Influence of Spontaneous Emission on Laser-Induced Autoionization

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A master equation that describes the effect of spontaneous emission on laser-induced autoionization is formulated and its solution is obtained for arbitrary laser strengths. The radiative decay is shown to affect drastically the nature of spectra near confluence. Analytic expressions for widths and positions are given to demonstrate the new features of spectra.

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Recent work[1-3] on laser-induced autoionization has shown several remarkable features of the autoionization spectra that arise as a result of the high intensity of the laser beam.[4] For example, the autoionization spectra have been shown to have an interesting "confluence[3] of coherences" near the Fano minimum. However, in such studies involving strong-field–induced autoionization, the effects of spontaneous emission and recombination and other incoherent processes have been ignored. In this Letter we report the effects of spontaneous emission on the photoelectron spectra. The spontaneous emission is treated by deriving a master equation[5] for the atomic system. The spontaneous emission mixes various Fano states,[6] just as the coupling by the laser field mixes Fano states. Even if "double diagonalization" has been carried out as far as the laser field interaction and the configuration mixing are concerned, the spontaneous emission again mixes such doubly diagonalized states and thus a further diagonalization is needed. We have been able to obtain the exact solution of the master equation and use this solution to show the extreme sensitivity of the photoelectron spectra to spontaneous decay rates, for energies near the confluence point. Analytic results for the height and width of the sharp feature are presented. The time dependence of the photoelectron spectra is also shown to change its character since the oscillations in the spectra (for a fixed energy) disappear as the radiative decay rate, γ, is increased.

The model we consider is schematically shown in Fig. 1, which is similar to the model considered in the literature,[2,3] but now we incorporate the vacuum coupling of the states $|\epsilon)$ and $|a)$ to some final state $|f\rangle$. All nonresonant interactions are ignored. The diagonalization of the configuration interaction between $|\epsilon)$ and $|a)$ leads to the Fano states $|\epsilon\rangle$,[6] as shown in Fig. 1. In the following we take the Fano states as basis states and all the elements of the density matrix will be taken between Fano states $|\epsilon\rangle$ and $|i\rangle$ and $|f\rangle$. The vacuum radiation field coupling of the states $|\epsilon\rangle$ can be treated in the framework of master equations,[5] i.e., from the total Hamiltonian of the system one can eliminate the vacuum field degrees of freedom by assuming that the coupling

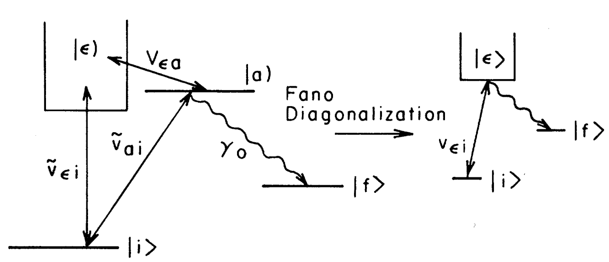

FIG. 1. Schematic diagram of the energy levels with various radiative and configuration interactions.

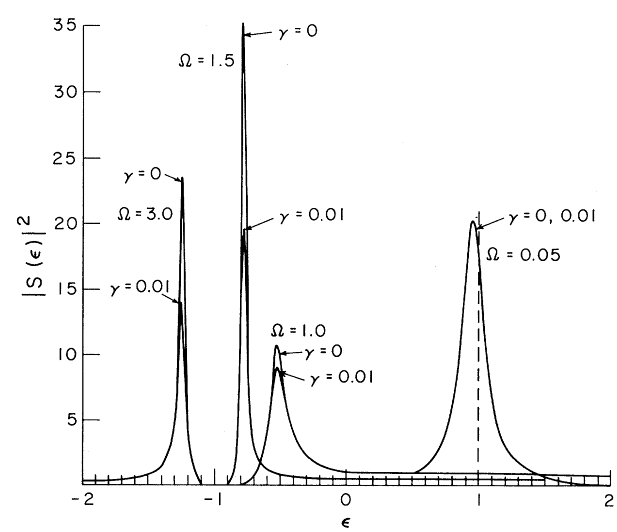

FIG. 2. Spectra $|S(\epsilon)|^2$ as computed from (7) and (9) in units of $1/\pi\Gamma$ for $q=1$, $\alpha=1$, $\sigma=20$ and for different values of field strengths $\Omega = 2\pi|\tilde{v}_{\epsilon i}|^2/\Gamma$. [Note that the parameter $\Omega = 4(q^2+1)\ (\Omega_0^2/\gamma_0^2)$ of Ref. 3.]

