

Effect of additional dc-field coupling on the long-time photoelectron spectrum from a system with double autoionizing levels

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The atomic model that contains two autoionization levels coupled to the discrete ground state by an external laser field is discussed. In addition both autoionizing levels are coupled mutually by the dc-electric-field gradient. The analytical formula for the long-time photoelectron spectrum is found for this system. Calculations are performed that are valid for any strength of the laser field and the dc-electric-field gradient. The effects of the additional coupling by the dc-electric-field gradient on the photoelectron spectrum are discussed and illustrated graphically.

INTRODUCTION

In recent years great attention has been paid to the problems of laser-induced autoionization. Its various aspects have been studied in numerous papers. For example, the effects of the narrowing of some lines in photoelectron spectra was studied by Lambropoulos and Zoller¹ and by Rzażewski and Eberly.² In Ref. 2 this effect was called a confluence of coherences. The influences of various incoherences have been studied, too. They included such mechanisms as spontaneous emission of radiation,³⁻⁵ photoabsorption from the autoionizing level,^{6,7} a finite laser bandwidth,⁸ and collisions.⁵ Moreover, the influence of the dc-field coupling on the systems that contain autoionizing levels has been investigated both theoretically and experimentally. For example, this effect was the subject of Refs. 9-13. The cited references are only examples of numerous studies of atomic systems that contain autoionizing levels. For extensive lists of references on this subject, see Refs. 5 and 13.

The discrete levels (with autoionization) that are parts of atomic systems can be grouped according to their various configurations. For example, they can be the lambda type, in which an excited autoionizing level is coupled to two discrete states that are located below it.¹⁴⁻¹⁶ A different group of systems have a ladder-type configuration, in which the lowest of the autoionizing levels is coupled to one discrete state that is situated below it, and, moreover, this autoionizing state is coupled to the next, higher, discrete state. This kind of atomic system was discussed in many papers.^{6,9,13,17} A third kind of atomic system of concern to us is the V type. This configuration contains two (or more) autoionizing levels coupled by an external field to one discrete level located below them.^{18,19} Obviously it is possible to find some isomorphisms and relations among these configurations. They have been discussed by Kyrola,¹⁶ Kyrola and Lindberg,²⁰ and by Agrawal *et al.*⁹

The present paper deals with a kind of mixture of V and ladder systems. We assume that the atomic system contains two autoionizing levels that are coupled by the same

laser field to the discrete ground level. Moreover, these autoionizing states are coupled mutually by a dc-electric-field gradient. This kind of system can be referred to as a delta or triangle type. Obviously one might replace the autoionizing levels, as well as the coupling between them, by two dressed states; this replacement would not affect the physical nature of our model. The dressed states are defined by the external factor (the gradient electric field), and their properties are determined by the parameters of the autoionizing levels and the external field. However, we employ this procedure to explain the properties of the photoelectron spectrum for some particular cases.

ATOMIC MODEL AND PHOTOELECTRON SPECTRUM

The atomic model discussed in this paper is presented in Fig. 1. This model contains two autoionizing levels |1> and |2>, of the same parity, that are coupled to the discrete ground state |0> by an external laser field. We assume that this coupling is of a dipole-electric nature. The ground state |0> is coupled to the continuum |c> by the same laser field. Moreover, both autoionizing levels are diluted in the same continuum as a result of the configuration interaction. We assume that the laser field is monochromatic. Its frequency is equal to E_L (we use units of $h/2\pi = 1$). The system is quite similar to that discussed in Ref. 18, but they differ in one important point: Namely, we assume that both autoionizing levels |1> and |2> are coupled mutually by the external dc-field gradient. Because the parity of the levels is the same, one should assume that the dc-field coupling can be realized as a coupling between the electric quadrupole moment of the atom and the dc-electric-field gradient. For brevity, below we use the term dc electric field, having in mind the dc-electric-field gradient. As a result, all discrete atomic levels present in the system form the delta-type configuration (for the system discussed in Ref. 18 they create the V configuration).

