Mixing of discrete and autoionizing states of an atom by a strong oscillating field

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The results are given of a numerical solution of the problem of mixing of discrete and autoionizing states of an atom by a strong electromagnetic field. The specific example of the barium atom is used in an analysis of the different ways in which the matrix element of the interaction of the atom with the field and the detuning from a resonance can be related. The states whose dynamic dipole moments vanish in a strong field are derived. A study is made of the influence of the field intensity on the configurational composition and width of the levels. The various processes which determine the profile of a preionization resonance in a strong oscillating field are considered.

1. INTRODUCTION

The change in the spectrum of an atom with a complex multiplet structure of terms under the action of a strong oscillating field is of interest because of the numerous experiments in which heavy atoms are investigated in laser fields.

The multiplet structure of the terms of heavy atoms is usually due to the spin–orbit splitting, which ranges from tens to hundreds of reciprocal centimeters. If the separation δE between the levels is considerably greater than the matrix element of the interaction of an atom with a field V, the multiplet structure of a term can be ignored. The problem then reduces to the thoroughly studied cases of resonant and quasiresonant two-level interactions with an external field or to an investigation of the behavior of an isolated level in a resonant field (see, for example, the review in Ref. 1). The role of the multiplet term structure increases when the separations δE become of the order of the matrix element V with increasing radiation field intensity. An interaction of this type cannot be considered using perturbation theory; moreover, it is a multilevel problem.

An analytic solution of the problem of modification of an atomic multiplet in a strong oscillatory field was obtained in Ref. 2. The cases of resonant mixing and nonresonant perturbation of discrete levels of an atom by a field were considered.

The present paper reports analogous numerical investigations for the case of mixing of discrete and autoionizing states of a complex atom. It follows from experimental investigations reported in Refs. 3–5 that resonant and quasiresonant mixing of such states may play a considerable role in the process of multiphoton ionization of atoms. A study of the characteristics of autoionizing and discrete states in a strong field should help in the interpretation of experimental data.

It is particularly interesting to investigate the physics of narrowing and broadening of autoionizing resonances in a strong oscillatory field. The possibility of a major change in autoionizing resonances in a strong field (particularly of narrowing) was pointed out by Geller and Popov.[6] The narrowing mechanism was assumed to be interference between the channels of decay of an autoionizing state: direct decay to the continuum and transition to the continuum via a discrete level. The conditions for narrowing a profile to the radiative width were considered and the results should be useful in a number of applications. However, the absence of any actual data on quantitative relationships between the amplitudes of the decay of autoionizing states in different channels prevented Geller and Popov[6] from identifying the main mechanism for the formation of a profile of an autoionizing line. We shall use numerical examples to show (Sec. 4) that interference usually plays a secondary role, compared with the process of resonant mixing of discrete and autoionizing states, in narrowing a resonance profile in a field.

It is well known that the narrowing of a line profile is hindered by ionization broadening, which becomes the dominant process if the field is sufficiently strong. A consistent allowance for these and other possible decay channels governing the profile of a preionization resonance was made in Ref. 7. General formulas were obtained for the parameters of a profile in a field. It was concluded that it is necessary to allow for noninterfering channels that limit line narrowing, and a description was given of the conditions under which interference between the channels is most effective. The general analytic conclusions of Ref. 7 were confirmed numerically in the present investigation for the specific autoionizing states of the barium atom when all the decay channels were allowed for.

Experimental studies of nonlinear ionization of alkaline earth atoms[4,8,9] yielded results which have not yet been fully interpreted. The following circumstance observed in a study of multiphoton ionization of Sr and Ba atoms[8] is worth noting. Although the majority of the resonances reported in Ref. 8 can be identified using the data on the spectra of a netural atom and of the corresponding ion, there are some cases when resonances with intermediate states located in the same range of energies and allowed by the selection rules are missing from the spectrum. We may assume that the characteristic features of such an ionization process are due to the complex structure of the spectrum of two-electron states of the Sr and Ba atoms.

Experimental studies of the multiphoton resonance widths of the Sr atom as functions of the laser radiation intensity[4] are also of major interest. The authors of Ref. 4 assumed that the appearance, with increasing field, of a narrow resonance superposed on a wide band arises because the field acts differently on autoionizing and discrete states. The former become broader and the latter are independent of the field. In our opinion, such an interpretation of the results requires fuller justification.

Finally, the spectrum of three-photon ionization of the Ba atom[9] includes a line corresponding to a resonance with the $5d\,7d\,{}^3D_2$ discrete state, found to have an anomalously large shift and broadening in a relatively weak field (10^{-4}–10^{-5} a.u.) compared with other lines in the spectrum. Ac-

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cording to Kelly *et al.*,[9] this is due to the influence of the spectrum of autoionizing states, the details of which are not yet clear.

By using the many-electron approximation we hope to construct a model of the interaction of an atom with radiation sufficiently realistic to yield specific answers to some of the problems raised by the experimental results. Calculations will be made on the basis of the many-configuration Hartree–Fock–Dirac method, which has made it possible to obtain accurate values of the matrix elements.

Our model is based on the following approximations.

1. Two multiplets of atomic energy levels with opposite parity, coupled by the dipole interaction, are considered.

2. The separations ΔE between the different levels of the multiplets and the frequency of the field ω satisfy the following relationships: $\Delta E > \omega$ and $\Delta E \leqslant \omega$.

3. The multiplets are characterized by large defects of the field frequency compared with other states of the atom.

4. The widths of the levels Γ and the fine-structure splitting δE are related in the present problem by $\delta E > \Gamma$, which makes it possible to allow for the broadening due to preionization and ionization in first-order perturbation theory.

The calculations are carried out for the case of circular polarization. The treatment for an arbitrary polarization presents no special problems and can be considered similarly.

