Resonant multiphoton ionization of atoms

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The excitation of an atom in the first stage of resonant multiphoton ionization is considered within the framework of the adiabatic passage through resonance. General formulas are obtained for the position of the resonance point on the curve representing the dependence of the ionization probability on the radiation intensity. The shape of the dispersion curve is found for a spatially homogeneous field with the Lorentzian profile of the spectrum. The representations developed in the present paper make it possible to explain qualitatively the experimental results on four-photon resonant ionization of the cesium atom.

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1. INTRODUCTION. FORMULATION OF THE PROBLEM

Multiphoton ionization of atoms is one of the elementary effects in nonlinear optics which are being investigated actively at present both theoretically and experimentally. It is usual to distinguish direct and resonant multiphoton ionization.[1] In the former case an atom undergoes a transition from an initial bound state to a state in the continuous spectrum as a result of the absorption of k_0 photons ($k_0 > 1$) and the intermediate excitation of quasistationary atomic states does not play any significant role in this process, as indicated by the smooth dependence of the ionization probability on the radiation frequency. Experimental investigations of direct ionization make it possible to determine the probability of the process and its dependence on the properties of radiation, in particular, on its polarization and correlation characteristics. The agreement with theoretical calculations is then on the whole satisfactory.[1]

In resonant multiphoton ionization an atom absorbs k photons ($k < k_0$) and thus acquires energy close to the energy of one of the excited quasistationary states. Consequently, the dispersion curve exhibits resonances which usually depend on the characteristics of radiation (polarization, intensity, etc.) because such quasistationary states are excited states of the "atom + strong optical field" system. Clearly, the experimental information obtained in studies of the resonant ionization is much more extensive and, in principle, it can be used to carry out direct spectroscopic investigations of the states of an atom in a strong field.

The influence of intermediate resonances on the ionization probability has been investigated also in the first theoretical papers on multiphoton ionization.[2,3] This description is based on the Breit–Wigner formulas:

$$W_a^{(k_0)} = \frac{|A_{ab}^{(k)}|^2 W_b^{(k_0-k)}}{\Delta^2 + {}^1/_4\Gamma^2}, \qquad (1)$$

where $A_{ab}^{(k)}$ is the amplitude of the k-photon excitation of a resonance state b from the ground state a; $W_b^{(k_0-k)}$ is the probability of $(k_0 - k)$-photon ionization of the resonance state; Δ is the detuning; Γ is the effective resonance width.

If Eq. (1) is valid, the theoretical description of the dispersion dependence of the ionization probability reduces to the calculation of Δ and Γ as a function of the characteristics of the incident radiation. If $k \geq 2$, the detuning from the resonance between the levels a and b in a field is governed by the dynamic polarizabilities of these levels $\alpha_{a,b}$:

$$\Delta = \mathrm{Re}[E_a - E_b + k\omega - {}^1/_4(\alpha_a - \alpha_b)F_0^2], \qquad (2)$$

where E is the energy of levels of a free atom creating given states of the "atom + field" system; F_0 is the amplitude of the electric field of the wave; ω is the radiation frequency; $\hbar = 1$.

Many investigations have been carried out since and in these Eq. (1) has been refined, particularly with the aim of allowing for the possiblity of saturation of a k-photon transition (see, for example, Refs. 4–6), but the results have been found to disagree even qualitatively with the experimental data obtained in strong ($\gtrsim 10^7$ V/cm) and weak (~10^5 V/cm) fields.

In the absence of saturation of the a–b transition, Eqs. (1) and (2) can be validated by Low equations describing the natural width of atomic states[7] if allowance is made for the mass operator not only of the photon vacuum but also of the laser radiation field.[8] However, it is then necessary to assume that the electromagnetic field is steady-state because otherwise the mass operator, which is a function of two four-points, depends on t and t' separately and not only on the difference $t - t'$, so that Eq. (1) cannot be derived.

