

# Interference effects in the photoionization of Rydberg atoms in a strong electromagnetic field

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An investigation is made of the photoionization of Rydberg atoms in a strong electromagnetic field. An allowance is made for the level degeneracy in respect of the orbital momentum, for the Stark splitting of the levels, and for the possibility of a resonant interaction with levels of lower energy. The complex quasienergies of the system, the spectrum of photoelectrons in the limit of a long interaction, and the time-dependent total ionization probability are found. It is shown that narrowing of quasienergy levels occurs in a strong field. The quasienergy spectrum consists of more or less narrow levels against the background of a quasicontinuum. The photoelectron spectrum acquires then many peaks. An increase in the field enhances the amplitudes of the peaks and reduces their width. The rate of ionization decreases on increase in the field. The presence of a continuum is responsible for the partly nonexponential nature of the decay of such atoms.

## 1. INTRODUCTION

We report here an investigation of the photoionization of Rydberg atoms in a strong electromagnetic field generalizing directly an earlier study<sup>1</sup> of the case of a weak field. The criterion of a strong field is determined by the magnitude of the matrix elements for bound-free transitions. In the semiclassical range, where  $n \gg 1$  and  $E \ll 1$  ( $n$  is the principal quantum number of a Rydberg level and  $E$  is the energy of an electron in the continuum), it is known<sup>2,3</sup> that

$$V_{nE} = V n^{-3/2}, \quad V \propto F_0 \omega^{-5/3}, \quad (1)$$

where  $V_{nE}$  are the matrix elements of the operator  $V = -\frac{1}{2} \mathbf{d} \cdot \mathbf{F}_0$ ;  $\mathbf{d}$  is the dipole moment of an atom;  $F_0$  and  $\omega$  are the amplitude and frequency of the field;  $\omega > n^{-2}/2$ ; here and later we shall use the atomic system of units. A field is strong if  $V > 1$  and we shall assume that this condition is satisfied. We shall adopt two formulations of the problem: the problem of complex quasienergies with real and imaginary parts governing the position and width of quasienergy levels in a strong field, and the initial problem with instantaneous activation of the interaction at  $t = 0$ . In the latter case we shall consider the dynamics of the ionization process and the energy spectrum of photoelectrons in the limit of a prolonged interaction.

## 2. POLARIZABILITY TENSOR AND IONIZATION WIDTH

It is well known<sup>4</sup> that in a weak field a perturbation of a level  $E_n$  that is degenerate in respect of the orbital momentum  $l$ , can be described by a matrix  $Q_{ll'}^{(n)} \propto F_0^2$ . Its real part is proportional to the polarizability tensor  $\alpha_{ll'}^{(n)}$ , ( $\text{Re} Q_{ll'}^{(n)} = -\frac{1}{4} \alpha_{ll'}^{(n)} F_0^2$ ), which governs the modification of a multiplet in a nonresonant field. The imaginary part of  $Q_{ll'}^{(n)}$ , which differs from zero for  $\omega > n^{-2}/2$ , determines the ionization broadening of the levels. If we write down explicitly the usual expression for  $\alpha_{ll'}^{(n)}$  in the form of a sum over intermediate states  $|n'\rangle$  and expand the energy denominators in powers of  $|E_n - E_{n'}|/\omega$ , we can readily estimate the first two nonzero terms of this expansion. The first of them is dominated by contributions characterized by  $n'$  close to  $n$ . Calculations carried out using the sum rule<sup>5</sup> give the familiar result

$$[\text{Re} Q_{ll'}^{(n)}]^{(0)} = \frac{1}{4} \delta_{ll'} F_0^2 \omega^{-2}.$$

In this approximation the polarizability tensor is diagonal and it determines the general shift of all the Rydberg levels by an amount equal to the average energy of oscillations of an electron in the field of a wave. This shift does not affect the dynamics of the ionization process. It can be included in the definition of the energy  $E_n$ , which we will henceforth assume. A calculation of the correction to  $[\text{Re} Q_{ll'}^{(n)}]^{(0)}$  by means of an expansion in terms of  $|E_n - E_{n'}|/\omega$  meets with certain difficulties because the corresponding sum over  $n'$  diverges at high values of  $|n' - n|$ . This can easily be demonstrated if the matrix elements  $V_{nn'}$  are described by semiclassical expressions analogous to those given by Eq. (1) [see Refs. 2 and 3]:

$$V_{nn'} \propto F_0 (nn')^{-3/2} |E_n - E_{n'}|^{-1/2}. \quad (2)$$

The divergence of the sum which gives  $[\text{Re} Q_{ll'}^{(n)}]^{(1)}$  demonstrates that in fact the expansion  $|E_{n'} - E_n|/\omega$  is not correct and  $[\text{Re} Q_{ll'}^{(n)}]^{(1)}$  is dominated by the contribution of the range  $|E_{n'} - E_n| \sim \omega$ . Nevertheless, we can estimate  $[\text{Re} Q_{ll'}^{(n)}]^{(1)}$  using this incorrect expansion, replacing the sum over  $n'$  with an integral which we cut off at  $|E_{n'} - E_n| \sim \omega$ . The result is then

$$[\text{Re} Q_{ll'}^{(n)}]^{(1)} \propto V^2 n^{-3} \propto \text{Im} Q_{ll'}^{(n)}. \quad (3)$$

Therefore, the main contribution to  $[\text{Re} Q_{ll'}^{(n)}]^{(1)}$  comes from transitions via levels which are far from the initial one:  $|n' - n| \sim n$ . If  $n \gg 1$  the matrix elements of such transitions are semiclassical [Eqs. (1) and (2)] and it follows from Ref. 3 that they depend weakly on the quantum numbers  $n$  and  $l$ . A weak dependence on  $n$  is observed if the characteristic number  $\Delta n$  of the Rydberg levels of interest in the vicinity of the initial value  $n_0$  is small compared with  $n_0$ :  $\Delta n \ll n_0$ . The weak dependence on  $l$  occurs for  $l < n_0^{2/3}$  (Ref. 3). It follows also that if  $n \gg 1$ , the matrix  $[\text{Re} Q_{ll'}^{(n)}]^{(1)} + i \text{Im} Q_{ll'}^{(n)}$  can be described by a single complex constant  $Q_0$ :

$$[\text{Re} Q_{ll'}^{(n)}]^{(1)} + i \text{Im} Q_{ll'}^{(n)} = Q_0 [(1 - \delta_{l0}) \delta_{l', l-2} + (2 - \delta_{l0}) \delta_{l', l} + \delta_{l', l+2}], \quad (4)$$

where the structure of the factor in the square brackets is governed by the selection rules given in Ref. 6.

