Isomorphism of dc-Field–Induced Interference and Laser-Induced Effects in Autoionization

G. S. Agarwal

Department of Mathematics, University of Manchester Institute of Science and Technology, Manchester, United Kingdom, and School of Physics, University of Hyderabad, Hyderabad 500134, India

J. Cooper

Department of Physics, University of Colorado, Boulder, Colorado 80309, and Joint Institute for Laboratory Astrophysics, University of Colorado and National Bureau of Standards, Boulder, Colorado 80309

S. L. Haan

Department of Physics, Calvin College, Grand Rapids, Michigan 49506

and

P. L. Knight

Physics Department, Imperial College, London SW7 2BZ, United Kingdom

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Recent observations of dc-field–induced interferences in autoionization by several groups can be understood by use of the theoretical work on laser-induced autoionization, which also enables one to study the influence of radiative effects, such as recombination.

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The purpose of this Letter is twofold: firstly, to establish the isomorphism of two apparently disjoint pieces[1-5] of work concerning autoionizing states—(A) laser-induced effects[4,5] arising because of strong coupling between a bound state lying below the ionization threshold and the autoionizing state and (B) dc-field–induced interference[1-3] arising from the strong coupling of the autoionizing states—and, secondly, to show how radiative effects may be included in the latter. We show how many of the results obtained in the context of laser-induced autoionization can be used to predict the behavior of the system involving dc fields.

Two important features of the laser-induced autoionization (system A) spectra were that (i) a single peak (corresponding to a single autoionizing state) goes over to a doublet as the field intensity is increased and that (ii) for certain values of field strength and detuning, one of the dressed states does not decay by autoionization but can decay via radiative effects, leading to a narrow resonance. This is a manifestation of "population trapping."[6] The experiments of Saloman, Cooper, and Kelleher[1] (on system B) essentially see effect (i). Conditions for (ii) are not met in their experiments. Conditions for (ii) are, however, fulfilled in the experiments of Liu *et al.*,[2] and hence they see considerable narrowing in the spectra.

Let us now establish the isomorphism of systems A and B. For system A (Fig. 1), the Hamiltonian is (with use of the notation of Ref. 4)

$$H = E_a|a\rangle\langle a| + \int E|E\rangle\langle E|dE + E_f|f\rangle\langle f| + E_i|i\rangle\langle i| + \int (V_{Ea}|E\rangle\langle a| + \text{H.c.})dE + \int (\tilde{v}_{Ei}|E\rangle\langle i|e^{-i\omega_l t} + \text{H.c.})dE + (\tilde{v}_{ai}|a\rangle\langle i|e^{-i\omega_l t} + \text{H.c.}) + \dots, \quad (1)$$

where ... denotes the terms responsible for spontaneous emission and radiative recombination and ω_l is the laser frequency. The meaning of the various matrix elements is clear from Fig. 1. On making a canonical transformation with $\omega_l|i\rangle\langle i|$, the effective Hamiltonian becomes

$$H = E_a|a\rangle\langle a| + \int E|E\rangle\langle E|dE + E_f|f\rangle\langle f| + (E_i+\omega_l)|i\rangle\langle i| + \int (V_{Ea}|E\rangle\langle a| + \tilde{v}_{Ei}|E\rangle\langle i| + \text{H.c.})dE + (\tilde{v}_{al}|a\rangle\langle i| + \text{H.c.}) + \dots. \quad (2)$$

We assume that $E_i+\omega_l$ is greater than the ionization energy. The Hamiltonian Eq. (2) is just the Hamiltonian for system B with the following identifications: (1) $\tilde{v}_{Ei}$ is responsible for the dc-field ionization of $|i\rangle$ which now lies in the continuum and has energy $E_i+\omega_l$. The parameter $2\pi|\tilde{v}_{Ei}|^2$ can be identified with the dc-field ionizing width Γ_i of $|i\rangle$. (It is often small; see below.) (2) $\tilde{v}_{ai}$ is the dc-field interaction between $|a\rangle$ and $|i\rangle$.