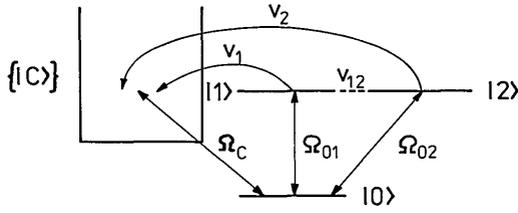


Fig. 1. The atomic-level scheme. Both autoionizing states, $|1\rangle$ and $|2\rangle$, are mutually coupled by the dc-electric-field gradient. These levels are diluted in the same continuum $|c\rangle$. The laser field of frequency E_L couples the ground state $|0\rangle$ to the continuum $|c\rangle$ and to the autoionizing levels $|1\rangle$ and $|2\rangle$.

The Hamiltonian that describes the atomic system discussed in this paper has the following form:

$$H = \sum_{i=0}^2 |i\rangle E_i \langle i| + \int dE |c\rangle E \langle c| + \langle 1| V_{12} \langle 2| + \sum_{i=1,2} |0\rangle \Omega_{0i} \langle i| \exp(iE_L t) + \sum_{i=1,2} \int dE |i\rangle V_i \langle c| + \int dE |0\rangle \Omega_c \langle c| \exp(-iEt) + \text{H.c.}, \quad (1)$$

where Ω_c , Ω_{01} , and Ω_{02} denote matrix elements that correspond to the coupling of the ground level $|0\rangle$ to the continuum $|c\rangle$ and to the autoionizing levels $|1\rangle$ and $|2\rangle$, respectively. The matrix elements V_i ($i = 1, 2$) describe configuration interaction between the continuum $|c\rangle$ and the autoionizing levels $|i\rangle$ ($i = 1, 2$), and V_{12} is the matrix element for transitions between both autoionizing states. We refrain at this moment from determining V_{12} . It should be a product of the electric-quadrupole moment and the gradient of the electric field (our model is universal enough to allow us to replace these two quantities, e.g., by the product of a magnetic field and the dipole magnetic moment). In addition, we assume that all matrix elements that appear above are real. The quantities E and E_i ($i = 0$ to $i = 3$) are energies of the levels $|c\rangle$, $|i\rangle$ ($i = 0$ to $i = 2$), respectively. Moreover, we define the wave function $|\Phi(t)\rangle$, characterizing evolution of the system in the form

$$|\Phi(t)\rangle = a(t)|0\rangle + b_1(t)|1\rangle + b_2(t)|2\rangle + \int dE b_c(t)|c\rangle. \quad (2)$$

We have now introduced the probability amplitudes a , b_1 , b_2 , and b_c that are functions of the time t . One should keep in mind that the amplitude b_c is a function of the continuum energy E . Assuming that the laser and dc-field amplitudes are turned on as the step function, and using the Schrödinger picture, one can find equations for the probability amplitudes. The equations are

$$i \frac{da}{dt} = aE_0 + \sum_{i=1,2} \Omega_{0i} \exp(iE_L t) b_i + \int dE \Omega_c b_c \exp(iE_L t), \quad (3a)$$

$$i \frac{db_1}{dt} = b_1 E_1 + \Omega_{01} \exp(-iE_L t) a + V_{12} b_2 + \int dE V_1 b_c, \quad (3b)$$

$$i \frac{db_2}{dt} = b_2 E_2 + \Omega_{02} \exp(-iE_L t) a + V_{12} b_1 + \int dE V_2 b_c, \quad (3c)$$

$$i \frac{db_c}{dt} = E b_c + \Omega_c \exp(-iE_L t) a + V_1 b_1 + V_2 b_2. \quad (3d)$$