In reality, naturally there is no such thing as steady-state laser radiation if only because of the existence of a period in which it is turned on. When the field amplitude depends on time, the probability of ionization of an atom given by Eq. (1) is also a function of t and the total number of ions formed by a laser pulse can be found by integration of Eq. (1) with respect to time. This procedure corresponds to the adiabatic limit and it is generally not objectionable in the absence of saturation, at least for sufficiently smooth functions $F_0(t)$, provided there is no crossing of levels with $\Delta \approx 0$ when the field is varied. In the latter case the situation is radically different from that described by the resonance formula (1) and it is close to the cases of adiabatic

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transitions between quasimolecular terms in collisions of atoms[9] and to spin flip particles as a result of adiabatic fast passage through resonance.[10]

The problem of level crossing in an alternating field was considered by Melikyan and Saakyn[11] and by Yakovlenko,[12] the treatment in the latter case being applied to resonant multiphoton ionization. Yakovlenko was the first to study the analogy between multiphoton excitation of an atom when the field amplitude varies with time and the problem of the adiabatic transitions in atomic collisions.

We shall investigate the influence of level crossing on the probability of multiphoton ionization. This method makes it possible to derive adiabatic quasienergy terms without restriction to the two-level approximation. It is shown that the frequency dependence of resonant ionization is large governed by the spatial inhomogeneity of the field which occurs under real experimental conditions and, therefore, information on the resonance position deduced from measurements of the dependence of the ionization on the intensity of radiation is physically more meaningful. The general formulas obtained will be illustrated by a numerical calculation for four-photon ionization of a cesium atom in the case of a three-photon intermediate resonance investigated experimentally by Morellec *et al.*[13] The results of Morellec *et al.* are in qualitative conflict with Eq. (1) but they can be accounted for within the framework of the representations described below.

2. ADIABATIC APPROXIMATION

We shall assume that after the beginning of action of laser radiation an atom is in one of the states $|a\rangle$ which belongs to a fine or hyperfine multiplet. Let us assume that the states of this multiplet resonate at a frequency $k\omega$ with states $|b\rangle$ of another higher multiplet. The two multiplets are localized in the atomic spectrum: the multiplet splitting allowing for the perturbation by the radiation field is much less than the separation to the next multiplet.

In this case we may consider resonance between a finite number of atomic levels. The quasienergy operator $\hat{H}$, which is the effective Hamiltonian of the atomic states belonging to the multiplets a and b, is[14]

$$\left.\begin{aligned} \hat{H}_{aa'}&=E_a\delta_{aa'}-{}^1/_4F_0^2\langle a'|d_z[G(E_a+\omega)+G(E_a-\omega)]d_z|a\rangle,\\ \hat{H}_{bb'}&=(E_b-k\omega)\delta_{bb'}-{}^1/_4F_0^2\langle b'|d_z[G(E_b+\omega)+G(E_b-\omega)]d_z|b\rangle,\\ \hat{H}_{ab}&=\hat{H}_{ba}=-\frac{F_0^k}{2^k}\langle b|d_z\prod_{j=1}^{k-1}\{G(E_a+j\omega)d_z\}|a\rangle, \quad k\geqslant 2. \end{aligned}\right\} \tag{3}$$

Here, $\mathbf{d}$ is the dipole moment operator,

$$G(E)=\sum_n\frac{|n\rangle\langle n|}{E-E_n}$$

is the Green function of a free atom, and the wave is assumed to be linearly polarized along the z axis.

The diagonal matrix elements $\hat{H}$ allow for the contribution of the conventional dynamic polarizability and perturbation of the atomic spectrum. The quantities $\hat{H}_{ab}$, assumed to be real, are identical with the amplitudes of the k-photon excitation of $A_{ab}^{(k)}$.

The wave functions of quasistationary states are of the form:

$$\Psi_\varepsilon(t)=\exp\left[-i\int^t\varepsilon(t')\,dt'\right]\left[\sum_{j=1}^{N}f_\varepsilon^{(j)}(t)\,|a_j\rangle+e^{-ik\omega t}\sum_{j=1}^{N'}f_\varepsilon^{(j+N)}(t)\,|b_j\rangle\right], \tag{4}$$