In the context of the experiment of Saloman, Cooper, and Kelleher,[1] we can take $|a\rangle = 5d9p\ {}^3P_1{}^\circ$; $|i\rangle = 5d8d$, $J=2$. Thus at the Hamiltonian level, systems A and B are completely equivalent for ionization into the odd-parity continuum. (We note that, in this case, since $\tilde{v}_{Ei}$ corresponds to a two-electron transition it will be very small. In addition, there may be a background autoionization from state $|i\rangle$ to an even-parity continuum.) The ionization spectra for system B can be read from the ionization spectra for system A. The only difference is in the initial condition; specifically, for system B, the population is in $|f\rangle$, which is equivalent to a nonzero a_E in Eq. (3.36) of paper I.

Assuming that at $t=0$ all the population is in $|f\rangle$, and using second-order perturbation theory for the interaction $\tilde{v}_{Ef}$ and the results of paper I, we can show that the long-time ionization spectra $p(\epsilon)$ are given by

$$p(\epsilon)=\frac{|\tilde{v}_{Ef}|^2}{\pi}\left|\left\{(\epsilon-\alpha)\left[\epsilon+q_f-\frac{i\gamma_i}{\Gamma}\left(1-\frac{q_f}{q_i}\right)\right]-\Omega q_i^2\left(1-\frac{q_f}{q_i}\right)\right\}\frac{1}{\psi(\epsilon-\epsilon_+)(\epsilon-\epsilon_-)}\right|^2, \tag{3}$$

where $\epsilon_\pm$ are the solutions of the quadratic equation [Eq. (5.11) of I]

$$\epsilon^2-\left[\alpha-i\left(\eta+\frac{\Omega}{\psi}\right)\right]\epsilon-\frac{2(\alpha-\epsilon)\gamma_f}{\psi\Gamma q_f}-\frac{\Omega}{\psi}q_i^2\left[1+\frac{\gamma_f}{\Gamma}\left(\frac{1}{q_i}-\frac{1}{q_f}\right)^2\right]-i\left(\alpha\eta-\frac{2\Omega}{\psi}q_i\right)=0, \tag{4}$$

$$\eta=\frac{1}{\psi}\left(1+\frac{\gamma_f}{\Gamma}\right),\quad \psi=1+\frac{\gamma_f}{\Gamma q_f^2}.$$

Other symbols are defined as in I:

$$\epsilon=(2/\Gamma)(E-E_a),\quad \Gamma=2\pi|V_{Ea}|^2,\quad \alpha=(2/\Gamma)(E_i+\omega_l-E_a),\quad \Omega=2\pi|\tilde{v}_{Ei}|^2/\Gamma, \tag{5}$$

and q_i and q_f are the Fano q parameters relative to $|i\rangle$ and $|f\rangle$. Note that the radiative recombination is included in these equations ($q_i=q_i'$; $q_f=q_f'$).

The following parameter identification is to be made for system B:

$$\Omega=\frac{\Gamma_i}{\Gamma},\quad q_i=\frac{2}{(\Gamma\Gamma_i)^{1/2}}\tilde{v}_{ai},\quad \gamma_i\approx 0. \tag{6}$$

