DEPENDENCE OF THE PROBABILITY OF MULTIPHOTON IONIZATION OF ATOMS ON THE PHOTON FLUX INTENSITY

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We discuss the reasons for the existence of different values of the exponent in the formula for the probability of multiphoton ionization of atoms by radiation (whose quantum energy is much less than the ionization potential) as a function of the number of quanta absorbed during the ionization. The effect of a strong radiation field on the quasiresonant energy levels of an atom is considered, and the results of calculations of this effect are given in an approximation in which the broadening and shift of the level energy under the action of the field are comparable to the departure from resonance for only one level.

In an experimental investigation[1] of multiphoton ionization of atoms in a strong field of an electromagnetic wave with $\hbar\omega < I$, it was observed that the probability of ionization of the Xe atom is proportional to the sixth power of the photon beam intensity, whereas the ratio of the ionization potential (I = 12.13 eV) and the quantum energy ($\hbar\omega$ = 1.785 eV) shows that the number of quanta necessary for ionization is $k = \langle I/\hbar\omega + 1\rangle = 7$. Here $\langle x\rangle$ denotes the integer part of x.

One of the possible explanations of this effect is that the smearing of the upper energy levels in a strong field ($\sim 10^7$ V/cm), in which multiphoton ionization is observed, leads to the overlap and to their merging into a quasi-continuous spectrum. An estimate[2] shows that this effect can lead to a decrease in the effective ionization potential by an amount ~ 1 eV.

We discuss below another possible mechanism for lowering the slope of the plot of the ionization probability W against the photon beam intensity F, due to the action of the field on the intermediate states of the atom. The action of the field consists in a shift of the level energy as a result of the Stark effect and a broadening due to the large probability of transition from this level to the continuous spectrum.

When the atom is ionized via intermediate excited states, an important role is played by quasiresonant levels, the distance of which from the ground state is close to the energy of an integral number of radiation quanta:

$$|E_0 - E_n + m\hbar\omega| \ll \hbar\omega. \tag{1}$$

In order to consider the effect of the action of the field, we separate in the spectrum of the atom the level E_n closest to resonance and making the largest contribution to the ionization probability of the atom, and the level E_s which is closest to E_n and whose interaction with the latter determines in the main the Stark shift of the level E_n.

Calculation by the method of Weisskopf and Wigner[3] gives for the probability of ionization via an intermediate level E_n an expression

$$W = \frac{2\pi}{\hbar}\rho_\nu \left| \sum_{n'n''\ldots} \frac{H_{\nu n'}\ldots H_{n''n}\ldots H_{n'''0}}{[E_0 - E_{n'} + (k-1)\hbar\omega]\ldots[E_0 - E_n + m\hbar\omega]\ldots[E_0 - E_{n'''} + \hbar\omega]} \right|^2 \frac{(E_0 - E_n + m\hbar\omega)^2}{(E_0 - E_n + m\hbar\omega - \Delta E_n)^2 + \hbar^2\gamma_n^2/4}, \tag{2}$$

in which we have specially separated the factor containing the Stark shift ΔE_n and the width γ_n of the level E_n. The remaining part of (2) coincides with the usual expression for the ionization probability, as obtained by perturbation theory, and can be written in the form αF^k.

Thus,

$$W = \frac{\alpha F^k (E_0 - E_n + m\hbar\omega)^2}{[E_0 - E_n + m\hbar\omega - \Delta E_n(F)]^2 + \hbar^2[\gamma_n(F)]^2/4}. \tag{3}$$

As can be seen from (3), at low photon beam intensity, when the width and the Stark shift of the level

Atom	k	Resonant level	$E_0 - E_n + m\hbar\omega$, cm^{-1}	m	Dipole level	$E_n - E_s$, cm^{-1}	$\frac{\lvert D_{ns}\rvert^2}{e^2a_0^2}$	$10^{16}\gamma_n$, sec^{-1}
Ne	13 (12)	$2P^5(^2P_{3/2})4S$	—300	11	$2P^5(^2P_{3/2})4P$	—4252	38,7	0,374 F
Ar	9	$3P^5(^2P_{3/2})5P$	—1883	8	$3P^5(^2P_{3/2})5S$	3528	65	1,37 F
Kr	8	$4P^5(^2P_{3/2})6S$	1040	7	$4P^5(^2P_{3/2})6P$	—3500	52,7	0,586 F
Xe	7	$5P^5(^2P_{3/2})7P$	—1440	6	$5P^5(^2P_{3/2})6d$	—1160	64	1,37 F