where N and N' are the numbers of states in the multiplets a and b, respectively, and the quantities $f_\varepsilon^{(j)}$ are the components of the eigenvector of the matrix $\hat{H}$ corresponding to the eigenvalue ε. The time dependences of ε and f_ε are related to the time dependence of the field amplitude $F_0(t)$. When the field is turned off slowly, N eigenvalues $\varepsilon_1,\ldots,\varepsilon_N$ tend to $E_{a1},\ldots,E_{aN}$ and N' eigenvalues $\varepsilon_{N+1},\ldots,\varepsilon_{N+N'}$ tend to $E_{b1}-k\omega,\ldots,E_{bN'}-k\omega$. Consequently, we find that the coefficients transform in accordance with $f_{\varepsilon\lambda}^{(j)}$. Thus, the function (4) changes after the end of action of the field into eigenfunctions of a free atom, so that it can be classified in terms of the same quantum numbers: $\Psi_{a,b}(t)$ are the functions (4) which transform after the end of action of the field into the functions $\exp(-iE_{a,b}t)\,|a,b\rangle$.

If the amplitude of the field is constant, the functions (4) are the quasienergy solutions of the Schrödinger equation with a time-periodic Hamiltonian.[15] In accordance with the adopted resonance approximation, each quasienergy state has zeroth and k-th harmonics. However, if the amplitude is time-dependent, the functions (4) are the approximate solutions of the secular Schrödinger equation. They are the quasienergy analogs of the "stationary" states of the Hamiltonian whose parameters vary adiabatically with time.

The function Ψ_ε describes the time evolution of the system if there is no second state $\Psi_{\varepsilon'}$ violating the inequality

$$\xi\equiv|\varepsilon(t)-\varepsilon'(t)|\,\tau\gg 1, \tag{5}$$

where τ is the characteristic time for the transit of an atom across the region where the quasienergy levels come closer together and the parameter ξ acts as the Massey parameter in the theory of atomic collisions.[9]

We shall assume that one-photon ionization is possible from the multiplet b. Then, the matrix elements (3) are complex and the quasienergies ε experience ionization broadening. If this broadening predominates over other possible decay mechanisms, the probability of ionization of an atom which is in the state a before turning on of a field is

$$W_a^{(k+1)}=1-\exp\left[2\int_{-\infty}^{\infty}\operatorname{Im}\varepsilon_a(t)\,dt\right]. \tag{6}$$

Thus, if the condition (5) is satisfied, Eq. (6) represents the solution, in principle, of the problem of probability of resonant multiphoton ionization of an atom allowing for the field nonchromaticity when a complex structure of the resonance depends, in its turn, on the radiation parameters.

In considering the multiphoton ionization of an atom from states in the b multiplet ($k_0>k+1$) it is necessary to allow additionally for the multiphoton ionization

broadening in the matrix elements $\tilde{H}_{bb'}$, which presents no additional difficulties. In particular, such broadening may be associated with resonant multiphoton ionization of the states b in the presence of some third multiplet c. If the multiplet c is localized in the atomic spectrum, the matrix (3) should be expanded to allow for the states of this multiplet. We shall not consider this problem here.

We shall discuss the simplest example when only one state from each multiplet, $|a\rangle$ and $|b\rangle$, can resonate. This may occur if the energy of the interaction between an atom and the field is low compared with the multiplet splitting. We then have

$$\tilde{H}_{aa}=E_a-{}^1/_4\alpha_a I, \quad \tilde{H}_{bb}=E_b-k\omega-{}^1/_4\alpha_b I,$$
$$\tilde{H}_{ab}=A_{ab}^{(k)}, \quad I=F_0^2.$$

Assuming that one-photon ionization from the state $|b\rangle$ is possible, we shall separate in α_b the real and imaginary parts: $\alpha_b=\beta+i\gamma$. Then, ignoring the imaginary parts $E_{a,b}$, we obtain

$$\varepsilon_{a,b}={}^1/_2[E_a+E_b-k\omega-{}^1/_4(\alpha_a+\alpha_b)I]$$
$$\pm\{\Delta^2+4A_{ab}^{(k)2}-{}^1/_{16}\gamma^2I^2+{}^1/_2i\gamma\Delta I\}^{1/2}, \tag{7}$$

where Δ is defined in Eq. (2).