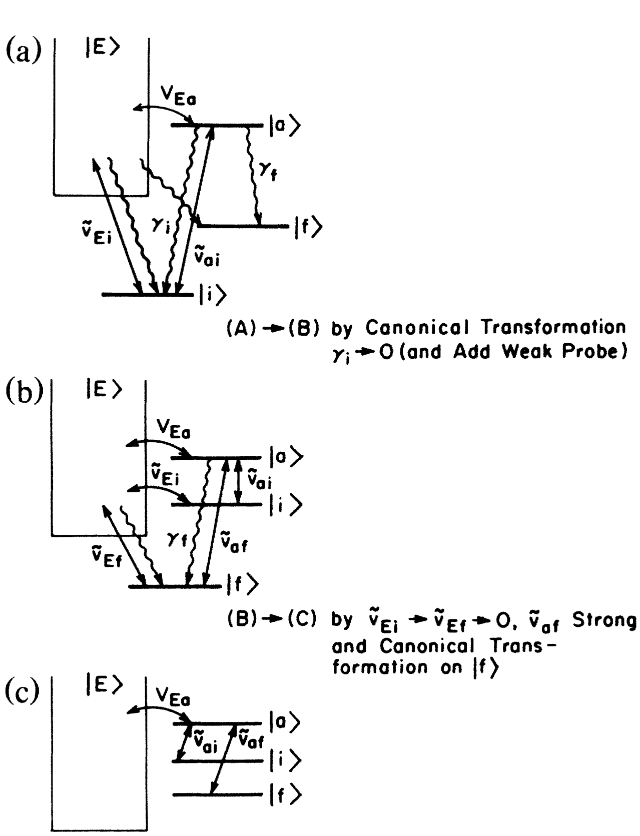

FIG. 1. Schematic representation of the equivalence of laser-induced autoionization (system A) and dc-field–induced interference (systems B and C). Here B describes the experiments of Saloman, Cooper, and Kelleher and of Liu *et al.*, and C, those of Feneuille *et al.*

An explicit expression for $p(\epsilon)$ can be obtained by use of Eq. (3) and the roots of the quadratic equation. The behavior of the complex roots for various values of Ω, q_i, q_f, γ_f, etc., is discussed in detail in I and in Ref. 5. A change in the strength of the dc field is equivalent to a change of q_i and Ω (or $q_i^2\Omega$ when Γ_i is small).

For no spontaneous emission $\gamma_f=0$, and if

$$\Omega=\Gamma_i/\Gamma=1+\alpha(\Gamma\Gamma_i)^{1/2}/2\tilde{v}_{ai}, \tag{7}$$

then one of the roots of Eq. (4) is *real* corresponding to a state which does not decay by autoionization. Thus for system B, if the dc fields and the energy separation between two states in the continuum are such that Eq. (7) is satisfied, then this will result in very narrow spectra. The width will essentially be determined by radiative effects, and it turns out to be the order of [cf. Eq. (5.9) of I with Eq. (7) satisfied] $\gamma_f\Gamma_i/\Gamma(\Gamma+\Gamma_i)$.

For the experiment of Saloman, Cooper, and Kelleher, $\Gamma_i \ll \Gamma$ and $\alpha > 0$; hence Eq. (7) cannot be satisfied. However, two well-separated complex roots of Eq. (4) do exist, leading to doublets.

For the experiment of Liu *et al.*,[2] $\alpha\approx 0$, and the line narrowing will be observed for $\Gamma_i\approx\Gamma$. The parameter α in their case is itself field dependent. The situation of Liu *et al.* is more complex as they have to consider coupling to two overlapping continua where the overlapping is large but not quite complete. The partial overlap will change the condition of Eq. (7), thus allowing more flexible values of the ratio Γ_i/Γ. It should, however, be noted that narrowing occurs over a range of parameters (cf. I, Figs. 4 and 5) with maximum narrowing when Eq. (7) holds. Generalization to two continua is straightforward.

We next discuss explicitly the photoelectron spectra for system B to illustrate the effects of radiative decay. For the experimental system of the *type* studied by Saloman, Cooper, and Kelleher, we can take the limits $q_i \to \infty$, $\gamma_i \to 0$, $\Omega q_i^2 = \Omega_0 = \text{const}$, $\Gamma_i \to 0$, and then, using Eq. (3), we obtain

$$p(\epsilon) = \frac{|\tilde{v}_{Ef}|^2}{\pi} \left| \frac{(\epsilon-\alpha)(\epsilon+q_f) - \Omega_0}{\psi[(\epsilon-\alpha)(\epsilon-\Delta_0+i\eta) - \Omega_0/\psi]} \right|^2, \quad \Delta_0 = -\frac{2}{\psi}\frac{\gamma_f}{\Gamma q_f}. \tag{8}$$