It is convenient to extend integration over continuum energies from minus to plus infinity. Physically this assumption is equivalent to that of neglecting threshold effects. In other words, we assume that both autoionizing levels are located far above the ionization threshold; in consequence, this threshold perturbs the photoelectron spectrum negligibly. Moreover, we assume that all matrix elements that correspond to the transitions to (from) the continuum $|c\rangle$ do not depend strongly on E . One can solve Eqs. (3) quite easily by using the Laplace transform procedure. Assuming that for the initial time $t = 0$ the atomic system was in its ground state $|0\rangle$ ($a = 1$, $b_1 = b_2 = b_c = 0$ for $t = 0$), one can write

$$izA - i = (E_0 + E_L)A + \sum_{i=1,2} \Omega_{0i} B_i + \int dE \Omega_c B_c, \quad (4a)$$

$$izB_1 = E_1 B_1 + \Omega_{01} A + V_{12} B_2 + \int dE V_1 B_1, \quad (4b)$$

$$izB_2 = E_2 B_2 + \Omega_{02} A + V_{12} B_1 + \int dE V_2 B_2, \quad (4c)$$

$$izB_c = E B_c + \Omega_c A + V_1 B_1 + V_2 B_2, \quad (4d)$$

where $A(z)$, $B_1(z)$, $B_2(z)$, and $B_c(z)$ are the Laplace transforms of the amplitudes $a(t)$, $b_1(t)$, $b_2(t)$, and $b_c(t)$, respectively. We have now also introduced the amplitude $\alpha(t) = a(t) \exp(iE_L t)$. The next step is to eliminate the transformed continuum amplitude $B_c(z)$ from Eqs. (4). By application of the pole approximation,^{1,21} the system of Eqs. (4) becomes

$$[z + i(E_0 + E_L) + \Gamma_0]A(z) + (i\Omega_{01} + \Gamma_{01})B_1(z) + (i\Omega_{02} + \Gamma_{02})B_2(z) = 1, \quad (5a)$$

$$(i\Omega_{01} + \Gamma_{01})A(z) + (z + iE_1 + \Gamma_1)B_1(z) + (iV_{12} + \Gamma_{12})B_2(z) = 0, \quad (5b)$$

$$(i\Omega_{02} + \Gamma_{02})A(z) + (iV_{12} + \Gamma_{12})B_1(z) + (z + iE_2 + \Gamma_2)B_2(z) = 0. \quad (5c)$$

We define here the following widths:

$$\begin{aligned} \Gamma_0 &= \pi \Omega_c^2, \\ \Gamma_{12} &= \pi V_1 V_2, \\ \Gamma_i &= \pi V_i^2, \\ \Gamma_{0i} &= \pi \Omega_c V_i, \end{aligned} \quad (6)$$

where $i = 1, 2$. The set of Eqs. (5) can be solved easily, and its solution has the form

$$\begin{aligned} A(z) &= [(\Gamma_1 - i\delta_1)(\Gamma_2 - i\delta_2) - \mathbf{V}_{12}^2]/D(z), \\ B_1(z) &= [\mathbf{V}_{12}\Omega_{01} - (\Gamma_1 - i\delta_1)\Omega_{02}]/D(z), \\ B_2(z) &= [\mathbf{V}_{12}\Omega_{02} - (\Gamma_2 - i\delta_2)\Omega_{01}]/D(z), \end{aligned} \quad (7a)$$

where

$$D(z) = [z + i(E_0 + E_L) + \Gamma_0](z + iE_1 + \Gamma_1) \\ \times (z + iE_2 + \Gamma_2) + 2\Omega_0\Omega_0\mathbf{V}_{12} \\ - [z + i(E_0 + E_L) + \Gamma_0]\mathbf{V}_{12}^2 - (z + iE_1 + \Gamma_1)\Omega_{02} \\ - (z + iE_2 + \Gamma_2)\Omega_{01}. \quad (7b)$$