The most stringent of the above conditions of validity of the approximation described by Eq. (4) is  $l < n_0^{2/3}$ . If  $n_0$  is not too large, then  $n_0^{2/3}$  is slightly greater than unity ( $n_0^{2/3} \sim 6$  when  $n_0 \sim 10$ ). This is clearly the reason for the dependence of the elements of the matrix  $Q$  on  $l$ , detected in numerical calculations in Ref. 7. In the subsequent analysis the assumption of a weak dependence of the elements of the matrix  $Q$  on  $l$  is unnecessary and we shall therefore describe the matrix  $Q$  by a set of complex constants  $Q_{ll'}^{(n)} \propto V^2 n_0^{-3} \propto F_0^2$ , where the relationship between  $l'$  and  $l$  is given by the selection rules  $l' - l = 0, \pm 2$ . However, many formulas and expressions are generally very cumbersome. We can simplify them using the approximation of Eq. (4), which is formally valid only if  $n_0$  is very large.

The weak dependence of  $Q_{ll'}^{(n)}$  on  $n$  is on the other hand extremely important in our subsequent analysis. We shall assume that  $Q_{ll'}^{(n)} \approx Q_{ll'}^{(n_0)}$ . The dependence  $Q_{ll'}^{(n)}(n)$  has not yet been investigated numerically for Rydberg levels. Naturally, this dependence cannot be weak under conditions of a resonance between a level  $E_{n_0}$  and a lower Rydberg level  $E_{m_0} \approx E_{n_0} - \omega$ . The resonant interaction will be included explicitly in the equations and solutions; we shall assume that the matrix  $Q$  is governed by the sum of just the nonresonant terms.

In a strong field characterized by  $V > 1$  we have to include also the elements of the matrix  $Q$  between Rydberg states with different values of  $n$ . We shall assume that all such matrix elements  $Q_{n'l',n}$  are characterized, irrespective of the values of  $n$  and  $n' \sim n_0$ , by the same set of complex constants  $Q_{ll'}$ . Moreover, in a strong field when  $V > 1$ , the corrections to  $Q_{ll'}$ , of higher order in  $V^2$  may become important and in the final analysis this results in renormalization of the constants  $Q_{ll'}$ . One of the models describing the renormalization effect  $V^2 \rightarrow \pi^{-1}V$ , based on the method of "essential states"<sup>8</sup> and allowing for multiple transitions to the continuum, was discussed in Refs. 9 and 10. We shall not specify here the renormalization model. We shall assume that the constants  $Q_{ll'}$  are determined allowing for the renormalization effect and that they are growing functions of the parameter  $V$  of Eq. (1), which are not necessarily quadratic, in contrast to the weak field case described by Eq. (3).

### 3. COMPLEX QUASIENERGIES

We shall assume that an atom at a Rydberg level  $E_{n_0}$  experiences a field of frequency  $\omega > \frac{1}{2} n_0^{-2}$ . The ionization process results in secondary population of the Rydberg levels  $E_n$  close to the initial level  $E_{n_0}$ . We shall assume that there may be a resonance with lower Rydberg levels  $E_m$  (in the vicinity of  $E_{m_0} \approx E_{n_0} - \omega$ ). Expanding the wave function of an atom in terms of the basis functions of the levels  $E_n$  and  $E_m$  and of the continuum, and excluding from the equations for the amplitudes the probabilities of finding an atom in the continuum (employing the standard method of Refs. 11–13), we obtain equations with constant complex coefficients for the amplitudes of the probabilities of finding an atom at the levels  $E_n$  and  $E_m$ . We shall seek the solution of these equations in the form  $\exp(-i\gamma t)$ , where  $\gamma$  is a complex quasienergy. The resultant equations for the amplitudes  $C_{nl}$  and  $C_{ml}$  are

$$\begin{aligned} (\gamma - E_n) C_{nl} &= \sum_{n'l'} Q_{ll'} C_{n'l'} + (n_0 m_0)^{-3/2} \sum_{m', l' = l \pm 1} V_{ll'} C_{m'l'}, \\ (\gamma - E_m - \omega) C_{ml} &= \sum_{m'l'} \tilde{Q}_{ll'} C_{m'l'} + (n_0 m_0)^{-3/2} \sum_{n', l' = l \pm 1} V_{ll'} C_{n'l'}, \end{aligned} \quad (5)$$

where  $Q_{ll'}$  and  $\tilde{Q}_{ll'}$  are the elements of the matrix  $Q$  for the Rydberg levels in the vicinity of  $E_{n_0}$  and  $E_{m_0}$ , respectively, and  $V_{ll'} \sim V$  in Eq. (1).

The assumption that the constants  $Q_{ll'}$ ,  $\tilde{Q}_{ll'}$ , and  $V_{ll'}$  are independent of  $n$ ,  $m$ ,  $n'$ , and  $m'$  simplifies the system of equations (5). Multiplying the first of these equations by  $(\gamma - E_n)^{-1}$  and the second by  $(\gamma - E_m - \omega)^{-1}$ , summing them over  $n$  and  $m$ , respectively, and introducing

$$\begin{aligned} \sum_n (\gamma - E_n)^{-1} &= X(\gamma), \quad \sum_m (\gamma - E_m - \omega)^{-1} = \tilde{X}(\gamma), \\ \sum_n C_{nl} &= A_l, \quad \sum_m C_{ml} = B_l, \end{aligned} \quad (6)$$

we obtain the following equations for  $A_l$  and  $B_l$ :

$$\begin{aligned} A_l &= X(\gamma) \left\{ \sum_{l'} Q_{ll'} A_{l'} + (n_0 m_0)^{-3/2} \sum_{l' = l \pm 1} V_{ll'} B_{l'} \right\}, \\ B_l &= \tilde{X}(\gamma) \left\{ \sum_{l'} \tilde{Q}_{ll'} B_{l'} + (n_0 m_0)^{-3/2} \sum_{l' = l \pm 1} V_{ll'} A_{l'} \right\}. \end{aligned} \quad (7)$$

The first terms on the right-hand sides of the system (7) describe modification and broadening of Rydberg levels because of the dynamic Stark effect and because of ionization. The second terms on the right-hand sides of the system (7) represent a resonant interaction of Rydberg multiplets. Since the levels  $E_m$  are not coupled directly to the continuum, we have  $\text{Im} \tilde{Q}_{ll'} = 0$ .

