Frequency dependence of multiphoton ionization of Xe and Kr atoms

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The wavelength dependence of multiphoton ionization of Xe and Kr atoms at an electric field strength $E \sim 5 \cdot 10^7$ V/cm is investigated. It is found that on weak variation ($\sim 10^{-3}$) of the wavelength around $\lambda = 1.06\ \mu$, the dependence of the number of ions produced on radiation intensity changes sharply. Approximation of the dependence by a power law yields values of the exponent K that vary from K_0, where K_0 is the number of quanta required for ionization, to values about half as large. The variation of K is of a resonant nature. The resonances observed are interpreted as a manifestation of the spectrum of electron bound states in an atom strongly perturbed by a light field.

1. INTRODUCTION

The probability of the process of multiphoton ionization of an atom is connected with the radiation intensity F by the relation

$$W = \alpha F^{K_0}, \tag{1}$$

if no resonance intermediate between the energy of several quanta and the energy of the bound state of the electron in the atom is produced upon ionization[1,2]. The exponent $K_0 = \langle I/\hbar\omega + 1 \rangle$ in (1) is the number of quanta absorbed upon ionization, while the constant α describes the cross section of the multiphoton process. The multiphoton ionization processed by relation (1) was observed experimentally in the ionization of alkali-metal atoms by neodymium-laser radiation and by its second harmonic, under conditions when there were no resonances[3,4].

When atoms of noble gases are ionized by neodymium-laser radiation, relation (1) does not hold[5,6]. In practically all the investigated cases, the approximation of the experimental data by a power law yields a value $K < K_0$. The nonsystematic character of the difference between the observed value of K and the corresponding values of K_0 had led to the conclusion that an important role is taken in this case by the spectrum of the bound states of the electron in the atom[5]. One of the causes of the change in the functional dependence may be the onset of intermediate resonance between the energy of several quanta and the energy of the bound state of the electron in the atom[1]. Experiments on multiphoton ionization of a potassium atom[7] have shown that when the intermediate resonance sets in, an abrupt change takes place in the form of the functional dependence of the number of ions produced on the radiation intensity. If the functional dependence is approximated by a power law, exponents $K < K_0$ are obtained.

Unlike the case of multiphoton ionization of potassium atoms, we cannot uniquely interpret the results of the experimental investigations of multiphoton ionization of noble-gas atoms, and for a number of reasons. From a comparison of the spectra of the unperturbed levels of the atoms of the noble gases with the energies of n quanta ($n < K_0$) we see that the situations are much more complicated in these cases than in potassium. In a number of cases, the energy of several quanta differs negligibly from the energy of the bound states of the electron in the atom. Taking into account the relatively large width, ~10—15 cm^{-1}, of the emission spectrum of the laser radiation used in[5,6], it can be assumed that in many cases resonance sets in between the energy of several quanta and the energy of the bound state of the electron in the atom.

From our point of view, however, such comparisons do not yield significant information. The point is that the multiphoton ionization of noble gases is observed at a relatively large electric-field intensity, ranging from 10^7 to 10^8 V/cm. In such a strong field, the spectrum of the atom is appreciably perturbed. Unfortunately, we do not have at present a theoretical description of the perturbation of the spectrum of the levels of noble-gas atoms in a strong optical field. The principal difficulty is that perturbation theory cannot be used in this case. The criterion for the validity of the use of perturbation theory for the description of the final state of an electron extracted from an atom is given by[8]

$$E/E_0 \ll K_0^{-3/2}, \tag{2}$$

where E is the intensity of the optical field and E_0 is the intensity of the atomic field.

The criterion (2) is based on the assumption that the electron is emitted from the atom with an energy $\hbar\omega$, which is the upper bound of the electron energy; accordingly, Eq. (2) yields an estimate of the upper limit of applicability of perturbation theory. From (2) at $K_0 \sim 10$ it follows that perturbation theory is applicable at field intensities $E \ll 10^8$ V/cm.

The establishment of the criterion for a bound state of an electron in an atom reduces to a solution of the problem of the Stark shift of the atomic levels in a nonresonant alternating field. It is known that in this case one can write in analytic form only a general relation[9], from which one can conclude neither the sign nor the magnitude of the shift of any concrete level. To obtain such data it is necessary to perform complicated numerical calculations[10]. Rough estimates of the situation can be made, however, by calculating the perturbation of the first levels in the hydrogen atom[11]. From the results of that paper it is seen that the perturbation increases sharply with the principal quantum number, like n^6. A numerical estimate shows that at a field intensity $E \sim 10^7$ V/cm the change of the energy of the state reaches a value on the order of the distance to the nearest state already at n = 2.