For convenience we have introduced here the complex matrix elements:

$$\Omega_{01} = i\Omega_{01} + \Gamma_{01}, \\ \Omega_{02} = i\Omega_{02} + \Gamma_{02}, \\ \mathbf{V}_{12} = i\mathbf{V}_{12} + \Gamma_{12} \quad (8)$$

and the detunings $\delta_i = E - E_i$ ($i = 1, 2$) and $\delta_L = E - E_0 - E_L$. Now we can define the long-time photoelectron spectrum $W(E)$ as

$$W(E) = \lim_{t \rightarrow \infty} |b_c(t)|^2. \quad (9)$$

It is possible to find a fully analytical solution for the probability amplitudes and, consequently, for the photoelectron spectrum [Eq. (9)]. Since $D(z)$ is a third-order polynomial in z , it is necessary to find three complex roots of the cubic equation. However, we restrict our considerations to the long-time limit, and only the imaginary root plays a significant role in the solutions. Therefore, after some trivial algebra, one can write $W(E)$ as follows:

$$W(E) = |\Omega_c A(z) + V_1 B_1(z) + V_2 B_2(z)|_{z=-iE}^2, \quad (10)$$

where A and B_i ($i = 1, 2$) are easily obtainable from Eqs. (7). From the form of Eq. (10) and the solutions of Eqs. (7), one can see that the photoelectron spectrum $W(E)$ differs from that discussed in Ref. 18. It is apparent that the spectrum also has two zeros, but their positions differ from those in Ref. 18. They are modified by the additional coupling from the external dc field. Moreover, positions and widths of the peaks should differ from those for the atomic system presented in Ref. 18. It is highly convenient to examine particular features of the spectrum by using the usual Fano q parameters.²² For the system discussed in this paper, they are defined as

$$q_i = \frac{\Omega_{0i}}{\pi V_i \Omega_c} = \frac{\Omega_{0i}}{\Gamma_{0i}} \quad (i = 1, 2). \quad (11)$$

Moreover, for easier comparison of our results with those of Refs. 2 and 18, we define the Rabi frequency Ω in the same way as Rzążewski and Eberly²:

$$\Omega = (4\pi\Gamma)^{1/2}(Q + i)\Omega_c \exp(if), \quad (12)$$

where Q and Γ are the effective asymmetry parameter and the effective autoionizing width, respectively. They can be expressed in the following way:

$$Q = \frac{q_1\Gamma_1 + q_2\Gamma_2}{\Gamma_1 + \Gamma_2}, \\ \Gamma = \Gamma_1 + \Gamma_2. \quad (13)$$

Now we are in a position to examine the long-time photoelectron spectrum for various parameters that characterize the atomic system.

DISCUSSION

Figure 2 shows the long-time photoelectron spectrum when the dc-field coupling is absent. This case corresponds to that in which the double autoionizing levels $|1\rangle$ and $|2\rangle$ are coupled only to the ground state $|0\rangle$ by the external laser field (this situation was discussed by Leoński *et al.*¹⁸). For large Fano parameters q_1 and q_2 the usual Fano-type zero²² does not perturb the spectrum in a significant way. When both of the above parameters are identical ($q_1 = q_2$) and the two autoionizing states $|1\rangle$ and $|2\rangle$ have the same energy ($E_1 = E_2$), the system becomes identical to one containing one autoionizing level. This kind of system was discussed extensively by Rzążewski and Eberly² and by Lambropoulos and Zoller.¹ This single autoionizing state is characterized by the effective autoionizing width Γ and the effective asymmetry parameter Q [Eq. (13)].

For the case of weak-laser-field coupling ($\Omega = 1$), one can observe the one-peak spectrum. This peak is located at the position of the two autoionizing levels $|1\rangle$ and $|2\rangle$. When the two autoionizing states have different parameters, the spectrum changes significantly. An additional zero appears in the spectrum and splits the existing peak. This zero is situated at the same position as the position of levels $|1\rangle$ and $|2\rangle$. It is a result of the interference between two channels of autoionization. These channels correspond to the transitions to (from) autoionization level $|1\rangle$ and to (from) level $|2\rangle$.

For the case of strong-laser-field coupling ($\Omega = 3$) and small Fano q parameters, the spectrum changes dramatically. The Autler-Townes peaks and the Fano zero are visible. One of these peaks falls to the Fano zero and becomes extremely sharp. This feature was discussed by Rzążewski and Eberly² and was called the confluence of coherences. However, in our case the confluence effect has a different nature. It corresponds to the interference between two autoionizing channels, but not to the presence of usual Fano zero.