2. RELATIVISTIC FORM OF THE METHOD OF QUASIENERGIES FOR CALCULATING THE BEHAVIOR OF A MANY-ELECTRON ATOM IN AN EXTERNAL FIELD

In numerical solution of the problem of the interaction of an atom with an oscillating field of circular polarization we shall use the method of rotation of coordinates which has been employed successfully in various problems dealing with quasistationary and discrete states in static[10] and time-dependent[11] fields. The relativistic variant of this method makes it possible to carry out the fullest and most consistent calculation dealing with the behavior of a many-electron atom in an external field.

An investigation of the role of relativistic effects in the case of excited states of heavy atoms ($56 \leqslant z \leqslant 70$) demonstrated[12] that corrections to these effects can have a significant influence on the motion of excited electrons, primarily because of a strong influence on the states of the core electrons, i.e., on the core potential which is experienced by excited electrons.

In this section we shall obtain the relativistic matrix element of the interaction of an atom with a field in a form suitable for numerical computations and describe the main assumptions made in the calculation method.

We shall consider a many-electron atom in a circularly polarized oscillatory field. The Dirac Hamiltonian in an external field (in the atomic system of units) is

$$H_D = \sum_i \left[c\alpha_i \left(\mathbf{p}_i - \frac{\mathbf{A}_i(t)}{c} \right) + \beta_i c^2 - \frac{Z}{r_i} \right] + \frac{1}{2} \sum_{i \neq j} \frac{1}{|r_i - r_j|}, \tag{1}$$

where α and β are the Dirac matrices, $\mathbf{p}$ is the electron momentum, Z is the charge of the atomic nucleus, and $\mathbf{A}(T)$ is the vector potential of the external field:

$$\mathbf{A}(t) = A_0(\mathbf{n}_x \cos\alpha + \mathbf{n}_y \sin\alpha), \quad A^2 = A_0^2, \quad \alpha = \omega t - \mathbf{kr}, \quad k = \omega/c. \tag{2}$$

The corresponding wave function of an atom in a field which varies periodically with time can be written as follows:

$$\Psi_E(t+T) = e^{-iET} \Psi_E(t). \tag{3}$$

The function (3) describes what is known as the quasienergy state, corresponding to a quasienergy E. It follows from Eq. (3) that the quasienergy is given to the nearest integer by $\omega = 1/T$. Simple transformations yield the Dirac equation for calculating the quasienergy states of an atom in a system of coordinates rotating at the field frequency ω:

$$H_D^f = H_D^{(0)} - \alpha \mathbf{A}(0) - \omega n J_z, \tag{4}$$

where $H_D^{(0)}$ is the Dirac Hamiltonian in the absence of a field.

The quasienergies in a strong field are obtained when Eq. (4) is solved exactly by diagonalization of the corresponding energy matrix $\{H_{\alpha\beta}\}$. The wave function $\varphi(\mathbf{r})$ is represented by a linear combination of determinants:

$$\varphi(\mathbf{r}) = \sum_\alpha c_\alpha \det_\alpha(m), \tag{5}$$

where α labels both the states of one configuration the mixing of which gives an intermediate type of coupling, and states of different configurations which describe correlation effects in the case of states of one symmetry and mixing of the states by an external field if they have different symmetries; $\det(m)$ is the eigenfunction of the operator $\hat{J}_z$ and it is composed of one-electron wave functions $f(r)$ found by solving the Hartree–Fock–Dirac equations in the absence of the field[13]:

$$f(\mathbf{r}) = \frac{1}{r} \begin{pmatrix} P_{n\tilde{k}}(r)\, \chi_{\tilde{k}m}(\theta, \varphi) \\ -iQ_{n\tilde{k}}(r)\, \chi_{-\tilde{k}m}(\theta, \varphi) \end{pmatrix}, \tag{6}$$

$P_{n\tilde{k}}$, $Q_{n\tilde{k}}$ are the large and small components of the radial wave function; $\chi(\theta,\varphi)$ is the spin–orbit wave function.[13] The coefficients c_α can be found from a variational principle based on the following system:

$$\sum_\beta H_{\alpha\beta} c_\beta = E c_\alpha, \quad H_{\alpha\beta} = \langle \alpha | H_D | \beta \rangle. \tag{7}$$

The system (7) constitutes an eigenvalue problem, which can be solved by diagonalization of the matrix $\{H_{\alpha\beta}\}$. It follows from Eq. (3) that the eigenvalues E represent quasienergies of the system under discussion. The matrix elements of the operator $H_D^{(0)}$ occurring in Eq. (4) are given in a form convenient for numerical calculations in our earlier papers.[13,14] Here we shall write down only the matrix element of the interaction of an atom with the field. We shall assume that a and b are unperturbed states belonging to different multiplets with magnetic quantum numbers m_a and m_b, differing by unity in a circularly polarized field, so that the general form of the matrix element is

$$\sum_i \langle a | \alpha_i \mathbf{A}_i(0) | b \rangle = \sum_i \sum_{\mu = \pm 1} (-1)^\mu \frac{A_0}{2^{1/2}} \langle a | \alpha_{i\mu} e^{i\mathbf{kr}} | b \rangle. \tag{8}$$

Substituting

$$e^{ikr}=4\pi\sum_i\sum_{l,Q} i^l j_l\left(\frac{\omega_{ab}}{c}r\right)Y_{lQ}^*(\mathbf{r}_i)\,Y_{lQ}(\mathbf{k}),$$

we obtain

$$\sum_i\langle a|\alpha_i\mathbf{A}_i(0)|b\rangle$$

$$=2^{1/2}A_0\sum_{\mu=\pm1}(-1)^\mu\sum_{l,Q} i^l Y_{lQ}(\mathbf{k})\sum_i\langle a|\alpha_i j_l Y_{lQ}^*(\mathbf{r}_i)|b\rangle, \quad (9)$$

where

$$\sum_j\langle a|\alpha_i j_l Y_{lQ}^*(\mathbf{r}_i)|b\rangle$$

is the many-electron element which can be represented by a linear combination of one-electron matrix elements:

$$V_\mu(a,b)=\sum_{\alpha,\beta} c_\alpha^*(a)\,c_\beta(b)\langle\alpha|V_\mu|\beta\rangle, \quad (10)$$

where $j_l(\omega_{ab}r/c)$ is a Bessel function; ω_{ab} is the transition frequency of an unperturbed atom; l assumes the values $k-1$, k, and $k+1$; if $l=k$, the photon is of the magnetic type, whereas for $l=k-1$ and $k+1$, it is of the electric type. In the dipole approximation, we find that

$$\frac{\omega_{ab}}{c}r\to 0, \quad j_l\left(\frac{\omega_{ab}}{c}r\right)\to\delta_{l0}$$

for $l=0$ and $j_0=1$.