A number of transformations can convert the system (7) into equations containing only the constants  $A_l$  (but not  $B_l$ ). With this in mind we begin with the first equation in the system (7) and find the sum

$$\tilde{X}(\gamma) X^{-1}(\gamma) (n_0 m_0)^{3/2} [A_{l-1} \tilde{Q}_{l, l-2} V_{l-1, l-2}^{-1} + A_{l+1} \tilde{Q}_{l, l+2} V_{l+1, l+2}^{-1}].$$

Subtracting the result from the second equation in the system (7), we obtain an explicit expression for  $B_l$  in terms of the constants  $A_l$ . Then, substituting the resultant expression for  $B_l$  into the first equation in the system (7), we find the required equations for  $A_l$ . In general, these equations are cumbersome. We shall therefore write down only a simplified variant corresponding to the approximation described by Eq. (4), i.e., we shall replace  $Q_{ll'}$  and  $\tilde{Q}_{ll'}$  with  $Q_0$  and  $\tilde{Q}_0$ :

$$\begin{aligned} A_l &= [X(\gamma) Q_0 + \tilde{X}(\gamma) \tilde{Q}_0 + (n_0 m_0)^{-3} V^2 X(\gamma) \tilde{X}(\gamma)] \cdot \\ &\times [A_{l-2} (1 - \delta_{l0}) + A_l (2 - \delta_{l0}) + A_{l+2}] - Q_0 \tilde{Q}_0 X(\gamma) \tilde{X}(\gamma) \cdot \\ &\times [A_{l-4} (1 - \delta_{l2}) (1 - \delta_{l2}) + A_{l-2} (1 - \delta_{l0}) (4 - \delta_{l2}) \\ &+ A_l (6 - 4\delta_{l0}) + A_{l+2} (4 - \delta_{l0}) + A_{l+4}], \end{aligned} \quad (8)$$

where  $l < l_{\max} \propto n_0^{2/3}$ .

In a strong field ( $V > 1$ ) the last term in the first square brackets on the right-side of Eq. (8) can be omitted since it is small compared with the coefficient in front of the third set of square brackets ( $Q_0 \propto V^2 n_0^{-3}$ ,  $\tilde{Q}_0 \propto V^2 m_0^{-3}$ ). Although the products  $XQ_0$  and  $\tilde{X}\tilde{Q}_0$  occur quite symmetrically in Eq. (8), their meaning and role are very different. The function  $\tilde{X}(\gamma)$  is much smoother than  $X(\gamma)$  because  $m < n$ . The struc-

ture of the function  $X(\gamma)$  essentially determines the quasienergy spectrum. In a strong field ( $V > 1$ ) the quasienergies of the system are localized in those regions where the function  $X(\gamma)$  is small and we have  $\tilde{X}(\gamma)Q_0 \sim 1$  (see below). The product  $\tilde{X}(\gamma)\tilde{Q}_0$  then can have any value and it determines the degree of resonant mixing of the multiplets. If  $\tilde{X}(\gamma)\tilde{Q}_0 \ll 1$  then Eq. (8) can be simplified by dropping all the terms proportional to  $\tilde{X}Q_0$ , which eases the situation greatly. We shall write down the resultant nonresonant system of equations for  $A_l$  in its general form (with arbitrary constants  $Q_{ll'}$ ) as well as in the approximation represented by Eq. (4):

$$A_l = X(\gamma) \sum_{l'} Q_{ll'} A_{l'} \\ = X(\gamma) Q_0 [(1-\delta_{l0})A_{l-2} + (2-\delta_{l0})A_l + A_{l+2}]. \quad (9)$$

If  $\tilde{X}\tilde{Q}_0 \gg 1$ , then Eq. (8) can be simplified by dropping all the terms that do not contain a large factor  $\tilde{X}\tilde{Q}_0$ , when Eq. (8) becomes

$$A_{l-2}(1-\delta_{l0}) + (2-\delta_{l0})A_l + A_{l+2} \\ = X(\gamma) Q_0 [(1-\delta_{l0})(1-\delta_{l2})A_{l-4} + (1-\delta_{l0})(4-\delta_{l0})A_{l-2} \\ + (6-4\delta_{l0})A_l + (4-\delta_{l0})A_{l+2} + A_{l+4}]. \quad (10)$$

In general, for arbitrary values of  $Q_{ll'}$  and  $\tilde{Q}_{ll'}$  we find that the approximation of strong resonant mixing still leaves the equations for  $A_l$  very cumbersome and, therefore, we shall not give them here.

Although at first sight Eq. (10) is much more complex than Eq. (9), we can show that their solutions are identical. This is a general result associated not with the use of the approximation represented by Eq. (4), but simply with the fact that the Stark mixing and ionization broadening of the levels are much more significant in a strong field than the effects of resonant mixing. We shall therefore consider a simpler system than Eq. (9) and adopt the approximation of Eq. (4). Introducing

$$XQ_0 = \lambda^{-1}, \quad (11)$$

we find that we are facing the eigenvalue problem

$$(1-\delta_{l0})A_{l-2} + (2-\delta_{l0})A_l + A_{l+2} = \lambda A_l, \quad l < l_{\max} \sim n_0^{3/2}. \quad (12)$$