Let us assume that the detuning from resonance in the absence of the field is, for example, positive: $\Delta_0\equiv E_a-E_b+k\omega>0$. Then the eigenvalue ε_a corresponds to the plus sign in front of the square root. If $\frac{1}{4}\gamma I$ is a small quantity, we obtain

$$\operatorname{Im}\varepsilon_a\approx-\frac{1}{8}\gamma I\left\{1-\frac{\Delta}{(\Delta^2+4A_{ab}^{(k)2})^{1/2}}\right\}. \tag{8}$$

For $\Delta>0$, which is true for any field intensity if $\alpha_a-\beta<0$ or in a limited range of intensities where $\alpha_a-\beta>0$, we find assuming that $4A_{ab}^{(k)2}\ll\Delta^2$, that

$$\operatorname{Im}\varepsilon_a\approx-{}^1/_4\gamma I A_{ab}^{(k)2}/\Delta^2. \tag{9}$$

Since $\frac{1}{2}\gamma I$ is the probability of ionization of an atom from the state $|b\rangle$ per unit time, Eqs. (6) and (9) give the Breit–Wigner formula (1) corresponding to large detuning, naturally if the total ionization probability of an atom is low, so that the exponential function in Eq. (6) can be expanded as a power series. On the other hand, in the range $\Delta<0$, we have

$$\operatorname{Im}\varepsilon_a\approx-{}^1/_4\gamma I \tag{10}$$

and the probability of ionization of an atom no longer depends on the probability of excitation of the state b from the state a. The change in the sign of Δ occurs in the region of the exact resonance when the field amplitude varies with time in such a way that $I(t)\approx 4\Delta_0/(\alpha_a-\beta)$. When the field continues to rise away from the resonance, an atom is found in the state b, in accordance with the adiabatic term.

Consequently, if, according to Eq. (1), the probability of ionization of an atom assumes high values of the order given by Eq. (10) only in a narrow time interval when $\Delta\approx 2A_{ab}^{(k)}$, the probability (10) during adiabatic motion applies in that time interval as long as $\Delta<0$, which is shown in Fig. 1. At a moment t_1 the terms come closer together and for $t>t_1$ an atom is excited to the state b and then its probability of ionization is high. At a moment t_0 the field reaches its maximum value, which corresponds to the extreme right-hand point of $\varepsilon(I)$. For $t>t_0$ an atom moves between the terms from right to left and for $t>t_2$, after the second approach of the terms, the probability of ionization is again low.

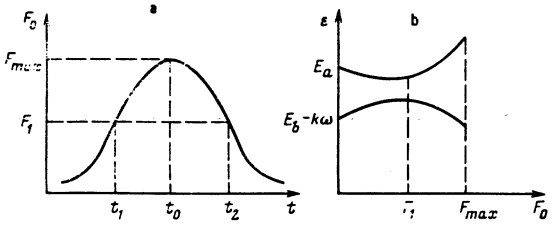

FIG. 1. Time dependence of the field (a) and behavior of the terms as a function of the field intensity (b).

3. PROBABILITY OF A NONADIABATIC TRANSITION

The adiabatic approximation discussed above is disturbed by transitions between quasienergy levels. The probability of such transitions can be calculated, in particular, by determining the time τ in Eq. (5) from the familiar results of the theory of atomic collisions. We shall assume that at a moment t_1 a pair of terms comes closest together and the square of the separation between them is found from Eq. (7):

$$\Omega^2(t)=\Delta^2(t)+4A_{ab}^{(k)2}(t). \tag{11}$$

Line broadening in the quasicrossing region is ignored because, as shown by several authors[16] the quasistationary nature of crossing terms has practically no influence on the probability of a transition between them. The probability of a nonadiabatic transition is given by[17]

$$w=\exp\left[i\int_{t_c^*}^{t_c}\Omega(t)\,dt\right], \tag{12}$$

where t_c is the root of $\Omega^2(t)$ lying in the upper half-plane of the complex quantity t.