Here Δ_0 is a small frequency shift term which typically occurs in weak-field profiles[4] if the radiative decay processes are accounted for. Note further that Ω_0 is a measure of the strength of the dc field coupling $|i\rangle$ and $|a\rangle$. Setting $\Omega_0 = 0$ gives the profiles of paper I and the usual Fano profiles are recovered in the limit $\Omega_0 \to 0$, $\gamma_f \to 0$. In Fig. 2 we display the photoelectron profiles as given by Eq. (8) for a range of parameters. Note the presence of a zero in the spectra which is approximately at the position $\epsilon = \alpha$ for large q_f and very small Ω_0. The experimental signals of Saloman, Cooper, and Kelleher do not dip to zero, in contrast to the theoretical profiles. This is related to the nonzero but small value of Γ_i which has been set equal to zero in our computations. The extremely sharp minimum for small Ω_0 [Fig. 2(b)] is also connected with zero Γ_i since, near the resonance $\epsilon = \alpha$, a finite width can result from nonzero values of Ω_0 and Γ_i.

For nonzero Ω_0 the numerator of Eq. (8) has two zeros, and these occur at

$$\epsilon = \tfrac{1}{2}\{\alpha - q_f \pm [(\alpha+q_f)^2 + 4\Omega_0]^{1/2}\}. \tag{9}$$

For the case $4\Omega_0 << (\alpha+q_f)^2$ a first-order expansion gives

$$\epsilon = -q - \Omega_0/(\alpha+q), \quad \alpha + \Omega_0/(\alpha+q).$$

The first zero corresponds to the usual zero-field Fano minimum, while the second zero is a new, dc-field minimum, and corresponds to the dip of Saloman, Cooper, and Kelleher.

The zeros of the expression in the square brackets in the denominator of Eq. (8) represent complex dressed states of the system; real parts represent the energies (relative to E_a in units of $\Gamma/2$) and imaginary parts, the decay rates (in units of Γ). For small Ω_0/ψ the zeros lie near $\epsilon = \alpha$ (the energy of state $|i\rangle$) and $\epsilon = \Delta_0 - i\eta$ (representing the autoionizing state). The effects of spontaneous radiative decay are to shift the zero-field autoionizing state from $0 - i$ to $\Delta_0 - i\eta$, to decrease the effective dc-field mixing parameters from Ω_0 to Ω_0/ψ, and to reduce the profile by an overall scaling factor of ψ^2. In Fig. 3 we display photoelectron profiles for the case when $|i\rangle$ lies near the usual in-

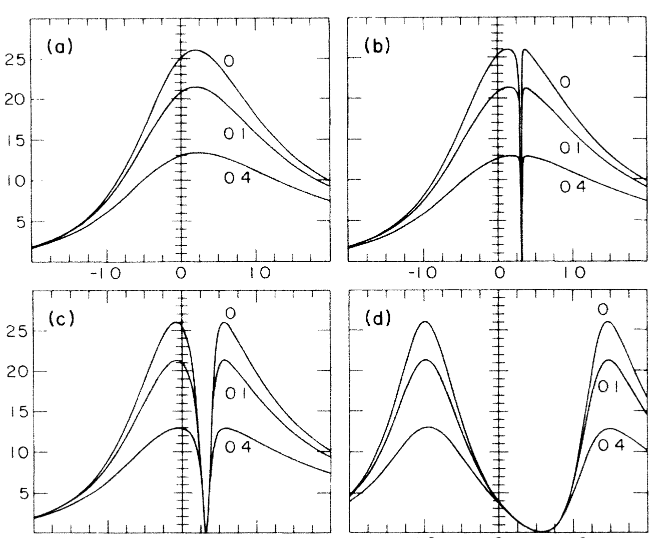

FIG. 2. Photoelectron spectra for system B for increasing values of the dc-field strength: (a) $\Omega_0 = 0$, (b) 0.01, (c) 0.1, (d) 1.5. Curves are marked by the values of the radiative decay parameter γ_f/Γ, and other relevant parameters are $q_f = 5.00$ and $\alpha = 0.30$.