In the system (12) there is no dependence whatever on the field  $V$ . In the limit  $l_{\max} \rightarrow \infty$ , the system (12) has the solution

$$A_l = [-1 + \lambda/2 \pm (\lambda^2/4 - \lambda^{3/2})^{1/2}], \quad (13)$$

which is defined for  $\lambda < 0$  and  $\lambda > 4$ . The value of  $\lambda$  is then obtained from Eq. (12) with  $l = 0$ :

$$\lambda - 1 = -1 + \lambda/2 \pm (\lambda^2/4 - \lambda)^{1/2}, \quad (14)$$

which gives  $\lambda = 0$ . This is the only eigenvalue of the system of equations (12) when  $l_{\max} \rightarrow \infty$ . In the case of a finite but large value of  $l_{\max}$  the solution (13) is valid for  $\lambda < 0$  and  $\lambda > 4$ , where according to Eq. (14) the system (12) has no eigenvalues. In the interval  $[0, 4]$  when  $l_{\max}$  is finite, we find that the right-hand side of Eq. (14) is replaced by a function which varies many times between  $-\infty$  and  $+\infty$ , and which passes through zero. This gives rise to  $l_{\max}$  with eigenvalues  $\lambda_\alpha$  ( $\alpha = 1, 2, \dots, l_{\max}$ ) concentrated in the interval  $[0, 4]$ . For our purpose it is important to note that all the eigen-

values of the system (12) are limited to the range  $0 < \lambda_\alpha \leq 4$  and they are different.

It follows from Eq. (11) that in a strong field ( $V > 1$ ) the value of  $|Q_0|$  is large ( $|Q_0| > n_0^{-3}$ ) so that for all  $\lambda_\alpha \sim 1$  the corresponding quasienergies  $\gamma_\alpha$  are localized in those regions where the function  $X(\gamma)$  is small, i.e., near the points described by

$$E^{(n)} = 1/2(E_n + E_{n+1}) \approx E_{n_0} + n_0^{-3}(n - n_0 + 1/2) \quad (15)$$

and located between the energies of the neighboring Rydberg levels of an unperturbed atom. In the last approximate equality [Eq. (15)] we are making use of the approximation of equidistant levels, which is valid if  $|n - n_0| \ll n_0$ . In this approximation the quasienergies of the system in a strong field become

$$\gamma_{\alpha n} = E^{(n)} - (\pi^2 n_0^6 Q_0 \lambda_\alpha)^{-1}, \quad (16)$$

where we have allowed for the fact that

$$\sum_{n=-\infty}^{+\infty} (n+1/2)^{-2} = \pi^2.$$

The result (16) is valid if the second term in the definition (16) of  $\gamma_{\alpha n}$  is small compared with the separation between the neighboring levels  $n_0^{-3}$ , i.e., if

$$|Q_0| \lambda_\alpha > n_0^{-3}. \quad (17)$$

Consequently, the conditions for transition to the strong-field asymptote depend on  $\lambda_\alpha$ . For low values of  $\lambda_\alpha$  ( $\lambda_\alpha \ll 1$ ) there is no transition to the strong field case at values of  $V$  higher than in the case when  $\lambda_\alpha \sim 1$ . If the reverse of the condition (17) is obeyed, the solutions of Eq. (11) are close to  $E_n$ :

$$\gamma_{\alpha n} = E_n + Q_0 \lambda_\alpha. \quad (18)$$

Equation (18) is quite conventional and natural. It describes the splitting and broadening  $\lambda_\alpha \operatorname{Re}Q_0$  and  $\lambda_\alpha |\operatorname{Im}Q_0|$  of the levels. According to the estimates given by Eq. (3), the shift and broadening are of the same order of magnitude. As long as the condition which is the opposite to the inequality of Eq. (17) is satisfied, the splitting and broadening are small compared with the separation between the neighboring levels of an unperturbed system.

On the other hand, Eq. (16) is quite unusual and it describes a new result on narrowing of quasienergy levels in a strong field and localization of such levels near the values  $E^{(n)}$  of Eq. (15), which are thus quasienergies of an atom in a strong field.

Since  $\lambda_\alpha$  is finite ( $\lambda_\alpha \leq 4$ ), there is a minimum field ( $V \sim 1$ ) beginning from which the above-described narrowing and localization effects occur in the quasienergy spectrum. On the whole, the pattern of its evolution is as follows. In a weak field ( $V \ll 1$ ) all the levels of an atom are weakly split and broadened [Eq. (18)]. If  $V \sim 1$ , the broadening and splitting of the levels with the highest values of  $\lambda_\alpha$  (of the order of unity) become comparable with the separation between the levels. For this group of quasienergy levels a discrete structure of the spectrum is lost completely and a quasicontinuum replaces it. Against the background of this quasicontinuum there are still weakly split and broadened lines corresponding to small values of  $\lambda_\alpha$ . In a stronger field

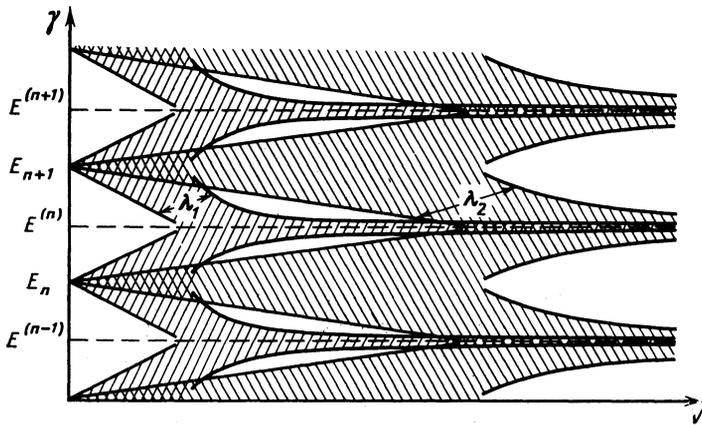


FIG. 1. Quasienergy spectrum of an atom: the shading slanting to the right corresponds to high values of  $\lambda$  ( $\lambda_1 \approx 4$ ) and that slanting to the left corresponds to small values of  $\lambda$  ( $\lambda_2 \ll 1$ ).

( $V > 1$ ) quasienergy levels with the highest values of  $\lambda_\alpha$  become narrower and they are localized near  $E^{(n)}$  [see Eq. (16)]. There are also levels characterized by  $|Q_0|\lambda_\alpha \sim n_0^{-3}$  and which consequently form a quasicontinuum. There are also levels with very small values of  $\lambda_\alpha$  for which we have to assume the reverse of Eq. (17). Such levels are weakly split and broadened even for  $V > 1$  and are close to the corresponding values for  $E_n$  of Eq. (18). Consequently, for all the values of  $V > 1$  the quasienergy spectrum has a quasicontinuum with two groups of narrow levels superimposed on its background: quasienergy levels with  $\lambda_\alpha$  satisfying the condition (17) and localized near  $E^{(n)}$  of Eq. (15), whereas the levels with very small values of  $\lambda_\alpha$  [obeying a condition which is the opposite of Eq. (17)] differs little from  $E_n$ . Figure 1 shows qualitatively the dependence of the structure of the quasienergy spectrum of an atom on a field.