between the vacuum field and the atomic system is weak and that the emitted photon does not react back with the system. The derivation of the master equation is analogous to that for two-level systems[5]; however, the structure of the equation is rather complicated because of the structure of the continuum $|\epsilon\rangle$. We have shown that the reduced density matrix for the atomic system, in the Born and Markov approximations as far as the vacuum field coupling is concerned but with laser field coupling treated exactly, obeys the equations of motion of the form[7] (taking $\hbar = 1$ and $\epsilon_i = 0$, and denoting the laser frequency by ω_l and dipole matrix elements by $\vec{d}_{ij}$)

$$\partial\rho_{ii}/\partial t = -i\int v_{\epsilon i}{}^*\rho_{\epsilon i}\,d\epsilon + i\int v_{\epsilon i}\rho_{i\epsilon}\,d\epsilon, \tag{1}$$

$$\partial\rho_{\epsilon_1 i}/\partial t = -i(\epsilon_1-\omega_l)\rho_{\epsilon_1 i} - i\{v_{\epsilon_1 i}\rho_{ii} - \int d\epsilon\,\rho_{\epsilon_1\epsilon}v_{\epsilon i}\} - (2/3c^2)\int d\epsilon\,\vec{d}_{\epsilon_1 f}\cdot\vec{d}_{\epsilon f}{}^*\omega_{\epsilon f}{}^3\rho_{\epsilon i}; \tag{2}$$

$$\partial\rho_{\epsilon_1\epsilon_2}/\partial t = -i(\epsilon_1-\epsilon_2)\rho_{\epsilon_1\epsilon_2} - (2/3c^3)\int d\epsilon\,\vec{d}_{\epsilon_1 f}\cdot\vec{d}_{\epsilon f}{}^*\omega_{\epsilon f}{}^3\rho_{\epsilon\epsilon_2} - (2/3c^3)\int d\epsilon\,\omega_{\epsilon f}{}^3\vec{d}_{\epsilon_2 f}{}^*\cdot\vec{d}_{\epsilon f}\rho_{\epsilon_1\epsilon} - iv_{\epsilon_1 i}\rho_{i\epsilon_2} + iv_{\epsilon_2 i}{}^*\rho_{\epsilon_1 i};\quad \omega_{\epsilon f}\equiv(\epsilon-\epsilon_f). \tag{3}$$

Here $v_{\epsilon i}$ is, as shown in Fig. 1, the matrix element of the laser interaction with the atomic system between the Fano state $|\epsilon\rangle$ and the initial state $|i\rangle$. Equations (1)–(3) show how various Fano states get mixed because of spontaneous emission. Nevertheless, an exact solution is still possible since the above set turns out to have the factorization property

$$\rho_{\epsilon_1\epsilon_2}(t) = \psi_{\epsilon_1}(t)\psi_{\epsilon_2}{}^*(t),\quad \rho_{\epsilon_1 i} = \psi_{\epsilon_1}(t)\psi_i{}^*(t),\quad \rho_{ii}(t) = \psi_i(t)\psi_i{}^*(t), \tag{4}$$

with

$$\dot\psi_{\epsilon_1} = -i\Delta_{\epsilon_1}\psi_{\epsilon_1} - (2/3c^3)\int d\epsilon\,\vec{d}_{\epsilon_1 f}\cdot\vec{d}_{\epsilon f}{}^*\omega_\epsilon{}^3\psi_\epsilon - iv_{\epsilon_1 i}\psi_i,\quad \Delta_{\epsilon_1} = (\epsilon_1-\omega_l), \tag{5}$$