We shall now consider the dependence of the excitation amplitude on the field intensity: $A_{ab}^{(k)}=\eta_{ab}^{(k)}F_0^k$. In the Landau–Zener approximation we can expand $\Omega^2(t)$ as a series and retain only two terms:

$$\Omega^2(t)=\Omega_1^2+v_1^2(t-t_1)^2,$$
$$v^2(t)=[{}^1/_{16}\tilde{\alpha}^2I^2+2k(k-1)\eta_{ab}^{(k)2}I^k](I/I)^2, \tag{13}$$
$$\tilde{\alpha}=\alpha_a-\beta, \quad \Omega_1=\Omega(t_1), \quad v_1=v(t_1).$$

Here, t_1 is the root of the equation

$$\tilde{\alpha}[\Delta_0-{}^1/_4\tilde{\alpha}I(t)]-8k\eta_{ab}^{(k)2}I^{k-1}(t)=0. \tag{14}$$

Substituting Eq. (13) into Eq. (12), we find that

$$w=e^{-\xi}, \quad \xi=\pi\Omega_1^2/2v_1. \tag{15}$$

The value of v is the rate of passage through the region where the terms approach time is

$$\tau=\Omega_1/v_1. \tag{16}$$

In the limit $\tilde{\alpha}I(t_1)\gg A_{ab}^{(k)}(t_1)$, when

$$\Omega_1^2 \approx 4A_{ab}^{(k)2}(t_1), \quad v_1 \approx {}^1/_4 \tilde{a} |I(t_1)|, \tag{17}$$

Eq. (15) gives the same result as that obtained by Yakovlenko.[12]

The relationships in Eq. (17) can be used to obtain qualitative estimates of the probabilities of nonadiabatic transitions. If $I \approx \theta I$, where θ is the width of the radiation spectrum, we find that

$$\xi \approx 2\pi A_{ab}^{(k)2} \Delta_0 \theta = \pi \Gamma_f^2 / 2\Delta_0 \theta. \tag{18}$$

This formula contains the field width Γ_f, which determines the width of the dispersion curves of resonant ionization under saturation conditions.[6]

For $k = 2$, the atomic matrix elements governing the polarizability and amplitude of the two-photon excitation can be regarded as quantities of the same order of magnitude. We then have $\xi \approx \Delta_0/\theta$ and we can justifiably ignore nonadiabatic transitions because the static detuning from resonance is considerably greater than the spectral width of the incident radiation. For $k > 2$, the quantity ξ depends on the actual atomic transition because multiphoton matrix elements generally vary over very wide limits. For the case of the cesium atom considered in Sec. 5, we have $k = 3$ and $\xi \approx 1$ for fields $F_0 \gtrsim 3 \times 10^5$ V/cm.

4. SPATIAL INHOMOGENEITY OF THE FIELD

The spatial inhomogeneity associated with the need to focus laser radiation under real experimental conditions complicates the relatively simple physical picture of ionization of an atom moving between adiabatic terms. We shall find the resultant differences by considering the simple case of a resonance between two isolated states. We shall also assume that only excited atoms become ionized; this corresponds to the time interval $t_1 < t < t_2$ in Fig. 1. This approximation is quite satisfactory because the probabilities of ionization from the ground and excited states differ by an order of magnitude (see a numerical sample in Sec. 5).

In general, the field amplitude is a function of time and of spatial coordinates. If the widths of the levels are ignored, the square of the separation between the terms a and b is given by Eq. (11). The surface S_1 in the four-space on which the approach of the terms takes place is given by [compare with Eq. (14)]:

$$\Delta_0 - \frac{1}{4}\tilde{a} I(\mathbf{r}, t) = \frac{8k}{\tilde{a}} \eta_{ab}^{(k)2} I^{k-1}(\mathbf{r}, t). \tag{19}$$

The excitation of atoms on passing through this surface occurs if the condition (5) is satisfied. Qualitatively correct results can be obtained by assuming that the excitation occurs in a region whose boundary is a different surface S_2:

$$[\Delta_0 - {}^1/_4 \tilde{a} I(\mathbf{r}, t)]^2 + 4\eta_{ab}^{(k)2} I^k(\mathbf{r}, t) \geqslant v(\mathbf{r}, t). \tag{20}$$

Several possible situations are shown in Fig. 2, where x is some spatial coordinate. It is assumed that the field is in the form of a single pulse whose maximum corresponds to $x = 0$ and $t = t_0 = 0$.