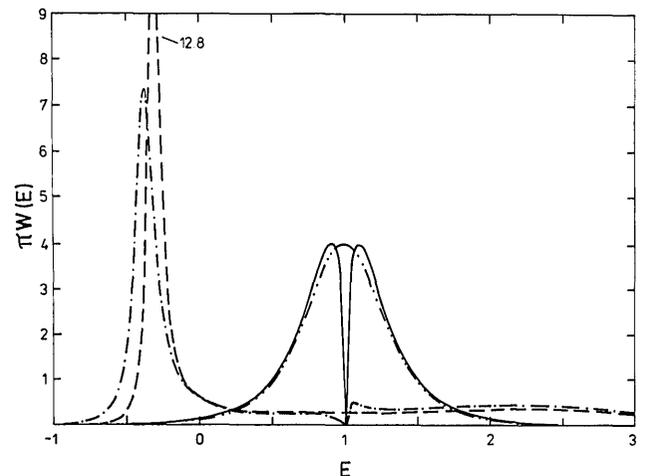


Fig. 2. Long-time photoelectron spectrum in the degenerate case ($E_1 = E_2 = 1$) for two strengths of the laser field ($\Omega = 1, 3$). The laser is tuned to the positions of autoionizing levels, and autoionizing widths are $\Gamma_1 = \Gamma_2 = 0.5$. The double-dotted-dashed curve represents $\Omega = 1$, $q_1 = q_2 = 100$; solid curve, $\Omega = 1$, $q_1 = 100$, $q_2 = 75$; dashed curve, $\Omega = 3$, $q_1 = q_2 = 2$; dashed-dotted curve, $q_1 = 2$, $q_2 = 3$. The coupling $V_{12} = 0$.

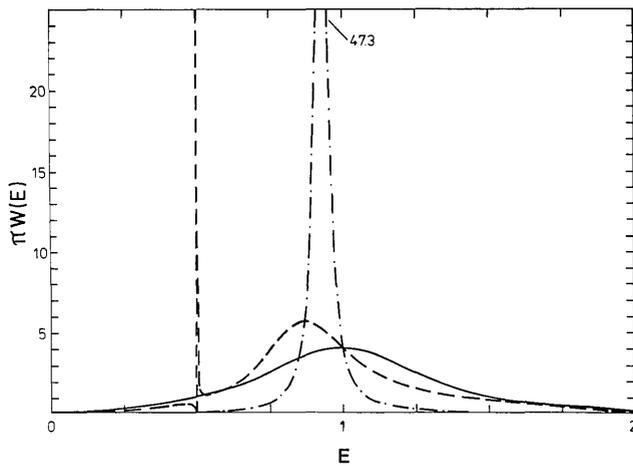


Fig. 3. Weak-laser-field ($\Omega = 1$) photoelectron spectrum for various values of V_{12} . The solid line represents $V_{12} = 0$; dashed curve, $V_{12} = 0.5$; dashed-dotted curve, $V_{12} = 3$; asymmetry parameters are the same ($q_1 = q_2 = 100$). The autoionizing widths, the positions of autoionizing levels, and the laser frequency are identical to those in Fig. 2.