The quantity $\langle\alpha|V_\mu|\beta\rangle$ is described by

$$\langle\alpha|V_\mu|\beta\rangle=\frac{F}{2^{1/2}c}\frac{(2j_b+1)^{1/2}}{(2j_a+1)^{1/2}}C^{j_a\,1/2}_{10,\,j_b\,1/2}C^{j_a m_a}_{j_b m_b,\,1-1}$$
$$\cdot\sum_{\nu=\pm1}\langle\Phi_\alpha(\nu)|\Phi_\beta(-\nu)\rangle[k_\alpha-k_\beta-\nu], \quad (11)$$

where $C^{jm}_{j_1m_1,j_2m_2}$ is a Clebsch–Gordan coefficient,

$$\Phi(\nu)=\begin{cases}P(r), & \nu=1,\\ Q(r), & \nu=-1,\end{cases}$$

and $F=A_0/c$ is the field intensity.

3. SPECIFIC MODEL FOR NUMERICAL CALCULATIONS

A numerical calculation of the mixing of discrete and autoionizing states in a strong oscillatory field was carried out for the specific case of the barium atom. Extensive experimental data are now available on the process of nonlinear ionization of Ba atoms (see, for example, Refs. 8 and 9) and interpretation of these data requires information on the influence of strong laser radiation on the atomic spectrum.

The discrete spectrum of the barium atom includes not only one-electron, but also two-electron states formed as a result of excitation of the outer $6s^2$ shell. The preionization spectrum of Ba between the first ($5p^66s$) and second ($5p^6$) ionization potentials include states with two-electron excitation $5p^6nln'l'$. Our calculations included two groups of levels belonging to the discrete $5d\,6d$ and autoionizing $6p6d$ configurations, the latter formed by excitation of the $5d$ valence electron. The physical justification for considering these two groups of levels is that in the process of multiphoton ionization of Ba atoms by radiation representing the second harmonic of a Nd laser ($\omega=16\,800$–$18\,000\ \mathrm{cm}^{-1}$) the levels of

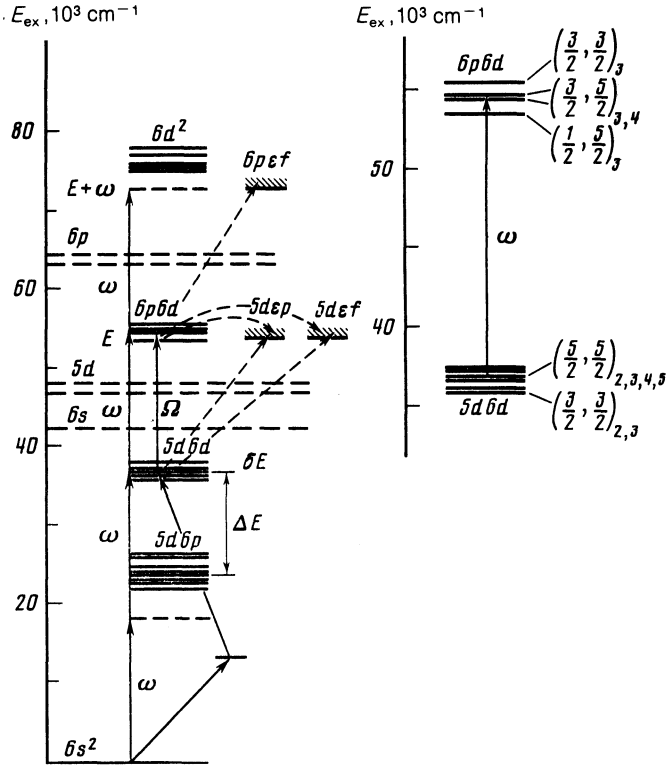

FIG. 1. Level diagram showing the terms of the barium atom: ω and Ω are the frequencies of strong and weak laser radiation fields, respectively; ΔE is the energy separation to the next multiplet; δE is the fine-structure splitting.

these configurations can give rise to intermediate resonances: a two-photon resonance in the bound $5d\,6d$ state followed by a one-photon resonance with the autoionizing state.[3] The excitation scheme of Ba is shown schematically in Fig. 1. The radiation frequency $\omega_T=18\,000\ \mathrm{cm}^{-1}$ used in the calculations was selected from the range of the second harmonics of an Nd laser in such a way as to ensure the excitation scheme of Fig. 1.

In the absence of a field the multiplet structure is characterized by the following relationships: 1) six levels of the $5d\,6d$ configuration ($J=5$–2) and four levels of the $6p6d$ configuration ($J=4$–2) are considered; 2) the separations δE between the levels obtained from $\delta E_{\max}$ to $\delta E_{\min}$ (Fig. 1) satisfy the inequality $\delta E\ll\omega$; 3) the separation from a multiplet to neighboring levels of other multiplets is $\Delta E\gg\delta E$; 4) in the case of the selected frequency ω_T there are near-resonances with the levels $5d\,6d(J=5)$–$6p6d(J=4)$ where $\Delta=0.11\ \mathrm{cm}^{-1}$. The remaining levels are coupled by the field quasiresonantly to a greater or lesser extent (Δ varies form $100\ \mathrm{cm}^{-1}$ to $1700\ \mathrm{cm}^{-1}$).

