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DC-field effects on the photoelectron spectrum from a system with two autoionising levels

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Abstract. A system with two autoionising states of opposite parity that are embedded in two orthogonal continua and coupled mutually by a DC electric field is discussed from the viewpoint of laser-induced ionisation. The long-time photoelectron spectrum from such a system is calculated for arbitrary strengths of the DC as well as the laser fields. The effects of the DC field and the finite autoionising width of the second level on the spectrum are discussed in detail and illustrated graphically.

1. Introduction

The problem of laser-induced autoionisation has been a subject of intense research in recent years (for extensive literature on the subject, see Agarwal *et al* (1984)). Lambropoulos and Zoller (1981) have studied autoionising states in a strong laser field and have noticed a narrowing of the autoionising resonance when the field strength approaches certain values. Rzążewski and Eberly (1981) have studied this narrowing effect, which they called the 'confluence of coherences' effect, by considering a simple, exactly solvable model consisting of a ground state, one autoionising state and the continuum. Andryushin *et al* (1982) have considered a model with two autoionising states coupled by way of an intense laser field. Their model, moreover, accounted for the transitions to the higher continuum states. Various incoherences, such as the finite laser bandwidth (Rzążewski and Eberly 1983), spontaneous emission of radiation (Lewenstein *et al* 1983, Haus *et al* 1983, Agarwal *et al* 1984), photoabsorption from the autoionising state (Andryushin *et at* 1984) as well as collisions (Agarwal *et al* 1984) were also included in the model.

Deeper insight into strong coupling between a bound state and an autoionising state can be achieved in terms of dressed states, the creation and stability of which have been recently discussed (Lami and Rahman 1986, Haan and Agarwal 1987). Dressed states are also useful in the interpretation of other effects involving bound-continuum transitions (Coleman *et al* 1982, Coleman and Knight 1983, Radmore and Knight 1984, Deng and Eberly 1984).

Recently, DC-field-induced interferences in autoionising resonances have been studied both experimentally and theoretically (Saloman *et al* 1985, Lecomte and Luc-Koenig 1985, Liu *et al* 1985, Agarwal *et al* 1986). Saloman *et al* (1985) have shown that the DC coupling between two autoionising states leads to a minimum in

the photoelectron spectrum from barium if the strength of the DC field becomes sufficiently high. Agarwal *et al* (1986) have proved the isomorphism of this effect and the laser-induced effects in autoionisation. A different model with two closely lying autoionising levels of the same parity, both coupled to the ground state by the same laser, has been recently considered by Leoński *et al* (1987).

In this paper we consider a system with two closely lying autoionising states of opposite parity mutually coupled by an external DC electric field. The long-time photoelectron spectrum from such a system is calculated for arbitrary strengths of the exciting laser field as well as of the DC field mixing the two autoionising states. The spectrum is illustrated graphically for various sets of the parameters. It is shown that the shape of the spectrum is strongly dependent on the autoionising width of the second autoionising level (the one that cannot be directly excited by the laser field due to parity considerations). The interference dip in the spectrum is completely washed away as the width becomes comparable to the width of the other autoionising level.

2. The model and equations of motion

The atomic model that we consider in this paper is shown in figure 1. The ground state $|0\rangle$ is coupled to the continuum $|\omega_1\rangle$ and to the autoionising state $|1\rangle$ by an external laser field of frequency ω_L. The autoionising state $|1\rangle$ is diluted in the continuum $|\omega_1\rangle$ and is coupled to the second, closely lying autoionising state $|2\rangle$ of opposite parity by way of the DC electric field. The state $|2\rangle$ is embedded in the second continuum $|\omega_2\rangle$ (orthogonal to $|\omega_1\rangle$). Since the two continua $|\omega_1\rangle$ and $|\omega_2\rangle$ are of different parity, there is no direct coupling of the continuum $|\omega_2\rangle$ to the ground state $|0\rangle$. We also neglect transitions $|1\rangle \leftrightarrow |\omega_2\rangle$ and $|2\rangle \leftrightarrow |\omega_1\rangle$. However, we put no restriction on the strengths of either the laser or the DC field. Within the above approximation our system is