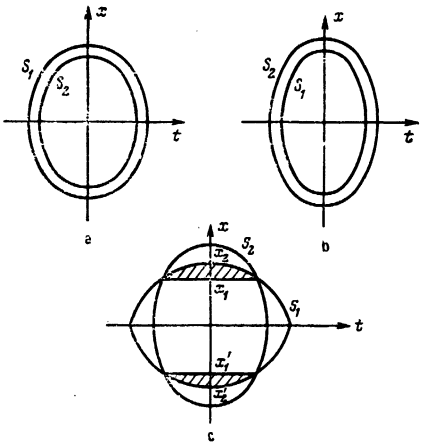

FIG. 2.

a) The surface S_2 is contained entirely inside the surface S_1. In this case the parameter ξ is small in the quasicrossing region, the adiabatic approximation does not apply, and on passing through the term-approach point an atom jumps from one adiabatic term to another. The excitation does not occur and the probability of ionization of an atom is relatively low.

b) The surface S_1 is entirely inside the surface S_2. In this case we may assume that the condition (5) is satisfied and that an atom is excited when the terms approach one another. The ionization occurs inside the surface S_1.

c) The surfaces S_1 and S_2 intersect. In this case the atoms in the parts of space shown shaded in Fig. 2*c* are excited and then ionized. We shall calculate the total number of ions formed in this case. For each value of r we have to find the moments of approach of the terms $t_1(\mathbf{r})$ and $t_2(\mathbf{r})$ which lie inside S_2. Then, the number of ions is given by

$$N_i = n \int d\mathbf{r} \left\{ 1 - \exp\left[-\frac{\gamma}{2} \int_{t_1(\mathbf{r})}^{t_2(\mathbf{r})} I(\mathbf{r}, t)\, dt \right] \right\}, \tag{21}$$

where n is the density of neutral atoms assumed to be constant in the focusing region and the integral with respect to $d\mathbf{r}$ is calculated within the limits $x_1 < x < x_2$, $x_1' < x < x_2'$, and within similar limits for other coordinates on which the field may depend (Fig. 2*c*).

General relationships governing the dispersion dependence of the probability of resonant multiphoton ionization can be deduced from the above discussion. The maximum detuning at which the excitation still takes place follows from Eq. (19):

$$\Delta_{0\,max} = {}^1/_4 \tilde{a} I_0 + 8k\eta_{ab}^{(k)2} I_0^{(k-1)}/\tilde{a}, \tag{22}$$

where I_0 is the maximum intensity, i.e., the value of $I(\mathbf{r}, t)$ corresponding to $\mathbf{r} = 0$ and $t = 0$. We recall the assumptions that $\Delta_0 > 0$ and $\tilde{a} > 0$.

The surface S_1 expands with decreasing Δ_0 and the number of ions formed the increases. The value of $\tilde{\Delta}_0$ corresponding to the maximum ion yield can be found for a spatially homogeneous field. In this case the sur-

faces S_1 and S_2 are planes orthogonal to the t axis. The following equation is obtained for t_{cr} from the condition that these planes coincide:

$$\left[\frac{8k}{\tilde{a}}\eta_{ab}^{(k)2} I^{k-1}(t_{cr})\right]^2 + 4\eta_{ab}^{(k)2} I^k(t_{cr}) = v(t_{cr}). \quad (23)$$

The substitution of t_{cr} into Eq. (19) gives the value of $\tilde{\Delta}_0$. If the conditions of Eq. (17) are satisfied, the first term on the left-hand side of Eq. (23) is much less than the second. If, moreover, we can assume an exponential time dependence of the intensity, $I(t) = I_0 e^{-\theta|t|}$, then

$$\tilde{\Delta}_0 = \frac{\tilde{a}}{4}\left(\frac{\tilde{a}\theta}{16\eta_{ab}^{(k)2}}\right)^{1/(k-1)}.$$

Figure 3 shows qualitatively the corresponding dispersion curve. It is interesting that in this model the value of $\tilde{\Delta}_0$ is independent of I_0. Naturally, we should bear in mind that for other types of the dependences of the radiation intensity on the coordinates and time the curve, and particularly the value of $\tilde{\Delta}_0$, may change. However, the value of Δ_{0max} given by Eq. (22) remains unaltered.