It is seen from the foregoing estimates that the use of perturbation theory to describe multiphoton ioniza-

tion of an atom at a field intensity $E > 10^7$ V/cm can lead to incorrect results.

Most interesting from our point of view is the fact that in a strong optical field one should expect a qualitative change in the spectrum of the bound states of the electron in the atom[11,12]. It should be noted that a strong field is also typified by the fact that the probability of ionization from excited states is very large. It is therefore practically impossible to use the traditional methods of investigating the atomic spectrum, methods connected with the finite lifetime in a fixed state or with the spontaneous decay of this state. It is therefore just the multiphoton ionization process which is the most promising for the study of the spectrum of an atom in a strong field. One can expect an experimental investigation of this process to yield additional information on the character of the spectrum of bound states of an electron in a strong optical field.

2. THE EXPERIMENT

As the first stage in investigations we decided to ascertain the frequency dependence of the process of multiphoton ionization of noble-gas atoms in a strong optical field. We chose the Xe and Kr atoms for the investigation. The experiment consisted of measuring the dependence of the ion yield on the radiation intensity at different fixed frequencies of the neodymium laser.

We used a multimode neodymium laser with variable generation frequency. The Q-switch was a rotating prism. The generation frequency was varied with the aid of two Fabry-Perot interferometers placed in the laser resonator[7]. The frequency could be tuned from 9424 to 9448 cm^{-1}. The intensity distribution in the generation line was close to Gaussian with a width ~ 3 cm^{-1} at half height. The radiation energy in the pulse was ~ 1 J. We used an amplifier with an energy gain ~ 3, operating in a traveling-wave regime. The laser radiation was focused into the interior of the vacuum chamber by a lens of focal length ~ 60 mm and relative aperture $\sim 1/5$. The distribution of the radiation in the focusing region was measured by the standard procedure, using an analogous lens mounted the same distance from the laser. Independent measurements were made of the spatial distribution, by a photometric method, and of the temporal distribution, with a high-speed photocell[13].

The laser radiation entering the vacuum chamber was attenuated with filters of colored optical glass. The linearity of the filter attenuation was verified against the laser radiation. The radiation energy passing through the focusing region was measured with a calorimeter. A typical diameter of the focusing circle was $\sim 10^{-3}$ cm; the corresponding volume effective in the production of ions in the process connected with the absorption of ~ 10 quanta was $V_{10} \sim 10^{-8}$ cm^3. The typical duration of the giant pulse was $\tau \sim 30$ nsec, corresponding to $\tau_{10} \sim 3$ nsec.

The vacuum chamber was filled with the investigated gas to a pressure 10^{-4} Torr, corresponding to a density $n_0 \sim 10^{12}$ cm^{-3}. The ions produced in the light-focusing region were accelerated with a constant electric field $\sim 10^3$ V/cm, mass-analyzed with a time-of-flight mass spectrometer, and registered with an electron multiplier. The gain of the electron multiplier was monitored against a Faraday cylinder.

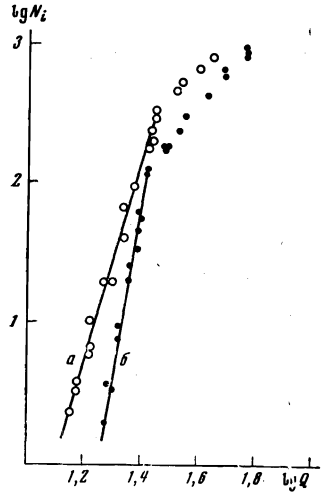

FIG. 1.

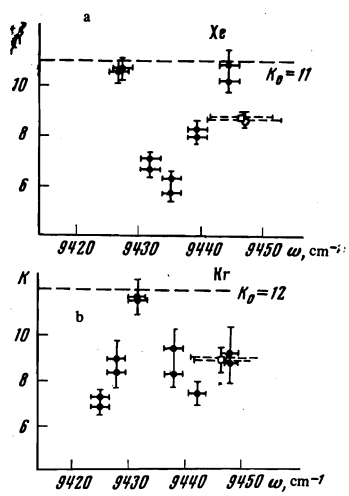

FIG. 2.

FIG. 1. Dependence of the ion-signal amplitude N_i on the radiation energy Q (in relative units). passing through the gas (Kr) in the laser pulse. Data are given for two radiation frequencies: a—for $\omega = 9425$ cm^{-1} ($K = 6.9 \pm 0.3$); b—for $\omega = 9431$ cm^{-1} ($K = 11.6 \pm 0.8$).

FIG. 2. Exponent K of the power law (1) vs. the radiation frequency for the xenon (a) and krypton atoms (b). For comparison we show the data obtained earlier in[5] (○) and in [6] (□); the half-widths of the generation line are indicated.