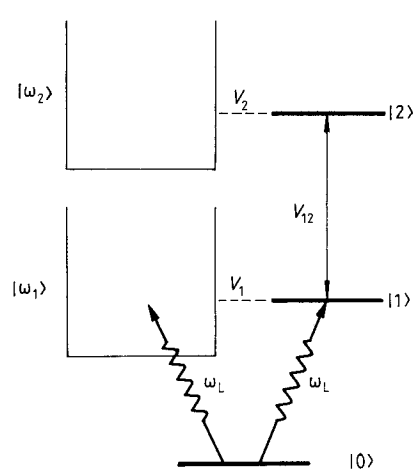

Figure 1. The atomic-level scheme. Both autoionising levels, $|1\rangle$ and $|2\rangle$, are mutually coupled by the DC electric field. These states interact with two orthogonal continua $|\omega_1\rangle$ and $|\omega_2\rangle$. The laser field of frequency ω_L couples the ground state to the lower continuum and autoionising states.

described by the following Hamiltonian ($\hbar = 1$ units are used):

$$H = [E_0 + \omega_L]|0\rangle\langle 0| + E_1|1\rangle\langle 1| + E_2|2\rangle\langle 2| + \int d\omega_1\, \omega_1|\omega_1\rangle\langle\omega_1|$$
$$+ \int d\omega_2\, \omega_2|\omega_2\rangle\langle\omega_2| + [|1\rangle V_{12}\langle 2| + |0\rangle\Omega_{01}\langle 1|$$
$$+ \int d\omega_1|0\rangle\Omega_0(\omega_1)\langle\omega_1| + \int d\omega_1|1\rangle V_1(\omega_1)\langle\omega_1|$$
$$+ \int d\omega_2|2\rangle V_2(\omega_2)\langle\omega_2| + \text{HC}] \tag{1}$$

where we have used the following basis states, which are products of the atomic and number of laser photon states:

$$\begin{aligned} |0\rangle &\equiv |0;\, n\rangle \\ |1\rangle &\equiv |1;\, n-1\rangle \\ |2\rangle &\equiv |2;\, n-1\rangle \\ |\omega_1\rangle &\equiv |\omega_1;\, n-1\rangle \\ |\omega_2\rangle &\equiv |\omega_2;\, n-1\rangle. \end{aligned} \tag{2}$$

The energies E_0, E_1, E_2 are those of the corresponding atomic states, and in (1) we have taken advantage of the fact that only the differences between the energies are relevant to our calculations. We also assume that the field couplings are of the electric dipole type only. In this paper we treat all the interactions on an equal footing. We perform no Fano diagonalisation (Fano 1961) at the start. However, it is also feasible to describe the continuum $|\omega_2\rangle$ together with the state $|2\rangle$ coupled to it as a continuum with Lorentzian structure (Leoński and Knight 1988). Since we will not discuss any incoherences such as spontaneous emission, our system can be described by the following state function:

$$|\Psi(t)\rangle = e^{-i(E_0+\omega_L)t}\Big[a|0\rangle + b_1|1\rangle + b_2|2\rangle$$
$$+ \int d\omega_1\, c_1(\omega_1)|\omega_1\rangle + \int d\omega_2\, c_2(\omega_2)|\omega_2\rangle\Big] \tag{3}$$

where all the amplitudes are time dependent and, according to the Schrödinger equation, fulfil the following equations:

$$i\frac{da}{dt} = b_1\Omega_{01} + \int d\omega\, c_1(\omega)\Omega_0(\omega) \tag{4a}$$

$$i\frac{db_1}{dt} = b_1\delta_1 + a\Omega_{01} + b_2V_{12} + \int d\omega\, c_1(\omega)V_1(\omega) \tag{4b}$$

$$i\frac{db_2}{dt} = b_2\delta_2 + b_1V_{12} + \int d\omega\, c_2(\omega)V_2(\omega) \tag{4c}$$

$$i\frac{dc_1(\omega)}{dt} = \Delta c_1(\omega) + a\Omega_0(\omega) + b_1V_1(\omega) \tag{4d}$$

$$i\frac{dc_2(\omega)}{dt} = \Delta c_2(\omega) + b_2V_2(\omega) \tag{4e}$$

where the detunings are defined as follows:

$$\Delta = \omega - E_0 - \omega_L$$
$$\delta_{1,2} = E_{1,2} - E_0 - \omega_L. \tag{5}$$

