Photoemission spectra in intense laser field induced autoionization

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Photoemission spectra from autoionizing states in the presence of a strong laser field are calculated. Such spectra have very sharp features near "confluence" (at the Fano minimum) and are very similar, in structure, to photoelectron spectra. The total intensity, as a function of laser intensity or detuning, has a peak near confluence, which can be used, among other things, for the accurate determination of Fano asymmetry parameter q.

In this Communication, we report a systematic calculation of the photoemission spectra[1] in strong laser field induced autoionization.[2] We demonstrate that the photoemission spectra have features very similar to photoelectron spectra[2,3] and provide us with a high-resolution method of studying the features of autoionizing states in strong fields. The total intensity of the emitted radiation has a very sharp feature for relatively small values of radiative decay constant as a function of the laser field strength and/or detuning and thus could be efficiently used to study the nature of "confluence"[2] which occurs close to the Fano minimum. In fact, this also provides us with a way to measure the Fano[4] asymmetry parameter q.

The calculations are based on a recently derived master equation[3] describing the laser induced autoionization. The master equation is transformed into quantum Langevin equations[5,6] for system operators, which are then solved exactly to obtain the two-time correlations involving the dipole operators. Such two-time correlations are then used to evaluate the photoemission spectra. Our treatment takes into account both the decay of the unperturbed continuum by radiative recombination and the autoionizing state. As we will discuss at the end of the paper, the master equation approach also enables one to calculate the properties of the emitting radiation when the system can emit many photons.

Our model consists of a continuum of states $|\epsilon)$ and the autoionizing state $|a\rangle$ with the configuration interaction V_ϵ. The state $|a\rangle$ radiatively decays to some final state $|f\rangle$ with the decay rate γ_0. A strong laser field of frequency ω_l couples the states $|\epsilon)$ and $|a\rangle$ [or equivalently Fano states $|\epsilon\rangle$ which are linear superpositions of $|a\rangle$ and $|\epsilon)$] to some initial state $|i\rangle$ with the matrix element $\tilde{v}_{\epsilon i}$ (or $v_{\epsilon i}$). These two matrix elements are different since $v_{\epsilon i}$ is the matrix element between Fano states rather than original continuum and $|i\rangle$. The master equation, obtained in the previous work, can be written in a more transparent form

$$\frac{\partial\rho}{\partial t}=-i[H,\rho]-\frac{\gamma_0}{2}(A^\dagger A\rho-2A\rho A^\dagger+\rho A^\dagger A) \ , \quad (1)$$

where the operator A is defined by

$$A=\int d\epsilon|f\rangle\langle\epsilon|B_{\epsilon a}, \quad B_{\epsilon a}\equiv\langle a|\epsilon\rangle\left[1+\frac{2(\epsilon-\epsilon_a)}{\Gamma q_f}\right] \ , \quad (2)$$

and H contains all the coherent interactions

$$H=\int v_{\epsilon i}|\epsilon\rangle\langle i|\,d\epsilon+\text{H.c.}+\int(\epsilon-\omega_l)|\epsilon\rangle\langle\epsilon|\,d\epsilon \ . \quad (3)$$

In writing (1), we have eliminated all the fast dependence from the Hamiltonian which is equivalent to making a canonical transformation with the Hamiltonian $\omega_l\int|\epsilon\rangle\langle\epsilon|\,d\epsilon+\epsilon_f|f\rangle\langle f|$. In Eq. (2), q_f denotes the Fano asymmetry parameter associated with the final state $|f\rangle$. The terms involving q_f arise due to the decay by possible radiative recombination while the electron is in the vicinity of the atom which is introduced via $v_{\epsilon f}$ (in contrast to q which has $v_{\epsilon i}$) of the unperturbed continuum. Of course, if q_f is large, then the decay of the unperturbed continuum is not important and then $B_{\epsilon a}\rightarrow b_{\epsilon a}\equiv\langle a|\epsilon\rangle$. In such a case, the operator A would essentially be the dipole moment operator associated with the transition $|a\rangle\rightarrow|f\rangle$.