5. COMPARISON WITH EXPERIMENTAL RESULTS

The spatial inhomogeneity and the multipeak time structure of laser radiation complicate considerably the interpretation of the experimental data on resonant multiphoton ionization and, therefore, the most suitable results for comparison with the theory are those obtained using single-mode laser radiation. Such results are now available for the four-photon ionization of the cesium atom from the ground state $6s$ by neodymium laser radiation involving a three-photon intermediate resonance of the $6f$ level.[13,18] We shall compare the results of Morellec *et al.*[13] with the theory because, in contrast to Grinchuk *et al.*,[18] the spatial inhomogeneity of the field was less in the former case and the data on the resonance position were deduced from the dependence of the ionization probability on the radiation intensity and not on the radiation frequency. This was most important in view of the discussion given in the preceding section.

The ^{133}Cs isotope has the nuclear spin $\frac{7}{2}$ and cesium has no other stable isotopes. The hyperfine splitting of the ground state is ≈ 0.3 cm^{-1}. The $6f$ resonant level has a doublet structure because of a fine splitting amounting to ≈ 0.1 cm^{-1}. We shall calculate the matrix elements of the quasienergy operator (3). In a linearly polarized field[13] the quasienergy levels are degenerate

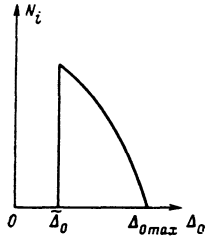

FIG. 3. Dispersion dependence of resonant multiphoton ionization for a spatially homogeneous pulse with the Lorentzian profile.

in respect of the sign of the M projection of the total momentum of an atom F. The polarizabilities of both components of the hyperfine doublet are the same and independent of M:

$$\tilde{H}_{FF} = E_F - {}^1/_4\alpha_{6s}F_0^2.$$

Here, $F = 3$ or 4. The nondiagonal matrix element $\tilde{H}$ between different hyperfine sublevels vanishes.

The polarizabilities of resonant levels depend on the total momentum of the electron shell $J = \frac{5}{2}, \frac{7}{2}$:

$$\tilde{H}_{JJ} = E_J - 3\omega - {}^1/_4\alpha_J F_0^2, \quad \alpha_{5/2} = \alpha_{6f}^{(S)} - {}^{24}/_{35}\alpha_{6f}^{(T)}, \quad \alpha_{7/2} = \alpha_{6f}^{(S)} - {}^3/_7\alpha_{6f}^{(T)},$$

where $\alpha_{6f}^{(S,T)}$ are the scalar and tensor polarizabilities of the resonant level.[19] The spin of the nucleus in the $6f$ state can be ignored because of the smallness of the hyperfine splitting in the state with a large orbital momentum of the valence electron. The projection of the momentum of the electron shell of an atom in a resonant state is assumed to be $m = \pm\frac{1}{2}$ in accordance with the selection rules applicable to a linearly polarized field.

The values of the nondiagonal matrix element $\tilde{H}$ between the components of the fine doublet depend on the sign of m:

$$\tilde{H}_{5/2\pm 1/2,\,7/2\pm 1/2} = \mp\sqrt{3}\,\alpha_{6f}^{(T)}F_0^2/70.$$

The matrix elements $\tilde{H}$ relating the ground and resonant sublevels depend on M and m:

$$\tilde{H}_{FM,Jm} = -{}^1/_8 F_0^3 C^{FM}_{1/2\,m';\,M-m} C^{Jm}_{30\,1/2\,m} D_{6s,6f},$$

$$D_{6s,6f} \equiv \langle 6f|d_z G(E_F+2\omega)d_z G(E_F+\omega)d_z|6s\rangle.$$

The above matrix element can be calculated ignoring the spins of the nucleus and valence electron: $|6s\rangle$, $|6f\rangle$ are the coordinate numbers of the wave functions of the atom.

The matrix $\tilde{H}$ is thus characterized by the values of M and for each M there are two possible values of m. In general, when $|M| \neq 4$, it is found that $\tilde{H}$ is a symmetric six-order matrix. For $|M| = 4$, we find that $\tilde{H}$ is a third-order matrix.