Figure 3 shows the weak-laser-field photoelectron spectrum for various strengths of the dc-field coupling. The two autoionizing levels $|1\rangle$ and $|2\rangle$ are positioned at the same place. Both q parameters are identical and are assumed to be large ($q_1 = q_2 = 100$). This means that we have neglected direct ionization. For this case the Fano zero is not visible in the spectrum. When the dc-field coupling is equal to zero the spectrum consists of one peak located at the same energy as the energy of autoionizing levels $|1\rangle$ and $|2\rangle$. It is identical to the spectrum described in Refs. 2 and 18. The spectrum discussed here corresponds to the case of a single autoionizing level that is characterized by the effective width Γ and the effective Fano parameter Q . When the dc-field coupling has a nonzero value ($V_{12} = 0.5$) the photoelectron spectrum changes in a visible way. A new kind of zero appears in the spectrum. This zero is associated with an extremely sharp peak, as for the confluence of coherences effect.² The zero is a result of the dc-field-induced shift of the autoionizing levels $|1\rangle$ and $|2\rangle$. Consequently the levels have different energies, and interference between the two channels of autoionization can occur. When V_{12} increases the zero and the peak are shifted toward negative energies. For strong dc-field coupling ($V_{12} = 3$) the zero and the peak become negligible and only one elastic peak remains in the spectrum. One can explain this behavior as a consequence of a large value of the dc-field-induced shift. For this case the transitions between the ground state $|0\rangle$ and the autoionizing levels $|1\rangle$ and $|2\rangle$ become nonresonant.

The strong-laser-field spectrum ($\Omega = 3$) is shown in Fig. 4. We assume that the two autoionizing levels have the same energy and identical large q parameters. For $V_{12} = 0$ one can observe the Autler-Townes doublet that is induced by the strong laser field. Because of the large values of q_1 and q_2 the Fano-type zero does not perturb the spectrum. This spectrum corresponds to that of an atomic system that contains a single autoionizing level described by the effective width Γ and the effective parameter Q . When the value of the dc-field coupling differs

from zero, the zero value appears in the spectrum. For $V_{12} = 1$ this zero falls onto one of the Autler-Townes peaks. As a consequence this peak becomes split into two peaks. One of them is quite sharp and has the confluence of coherences character. Again, this confluence originates from the interference between the two autoionizing channels induced by the dc-field coupling. For extremely large values of V_{12} ($V_{12} = 5$) the zero is shifted far from the peaks. In this case the photoelectron spectrum consists of two peaks. One of them is quite sharp and corresponds to the above-mentioned confluence of coherences effect. The second peak is much wider and reflects the nonresonant excitation of the ground state $|0\rangle$ by the laser field.

The case illustrated in Fig. 5 is similar to that presented in Fig. 3, but it corresponds to the small values of q parameters. For $V_{12} = 0$ one can see that the one-peak spectrum is identical to that for a single autoionizing level characterized by the effective asymmetry parameter Q and the effective autoionizing width Γ . Although q parameters are small ($q_1 = q_2 = 2$), the Fano-type zero does not perturb the photoelectron spectrum significantly. The peak present in the spectrum is distorted

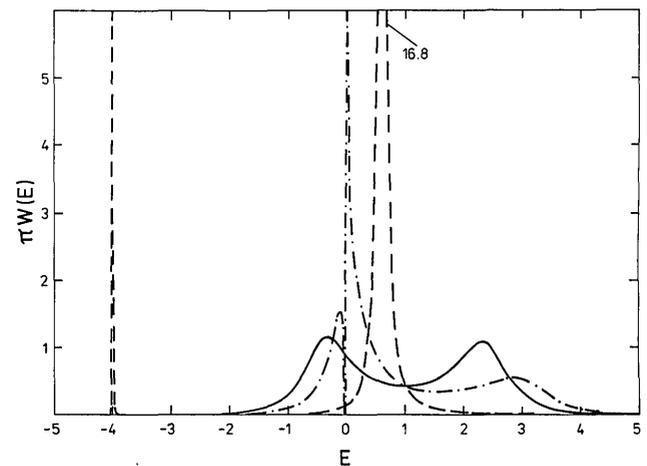


Fig. 4. Same as Fig. 3 but for a strong laser field ($\Omega = 3$). The solid curve represents $V_{12} = 0$; dashed-dotted curve, $V_{12} = 1$; dashed curve, $V_{12} = 5$.