The large number of levels in the multiplets makes it possible to consider in the same problem a whole range of different ways an atom on the field characterized by a very wide range of relationships between the matrix element V and the detuning Δ.

In the case of resonant multistage excitation of alkaline earth atoms by radiation from a tunable laser it is possible to fill effectively discrete two-electron states and then ionize them via the autoionizing states. We shall consider the effects of a strong oscillating field of frequency ω on a system of discrete and autoionizing multiplet terms between which

TABLE I. Notation used for the quasilevels of Ba investigated.

Level No.	Principal component in weak field	J	m	Level No.	Principal component in weak field	J	m
1	$6p_{1/2}6d_{5/2}$	3	3	6	$6p_{3/2}6d_{5/2}$	4	3
2	$5d_{3/2}6d_{3/2}$	3	2	7	$5d_{5/2}6d_{5/2}$	5	2
3	$5d_{3/2}6d_{3/2}$	2	2	8	$5d_{5/2}6d_{5/2}$	4	2
4	$6p_{3/2}6d_{3/2}$	3	3	9	$5d_{5/2}6d_{5/2}$	2	2
5	$5d_{5/2}6d_{3/2}$	3	2	10	$6p_{3/2}6d_{3/2}$	3	3

transitions take place under the influence of an additional weak field of frequency Ω which can be varied. This makes it possible to study the line profiles at different intensities of the strong field.

In view of the complex structure of such an atom, we shall confine our attention to transitions between the levels which have the magnetic quantum numbers $m = 2$ and $m' = 3$ in a strong field and which in the limit of a weak field ($F = 0$) correspond to the levels $J = 2$ and $J = 3$. It is these states that are filled effectively under real experimental conditions. The frequency Ω of the weak field, which induces the transitions, is selected in the range of the transition frequencies between the levels of multiplets allowing for the splitting and shifts in the strong field.

In the limit $F \to 0$ the width of an autoionizing state is governed by spontaneous decay into an ion and a free electron due to a correlating electron–electron interaction. In the case of the $6p6d$ states ($J = 3$) the decay is to the $5d\varepsilon p$ and $5d\varepsilon f$ continua. The application of the strong field gives rise also to transitions from the autoionizing states to higher excited states in the continuum. The greatest contribution to broadening of this line is made in our model by transitions of the $6p6d \to 6p\varepsilon f$ type with an energy $E + \omega$. The strong field also results in interference between the decay channels of the autoionizing states: direct decay to a continuum and decay to a continuum via a discrete level.

4. DEPENDENCE OF QUASIENERGY CHARACTERISTICS ON THE OSCILLATORY FIELD STRENGTH

In the present section we solve the problem of mixing of atomic multiplets by an oscillatory field for different relationships between V and Δ, allowing for the multilevel nature of the interaction. It is known that circularly polarized radiation splits the levels of an atom and mixes the states of opposite parity characterized by different quantum numbers. However, the problem can be simplified by selecting a specific direction of polarization, such as $\Delta m = +1$. Then all the sublevels split into groups of states interacting in the field, characterized by $m_b - m_a = 1$. We shall consider the specific case of one of these groups when $m_{5d\,6d} = 2$ and $m_{6p6d} = 3$. Table I gives the notation used for such states. The eigenvalues of the matrix $H_{\alpha\beta}$ of Eq. (7) are plotted as a function of the field strength F in Fig. 2. In accordance with the quasienergy representation, the energies of the sublevels $5d\,6d$ and $6p6d$ are shown in the same region, so that the distances between them determine the detuning from resonances. The average energy of a quasimultiplet is regarded as zero in measuring the energies.

An analysis of the results obtained shows that in the range $0 \leqslant F \leqslant 5 \times 10^6$ V/cm there are interactions of two types:

a) a resonant two-level interaction $V > \Delta$ with a linear dependence $E(F)$ (levels 4–5 and 6–7);

b) a nonresonant interaction $V > \Delta$ with a quadratic dependence $E(F)$ (levels 1–3 and 8–10). In the latter case the energy structure differs little from the structure of an atom in the absence of the field.

Clear characteristics of the interaction effects are the coefficients c_α representing mixing of the states and obtained as a result of diagonalization of $H_{\alpha\beta}$ of Eq. (7). Figure 3 shows how the squares of these coefficients C^2 depend on the field strength F. They satisfy the identity

$$\sum_\alpha c_\alpha^2(5d6d) + \sum_\beta c_\beta^2(6p6d) = 1.$$

In the range 5×10^6 V/cm $< F \leqslant 5 \times 10^7$ V/cm all the levels have linear Stark shifts if we ignore the frequency defect $V > \Delta$. The linear behavior of $E(F)$ indicates the effective dynamic dipole moments of the atom in different states. An increase in the field intensity alters the quasienergy spec-

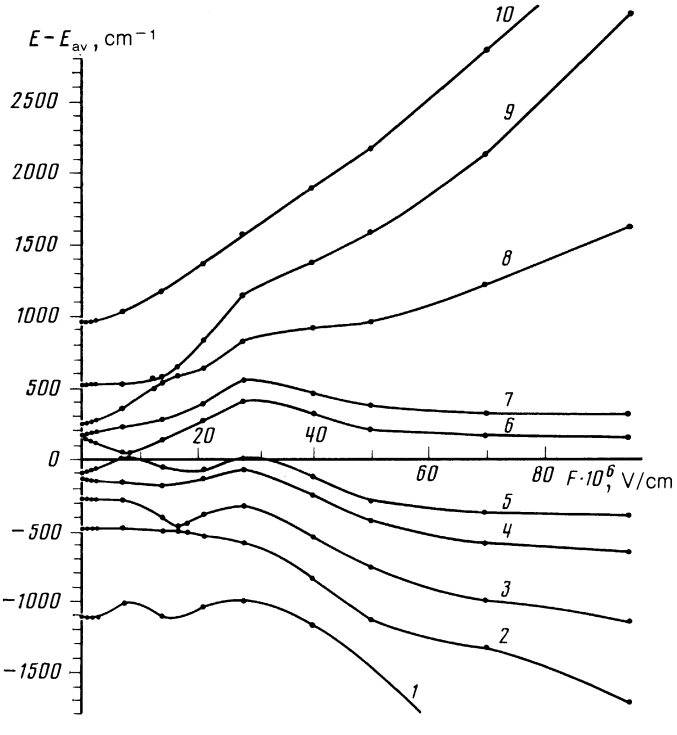

FIG. 2. Quasienergy structure of the Ba atom plotted as a function of an external oscillatory field F; here, e_{av} is the average energy of a quasimultiplet. The levels are numbered in the same way as in Table I.