We have assumed that all matrix elements appearing in equations (4) are real. The set of linear differential equations (4) can be transformed to a set of algebraic equations using the Laplace transform method. On performing the Laplace transformation the continua can be eliminated from the set of equations for the transformed variables. We assume here that the continua are flat and thus the matrix elements are flat functions of ω. We neglect ionisation threshold effects and extend the lower limit in the integrations over ω to minus infinity. The 'pole approximation' (Lambropoulos and Zoller 1981) is used to evaluate the integrals. Thus, assuming that initially the atom was in its ground state, i.e. $a = 1$ for $t = 0$ and the remaining amplitudes were zero, we obtain the following set of linear algebraic equations:

$$\begin{aligned}
&[z+\Gamma_0]a(z)+[\mathrm{i}\Omega_{01}+\Gamma_{01}]b_1(z)=1\\
&[\mathrm{i}\Omega_{01}+\Gamma_{01}]a(z)+[z+\mathrm{i}\delta_1+\Gamma_1]b_1(z)+\mathrm{i}V_{12}b_2(z)=0\\
&\mathrm{i}V_{12}b_1(z)+[z+\mathrm{i}\delta_2+\Gamma_2]b_2(z)=0
\end{aligned} \tag{6}$$

where we have introduced the widths

$$\begin{aligned}
&\Gamma_0 = \pi\Omega_0^2(\omega)\\
&\Gamma_{1,2} = \pi V_{1,2}^2(\omega)\\
&\Gamma_{01} = \pi\Omega_0(\omega)V_1(\omega)
\end{aligned} \tag{7}$$

and $a(z)$, $b_1(z)$, $b_2(z)$ are the Laplace transforms of $a(t)$, $b_1(t)$ and $b_2(t)$. The set of equations (6) is readily solved and the solutions are given in the appendix.

3. Photoelectron spectrum

In our case, the long-time photoelectron spectrum can be defined as

$$\begin{aligned}
W(\omega) &= \lim_{t\to\infty}[|c_1(\omega;t)|^2+|c_2(\omega;t)|^2]\\
&= [|(z+\mathrm{i}\Delta)c_1(\omega,z)|^2+|(z+\mathrm{i}\Delta)c_2(\omega;z)|^2]_{/z=-\mathrm{i}\Delta}\\
&= [|a(-\mathrm{i}\Delta)\Omega_0(\omega)+b_1(-\mathrm{i}\Delta)V_1(\omega)|^2+|b_2(-\mathrm{i}\Delta)V_2(\omega)|^2].
\end{aligned} \tag{8}$$

Since we have assumed the two continua to be orthogonal and have neglected all terms that could mix them, the photoelectron spectrum (8) is a simple sum of the spectra for transitions to the two separate continua. On insertion of (A1) into (8) and after some simple algebra we arrive at the following expression for the long-time photoelectron spectrum:

$$\pi W(\omega) = \Gamma_0 \frac{N(\omega)}{M(\omega)} \tag{9}$$

where

$$\begin{aligned}
N(\omega) &= |(\Delta_2+\mathrm{i}\Gamma_2)(\Delta_1+q\Gamma_1)-V_{12}^2|^2+\Gamma_1\Gamma_2|V_{12}(q-\mathrm{i})|^2\\
M(\omega) &= |(\Delta_2+\mathrm{i}\Gamma_2)[(\Delta+\mathrm{i}\Gamma_0)(\Delta_1+\mathrm{i}\Gamma_1)-\Gamma_0\Gamma_1(q-\mathrm{i})^2]-V_{12}^2(\Delta+\mathrm{i}\Gamma_0)|^2
\end{aligned} \tag{10}$$

and where we have introduced the notation

$$\begin{aligned} \Delta_1 &= \Delta - \delta_1 = \omega - E_1 \\ \Delta_2 &= \Delta - \delta_2 = \omega - E_2 \\ q &= \Omega_{01}/\Gamma_{01}. \end{aligned} \tag{11}$$