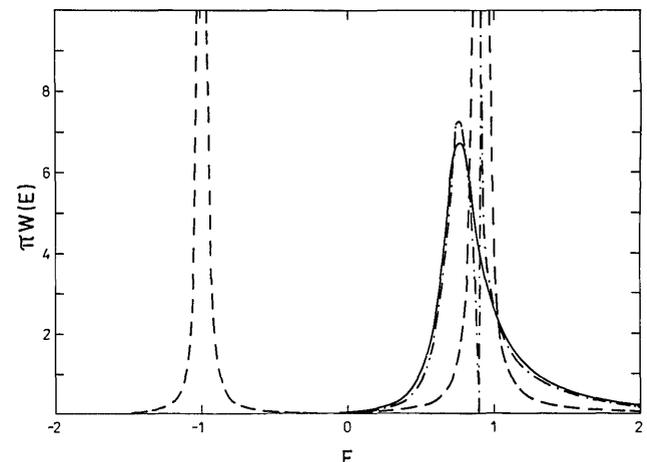


Fig. 5. Same as Fig. 3 but for small asymmetry parameters ($q_1 = q_2 = 2$). The solid curve represents $V_{12} = 0$; dashed-dotted curve, $V_{12} = 0.1$; dashed curve, $V_{12} = 2$.

only slightly by this zero. However, for $V_{12} = 0.1$ an additional zero becomes visible. This zero splits the peak in the spectrum. Consequently one can observe two peaks in the spectrum; they are extremely close to each other. One of them is quite sharp, showing its confluence of coherences character. For large value of the dc-field coupling ($V_{12} = 2$) two peaks are visible in the spectrum also. One can note that the distance between them is equal to the value of V_{12} ; this effect is a result of the splitting of discrete levels by the dc electric field.

The photoelectron spectra illustrated in Fig. 6 are plotted for the same parameters as those in Fig. 5 but for stronger laser-field couplings ($\Omega = 3$). When the dc-field coupling is assumed to be equal to zero, the spectrum shows the well-known form reported in Refs. 2 and 18. A strong laser field induces the Autler-Townes doublet, and the interference between direct ionization and autoionization leads to the occurrence of the Fano zero in the spectrum. This zero has a position identical to that of one of the Autler-Townes peaks. Thanks to this, the confluence of coherences occurs identically to that discussed in Refs. 2 and 18. For $V_{12} = 0.2$ the second zero appears in the spectrum. It is accompanied by the extremely sharp peak. However, this zero originates as a consequence of the interference between two autoionizing channels. This interference results from the dc-field coupling between states $|1\rangle$ and $|2\rangle$. When V_{12} becomes greater ($V_{12} = 0.45$) one can observe that the left peak in the spectrum becomes lower and wider. The remaining components of the spectrum behave as does the spectrum for an atomic system that contains one autoionizing level irradiated by a strong laser field. This part of the spectrum exhibits the confluence of coherences character, but like that for q parameters smaller than 2 ($q_1 = q_2 = 2$).

Figure 7 shows a situation similar to that presented in Fig. 4 but for different values of the energies of the autoionizing levels. One can see that for $V_{12} = 0$ a zero occurs in the spectrum (one should keep in mind that the asymmetry parameters have large values). Since it appears at the same position as the position of the Autler-Townes doublet, the confluence of coherences occurs and

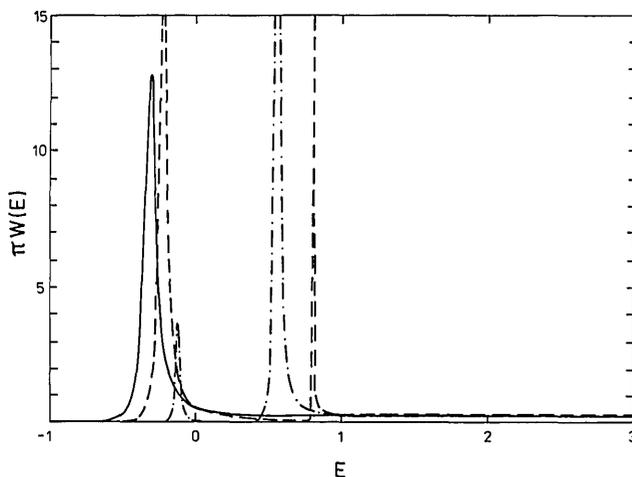


Fig. 6. Strong-laser-field photoelectron spectrum ($\Omega = 3$) for various values of the dc-field gradient coupling. The solid curve is for $V_{12} = 0$; dashed curve, $V_{12} = 0.2$; dashed-dotted curve, $V_{12} = 0.45$. The parameters that characterize the atomic system are the same as in Fig. 5.