The results described are qualitatively insensitive to the assumptions made: they are insensitive to the approximation (4) and to the absence of resonances  $|\tilde{X}(\gamma_{\alpha n})\tilde{Q}_0| < 1$ . In general, the matrix which governs the coupling between the components of multiplets characterized by different values of  $l$  also becomes more complex. However, even in the general case the field-independent eigenvalues of the corresponding matrices  $\lambda_\alpha$  are bounded, which was used above. Equation (11) for the determination of quasienergies remains valid also in the general case.

#### 4. SOLUTION OF THE INITIAL PROBLEM

In principle, formulation of the problem of complex quasienergies postulates that the interaction is activated instantaneously at the initial moment ( $t = 0$ ). However, it is far from simple to obtain information on the dynamics of the ionization process directly by solving the problem of quasienergies. Therefore, in fact, the initial problem requires separate formulation and solution.

We shall assume that the interaction with an electromagnetic field is instantaneous and it is activated at  $t = 0$ . We shall expand the wave function of an atom in terms of the wave functions of the states  $|nl\rangle$  and  $|ml\rangle$  and of the states in the continuum  $|El\rangle$ . The time-dependent amplitudes of the expansion will be denoted by  $C_{nl}(t)$ ,  $C_{ml}(t)$ , and  $C_{El}(t)$ . In view of the instantaneous nature of the application of the field, the problem can be solved most simply by the Laplace transformation. The Laplace transforms of the required functions will be labeled  $\tilde{C}_{nl}(p)$ ,  $\tilde{C}_{ml}(p)$ , and  $\tilde{C}_{El}(p)$ . If

$\tilde{C}_{El}(p)$  is omitted from the resultant equations, we obtain equations of the type described by Eq. (5) where  $\gamma$  is replaced with  $ip - \omega$  and the right-hand side of the first equation in the system (7) is supplemented by a term  $i\delta_{l0}\delta_{n,n_0}$ . Similar replacements occur also in Eqs. (6) and (7). The additional term on the right-hand side of the first equation in the system (7) is  $i\delta_{l0}(ip - E_{n_0} - \omega)^{-1}$ . We shall consider only the simplest nonresonant case (see above). We shall adopt the eigenvectors  $A^{(\alpha)}$  and the eigenvalues  $q_\alpha$  of the matrix  $Q(Q_{\alpha\alpha'})$ :

$$QA^{(\alpha)} = q_\alpha A^{(\alpha)}, \quad (19)$$

which yields the solution of the equations of  $\tilde{A}(p)$ :

$$\tilde{A}_l(p) = \sum_\alpha \frac{iA_l^{(\alpha)} A_0^{(\alpha)}}{[1 - q_\alpha \tilde{X}(ip - \omega)](ip - E_{n_0} - \omega)}. \quad (20)$$

In the approximation described by Eq. (4), we have  $q_\alpha = Q_0 \lambda_\alpha$ ,  $0 < \lambda \leq 4$ .

#### a) Photoelectron spectrum

The functions  $\tilde{A}_l(p)$  in Eq. (20) determine directly the photoelectron spectrum  $w(E)$  in the limit of infinitely long interactions  $t \rightarrow \infty$ :

$$C_{El}(t)|_{t \rightarrow \infty} = -ie^{-iEt} V n_0^{-3/2} [\tilde{A}_{l-1}(p) + \tilde{A}_{l+1}(p)]_{p = -iE}, \quad (21)$$

$$w(E) = \sum_l |C_{El}(t)|^2 = \frac{V^2 n_0^{-3}}{i \rightarrow \infty (E - E_{n_0} - \omega)^2} \times \sum_l \left| \sum_\alpha \frac{A_0^{(\alpha)} (A_{l-1}^{(\alpha)} + A_{l+1}^{(\alpha)})}{1 - q_\alpha \tilde{X}(E - \omega)} \right|^2. \quad (22)$$

If we ignore both the degeneracy of the levels and their Stark splitting ( $\text{Re } Q \rightarrow 0$ ), then Eq. (22) can be written in the simplest form:

$$w(E) = V^2 n_0^{-3} (E - E_{n_0} - \omega)^{-2} \times \left\{ 1 + \pi^2 V^4 n_0^{-8} \left[ \sum_n (E - E_n - \omega)^{-1} \right]^2 \right\}^{-1}. \quad (23)$$

This is the simplest model result first described in our brief communication<sup>9</sup> [see also Ref. 10].

According to Eq. (23) the photoelectron spectrum is very different for weak ( $V \ll 1$ ) and strong ( $V > 1$ ) fields. In a weak field the function  $w(E)$  is close to a Lorentzian curve

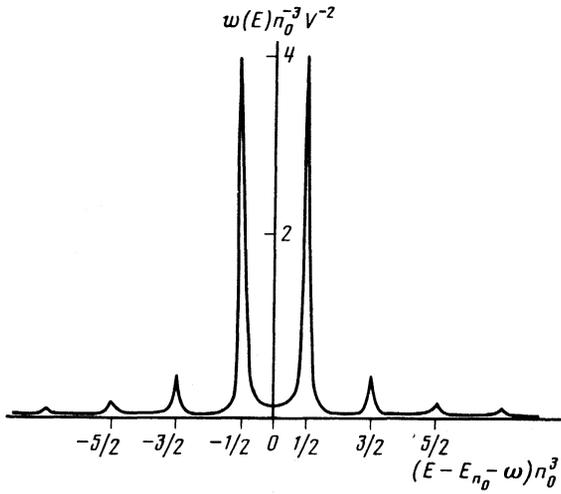


FIG. 2. Photoelectron spectrum deduced using a model of nondegenerate Rydberg levels.

with a maximum at  $E = E_{n_0} + \omega$ , whereas in a strong field the function  $w(E)$  has many peaks (Fig. 2). The maxima of  $w(E)$  are located near  $E^{(n)} + \omega$  of Eq. (15). The widths of the maxima are  $(\pi^3 V^2 n_0^3)^{-1}$  and the amplitudes of the first two maxima (closest to  $E_{n_0} + \omega$ ) are  $4V^2 n_0^3$ . An increase in the field causes narrowing and increases the maxima of the curve  $w(E)$ .