In the above, q is the well known Fano asymmetry parameter (Fano 1961) for the autoionising state $|1\rangle$.

The spectrum given by equations (9) and (10) is valid for any strengths of the laser field (any values of $\Gamma_0 = \pi\Omega_0^2(\omega)$) and of the DC electric field (any values of V_{12}^2). It is evident from (10) that $N(\omega)$ has two zeros. One is the usual 'Fano zero', whereas the other appears whenever the DC field is 'on' and V_{12} differs from zero. Thus, the DC field modifies the spectrum essentially. Even for high values of q, when the Fano zero disappears, this extra zero due to DC-field-induced interference still survives. If the autoionising width Γ_2 of the second level is very small and if the exciting laser field is weak ($\Gamma_0 \ll \Gamma_1$, V_{12}), the formula (9) for the photoelectron spectrum reduces to

$$\pi W(\omega) = \frac{\Gamma_0}{\Delta^2} \left| \frac{\Delta_2\Gamma_1(\varepsilon + q) - V_{12}^2}{\Delta_2\Gamma_1(\varepsilon + \mathrm{i}) - V_{12}^2} \right|^2 \tag{12}$$

where $\varepsilon = \Delta_1/\Gamma_1$. Formula (12) consists of two factors, one of which corresponds to the elastic peak at $\Delta = 0$ and the other is a generalised Fano profile for the system discussed here. This is just the Fano profile that describes the weak-field absorption of the system. Such a profile, modified by the DC electric field, was experimentally studied by Saloman *et al* (1985) for barium atoms. Agarwal *et al* (1986) have discussed theoretically the form of such a profile including the effects of radiative decay. They assumed $\Gamma_2 = 0$ in their calculations and obtained a profile that dips to zero because of the extra zero introduced by the DC field. On neglecting the radiative decay effects our profile (12) becomes identical to that of Agarwal *et al* (1986). The experimental signals of Saloman *et al* (1985) do not dip to zero, however, and Agarwal *et al* (1986) attribute this fact to the small though finite value of Γ_2 in the experiment. Our general formula (12) for the photoelectron spectrum describes, moreover, the cases of finite Γ_2 and strong laser excitation. Some of them are illustrated in figures 2–5.

For convenience and easy comparison with earlier work we introduce the Rabi frequency Ω_0 defined as (Rzążewski and Eberly 1981, 1983)

$$\Omega_0 = \sqrt{4\pi\Gamma_1}\,(q + \mathrm{i})\,\mathrm{e}^{\mathrm{i}\varphi}\,\Omega_0(\omega) \tag{13}$$

where the arbitrary phase φ can be chosen to make Ω_0 real. We will use the Rabi frequency Ω_0 as a measure of the strength of the exciting laser field. All energies will be given in units of Γ_1.

Figure 2 shows the generalised Fano profile $F(\omega)$ obtained from (9) by factoring out Γ_0/Δ^2, which can be done for weak exciting fields ($\Gamma_0 \ll \Gamma_1, \Gamma_2, V_{12}$). The positions of the autoionising levels are: $E_1 = 1$; $E_2 = 1.3$. The DC-field coupling between them is weak ($V_{12} = 0.3$). The asymmetry parameter is taken to be $q = 5$. For $\Gamma_2 = 0$ we obtain the same profile as that of Agarwal *et al* (1986) (for their $\gamma_f = 0$) with the additional zero due to DC-field coupling clearly visible. This extra zero is located at the position of the second autoionising level only slightly modified by the interaction V_1. As Γ_2 increases the dip in the profile becomes less and less pronounced, disappearing completely when Γ_2 approaches unity. This confirms the suggestion made by Agarwal *et al* (1986). For $\Gamma_2 = 1$ we have a broad one-peak profile. The shape of this profile has been discussed by Saloman *et al* (1985) in a slightly different way: they