$$\dot\psi_i = -i\int v_{\epsilon i}{}^*\psi_\epsilon\,d\epsilon. \tag{6}$$

It may be added that elements like ρ_{ff} do not admit factorization like (4) because of the radiative decay to $|f\rangle$ and hence ρ still represents a mixed state in spite of the partial factorization property (4). We have been able to solve the integro-differential equations (5) and (6) exactly with the following result (z being the Laplace variable):

$$\hat\psi_\epsilon(z) = (z+i\Delta_\epsilon)^{-1}(-i/z)\{(1+m_{11})(1+m_{22}) - m_{12}m_{21}\}^{-1}\{v_{\epsilon i}(1+m_{22}) - zm_{21}(\gamma_0/2)^{1/2}b_{\epsilon a}{}^*\}, \tag{7}$$

$$m_{11} = \int\frac{d\epsilon|v_{\epsilon i}|^2}{z(z+i\Delta_\epsilon)},\quad m_{22} = \left(\frac{\gamma_0}{2}\right)\int\frac{d\epsilon|b_{\epsilon a}|^2}{(z+i\Delta_\epsilon)},\quad m_{21} = \left(\frac{\gamma_0}{2}\right)^{1/2}\int\frac{d\epsilon\,v_{\epsilon i}b_{\epsilon a}}{z(z+i\Delta_\epsilon)},\quad m_{12} = \left(\frac{\gamma_0}{2}\right)^{1/2}\int\frac{d\epsilon\,b_{\epsilon a}{}^*v_{\epsilon i}{}^*}{(z+i\Delta_\epsilon)}. \tag{8}$$

Here γ_0 gives the spontaneous decay rate of the bound state $|a)$ and $b_{\epsilon a}$ gives the amplitude of the state $|a)$ in Fano state

$$\{|\epsilon\rangle = b(\epsilon,a)|a) + \int b(\epsilon,\epsilon')|\epsilon')d\epsilon'\}.$$

We have assumed that the decay of the unperturbed continuum to $|f\rangle$ is unimportant. It may be noted that the zeros of the denominator of the first curly bracket in (7) give the dressed-state energies of the atom for $\gamma_0 = 0$. In deriving (7) we have not made any approximation regarding either the structure of the continuum $|\epsilon)$ or the strength of the laser field but the off-resonant coupling of the laser between $|f\rangle$ and $|\epsilon)$ is ignored. The integrals in (8) can be done analytically for the Lorentzian model with width σ of the continuum:

$$m_{22} = \gamma L(1);\quad m_{11} = \frac{\Omega}{z}\frac{\Gamma}{2}\left\{(q-i)^2L(1) + \frac{(q-i\sigma)^2\sigma}{(1-\sigma^2)}L(\sigma)\right\};$$

$$m_{21}z\frac{V}{\bar v_{\epsilon i}} = m_{12}\frac{V^*}{\bar v_{\epsilon i}{}^*} = (q-i)L(1)\left(\frac{\gamma_0}{2}\right)^{1/2},\quad \bar v_{\epsilon i} = (\epsilon|v|i),\quad \gamma = \frac{\gamma_0}{\Gamma}; \tag{9}$$

$$\Omega = 2\pi|\bar v_{\epsilon i}|^2/\Gamma,\quad |V_{\epsilon a}|^2 = \Gamma/2\pi,\quad L^{-1}(\beta) \equiv \beta + (2/\Gamma)\{z + i(\epsilon_a - \omega_l)\}.$$

Here Γ is the autoionization rate, q is Fano's asymmetry parameter, and we have used Fano's relationship (16) (Ref. 6) to relate the matrix element $v_{\epsilon i}$ to the $\bar v_{\epsilon i}$ in terms of unperturbed continuum states. The photoelectron spectra are proportional to $\rho_{\epsilon\epsilon}(t)$ and hence these now could be computed using (7) and (9) in both the transient and steady-state regions which are shown in Figs. 2–5.