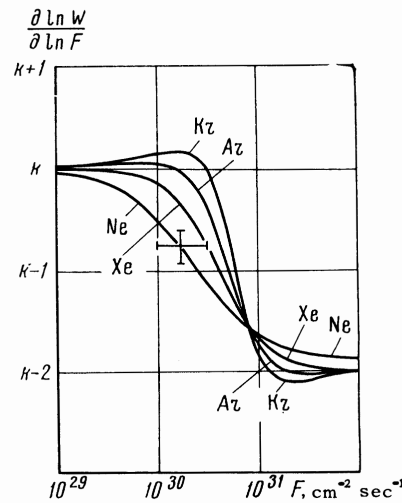
are small compared with the detuning from resonance, the denominator is constant and $W \sim F_k$, in accord with usual perturbation theory. At larger F, the denominator also depends on F. Depending on whether the denominator increases or decreases with increasing F, the slope $k' = \partial \ln W/\partial \ln F$ of the plot of W(F) will be larger or smaller than k.

The level width γ_n is determined in our case essentially by the ionization probability of the level E_n and is proportional to F^{k-m}. The broadening of the level leads to a decrease in the slope of the dependence of W(F), and at very large intensity this decrease may reach $2(k - m)$.

The Stark shift $\Delta E_n(F)$, depending on the ratio of the energies $E_0 + m\hbar\omega$, E_n, and E_s can decrease or increase the detuning from resonance, leading respectively to an increase or decrease in the slope of W(F).

The table lists the parameters of the quasi-resonant levels, separated as a result of an analysis of the spectra of several monotonic gases, and also the results of calculation of the ionization probability γ_n by the method of Burgess and Seaton[4], and of the dipole moment D_{ns} by the methods of Bates and Damgaard[5].

In the case of Ne, the energy of 12 emission quanta is only 0.1 eV lower than the ionization



potential. The energy levels in this place are located so close together, that as a result of their broadening and overlap in a strong field, they practically merge into a continuous spectrum. We can therefore put for neon $k = \langle I/\hbar\omega + 1\rangle = 12$.

The magnitude of the Stark shift in a rapid alternating field with $\hbar\omega \gg |E_n - E_s|$ is smaller by a factor $[(E_n - E_s)/\hbar\omega]^2$ than in a constant field, and is of opposite sign. The dependence of the Stark shift $\Delta E_n(F)$ can be represented in the form[6]

$$\Delta E_n = \frac{\hbar\omega - E_s + E_n}{2}\left(\sqrt{1 + \frac{16\pi|D_{ns}|^2(E_s - E_n)F\hbar\omega}{[\hbar^2\omega^2 - (E_s - E_n)^2](\hbar\omega - E_s + E_n)c}} - 1\right), \tag{4}$$

which describes the transition of the quadratic Stark effect into the linear one.

The figure shows the results of calculation of the effective slope $k' = \partial \ln W/\partial \ln F$ as a function of the photon beam intensity.

In the case when the Stark shift makes the resonance worse (Ne, Xe), the slope of W(F) decreases monotonically with increasing F. In the case when the Stark shift improves the resonance (Ar, Kr), the form of $k'(F)$ is more complicated, owing to passage through resonance.

In this calculation we took into account only one quasi-resonant level. The influence of other levels increases with increasing F and can lead to further decrease of k'.

The result of an experiment[1] on the measurement of k' for Xe, namely $k' = 6.23 \pm 0.14$ at $F = 10^{30.25 + 0.25}$ cm^{-2} sec^{-1}, coincides with the calculation made barely within the limits of the experimental error.

This solitary experiment does not enable us to decide which of the two mechanisms for the decrease of k' takes place, since reduction of k' by unity can also be obtained by overlap of the level. A decisive factor would be an experimental observation of a decrease of k' by more than unity.

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