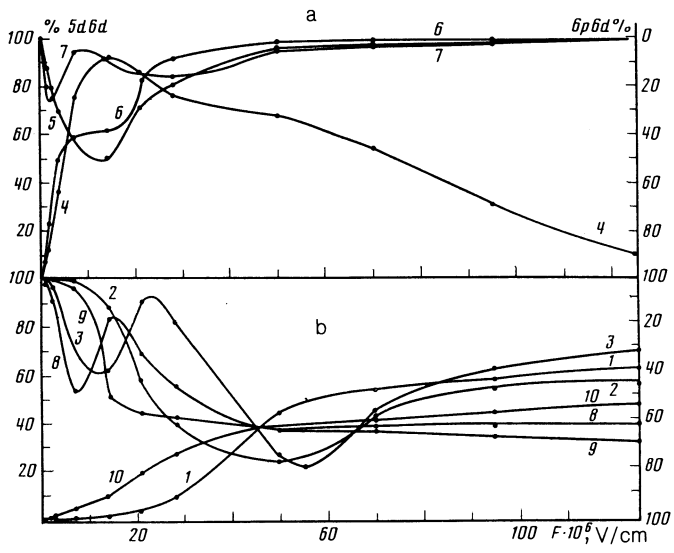

FIG. 3. Squares of the coefficients representing an admixture of states of different parity plotted as a function of the field intensity F: a) resonant interaction; b) nonresonant interaction. The levels are numbered in the same way as in Table I.

trum so that it becomes closer to the spectrum of eigenvalues of the dipole moment operator matrix multiplied by the field intensity F. The fine-structure splitting of the multiplets now plays an insignificant role. Since the reversal of the field F to $-F$ does not alter the nature of the spectrum governed by the dipole moment operator, it follows that in high fields F we can expect the appearance and increase in the symmetry of the spectrum of quasienergies relative to the average energy E_{av} (in Fig. 2 the value of E_{av} is taken as zero). Quasicrossing of the levels 2–3, 5–6, and 8–9 alters greatly their characteristics, particularly inducing jumps of the coefficients c_α^2 right up to complete exchange of the configurational compositions of the levels 5 and 6 (Fig. 3).

In the range of high values of $F > 5\times 10^7$ V/cm ($V \gg \Delta$) such symmetrization of the quasienergy structure gives rise to quasilevels with zero eigenvalues of the matrix of the dipole moment operator. Depending on the number of unperturbed states, the number of such quasilevels is even or odd. These quasilevels correspond to wave functions derived from the states of one multiplet of specific parity, as a result of which the average value of the dipole moment vanishes. It follows from the symmetry of the problem that in our case there are four such states (levels 4–7). The $E(F)$ curves for these states are parallel to the abscissa (Fig. 2) and an admixture of the states of the opposite parity tends to zero (Fig. 3a). In the case of the states with a finite value of the dipole moment (levels 1–3 and 8–10) in strong fields F the dependences $E(F)$ and $c_\alpha^2(F)$ are linear and an admixture of the states of different parity tends to 50%. In spite of the fact that in this range the interaction is of multilevel nature, it is similar to the two-level resonant interaction in relatively weak fields.

5. CHANGES IN LINE PROFILES IN A STRONG OSCILLATORY FIELD

The cross section of the transition between discrete and autoionizing states is described by an expression analogous to the Fano formula[15] modified to deal with the case of several decay channels of an autoionizing state in the presence of a strong field. The widths of the upper and lower levels of such a line include broadening due to ionization and preionization, calculated allowing for the mixing and interference of the states:

$$\Gamma^{(i)}=(\Gamma_{\text{авт}}+\Gamma_{\text{ион}})^{(i)}=2\pi\left[\sum_j\Big|F\sum_\alpha c_\alpha^{(i)}V_{\alpha j}^{(i)}+\sum_\beta c_\beta^{(i)}W_{\beta j}^{(i)}\Big|^2+\sum_{j'}\Big|F\sum_\beta c_\beta^{(i)}V_{\beta j'}^{(i)}\Big|^2\right],\quad i=1,2,\quad (12)$$

where j labels the states in the continuum with energy E and j' labels the states with energy $E+\omega$, to which dipole transitions from autoionizing and discrete states are possible; $W_{\beta j}=\langle \det_j|r_{12}^{-1}|\cdot\det_\beta\rangle$ is the matrix element of a nonradiative transition of the component β of the state i to the continuum j; $V_{\alpha j}^{(i)}=\langle \det_j|\hat V|\det_\alpha\rangle$ is the matrix element of the dipole transition of the component α of the state i to the continuum j, which can have the energies E or $E+\omega$. Since the operator $\hat V$ is diagonal in the one-electron approximation, it has no components between the wave functions with different values of m for the cores. This reduces greatly the number of channels participating in the interference process of Eq. (12). The matrix elements $W_{\beta j}$ and $V_{\alpha j}$ were calculated using methods developed earlier.[16,17] A comparison of the results of a calculation of the oscillator strengths and widths of autoionizing levels[16,17] with the corresponding experimental data showed that the average error of the calculations of such quantities was 10–15%. The matrix element of the radiative transition in a weak field from a discrete state to an autoionizing state is given by the following expression obtained in the multiconfigurational approximation:

$$V_{12}=\sum_{\alpha,\beta}c_\alpha c_\beta\langle\det_\beta|\hat V|\det_\alpha\rangle.\quad (13)$$