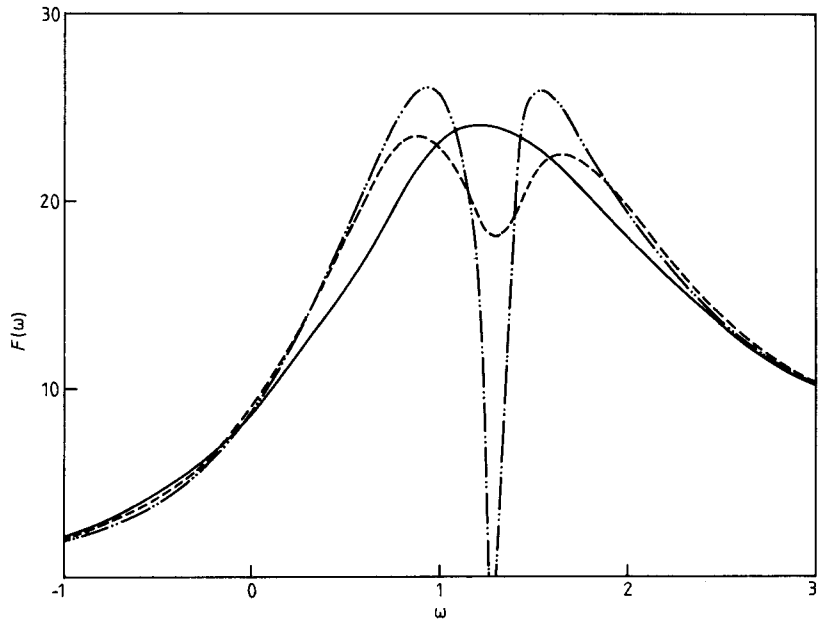

Figure 2. Generalised Fano profile for various autoionising widths Γ_2 and weak DC-field coupling ($V_{12} = 0.3$). The asummetry parameter $q = 5$ and the Rabi frequency $\Omega_0 = 1$. Chain curve, $\Gamma_2 = 0$; broken curve, $\Gamma_2 = 0.2$; full curve, $\Gamma_2 = 1$. The autoionising width $\Gamma_1 = 1$, and the laser is tuned to the lower level ($\delta_1 = 0$). Moreover, $E_0 = 0$, $E_1 = 1$ and $E_2 = 1.3$.

used a two-step diagonalisation leading to dressed states. The effect of Γ_2 on the profile is quite similar to that found by Leoński and Knight (1988) and can be attributed to a decrease in effective coupling between the levels $|1\rangle$ and $|2\rangle$ with increasing ω.

The long-time photoelectron spectrum, as given by equation (9) for a strong exciting laser field (the Rabi frequency $\Omega_0 = 3$), is shown in figure 3. The DC-field coupling V_{12} and the asymmetry parameter q are the same as in figure 2 ($V_{12} = 0.3$, $q = 5$). The Autler–Townes doublet is clearly visible in the spectrum, as it should be for a strong laser field. However, one of the Autler–Townes peaks is split if Γ_2 is small. For $\Gamma_2 = 0$ the DC-field-induced zero appears in the spectrum, and since it appears at the positions of the Autler–Townes peak the effect is similar to the 'confluence of coherences' effect discussed by Rzążewski and Eberly (1981, 1983). This zero in our photoelectron spectrum is due to interference between the transitions induced by the DC field and the transitions between the lower autoionising and ground levels. The latter transitions are induced by the laser field. A similar looking zero has been obtained by Deng and Eberly (1984) in their spectrum; however, theirs was an atomic model with one Fano continuuum coupled to two different discrete levels by two laser fields of two different frequencies.

