value can be calculated by semiempirical quantum defect methods using the Burgess – Seaton formula given in [5]. The resulting estimates give the following widths: for the level 5d – G_i = 8,3-10^{-5}sm^{-1}, for 6d – G_i= 4,1-10^{-5}sm^{-1}.

Thus, it follows from the derived widths that, under the conditions of our experiment, the level shift due to the dynamic Stark effect and the ionization width are an order of magnitude smaller than the field and natural widths, which are of the same order of magnitude. If for the level 5d - $G_\delta \approx 3\,\Gamma_f$, then already for 6d - $G_\delta \approx G_f$, and for levels with a large value of Ω, the field width becomes larger than the natural width $G_f \gg G_\delta$. This is due to the different dependence on the principal quantum number

$G_\delta \sim \Omega^{-3}$ [134], $G_f \sim \Omega^{-3[2}$ [13]. The performed analysis shows that in our case $G_f \geq G\delta$, $2\pi G\tau \sim 1$ and it would seem that the division into multi-quantum and cascade processes is not justified. However, in deriving criterion (4), the non-monochromaticity of the laser radiation was not taken into account. The dye lasers used in the experiment have a spectral emission width $\Delta w \sim 1$ sm^{-1}, which is much larger than all the widths calculated above. A detailed theoretical analysis of three-photon ionization with a two-photon intermediate resonance in a nonmonochromatic field was carried out in Ref [13].

2. Ionization of neighboring triplet and singlet states

As a result of the analysis, the authors of [13] come to the conclusion that in the limit of large widths and low intensities ($\Delta w \gg G$), the coupled-coupled transition to the resonant state is not statistically associated with the transition from the excited state to the continuum. That is, the ionization process can be considered as consisting of two stages - excitation and subsequent ionization.

3. Ionization level width

In other words, if the characteristic interaction time is longer than the laser radiation coherence time, then the whole process will be incoherent. Two-photon excitation will nevertheless occur coherently due to the short lifetime of the intermediate virtual state, and the ionization step is not statistically related to the excitation step.

Pulsed spectral width multimode dye laser $\Delta w \sim 1$sm^{-1} has a coherence time $\tau_{\text{ког}} \sim (\Delta w)^{-1}$ 33 Ps which is small compared to the pulse duration (10 ns) and, therefore, in our case ($\Delta w \gg G_\delta$, G_i, G_f, δE), the process of resonant ionization will certainly not be coherent. Thus, for nonmonochromatic radiation, criterion (4) must be rewritten in the form $\Delta w\tau \gg 1$, provided that the field is weak (i.e. $\Delta w \gg G_i, G_f, \delta E$). Taking into account all of the above, we have carried out numerical estimates of the total probability of resonant ionization through the level 5d - W = 6,7-10^{-2}см$^{-1}$ and through the level 6d - W = 7,9 · 10^{-2} см$^{-1}$. At a density of atoms $n_0 = 10^9$см$^{-3}$, focus volume 10^{-4}sm^{-3} and the ratio of the amplitude of the signal from the detector and the number of produced ions

1 мВ -10 ions, the obtained probabilities correspond to ion signals with amplitudes $A_{5d} \sim$ 700мВ, A_{6d} 800 мВ, which is in good agreement with experiment.

From the relative yield of ions to resonance with singlet and triplet states, provided that the strength of the oscillator of the transition to the singlet state is known, it is possible to estimate the strength of the oscillator of the transition to the triplet state.

Indeed, resonances with triplet and singlet states having the same principal quantum number occur at close frequencies, the values of the effective principal quantum numbers of these states differ little, and therefore it can be assumed that the ionization of neighboring triplet and singlet states occurs with almost equal weight -peakness.

Consequently, the difference in the amplitudes of the resonances is associated with the difference in the probabilities of excitation of triplet and singlet states. For the state 4s6d^{1}D$_2$ the strength of the transition oscillator is known 4s4p^{1}P$^0_1$-4sd^{1}D$_2$, f^{δ}_{pd} = 4,4-10^{-2} [48]. From the ratio of the amplitudes of resonances with triplet and singlet states 6d, we estimated the oscillator strength of the intercombination transition 4s4p^{1}P°-4sd^{3}D$_2$, f^{T}_{pd} = 2,3·10^{-3}. Unfortunately, for other resonances in the Ca atom and for all those registered in Sr atoms; Ba, we do not know the oscillator strengths of the allowed transitions. Therefore, to obtain the values of the oscillator strengths for other intercombination transitions without absolutizing the parameters of laser radiation, atomic beam and detector is not possible. But, as will be shown below, valuable

information can also be obtained from the relative values of the probability of intercombination transitions. For example, by tracing how this probability changes along the n0 and series of triplet states.

The degree of localization of such deviations from regularity contains information about the number of interacting levels of various configurations. The magnitude of the quantum defect can serve as a measure of how strongly the state is perturbed.

$\mu = n - n^*$ ($n^* = \sqrt{R_y / E_n}$, n^* - effective principal quantum number. Shows the dependences of the magnitude of the triplet (dashed line) and singlet (solid line) levels n0 and of the series of Ca, Sr, and Ba atoms on the value of the principal quantum number n. All three dependences demonstrate a sharp change in the magnitude of the quantum defect with the growth of p.

RESULTS. It is known that in the one-electron approximation, disregarding the interaction of configurations, the magnitude of the quantum defect weakly depends on n, decreasing monotonically with increasing n [13]: $\mu = \mu_0 + \mu_1 / n^2 + \mu_2 / \mu^4 + \ldots$. The jumps in the magnitude of the quantum defect with a change in n can only be caused by the effect of the interaction of configurations. The dependences shown in show that in the regions of the spectrum of Ca, Sr, and Ba atoms studied by us in the experiment, the one-electron states n0 and 1,3D2 are very strongly perturbed and the one-electron approximation is not applicable to their description.

The dependences μ (n) contain empirically obtained information on the nature and strength of the interaction of configurations and will be used by us in the future to interpret the experimental results.

CONCLUSION The results of an experimental study of the intermediate region of the Ca spectroatom provide information on the relative probability of spin-forbidden transitions and its dependence on the principal quantum number of the excited state. In the Sr atom, resonances with triplet states corresponding to the two-photon intercombination transitions 5s2 1S0 - 5snd3D2 are considered. As with Ca, the amplitudes of these resonances vary greatly depending on the value of the principal quantum number of excited states. The appearance of intense intercombination transitions coincides in n with a sharp change in the magnitude of the quantum defect. Since an abrupt change in the magnitude of the quantum defect of a state is caused by the perturbing action of an extraneous level from another configuration, therefore, the appearance of intercombination transitions is due to the effect of interaction of configurations.

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