Investigation of the perturbation of atomic states in an elliptic-polarization field

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We have observed, for the first time, multiphoton resonance in an atom in a field of elliptic polarization. The results yield important data concerning the composite matrix elements that determine the dynamic polarizability of the resonating levels.

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Investigation of the perturbation of atomic states in an elliptic-polarization field by the multiphoton-resonance method uncovers great possibilities for the experimental measurement of composit matrix elements of atomic states. We report here the first experiments in this direction.

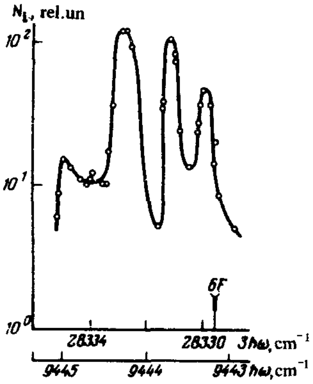

FIG. 1. Dispersion dependence of the number of ions $N_i(\omega)$ produced by four-photon resonant ionization of Cs atoms from the ground state 6s (three-photon resonance is produced in the transition 6s–6f) in the radiation field of a single-mode single-frequency neodymium laser with elliptic polarization (ellipticity angle $\theta = 32°$). The energy scale at triple the neodymium-laser frequency is also shown. The position of the 6f level (reckoned from the position of the 6s level) in the absence of a field is marked.

The hitherto-performed experimental investigations of multiphoton excitation of atoms were limited to the use of either linearly[1] or circularly[2] polarized radiation. Since the ground state of the atoms investigated in these studies has a zero orbital-momentum projection, the dependence of the excitation probability on the laser frequency was characterized by a single resonance peak corresponding to excitation of a single magnetic sublevel with zero angular-momentum projection in the case of linear polarization or projection $\pm n$ for right-hand (left-hand) circular polarization (n is the multiplicity of the resonance). Therefore the data obtained on the position of the resonance did not make it possible to draw unambiguous conclusions concerning either the relative shift of each of the resonating levels separately, or the splitting of the magnetic sublevels of the excited states of the atom in the laser field.

The use of elliptically-polarized radiation permits direct measurement of the two indicated quantities, for in this case the external field has no axial symmetry and the magnetic quantum number selection rules for successive absorption of several photons are much less stringent. The quasi-stationary states of the atom in such a field are characterized no longer by magnetic quantum numbers, but only by the parity of these numbers.[3] 1) When an S-state atom absorbs an even or odd number of photons, excitation takes place of all the quasi-stationary states made up of the initial states of the free atom with even or odd magnetic quantum numbers, respectively. As a result, multiphoton-excitation the probability dispersion curve has already many resonances, as many as there are quasi-stationary states of the atom with given parity of the magnetic quantum numbers.

We measured the change of the energy of the 6s–6f transition in the Cs atom in the elliptically-polarized field of the emission of a single-frequency single-mode neodymium laser. The frequency of the ionizing radiation was chosen such as to produce three-photon resonance on the 6s–6f transition. At the same time, four-photon ionization of the Cs atom took place. We measured the dependence of the yield $N_i(\omega)$, of the four-photon ionization of the Cs atom on the emission frequency near resonance, a dependence that reflected fully the structural features of the multiphoton-excitation probability dispersion.

The experimental setup was similar to that customarily used in investigations of multiphoton ionization.[1] The focused radiation of the single-mode single-frequency laser interacted with a beam of Cs atoms ($p \sim 10^9$ cm^{-3}). The radiation frequency was varied in a range of 60 cm^{-1}. The laser emission was line width ~ 10^{-2} cm^{-1}. The plane-polarized radiation of the neodymium laser passed through a quarter-wave quartz plate, which could be oriented to obtain the required degree of ellipticity of the radiation polarization.

The figure shows a plot of $N_i(\omega)$ measured in the experiment at a field intensity ~ 10^6 V/cm and an ellipticity angle $\theta = 32°$ (the ratio of the ellipse axes is equal to $\tan\theta$).

In a field of the indicated intensity, the perturbation of the 6f level greatly exceeds the spin-orbit interaction (0.1 cm^{-1}), which can therefore be completely neglected. When three photons are absorbed, only four odd magnetic states 6f (out of a total number $2J+1=7$) are excited: $M \pm 1$ and ± 3, as is fully confirmed by experiment.

We recall that measurements in fields with a linear and circular polarization in the same range of frequencies and field intensities yield dispersion curves having single resonance peaks, corresponding to three-photon resonance between the levels $6s_{m=0} - 6f_{m=0}$ in a linear-polarization field[1] and $6s_{m=0} - 6f_{m=\pm 3}$ (the sign depends on the direction of the photon helicity) in a circular-polarization field.[2]

In the investigated case, for each peak of the $N_i(\omega)$ plots we can calculate the polarizability $\alpha_{\Sigma}^{(k)} = 4\Delta_k/F$ (Δ_k is the position of the maximum relative to the unshifted position of the 6f level, and F is the radiation intensity), which is, however, the difference between the polarizabilities of the 6s level and one of the quasi-stationary states of the 6f level: $\alpha_{\Sigma}^{(k)} = \alpha_{6s} - \alpha_{6f}^{(k)}$; $k = 1, 2, 3, 4$. All the quantities $\alpha_{6f}^{(k)}$ are determined by the three atomic parameters U, V, and W which have been introduced in[3] and are expressed in terms of the composite matrix elements of the atom. Therefore, by using the measured positions of the peaks of the $N_i(\omega)$ plot, we can obtain these parameters of the 6f level as well as the polarizability of the 6s level. The results are shown in the first row of the table. The second row lists the theoretical values of the same parameters,

	α_{6s}	U_f	V_f	W_f
Experiment	1240	933	− 540	199
Theory	1500	674	− 883	19

*All the quantities are given in atomic units.

calculated with the aid of the Green's function of the atom in the approximation of the quantum-defect method.[4]

Comparing the results of the theory and experiment, we can state that they are in full agreement for all the quantities except the parameter W. It must be emphasized here that when the parameters U, V, and W are made up from the matrix elements of the atom, one or two significant figures are lost, a phenomenon typical of the dynamic polarizabilities of highly-excited states. In the actual experiment we therefore compare small differences between theoretically calculated parameters, and the theoretical model must therefore be sufficiently accurate. All these reasons can give rise to a discrepancy between the theoretical and experimental data.

The first experimental measurement, by the multiphoton-excitation method, of the perturbation of atomic states in an elliptically polarized field has shown that such investigations can be used in the future to measure the composite matrix elements of the atom, which play the same role in nonlinear spectroscopy as the oscillator strengths in classical optics.

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[1)] The conservation of the parity of magnetic quantum numbers is a consequence of the invariance of the Hamiltonian of the atom in an elliptic field to rotations through 180°, in analogy with the same properties of a paramagnetic ion in a crystal of rhombohedral symmetry.

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