Using Eqs. (12) and (13), we can write down the preionization cross section in atomic units:

$$\sigma_{12}(\Omega)=\frac{4\pi\Omega}{c}\Big[\frac{2}{\pi}\Big(\sum_i\Gamma^{(i)}\Big)^{-1}|V_{12}|^2\sin^2\delta-\frac{2^{3/2}}{\pi^{1/2}}\cdot V_{12}\Big(\sum_i\Gamma^{(i)}\Big)^{-1/2}\sum_j\sum_\alpha c_\alpha V_{\alpha j}\cos\delta\sin\delta+\Big|\sum_j\sum_\alpha c_\alpha V_{\alpha j}\Big|^2\cos^2\delta\Big],\quad i=1,2.\quad (14)$$

Here $\delta(\Omega)$ is the shift of the phase of the wave function of the state 2 because of the interaction with the continuum:

$$\delta(\Omega)=-\operatorname{arctg}\frac{1}{2}\frac{\Gamma^2}{\Omega-\Omega_0-F(\Omega)},\quad (15)$$

where Ω_0 is the position of the line maximum obtained ignoring the interaction of Eq. (15) with the continuum, and $F(\Omega)$ is the shift of the position of the maximum relative to Ω_0 governed by the principal value of an integral of the form

$$F(\Omega)=\frac{1}{2\pi}P\int d\Omega'\frac{\Gamma^{(2)}}{\Omega-\Omega'}.\quad (16)$$

An investigation of the dependence of the line profile on the field intensity first of all gives the changes in the parameters V_{12} and Γ, which occur in σ_{12} of Eq. (14), under the influence of a strong quasiresonant field.

TABLE II. Squares of matrix elements of dipole transitions in an atom of barium subjected to an external field of different intensities.

Levels participating in transition	Transition (principal components)	m	m'	Ω_T, cm^{-1}	$\lvert V_{12}\rvert^2$, (a.u.)^2			
					$F=0$	$F=7\cdot10^6$ V/cm	$F=1{,}4\cdot10^7$ V/cm	$F=7\cdot10^7$ V/cm
3—1	$5d_{3/2}6d_{3/2}-6p_{1/2}6d_{5/2}$	2	3	17 165	0.0045	0.00214	0.00018	0.00014
3—4	$5d_{3/2}6d_{3/2}-6p_{3/2}6d_{5/2}$	2	3	18 128	0,0112	0.0032	0,0040	0,0033
9—4	$5d_{5/2}6d_{5/2}-6p_{3/2}6d_{5/2}$	2	3	17 340	0,0154	0,0003	0.0080	0.0004
9—1	$5d_{5/2}6d_{5/2}-6p_{1/2}6d_{5/2}$	2	3	16 368	0.0026	0.0054	0.5100	0.0600
3—10	$5d_{3/2}6d_{3/2}-6p_{3/2}6d_{3/2}$	2	3	16 758	0,0940	0.0760	0,0025	0.0036

a. Influence of a field on the magnitudes of the matrix elements of a dipole transition

It is clear from Eq. (13) that the dependence $V_{12}(F)$ is governed by the coefficients $c_\alpha(F)$ and $c_\beta(F)$ which characterize different components included in the multiconfigurational wave functions of the initial and final states of the transition; $c_\alpha(F)$ and $c_\beta(F)$ vary with increase in F because of a change in the relative separation of levels in a quasimultiplet. We can see that modification of the structure of terms of the same parity changes the type of coupling, whereas modification of terms of the opposite parities alters the effective dipole moment.

The behavior of the coefficients c_α and c_β in a field was considered in the preceding section (Fig. 3) for different variables of the $5d\,6d$ and $6p6d$ configurations of Ba; here, we shall give the relevant values of the squares of the matrix elements V_{12}^2 at fixed field intensities for a number of transitions between the variables of a quasimultiplet (Table II). We can see from Table II that in the strong-field limit $F\to F_{at}$ the values of V_{12} decrease; this is due to equalization of the impurity composition of the levels (Fig. 3b). We can easily show that for a two-level system in a field we have $V_{12}=(c_1^2-c_2^2)\langle 2|\hat{V}|1\rangle$, where c_1 and c_2 are related by $c_1^2+c_2^2=1$. Clearly, the field increases we have $c_1^2\approx c_2^2$ and $V_{12}\to 0$. For a many-level system we cannot write down $V_{12}(F)$ in a simple form and , moreover, in the case of some transitions the value of V_{12} can increase considerably with F, followed by a fall in the limit $F\to F_{at}$ (transition $9\to 1$, Table II). The nonmonotonic behavior of $V_{12}(F)$ is due to interference between a maximum in the expansion of V_{12} given by Eq. (13) in the range of level quasicrossing, which appears when the shifts of the levels in the field are close to the fine-structure splitting of the terms. In our case level quasicrossing is observed in the range $F=10^7$ V/cm (Fig. 2).

b. Width of a level in a field

The width of an autoionizing state depends on the ratio of the coefficients $c_\alpha(F)$ and $c_\beta(F)$ of the matrix elements describing radiative and nonradiative decay to a continuum [first two terms in Eq. (2)]. Figure 4 shows how the widths of these autoionizing levels depend on the field F, allowing for the mixing of these levels by a strong field. The levels of the $6p6d$ configuration (1, 4, and 10 in Fig. 4) become narrower with increasing field in the range $F\leqslant 5\times10^7$ V/cm, whereas the levels of the $5d\,6d$ configuration (3, 5, 9) become broader. It is well known that in a closed system the sum of the squares of the mixing coefficients of the states remains constant. However, in the case of the curves shown in Fig. 4 this condition is not satisfied because this figure represents only some of the levels included in the calculation of a quasimultiplet.