As Γ_2 increases the minimum in the spectrum does not dip to zero and disappears completely for sufficiently large Γ_2. This behaviour of the photoelectron spectrum is similar to that of the generalised Fano profile discussed earlier. For large Γ_2 the presence of the second autoionising state does not essentially affect the photoelectron spectrum obtained from such a system.

Figure 4 shows again the photoelectron spectrum for strong laser excitation but this time for stronger DC coupling ($V_{12} = 1$) and small q values. In this case of small

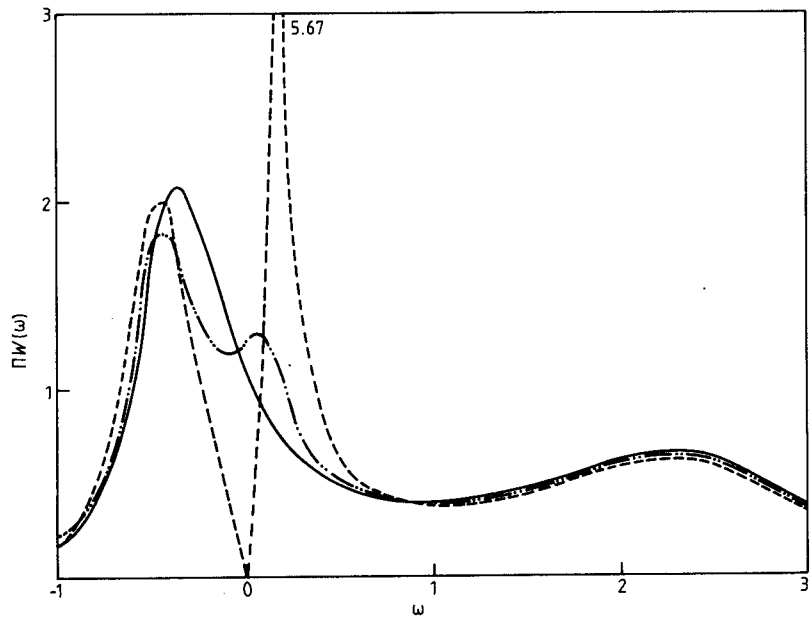

Figure 3. Strong laser-field photoelectron spectrum ($\Omega_0 = 3$) for various widths Γ_2. The autoionising $|2\rangle$ and ground $|0\rangle$ states have the same energies ($E_0 = E_2 = 0$); the energy $E_1 = 1$ and the laser frequency $\omega_L = 1$. DC coupling is weak ($V_{12} = 0.3$) and $\Gamma_1 = 1$, $q = 5$. Broken curve, $\Gamma_2 = 0$; chain curve $\Gamma_2 = 0.2$; full curve $\Gamma_2 = 1$.