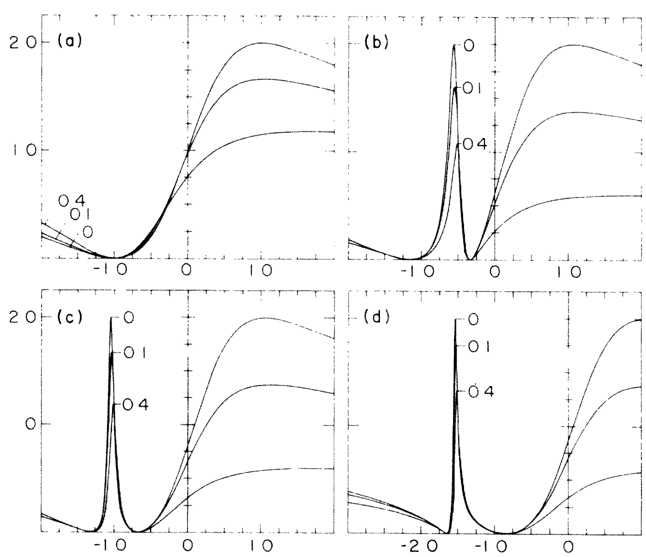

FIG. 3. (a) Photoelectron profile for $\Omega_0 = 0$, $q_f = 1$, and various γ_f/Γ. (b)–(d) Photoelectron profile for $\Omega_0 = 0.1$ and $\alpha = -0.5$, -1.0, and -1.5, respectively. Note the relative insensitivity of the interference maximum to γ_f/Γ. Note too from Eq. (8) that for $\Omega_0 \neq 0$, the profile is unity at $\epsilon = \alpha$ for all γ_f.

terference minimum at $\epsilon = -q$.

We also discuss an earlier experiment by Feneuille *et al.*[3] which produced evidence for the electric-field–induced stabilization of Rydberg states of rubidium. Their first model interpreted this stabilization in a single-channel bound-bound spin-orbit coupling with the upper bound level coupled much more strongly to the continuum states. This situation is immediately describable in our formalism by taking $\Omega \rightarrow 0$ and $q_i \rightarrow \infty$ in such a way that $q_i^2\Omega$ is held constant. The approximate stabilization in this first model is understandable in their limit $\Omega << \Gamma$, the tunneling width: Under these circumstances the initial-state–induced width is Ω^2/Γ and, as Γ exponentially rapidly increases in the tunneling region, this induced width decreases and the state is Stark stabilized. Essentially the weak bound-bound coupling can sample less and less of the broadened state. The model introduced by LeComte and Luc-Koenig[3] to explain Feneuille's experiment is also described transparently within our formalism. In this newer model, two lower-lying bound states are coupled to a higher-lying tunneling state in a lambda configuration. (The complication of the direct coupling between these two lower-lying states can be removed by a prior prediagonalization so that the system is described entirely as a λ system.) The Stark stabilization observed is then created entirely by destructive interference between the two channels of the lambda system.[7] If the coupling $\tilde{v}_{af}$ in Fig. 1 were strong, then again in the appropriate limit ($\gamma_f \rightarrow 0$, $\tilde{v}_{Ei} \rightarrow 0$, and zero coupling between $|f\rangle$ and $|E\rangle$), the model becomes that of Radmore and Knight.[7] A further comment concerning the adiabatic turn-on of the excitation in Ref. 3 warrants mention here. An adiabatic excitation will populate not both dressed states (of energies $\epsilon_{\pm}$) but only that which is adiabatically connected with the initial state; under these circumstances only a single line will be produced in the spectrum. For nonadiabatic excitation, the usual doublet is generated.

Here we have considered the isomorphism with problems in which electrons are created. The isomorphism carries over to similar inverse problems, e.g., dielectronic recombination in which an electron is captured and a photon is created. Here the radiative decay is crucial, and is best handled by the methods of papers I and II.

We mention that, in a manner similar to the experiments of Davis, Metcalf, and Phillips[8] which show the vanishing of bound-state dipole matrix elements, the dc-field interference effects on autoionizing states may be seen by optical methods, like monitoring of fluorescence.

Finally, it would be interesting to observe radiative processes in experiments similar to those discussed here. Since these features are most interesting when the parameter q is small and the ratio of the radiative to autoionizing rate, γ_F/Γ, is as large as possible, the search for suitable systems should concentrate on isolated but relatively weak autoionizing transitions.

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