The behavior of the width of quasiionizing levels Γ_{aut} in the range of fields $F>5\times10^7$ V/cm varies with the nature of the states. The states with zero dipole moment are described by a function of specific parity in the strong-field limit (Fig. 2). The width Γ_{aut} for these states either tends to the radiative width (level 5 in Fig. 4) or to the sum of the widths of the autoionizing states belonging to a given configuration (level 4). The states with a nonzero dipole moment can be represented by a superposition of the wave functions of different parities. In the limit $F\to F_{aut}$ the widths of these autoionizing states decrease monotonically, tending to a specific limit, which in our case lies within the interval 10–15 cm^{-1}. The dashed curves in Fig. 4 represent the F-dependence of the total widths of the levels in a field, which include widths due to preionization and ionization [Eq. (12)].

Table III gives the values of the autoionizing widths of the levels calculated allowing (Γ_{aut}^{int}) and ignoring (Γ_{aut}) the interference between the decay channels:

$$\Gamma_{aut}^{int}=2\pi\sum_j\left|F\sum_\alpha c_\alpha V_{\alpha j}+\sum_\beta c_\beta W_{\beta j}\right|^2, \tag{17}$$

$$\Gamma_{aut}=2\pi\sum_j\left|\sum_\beta c_\beta W_{\beta j}\right|^2, \tag{18}$$

where, as before, the summation over α and β allows for the configurational composition of a level. It is clear from Table

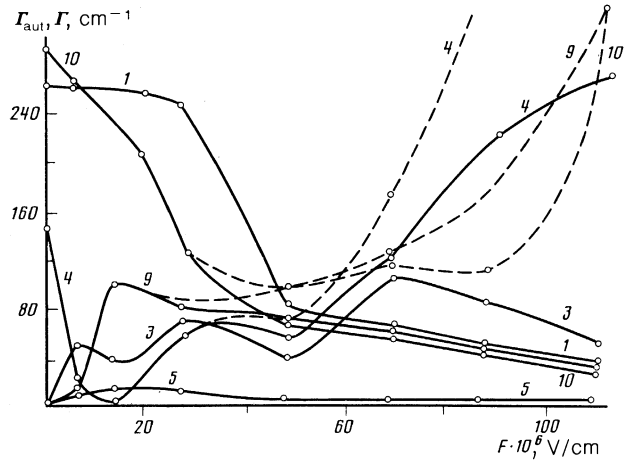

FIG. 4. Widths of the autoionizing levels Γ_{aut} (continuous curves) and total widths of the levels Γ (dashed curves) plotted for barium as functions of the external field intensity. The levels are numbered as in Table I.

TABLE III. Widths of discrete and autoionizing states of Ba calculated allowing and ignoring interference of channels of decay from the $5d_{3/2}p_{3/2}$, $5d_{5/2}p_{3/2}$, $5d_{5/2}p_{1/2}$, $5d_{3/2}f_{5/2}$, $5d_{3/2}f_{7/2}$, $5d_{5/2}f_{5/2}$, and $5d_{5/2}f_{7/2}$ states to the continuum in a field $F = 7\times10^7$ V/cm.

Transition (main components)	Allowing for interference		Ignoring interference	
	Γ_i, cm^{-1}	$\sum_i \Gamma_i$, cm^{-1}	Γ_i, cm^{-1}	$\sum_i \Gamma_i$, cm^{-1}
$5d\varepsilon p$	30.3		30.3	
$5d_{3/2}6d_{3/2}$		104.8		102.3
$5d\varepsilon f$	74.5		72.0	
$5d\varepsilon p$	8.0		8.6	
$6p_{3/2}6d_{5/2}$		121.8		123.2
$5d\varepsilon f$	113.8		114.6	
$5d\varepsilon p$	28.5		28.5	
$5d_{5/2}6d_{3/2}$		64.6		61.6
$5d\varepsilon f$	36.1		33.1	

III that the contribution of the interference term to Γ_{aut} amounts to 1–3% for $F = 7\times10^7$ V/cm. It follows from Table III and Fig. 4 that the main process governing the narrowing of the preionization resonances in a field is mixing of these resonances with a discrete spectrum. The interference between the decay channels then plays a secondary role.

Quite frequently the effect of a field on an atom is described using a two-level system with a pure coupling between the angular momenta. An allowance for the multiplet structure of the terms presupposes introduction of an intermediate type of coupling, which moreover can vary as a function of the external field. An increase in the field changes the relative positions of the components of a quasimultiplet, which is reflected in the type of coupling of the angular momenta of the configurations under discussion. We shall compare $\Gamma_{aut}(F)$ for two autoionizing levels with $J = 3$, calculated by two different methods: 1) for the intermediate type of coupling when the $6p_{3/2}6d_{5/2}$, $6p_{3/2}6d_{3/2}$, and $6p_{1/2}6d_{5/2}$ are superimposed; 2) for the JJ type of coupling. The results are presented in Fig. 5. The considerable discrepancies between the continuous (intermediate coupling) and dashed (JJ coupling) curves in Fig. 5 demonstrate how important it is to allow for the multiplet structure of the terms in calculations of different characteristics of an atom in a field.

c. Profile of a line in a field

We shall consider changes in the profiles of preionization resonances under the influence of a strong oscillatory field allowing for the dependence of the various spectral characteristics on F found above. Plotting the line profiles in accordance with Eq. (14) for a number of transitions (Table II) demonstrates the various ways the field can affect the line profile. Figure 6a shows the case of a monotonic reduction in the transition cross sections with F. The smoothing of preionization resonances in a strong field is observed also for other transitions (Fig. 6b). However, in the intermediate range of fields $F = 2\times10^{-3}$ a.u. there may be an anomalous increase in the transition cross section followed by a fall in the limit $F\to F_{at}$. The rise is due to two circumstances: an interference maximum in the expansion of the matrix ele-