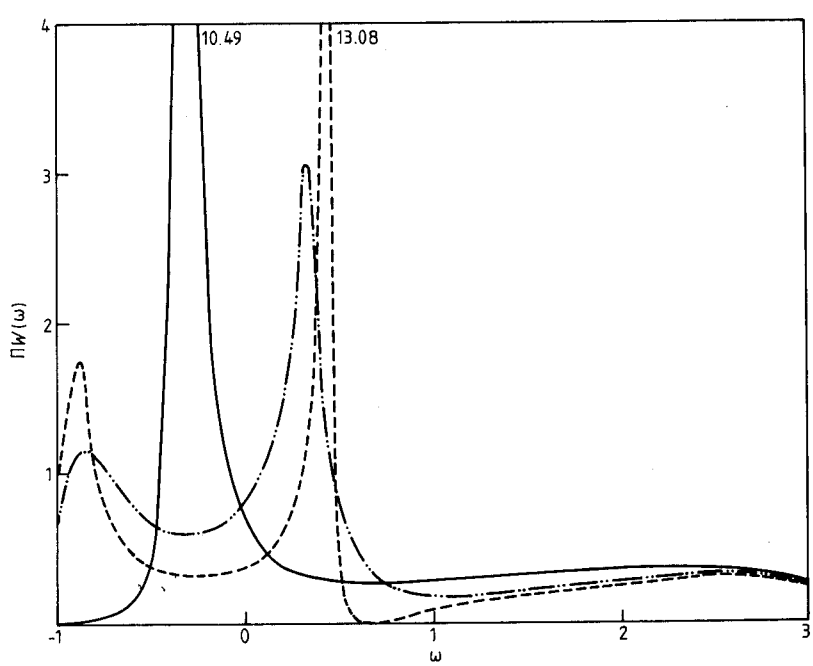

Figure 4. The same as figure 3, but for strong DC-field coupling ($V_{12} = 1$) and asymmetry parameter $q = 2$. Broken curve, $\Gamma_2 = 0$; chain curve, $\Gamma_2 = 0.25$; full curve, $\Gamma_2 = 50$.

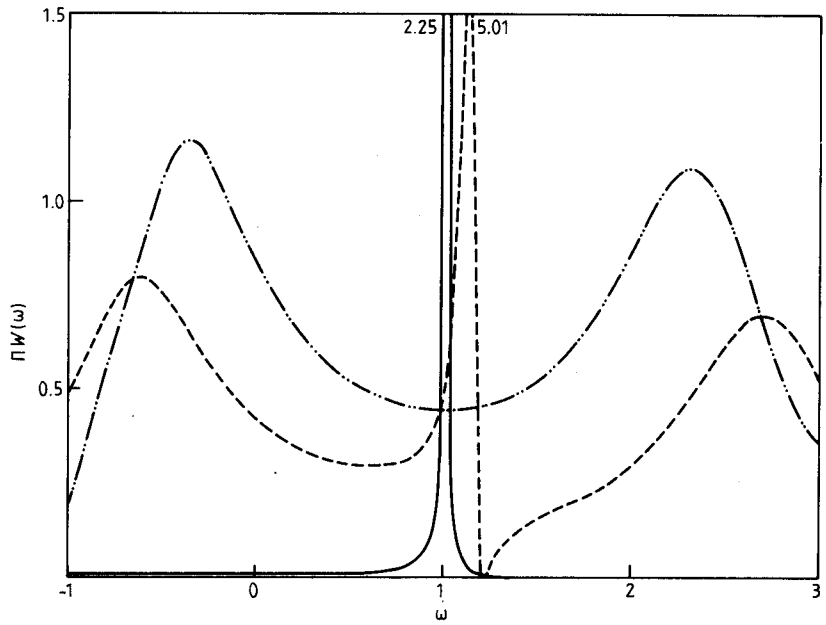

Figure 5. Strong laser field photoelectron spectrum for various strengths of the DC coupling. The autoionising widths are $\Gamma_1 = 1$ and $\Gamma_2 = 0$. The energies of the discrete levels are $E_0 = 0$, $E_1 = 1$ and $E_2 = 1.2$. Moreover, the parameter $q = 100$ and $\omega_L = 1$. Chain curve, $V_{12} = 0$; broken curve, $V_{12} = 1$; full curve, $V_{12} = 5$.

q the usual Fano zero occurs in the spectrum and, for large Γ_2 when the influence of the additional autoionising state is negligible, the confluence of coherences manifests itself as in the case of a single autoionising state, discussed by Rzążewski and Eberly (1981, 1983); for easier comparison, we have used the same values of the parameters. However, for $\Gamma_2 = 0$ the second zero due to the DC-field coupling does appear in the spectrum. Since this extra zero falls exactly on one of the peaks due to splitting of the Autler-Townes peak by the DC field, a very narrow peak appears in the spectrum signifying a 'second-order confluence of coherences' effect. Here, 'competition' between the DC and laser fields is clearly visible in the spectrum.