In Fig. 2 we essentially reproduce, in the limiting case $\gamma=0$, the spectra of Rzazewski and Eberly.[3] A small spontaneous-emission decay rate is shown to have a drastic effect on the spectra for field strengths Ω of order 2 (for $q=1$). Figure 3 is for large values of q. Figure 4 shows the behavior of the height of the sharp feature (spike) as a function of laser intensity. In all cases spontaneous emission has a significant effect. In order to understand these features, we analyze analytically the steady-state spectrum in the limit $\sigma\to\infty$. We find that

$$\rho_{\epsilon\epsilon}(t\to\infty)=\left|S\left(\frac{\epsilon-\epsilon_a}{\Gamma/2}\right)\right|^2(\pi\Gamma)^{-1},\quad S(\epsilon)=[2\Omega/(1+\epsilon^2)]^{1/2}(\epsilon+i)(\epsilon+q+i\gamma)P^{-1}(\epsilon), \tag{10}$$

where

$$P(\epsilon)=\epsilon^2+\epsilon[-\alpha+i(\Omega+1+\gamma)]-\Omega(\gamma+q^2)-i(\alpha+\alpha\gamma-2\Omega q),\quad \alpha=2(\omega_l-\epsilon_a)/\Gamma. \tag{11}$$

The behavior of this polynomial as Ω, γ, and α are changed determines the characteristic changes in the spectra.

(i) For $\gamma=0$ and at the Fano minimum $\epsilon=-q$, P vanishes exactly for $\Omega=1+\alpha/q$, and for this particular Ω one of the dressed states lies at $-q$. This is what leads to very sharp structures near $\Omega=1+\alpha/q$ (near $\Omega=2$ in Fig. 2). For $\gamma\neq 0$, $\Omega=1+\alpha/q$, the root $-q$ moves from the real axis to $-q-i\gamma(1+\alpha/q)(2+\alpha/q)^{-1}$ giving the peak a finite width and thus even a very small decay rate would lead to a substantial change in the spectra.

(ii) For field strengths in the vicinity of the confluence point $\Omega=1+(\alpha/q)+b$, the roots of (11) to lowest order in b and γ are

$$z_+\approx -q-\frac{q^2 b}{2q+\alpha}-\frac{i\gamma(q+\alpha)}{(2q+\alpha)},\qquad z_-\approx q+\alpha-i(2+\alpha/q). \tag{12}$$

The expressions (12) show that the imaginary part of z_+ is independent of b to first order in b and depends only on the spontaneous lifetime, and hence spontaneous emission will again affect drastically the sharp features of the spectra. In fact one can show from (10) that, for $\epsilon=\mathrm{Re}\,z_+$,

$$S\simeq\left(\frac{(1+\alpha/q+b)}{1+q^2+O(b)}\right)^{1/2}\left\{\frac{Ab+B\gamma+O(b,\gamma)^2}{C\gamma+Db^2+O(b^3,\gamma^2)}\right\} \tag{13a}$$

$$\xrightarrow[\gamma\neq 0]{b\to 0}\left(\frac{2q}{(q+\alpha)(q^2+1)}\right)^{1/2}\sim\frac{1}{q} \tag{13b}$$

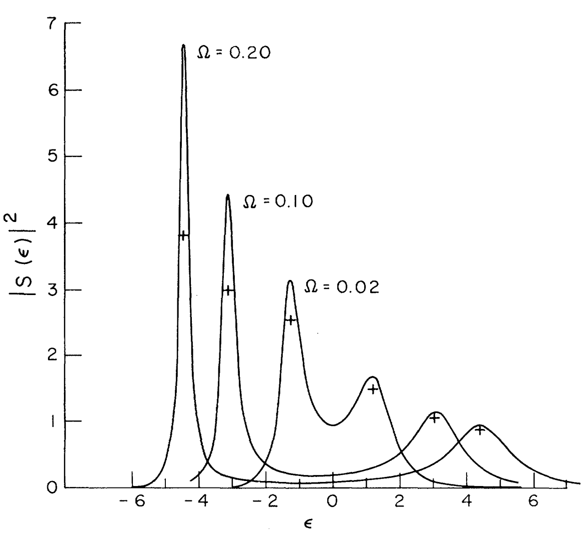

FIG. 3. Spectra $|S(\epsilon)|^2$ for large q (= 10), $\sigma\to\infty$, $\alpha=0$. The confluence occurs at $\Omega=1$. The crosses give the peak height for $\gamma=0.1$.