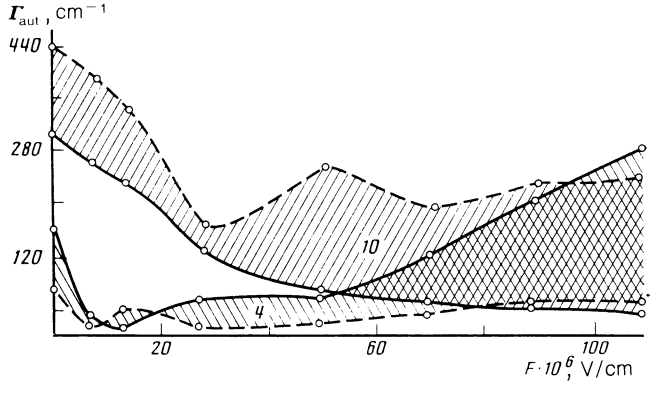

FIG. 5. Widths Γ_{aut} of the $6p_{3/2}6d_{5/2}$ and $6p_{3/2}6d_{3/2}$ levels ($J = 3$) of Ba plotted as a function of F and calculated for the intermediate type of coupling (continuous curves) and for the JJ type of coupling (dashed curves).

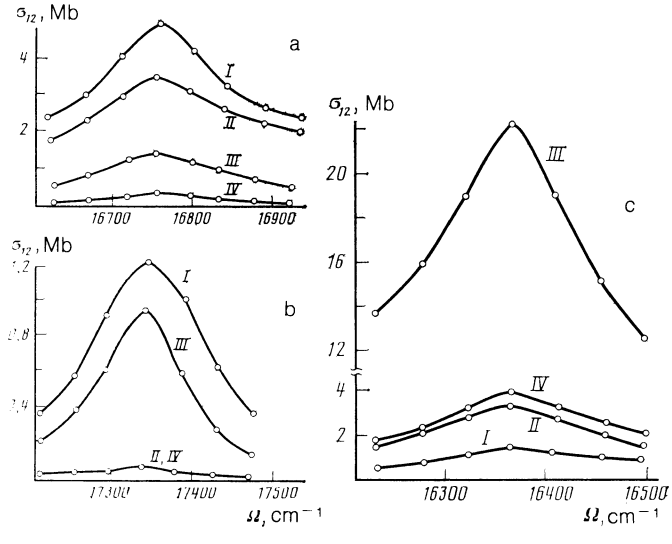

FIG. 6. Profiles of preionization resonances of the Ba atom in fields of different intensities: I) $F = 0$; II) $F = 7\times10^6$ V/cm; III) $F = 1.4\times10^7$ V/cm; IV) $F = 7\times10^7$ V/cm. a)–c) Transitions 3–10, 9–4, 9–1 (Table II).

ment of the dipole transition $V_{12}(F)$ of Eq. (13) and narrowing of the initial or final states in the quasicrossing region. Therefore, we may conclude that in an increasing field of moderate intensity a line profile can also behave in different ways as a function of the nature of the interaction of the initial and final states with the field. In the strong-field limit we can observe flattening of all the preionization resonances related in a greater or smaller degree to the nature of the transition. Universal flattening of the line profile in a strong field is due to the fact that the widths of the initial and final states of any transition increase in the limit $F \to F_{at}$ (Fig. 4) and the transition cross sections either remain constant (for transitions between states with near-zero dipole moments) or they decrease (for states with nonzero dipole moments).

6. GENERAL CONCLUSIONS

1. Mixing of atomic multiplets by fields of intensity $0 < F \leqslant 5 \times 10^6$ V/cm is described by the relations deduced using perturbation theory applied to an isolated level in a nonresonant field if $V < \Delta$ and by the theory of resonant two-level interaction if $V > \Delta$.

2. In fields in the range 5×10^6 V/cm $< F \leqslant 5 \times 10^7$ V/cm all the levels of a quasimultiplet satisfy $V \geqslant \Delta$. The interaction is of the many-level nature. The structure of multiplets in an isolated atom has little effect on the quasistructure as a whole. The different characteristics of a quasimultiplet are linear functions of F in various regions, but in the full range of F they are complicated and can be found by numerical calculations. The shifts of the levels in a field are of the order of the separations δE between the levels, which in some cases results in quasicrossing of the levels accompanied by major changes in all the characteristics right up to a transposition of the configurational composition.

3. In fields close to the atomic values, $F > 5 \times 10^7$ V/cm, if $V \gg \Delta$, it is found that all the states have constant (field-independent) values of the effective dipole moment. The structure of a multiplet is then symmetric relative to the average energy E_{av}. Resonantly coupled levels form pairs of states with zero momenta, so that their characteristics (quasienergies, configurational composition, and widths) are independent of the field. The levels which are not resonant with the field acquire constant nonzero values of the dipole moment so that the admixture of the states of the opposite parity tends to 50% for high values of F. The shifts of the levels are linear in F. The interaction is essentially of a many-level nature, but there are also features of a two-level resonant interaction.

4. An oscillating field may reduce the width of autoionizing states to a few reciprocal centimeters when these states become mixed with a discrete spectrum. Such narrowing may occur in moderate fields as long as the ionization broadening does not play a significant role. In the case of barium the compensation of such narrowing by the ionization broadening occurs in the range $F > 5 \times 10^7$ V/cm.

5. In the strong-field limit the width of a level determined by preionization (without allowance for the ionization broadening) depends strongly on the nature of the interaction with the strong field. A state with zero dipole moment may decrease in width to the radiative value or broaden to the maximum preionization width of the states of given configuration. The state with a nonzero dipole moment decreases in width monotonically with F down to some constant limit.

6. Flattening of preionization resonances occurs in a strong field primarily because of a reduction in cross sections of the transitions between states mixed by a strong field. Moreover, in the limit $F \to F_{at}$ such flattening is facilitated by strong ionization broadening of the line profiles.

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