Figure 5 shows the photoelectron spectrum for high $q (q = 100)$ and various values of the DC coupling V_{12}. In this case the only zero that can affect the spectrum is that induced by the DC field. In figure 5 we assume $\Gamma_2 = 0$. For $V_{12} = 0$ the spectrum is identical to that obtained by Rzążewski and Eberly (1983) with the Autler-Townes doublet clearly visible. At non-zero values of V_{12} the additional zero affects the spectrum. As the DC coupling V_{12} becomes much greater than the Rabi frequency Ω_0, the Autler-Townes doublet disappears and only the elastic peak remains in the spectrum. In the case of high q, however, the interplay between the DC field and the laser field is less spectacular.

4. Conclusions

We have considered the influence of a DC field on the photoelectron spectrum of a system with two closely lying autoionising states of opposite parity. An explicit

expression for the long-time photoelectron spectrum from such a system is derived without any restrictions whatsoever on the values of the DC as well as the laser field. It has been shown that the presence of the second autoionisation level affects the spectrum essentially only if the autoionisation width of this level is small. The effect is most pronounced if there is no coupling of this level to the continuum ($\Gamma_2 = 0$). In this case, an additional zero due to the DC coupling appears in the spectrum. The weak-field absorption profile (generalised Fano profile) has also been obtained. Our results for the profile agree with those of Saloman *et al* (1985) and Agarwal *et al* (1986). The extra zero due to the DC coupling between the two levels is shown to essentially affect the spectrum for a strong laser field also. The results are illustrated graphically in figures 2–5. The interplay between the effects of the DC field and the laser field becomes clearly visible if the two fields are strong. This can be explained in terms of dressed states of the atom, albeit with different dressing fields: the DC field and the laser field. The emergence of a narrow peak in the spectrum naturally leads us to ask the following question: which of the dressed states is the stable one? The stability of such dressed states against radiative decay has been recently discussed by Haan and Agarwal (1987).

It would be interesting to test these features experimentally. We suggest an experiment similar to that performed and discussed by Saloman *et al* (1985) with barium atoms multiply excited by the external laser field. The final excitation should be caused by the strong laser field, of a power upwards of 1 MW cm^{-2}. Moreover, we suggest the use of the DC electric field, about 10 kV cm^{-1} or more, to couple the autoionising levels. Under these conditions concerning the strength and power of the external fields the effects predicted in this paper should be accessible to experimental verifiication.

Acknowledgments

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Appendix

The solutions of the set of equations (6) are given by the following formulae:

$$a(z) = \frac{[z+\mathrm{i}\delta_1+\Gamma_1][z+\mathrm{i}\delta_2+\Gamma_2]+V_{12}^2}{D(z)}$$

$$b_1(z) = -\frac{[\mathrm{i}\Omega_{01}+\Gamma_{01}][z+\mathrm{i}\delta_2+\Gamma_2]}{D(z)} \tag{A1}$$

$$b_2(z) = \frac{\mathrm{i}V_{12}[\mathrm{i}\Omega_{01}+\Gamma_{01}]}{D(z)}$$

with

$$D(z) = (z+\Gamma_0)[(z+\mathrm{i}\delta_1+\Gamma_1)(z+\mathrm{i}\delta_2+\Gamma_2)+V_{12}^2]-(\mathrm{i}\Omega_{01}+\Gamma_{01})^2(z+\mathrm{i}\delta_2+\Gamma_2). \tag{A2}$$

Equations (4*d*) and (4*e*) give, for the Laplace transforms of the continua amplitudes,

the relations

$$c_1(\omega; z) = -\frac{\mathrm{i}}{z+\mathrm{i}\Delta}[a(z)\Omega_0(\omega)+b_1(z)V_1(\omega)]$$
$$c_2(\omega; z) = -\frac{\mathrm{i}}{z+\mathrm{i}\Delta}b_2(z)V_2(\omega) \tag{A3}$$

and on insertion of (A1) and (A3) we obtain explicit expressions for the Laplace transforms of the probability amplitudes of the two continua. To determine the time evolution of the system, the inverse Laplace transforms of (A1) and (A3) have to be taken. With this aim, the roots of the third-order polynomial occurring in (A2) are needed. Here, however, we are not going to discuss the time evolution but shall restrict our considerations to the long-time photoelectron spectrum from the system discussed in this paper.

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