Special electronic apparatus was used for automatic cyclic operation of the laser and maintained the principal parameters constant. Thus, we obtained a laser operating regime in which the radiation intensities in the focal region were identical to a high degree in a large number of successive pulses. This permitted us to measure, in each pulse, only the energy passing through the focal region, since the size of the focusing circle and the duration of the pulse remained constant with sufficient accuracy.

The direct result of the experiment consisted of determining the dependence of the number of produced ions on the energy passing through the focal region. This dependence was measured for a number of fixed laser-radiation frequencies. Figure 1 shows typical experimental results in a log-log scale. The change in the slope of the plot of $\log N_i = f(Q)$ with increasing radiation invensity is due to the ion-yield saturation that sets in when the effective ionization approaches 100%. Further increase of the ion signal with increasing radiation intensity is due to the uneven distribution of the radiation intensity in the focal region. In all cases, we separated the region of variation of the intensity in which the change of the ion signal was not connected with the onset of saturation: The logarithmic derivative K of the number of ions N_i with respect to the pulse energy Q was calculated by least squares for the ion-signal variation regions of interest to us (see Fig. 1). To increase the reliability of the results, K was measured in several independent experiments for each fixed value of the frequency. We note that, as already mentioned, the experimentally-measured function $N_i(Q)$ is analogous to the function $N_i(F)$.

The absolute value of the radiation intensity at which the measurements were performed was determined by the standard method[13]. All the experimental data were obtained at an electric field intensity $E \sim 5 \times 10^7$ V/cm. The typical change in the electric field intensity corre-

sponding to the change in the ion signal at a fixed frequency amounted to a factor ~1.3.

3. MEASUREMENT RESULTS AND DISCUSSION

The results of the experiments performed on the Xe atom[14] are shown in Fig. 2a, while the results for the Kr atom are shown in Fig. 2b in the form of plots of K against the radiation frequency. These figures also show data obtained earlier with a neodymium laser having a rather broad (~10—15 cm^{-1}) generation line with a frequency ~9446 cm^{-1} at the maximum of the distribution[5,6]. We see that these data agree satisfactorily with the results of the new measurements, with allowance for the differences in the width of the laser spectrum.

The results of the experiments demonstrate that K is a rapidly varying function of the radiation frequency. In both cases, a small change of the frequency (~10^{-3} of its absolute value) caused K to decrease from $K = K_0$ to approximately half this value. It should be noted that the shapes of the $K(\omega)$ plots are different for Xe and Kr. Whereas a minimum is observed for Xe, both a minimum and a maximum are observed for Kr.

Observation of a strong frequency dependence of K leads to the suggestion that the aggregate of the data on the values of K, as measured earlier[5,6], does not reflect qualitatively the main features of the process of multiphoton ionization of noble gas atoms, and constitutes an aggregate of particular values corresponding to the employed radiation frequency.

The different reasons why values $K < K_0$ are observed in multiphoton ionization of noble-gas atoms have been discussed earlier[5,6,8,15,17]. The present results, from our point of view, offer additional weighty arguments pointing to a decisive role of the boundstate spectrum. Apart from resonances, we cannot see at present any factors that could cause the observed change in the character of the ionization process with changing radiation frequency.

At the present time, however, we cannot indicate the concrete state in the spectrum of the atom with which resonance can take place. The main difficulty, which has already been discussed in the Introduction, is that it is impossible to estimate theoretically the perturbation of the atomic spectrum in a strong optical field. The experimental results enable us to estimate quantitatively the distances between those states in the spectrum with which resonance sets in. Indeed, since in both atoms the electron must absorb ~10 quanta in order to fall in the region of the bound states, and since the experimentally observed frequency change corresponding to the change of K from one extreme to the other is ~10 cm^{-1}, the resonant states in the spectrum are separated by ~100 cm^{-1}. The value $K = K_0$ obtained for certain values of the frequency obviously does not mean that the ionization proceeds for this frequency without an intermediate resonance. To be able to make such a statement it would be necessary to observe the value $K = K_0$ over a certain interval of frequency variation.

Summarizing, we can point to two principal conclusions that can be drawn from the described experiments: 1) the character of the multiphoton ionization of atoms of noble gases depends strongly on the radiation frequency; 2) the discreteness of the spectrum of the bound states of the electron becomes manifest under conditions of strong perturbation of the atom by the optical field.

These conclusions are qualitatively confirmed by the resonance effect observed in multiphoton ionization of the neon atom[16]. Just as in the case with the resonances observed in multiphoton ionization of the Xe and Kr atoms, we are unable to set the resonance observed in neon in one-to-one correspondence with a definite bound state of the electron in the atom.

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