Naturally, the model of nondegenerate and unsplit Rydberg levels is an idealization. However, we shall show that to a great extent it describes correctly also a more complex system with level degeneracy and splitting.

In a strong field ( $V > 1$ ) we find quasienergy levels, localized near  $E^{(n)}$  of Eq. (16) and  $E_n$  of Eq. (18), against the background of a quasicontinuum. We can therefore expect that the photoelectron spectrum will have maxima in the vicinity of the energies  $E_n + \omega$  and  $E^{(n)} + \omega$ . We shall concentrate our attention on maxima of the latter type, i.e., we shall assume that  $E \approx E^{(n)} + \omega$ . Expanding the function  $X(E - \omega)$  near  $E^{(n)}$ , we find that Eq. (22) considered in the approximation of Eq. (4) yields

$$w^{(n)}(E) = \frac{V^2 n_0^3}{(n - n_0 + 1/2)^2} \times \sum_l \left| \sum_{\alpha} \frac{A_0^{(\alpha)} (A_{l-1}^{(\alpha)} + A_{l+1}^{(\alpha)})}{1 + \pi^2 n_0^6 Q_0 \lambda_{\alpha} (E - E^{(n)} - \omega)} \right|^2. \quad (24)$$

The summation with respect to  $l$  and  $\alpha$  requires finding explicitly the eigenvectors  $A^{(\alpha)}$  in Eq. (19). In the approximation of Eq. (4) this problem can be solved in the range of small values of  $\lambda_{\alpha}$  and of  $\lambda_{\alpha}$  close to the maximum value  $\lambda_{\max} = 4$ . In the latter case the functions  $A_l^{(\alpha)}$  can be regarded as smooth fluctuations, so that  $A_{l \pm 2}^{(\alpha)}$  can be expanded in terms of small changes in  $l$  (by  $\pm 2$ ) and the difference equations of the system (12) can be replaced with the differential equations

$$4d^2 A_l^{(\alpha)} / dl^2 + (4 - \lambda_{\alpha}) A_l^{(\alpha)} = 0. \quad (25)$$

The boundary conditions for this equation are

$$A_{l_{\max}}^{(\alpha)} = 0, \quad A_l^{(\alpha)} \approx A_0^{(\alpha)} (2l+1), \quad l \rightarrow 0.$$

The second condition follows directly from the system (25)

when we substitute  $\lambda_{\alpha} \approx 4$ . The normalized solutions of the system (25), satisfying the conditions set out above, are

$$A_l^{(\alpha)} = \frac{2}{l_{\max}^{1/2}} \sin \left[ \frac{(4 - \lambda_{\alpha})^{1/2}}{4} (2l+1) \right],$$

$$\lambda_{\alpha} = 4 - \left( \frac{2\pi\alpha}{l_{\max}} \right)^2 \quad \alpha = 0, 1, 2, \dots \quad (26)$$

In the range of small values of  $\lambda_{\alpha}$  ( $\lambda_{\alpha} \ll 1$ ), the functions  $A^{(\alpha)}$  can no longer be regarded as weakly dependent on  $l$ . It follows directly from the system (12) that  $A_0^{(\alpha)} = -A_2^{(\alpha)} = A_4^{(\alpha)} = -A_6^{(\alpha)} = \dots$  in the limit  $\lambda_{\alpha} \rightarrow 0$ . Making the substitution  $A_{2k}^{(\alpha)} = (-1)^k B_{2k}^{(\alpha)}$ , we obtain in this case functions  $B_l^{(\alpha)}$  which can be regarded as smooth, so that it is possible to introduce a differential equation

$$4d^2 B_l^{(\alpha)} / dl^2 + \lambda_{\alpha} B_l^{(\alpha)} = 0. \quad (27)$$

The normalized solutions satisfying the boundary conditions

$$B_{l_{\max}}^{(\alpha)} = 0, \quad B_l^{(\alpha)} |_{l \rightarrow 0} \approx \text{const},$$

now are of the form

$$B_l^{(\alpha)} = \frac{2}{l_{\max}^{1/2}} \cos \left( \frac{l\lambda_{\alpha}^{1/2}}{2} \right),$$

$$\lambda_{\alpha} = \left[ \frac{2\pi(\alpha + 1/2)}{l_{\max}} \right]^2, \quad \alpha = 0, 1, 2, \dots \quad (28)$$

The square of the modulus of the sum over  $\alpha$  in Eq. (24) represents a double sum over  $\alpha$  and  $\alpha'$ . In the region of  $\lambda_{\alpha} \approx 4$  a weak dependence of  $A^{(\alpha)}$  on  $l$  could be used to ignore the difference between  $A_{l+1}^{(\alpha)}$  and  $A_{l-1}^{(\alpha)}$ . As a result, the summation over  $l$  gives  $\delta_{\alpha\alpha'}$  because of the orthogonality of the functions  $A^{(\alpha)}$  and  $A^{(\alpha')}$ , i.e., a square of the modulus of the sum over  $\alpha$  in Eq. (24) is converted into a sum of the squares of the moduli. At low values of  $\alpha$  we have an analogous situation, except for the important difference that  $A_{l+1}^{(\alpha)}$  and  $A_{l-1}^{(\alpha)}$  differ in respect of the sign and can compensate each other. After substitution of the quantities  $A_l^{(\alpha)}$ , defined by Eq. (28), in Eq. (24) we have to expand  $\cos[\lambda_{\alpha}^{1/2}(l \pm 1)/2]$  in terms of small corrections  $\pm 1$  to  $l \gg 1$  and then summation over  $l$  gives  $\lambda_{\alpha} \delta_{\alpha\alpha'}$ .