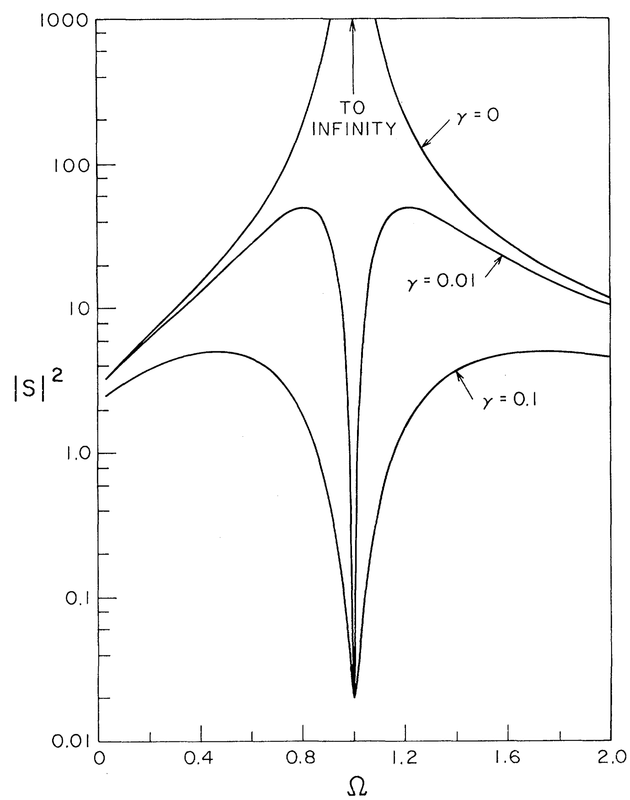

FIG. 4. Spike height vs Ω for $\sigma\to\infty$, $q=10$, $\alpha=0$. The minimum is consistent with (13b).

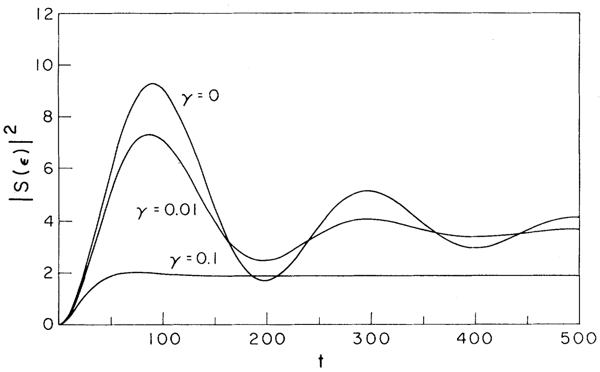

FIG. 5. Time-dependent spectra $|S(\epsilon, t)|^2$ for fixed $\epsilon = 0.75$, and for $\Omega = 1.5$, $q = 1$, $\alpha = 1$, $\sigma \to \infty$. In this case the spike occurs at $\epsilon_0 = -0.812$ and the oscillation frequency is consistent with the beat frequency between ϵ and ϵ_0. Note that time is in units of $1/\Gamma$.

whereas

$$S \xrightarrow[\gamma \neq 0]{b \to 0} O\left(\frac{1}{b}\right) \to \infty .$$

Expression (13) explains the behavior of the spike height shown in Fig. 4. Note the asymmetry in the spike height on the two sides of the confluence. The characteristic oscillations in time-dependent spectra (Fig. 5) occur at the beat frequency determined by ϵ and $\mathrm{Re}z_+$. The damping of oscillatory behavior depends on $\mathrm{Im}z_+$. As γ increases the oscillations die out very fast. It is clear from the foregoing discussion that a systematic study of the experimental photoelectron spectra (particularly their widths and heights) near the confluence can only be made if the spontaneous emission is properly incorporated in the theory.

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