A mechanism of two-electron multiphoton ionization of atoms

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A mechanism for the formation of doubly-charged ions, which were observed experimentally in the multiphoton ionization of a number of atoms, is proposed.[1] This mechanism is based on the diffusive ionization of excited atoms,[2] which in this case is realized in the optical frequency range.

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Recently, a new effect was observed experimentally—the formation of doubly-charged ions as a result of multiphoton ionization of strontium, samarium, europium, and barium atoms.[1] In all these cases, the doubly-charged ions were produced simultaneously with the singly-charged ions with a probability W, that was smaller by only one to two orders of magnitude. The nonlinearity in the formation of doubly-charged ions $K_e^{2+} = \partial \log W / \partial \log \mathscr{E}^2$ in all cases was much less than the number of quanta whose absorption was necessary to satisfy the law for conservation of energy and, in the case of the barium atom, was even less than the nonlinearity for singly-charged ions. Both experimental facts - the relatively large probability for formation of doubly-charged ions and the small nonlinearity - cannot be explained even qualitatively in terms of the perturbation theory. However, the experimental facts can be explained if it is assumed that ionization of the two-electron excited bound state occurs as a result of diffusion of the electron along the quasicontinuum of the highly excited levels. The diffusion mechanism for ionization of excited atoms which was substantiated by Delone *et al.*,[2] was used to describe the results of the experiments on ionization by a field in the SHF range.[3] Here we show that this mechanism is also valid in the optical-frequency range.

Three conditions must be satisfied for the diffusion mechanism to be effective. First, the intensity of the field must be high enough to completely eliminate degeneracy. Second, it is necessary that the electron transition from the state with a quantum number to the continuous spectrum have a multiphoton character. Third, there must be an independent broadening, which determines the formation of a quasicontinuum of excited levels. The broadening apparently must exceed the distance between the excited levels, i.e., it must have the value $\Delta\omega \gtrsim Z^2/Ryn^5$. The broadening may be attributable to the finite width of the spectrum of the exciting field, to the atomic collisions, and to other factors.

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In satisfying the indicated conditions the electron goes from the level with principal quantum number n to the continuous spectrum due to the large number of quanta of single-photon absorption and the emission of quanta in the quasicontinuum. The probability of this process, which is described by a diffusion equation,[2] is proportional to the first power of the radiation intensity.

For the atoms with the two outer electrons of interest to us, the diffusion mechanism for formation of doubly-charged ions in the optical-frequency range is realized at $n=6$, field intensity $\mathscr{E}=5\times10^{6}$ V/cm and $\Delta\omega\approx10$ cm^{-1}, i.e, under conditions corresponding to an experiment[1] in which multimode laser emission with a spectrum width $\Delta\omega=14$ cm^{-1} was used. As the calculations show, the probability of diffusion ionization is $W_{nE}\sim10^{15}$ sec^{-1}, i.e., during the laser pulse ($\sim10^{-8}$ sec) all the excited atoms are ionized. Consequently, the probability of atomic ionization is determined by the probability for excitation of the resonance state which causes a small nonlinearity and a high probability for formation of doubly charged ions, which is observed experimentally.

The singly charged ions are produced as a result of the competing process of single-electron multiphoton ionization which may be of a direct nature (for strontium and europium atoms) and of a resonance nature (for a barium atom). We note that for the onset of resonances with sufficiently high single-electron states, including $n=6$, the multi-quantum condition is not satisfied, and, therefore, diffusion does not occur. Thus, the process of multiphoton single-electron ionization can be described within the framework of perturbation theory.[4]

A quantitative description of the experimental data based on the proposed model was done by us for the barium atom. In this case the removal of one electron requires an absorption of $K_0^{+}=5$, and the removal of two electrons requires an absorption of $K_0^{2+}=13$ quanta of radiation. A nonlinearity of $K_e^{+}=4$ and $K_e^{2+}=3$, was observed experimentally and the probabilities for formation of the Ba^{+} and Ba^{2+} ions have the same order of magnitude at a field intensity of $\mathscr{E}\approx7\times10^{5}$ V/cm.[1] It can be seen from the spectrum of the barium atom that there is a four-photon resonance between the ground state and the two-electron excited state $5d\,6d$. Since the width of the excited state is determined by the diffusion ionization mechanism, i.e., it is equal to $\sim\mathscr{E}^{2}$, according to the well-known equation describing the resonance ionization,[4] we can see that $K_T^{2+}=3$, in agreement with the experiment. The four-photon resonance also occurs in the singly charged ions; moreover, since the single-photon transition from the resonance state to the continuous spectrum occurs in atomic time, it does not affect the total probability. The probabilities for production of the singly and doubly charged ions, which are determined by the four-photon excitation of the resonance states, were calculated according to the perturbation theory in the approximation of the quantum defect . They turned out to be equal at a field intensity $\mathscr{E}\approx5\times10^{5}$ V/cm, in good agreement with the experimental data.

In conclusion, we note that the assumption[5] that the diffusion-ionization model is not applicable to highly excited states is based on a misunderstanding. In making this claim the authors[5] assumed that the excitating field is highly monochromatic. Delone *et al.*[2] assumed, however, that external perturbations, which are responsible for the quasicontinuum, were present. In particular, a drift broadening of the atomic levels due to the motion of the atom in the standing-wave field of a microwave cavity occurs in the experiments on ionization of highly excited hydrogen in the SHF range,[3] which were discussed by Delone *et al.*[2]

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