It follows from the above discussion that the function  $w^{(n)}(E)$  of Eq. (24), which determines the structure of the maximum in the photoelectron spectrum in the vicinity of  $E^{(n)} + \omega$ , can be represented in the form

$$w^{(n)}(E) = \frac{2V^2 F(E - E^{(n)} - \omega)}{\pi^2 (n - n_0 + 1/2)^2 |Q_0|^2 n_0^6}, \quad (29)$$

where

$$F(x) = \frac{1}{2} \int_0^c \frac{\lambda^{1/2} d\lambda}{(\lambda x + a)^2 + b^2} + \int_c^4 \frac{d\lambda}{(4 - \lambda)^{1/2} [(\lambda x + a)^2 + b^2]}, \quad (30)$$

$$a = \text{Re } Q_0 / |Q_0|^2 \pi^2 n_0^6, \quad b = \text{Im } Q_0 / |Q_0|^2 \pi^2 n_0^6, \quad c \sim 1.$$

Separation of the interval  $[0, 4]$  into parts with large and small values of  $\lambda$  is naturally arbitrary, and the parameter  $c$  separating the two parts can be found to within a coefficient  $\sim 1$ . In writing down the function  $F(x)$  of Eq. (30) in the form of a sum of integrals, we replace summation over  $\alpha$  with

integration with respect to  $\lambda$  and use the dependences  $\lambda(\alpha)$  [Eqs. (26) and (28)] and also the assumptions  $l_{\max} \gg 1$ ,  $l \gg 1$ . The rapidly oscillating functions  $\cos^2(\lambda l^{1/2}/2)$  and  $\sin^2[(4-\lambda)^{1/2}(2l+1)/4]$  are replaced by their average values amounting to  $\frac{1}{2}$ .

Equations (29) and (30) allow us to describe qualitatively the shape of the  $w^{(n)}(E)$  curves. At  $x=0$ , we have

$$F = [1/3c^{3/2} + 2(4-c)^{3/2}] (a^2 + b^2)^{-1} \sim (a^2 + b^2)^{-1}.$$

The derivative  $F'(x)$  differs from zero at  $x=0$ :  $F'(x=0) \propto a(a^2 + b^2)^{-2}$ . Consequently, the point  $x=0$  is no longer the point of a maximum of the functions  $F(x)$  and  $w^{(n)}(E)$ . The maximum of  $w^{(n)}(E)$  is shifted relative to  $x=0$  ( $E = E^{(n)} + \omega$ ) by an amount of the order of  $-a$ , i.e., by a distance of the order of the width of the maximum ( $\sim b$ ). The amplitude of the maximum  $F(x)$  is slightly higher than  $F(0)$  and, consequently, the amplitude of the maximum of the  $w^{(n)}(E)$  curve of Eq. (29) is of the same order of magnitude as in the absence of the degeneracy and Stark splitting of the levels:  $[\omega^{(n)}(E)]_{\max} \propto V^2 n_0^3$ . The function  $F(x)$  of Eq. (30) falls monotonically on increase in  $|x|$  for  $x/a > 0$ . However, if  $x/a < 0$ , then as  $|x|$  increases the function  $F(x)$  first rises, reaches a maximum at  $x \sim -a$ , and then falls. The condition  $x \sim -a$  determines the shift of the maximum and the width of the  $w^{(n)}(E)$  curve, and it is of the order of the width of the curve described by Eq. (23) if the degeneracy and splitting of the levels are ignored:  $\sim (V^2 n_0^3)^{-1}$ . The law describing the fall of the functions  $w^{(n)}(E)$  [Eq. (29)] and  $F(x)$  [Eq. (30)] in the wings of the maximum were  $|x| \gg |a|$  can be estimated quite readily using Eq. (30):  $F(x) \propto |b|^{-1/2} |x|^{-3/2}$ . Therefore, if we allow for the degeneracy and splitting of the levels the structure of the maximum of the function  $w^{(n)}(E)$  of Eq. (29) differs from the structure of the maximum of the function (23) in the model of nondegenerate levels. The main differences are the non-Lorentzian nature of the  $w^{(n)}(E)$  curve of Eq. (29): the asymptote is  $|x|^{-3/2}$  and not  $|x|^{-2}$  and the maximum is shifted by an amount of the order of the width of the curve ( $\sim -a$ ) relative to the value  $E^{(n)} + \omega$ . On the other hand, such basic characteristics of the  $w^{(n)}(E)$  curve as its amplitude and half-width remain unchanged (to within an order of magnitude): they are  $\sim V^2 n_0^3$  and  $\sim (V^2 n_0^3)^{-1}$ , respectively.

## b) Dynamics of decay of an atom

The narrowing and localization of the quasienergy spectrum of an atom described in Sec. 3 are reflected not only in the photoelectron spectrum but also in the dynamics of the ionization process. The time-dependent amplitudes  $C_{Ei}(t)$  of the probability of finding of an electron in the continuum with an energy  $E$  are determined by the familiar Laplace transforms of the amplitudes  $A_i(t)$  of Eq. (20). We can readily show that

$$C_{Ei}(t) = iV(2\pi)^{-1} n_0^{-3/2} \int_{-\infty}^{+\infty} dz \frac{e^{-itz}}{z - E + i0} \times \sum_{\alpha} \frac{(A_{i-1}^{(\alpha)} + A_{i+1}^{(\alpha)}) A_0^{(\alpha)}}{[1 - q_{\alpha} X(z - \omega)] (z - E_{n\alpha} - \omega)}. \quad (31)$$

Equation (31) allows us to determine also the time-dependent total probability of ionization. The calculations

are largely similar to those described in Sec. 4a. Without going into details, we give the final result:

$$w(t) = \int dE \sum_i |C_{Ei}(t)|^2 = 1 + V^2 n_0^{-3} (Q_0'')^{-1} \int_0^{V^2} d\lambda \lambda^{-1/2} \exp(2tQ_0''\lambda) + V^2 n_0^{-3} (Q_0'')^{-1} \left( \int_{V^2}^c \lambda^{1/2} d\lambda + 2 \int_c^4 (4-\lambda)^{-1/2} d\lambda \right) \lambda^{-1} \times \exp\left(\frac{2tQ_0''}{\pi^2 n_0^3 |Q_0|^2 \lambda}\right), \quad Q_0 = Q_0' + iQ_0''. \quad (32)$$

The last two terms on the right-hand side of Eq. (32) determine the nature of the decay of an atom. The term containing the integral with respect to  $\lambda$  between 0 and  $V^2$  represents the contribution of the weakly split and broadened quasienergy states (corresponding to small values of  $\lambda$ ). The last term on the right-hand side of Eq. (32) corresponds to the contribution of the quasicontinuum and of the narrowed (in a strong field) quasienergy levels localized near  $E^{(n)}$ .