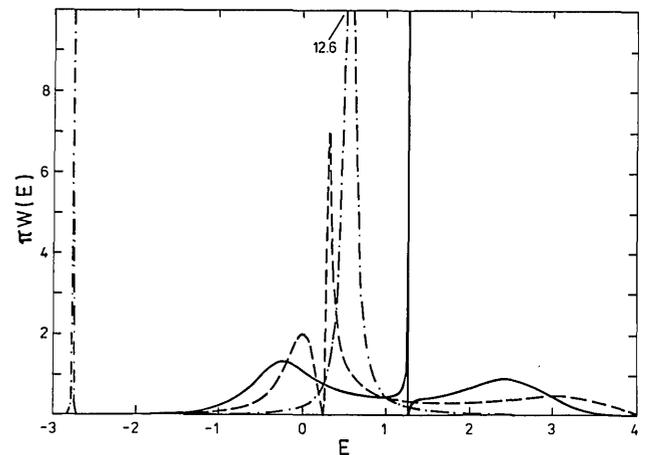


Fig. 7. Strong-laser-field photoelectron spectrum ($\Omega = 3$) for large asymmetry parameters ($q_1 = q_2 = 100$) and for different positions of the autoionizing levels $|1\rangle$ and $|2\rangle$ ($E_1 = 1$, $E_2 = 1.5$). The parameters for the system are the same as in Fig. 5.

three peaks are visible in the spectrum. When V_{12} increases the position of zero tends to negative energies. For $V_{12} = 1$ this zero splits one of the Autler-Townes peaks. The confluence-type peak loses its character and becomes wider and lower. Again, as in Fig. 4, two peaks remain in the spectrum for large values of V_{12} ($V_{12} = 4$). When V_{12} increases one of the peaks becomes quite sharp and moves toward negative energies, whereas the second starts to play a more significant role in the photoelectron spectrum. As in Fig. 4, the latter peak corresponds to the nonresonant excitation of the ground level $|0\rangle$.

CONCLUSIONS

Additional coupling by the dc electric field between two autoionizing levels leads to some new phenomena in the long-time photoelectron spectrum. This coupling can give an additional zero in the spectrum when the energies and the parameters that characterize autoionizing levels are identical. This zero has a different nature than the usual Fano-type zero. It does not vanish even for large values of the asymmetry parameters. One can explain this feature as a result of the dc-field-induced splitting of the autoionizing levels $|1\rangle$ and $|2\rangle$. As a consequence interference between two autoionizing channels, corresponding to the two autoionizing states, can occur. This zero can produce the confluence of coherences effect. For the case when states $|1\rangle$ and $|2\rangle$ are different or have different positions, the additional dc-field coupling shifts the positions of zeros in the spectrum. In addition, this coupling can split the single or the Autler-Townes peaks that are present in the spectrum. Moreover, the zero can also shift positions of the peaks. Some of these peaks can become quite sharp, and their contribution to the spectrum becomes less significant. In addition, it is possible to observe the two-peak spectrum that corresponds to the dc-field splitting only.

It would be interesting to test the above features experimentally. In this paper we have expressed all energies and widths in units of the effective autoionizing width $\Gamma = \Gamma_1 + \Gamma_2$. To observe the effects predicted in this paper, we would need an electric-field gradient of

$\sim 10^4$ kV/cm²; this value would correspond to $V_{12} = 1$ of the present paper. We have performed these calculations for barium atoms. The magnitudes for the electric-field gradient can differ for other types of atoms or molecules.

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