The numerical values of the polarizabilities and excitation amplitudes were found in the approximation of the quantum defect method.[19] The quantities expressed in atomic units are

$$\alpha_{6s} = 1500, \quad \alpha_{6f}^{(S)} = -800 + 10.5i, \quad \alpha_{6f}^{(T)} = 190 + 6.6i,$$

$$D_{6s,6f} = 1.2\cdot 10^5.$$

Morellec *et al.*[13] used fields $F_0 < 10^6$ V/cm. In such fields the second term in Eq. (22) is small and the rate of the field-induced shift of a singularity (kink) of the dependence of the ionization probability on the radiation intensity is $3.81 I_0$ cm$^{-1}\cdot$GW$^{-1}\cdot$cm^2 for the numerical parameters given above. The experimental value of this quantity is $4.3 I_0$ cm$^{-1}\cdot$GW$^{-1}\cdot$cm^2, which is in satisfactory agreement with the theoretical estimate.

An important qualitative result of the experiments of Morellec *et al.*[13] was the resolution of the structure of this singularity in weak fields $\lesssim 4\times 10^5$ V/cm. They found two kinks of the dependence of the ionization

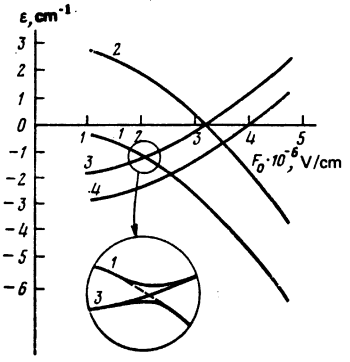

FIG. 4. Quasienergy terms of the cesium atom.

probability on the radiation intensity and these were separated by ≈ 0.17 cm^{-1}, whereas the theory predicted four such points because of the doublet structure of the lower and upper levels. The result obtained can be explained on the basis of the results derived above. Figure 4 shows the behavior of the quasienergy terms of cesium in a field. On application of the field, the levels 1 and 2 correspond to the initial state $6s$, $F=3$, and $F=4$, whereas the levels 3 and 4 correspond to the resonant state $6f$, $J=\frac{5}{2}$ and $\frac{7}{2}$; the resonant levels are doubly degenerate in respect of the sign of the projection of the electron-shell momentum m. The qualitative behavior of the levels at the quasicrossing points is the same for all values of M and it is shown at the bottom of Fig. 4 on an enlarged scale.

If the adiabaticity parameter ξ is large in the quasicrossing region, an atom is excited when the terms come closer together and it is then rapidly ionized. In this case for each of the hyperfine structure sublevels (terms 1 and 2) there is only one singularity and the dependence of the ionization probability on the radiation intensity has two kinks, as found experimentally. If the parameter ξ is large even for the first cross sections of the terms 1 and 2 with the term 3, the separation between the kinks is 0.3 cm^{-1}. However, we may assume that for the term 1 crossing the term 3 the parameter ξ is insufficiently large and there is no excitation of the atom to the term 3. In the case of the second crossing with the term 4, the parameter ξ—which is proportional to F_0^4—increases approximately by a factor of three and the transition $1-4$ takes place, whereas further increase of the field produces the transition $2-3$. In this case the separation between the kinks is 0.2 cm^{-1}, which is close to the experimental value 0.17 cm^{-1}.

The theoretical adiabaticity parameter ξ, obtained using the above numerical values of the atomic matrix elements, is 0.16 in a field of 4×10^5 V/cm if it is assumed that the field is an exponential function of time. This value increases and reaches the required interval $\xi \approx 1$ if allowance is made for the smooth time dependence of the radiation intensity at the maximum of a pulse.

This interpretation can be checked by investigating experimentally resonant multiphoton ionization of cesium in stronger fields of $\sim(5-6)\times 10^5$ V/cm, which can be done by selecting a somewhat greater initial resonance detuning. In this case there should be again two kinks but the separation between them should be 0.3 cm^{-1}. In weaker fields $\lesssim 10^5$ V/cm we may observe only one singularity: an atom in the term 1 jumps across the terms 3 and 4 without excitation.

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