In the simplest model which ignores the degeneracy and splitting of the levels there is only one quasienergy state with a given value of  $n$ , the integrals with respect to  $\lambda$  disappear, and the probability  $w(t)$  becomes<sup>9,10</sup>

$$w = 1 - \exp(-2t/\pi^3 V^2 n_0^3). \quad (33)$$

The ionization rate is governed by a constant equal to twice the width of narrow maxima in the photoelectron energy distribution (23). An increase in the field slows down the decay process, i.e., it stabilizes the investigated atom or the populations become trapped in a discrete spectrum. The ionization time of the atom  $t_i \propto V^2 n_0^3$  increases on increase in the field. These effects are due to coherent population of neighboring Rydberg levels in the process of ionization (in a time which is proportional to  $n_0^3$ ) and subsequent interference quenching of transitions from these levels to the continuum.

In a real system with multiple degeneracy of the levels we can expect deviations from the simple exponential dependence of Eq. (33). It follows directly from Eq. (32) that if  $t$  is large, the main contribution to the integrals with respect to  $\lambda$  comes from the range of small values of  $\lambda$  ( $\ll V^2$  between 0 and  $V^2$ ) and  $\lambda$  close to the maximum value ( $4 - \lambda \ll 1$  in the interval from  $c$  to 4). In this approximation the integrals of Eq. (32) are readily calculated and the function  $w(t)$  becomes

$$w(t) = 1 - 1/2 V^2 n_0^{-3} |Q_0''|^{-1} (\pi/2)^{1/2} (t|Q_0''|)^{-1/2} - 2^{1/2} \pi^{1/2} V^2 |Q_0''|^{-1} |Q_0| (t|Q_0''|)^{-1/2} \times \exp(-t|Q_0''|/2\pi^2 n_0^3 |Q_0|^2). \quad (34)$$

The last term on the right-hand side of this equation represents exponential decay similar to that described by Eq. (33). Since  $|Q_0'| \propto |Q_0''| \propto V^2 n_0^{-3}$ , the argument of the exponential function in Eq. (34) and the ionization time are of the same order of magnitude as in the absence of degeneracy:  $t_i \propto V^2 n_0^3$ . A new circumstance, which is due to the degeneracy and quasicontinuum of levels, is the appearance of a power-law pre-exponential dependence in the last term on

the right-hand side of Eq. (34). The corresponding additional factor is  $(t_i/t)^{1/2}$  and it is of the same order of magnitude as the coefficient in front of the exponential function. Consequently, the presence of the quasicontinuum accelerates somewhat the ionization process which becomes rather nonexponential. However, the earlier definition of the characteristic ionization time ( $t_i \propto V^2$ ) is retained and the stabilization of an atom in a strong field takes place.

The second term on the right-hand side of Eq. (34) describes completely nonexponential decay due to the presence of a large number of weakly split and broadened quasienergy levels (with small values of  $\lambda$ ). However, the fraction of such levels decreases on increase in the field and this reduces the nonexponential term in  $w(t) - 1$  [see Eq. (34)]. In fact, we can readily see that if  $t \gtrsim t_i$ , this term is of the order of

$$V^{-2}(t_i/t)^{1/2} \ll V^{-2} \ll 1.$$

The limits of validity of these results are governed mainly by the above-mentioned assumption that the matrix elements  $V_{n_l, n_l'}$  and  $Q_{n_l, n_l'}$  vary slowly with  $n$  and  $n'$ .

<sup>1</sup>A. M. Movsesyan and M. V. Fedorov, Zh. Eksp. Teor. Fiz. **94**, No. 3, 51 (1988) [Sov. Phys. JETP **67**, 462 (1988)].

I. Ya. Berson, Izv. Akad. Nauk SSSR Ser. Fiz. **45**, 2289 (1981).

<sup>3</sup>S. P. Goreslavskii, N. B. Delone, and V. P. Kraĭnov, Zh. Eksp. Teor. Fiz. **82**, 1789 (1982) [Sov. Phys. JETP **55**, 1032 (1982)].

<sup>4</sup>B. A. Zon, L. P. Rapoport, and N. L. Manakov, *Theory of Multiphoton Processes in Atoms* [in Russian], Atomizdat, Moscow (1978).

<sup>5</sup>H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Systems*, Springer Verlag, Berlin (1958); reprinted from: *Handbuch der Physik* (ed. by S. Flügge), Vol. 35, Springer Verlag, Berlin (1957), pp. 88–346.

<sup>6</sup>L. D. Landau and E. M. Lifshitz, *Quantum Mechanics: Non-Relativistic Theory*, 2nd ed., Pergamon Press, Oxford (1965).

<sup>7</sup>R. Grobe, G. Leuchs, and K. Rzazewski, Phys. Rev. A **34**, 1188 (1986).

<sup>8</sup>Z. Deng and J. H. Eberly, J. Opt. Soc. Am. B **2**, 486 (1985).

<sup>9</sup>M. V. Fedorov and A. M. Movsesian, J. Phys. B **21**, L155 (1988).

<sup>10</sup>M. V. Fedorov and A. M. Movsesyan, *Elementary Processes in a Laser Radiation Field* [in Russian], Council on Spectroscopy of the USSR Academy of Sciences, Moscow (1988), p. 56.

<sup>11</sup>V. S. Lisitsa and S. I. Yakovlenko, Zh. Eksp. Teor. Fiz. **66**, 1981 (1974) [Sov. Phys. JETP **39**, 975 (1974)]; S. I. Yakovlenko, Preprint No. 2824 [in Russian], Institute of Atomic Energy, Moscow (1977).

<sup>12</sup>M. V. Fedorov, J. Phys. B **10**, 2573 (1977).

<sup>13</sup>N. B. Delone and M. V. Fedorov, Tr. Fiz. Inst. Akad. Nauk SSSR **115**, 42 (1980).

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