The number of photons in any mode can be calculated from the knowledge of the dipole-dipole correlation function[7]

$$N_{Ks}(t)=\langle a_{Ks}^\dagger(t)a_{Ks}(t)\rangle=|\vec{d}_{af}\cdot\vec{u}_{Ks}|^2\int_0^t dt_1\int_0^t dt_2\,\langle A^\dagger(t-t_1)A(t-t_2)\rangle\exp[+i\omega_{Ks}(t_1-t_2)-i(\omega_l-\epsilon_f)(t_1-t_2)] \ . \quad (4)$$

The total number in all the modes is equal to

$$N(t)=\sum_{Ks}N_{Ks}(t)=\gamma_0\int_0^t d\tau\,\langle A^\dagger(\tau)A(\tau)\rangle \ . \quad (5)$$

We can sum (4) over all the directions and polarizations and rewrite the photon numbers with frequency ω as

$$N(\delta)\equiv\frac{\gamma_0}{2\pi}\int_0^t dt_1\int_0^t dt_2\langle A^\dagger(t_1)A(t_2)\rangle\exp[-i\delta(t_1-t_2)],\quad \delta=\omega-(\omega_l-\epsilon_f)\ . \tag{6}$$

The calculation of the two time correlation functions which appear in relation (4) turns out to be much easier by using quantum Langevin equations. Using (1), one can, for example, derive the following closed sets of Langevin equations:

$$\frac{d}{dt}|f\rangle\langle\epsilon_1|=-i\Delta_{\epsilon_1}|f\rangle\langle\epsilon_1|-iv_{\epsilon_1 i}|f\rangle\langle i|-\frac{\gamma_0}{2}\int d\epsilon\, B^*_{\epsilon_1 a}B_{\epsilon a}|f\rangle\langle\epsilon|+F_{f\epsilon_1}(t)\ ,\quad \Delta_\epsilon=(\epsilon-\omega_l)\ , \tag{7}$$

$$\frac{d}{dt}|f\rangle\langle i|=-i\int d\epsilon\, v^*_{\epsilon i}|f\rangle\langle\epsilon|+F_{fi}(t)\ . \tag{8}$$

The quantum Lagevin forces $F_{f\epsilon_1}(t)$, $F_{fi}(t)$ are found to have the properties

$$\langle F_{f\epsilon_1}(t)\rangle=\langle F_{fi}(t)\rangle=0\ ;$$

$$\langle F^\dagger_{f\epsilon_1}(t)F_{f\epsilon_2}(t')\rangle=\langle F^\dagger_{f\epsilon_1}(t)F_{fi}(t')\rangle=\langle F_{fi}(t)F^\dagger_{f\epsilon}(t')\rangle=\langle F^\dagger_{fi}(t)F_{f\epsilon_2}(t')\rangle=\langle F^\dagger_{fi}(t)F_{fi}(t')\rangle=0\ ; \tag{9}$$

$$\langle F_{fi}(t)F^\dagger_{fi}(t')\rangle=\gamma_0\delta(t-t')\left\langle\int d\epsilon'\int d\epsilon''\, B^*_{\epsilon' a}B_{\epsilon'' a}|\epsilon'\rangle\langle\epsilon''|\right\rangle_t,\ \text{etc.}$$

The higher-order correlations of such random forces can also be computed from (1), but in the following we do not need them. We have been able to solve quantum Langevin equations (7) and (8) exactly. Using these and the correlation functions (9) and the initial condition $\rho(t=0)=|i\rangle\langle i|$, the dipole correlation function $\langle A^\dagger(t_1)A(t_2)\rangle$ can be computed. This calculation finally leads to the following[8–10] result for photon spectrum $N(\delta)$, for long times $t\to\infty$,

$$N(\delta)=\frac{\gamma_0}{2\pi}\left|\frac{\sqrt{2/\gamma_0}\,m_{21}(z)}{[1+m_{11}(z)][1+m_{22}(z)]-m_{12}(z)m_{21}(z)}\bigg|_{z=-i\delta}\right|^2\ . \tag{10}$$

The m matrix is defined in terms of the basic parameters of the system as

$$m=\int\frac{d\epsilon}{z+i\Delta_\epsilon}\begin{pmatrix}|v_{\epsilon i}|^2/z & \sqrt{\gamma_0/2}\,B^*_{\epsilon a}v^*_{\epsilon i}\\ \frac{1}{d}\sqrt{\gamma_0/2}\,v_{\epsilon i}B_{\epsilon a} & \frac{\gamma_0}{2}|B_{\epsilon a}|^2\end{pmatrix}, \tag{11}$$

which reduces to the same matrix as that defined in a previous Communication[3] if we let $q_f\gg 1$ (i.e., $v_{\epsilon f}\to 0$). It can be shown that the effect of radiative recombination, i.e., decay of the unperturbed continuum, is unimportant if $\gamma/\Gamma q_f^2\ll 1$. In this paper, we only consider such a case. On simplification and taking the limit of flat continuum $|\epsilon)$, (10) leads to

$$N(\delta_0)=\frac{2\Omega\gamma}{\pi\Gamma}\frac{(q^2+1)}{|P(\delta_0)|^2},\quad \gamma=\frac{\gamma_0}{\Gamma},\quad \Omega=\frac{2\pi|\tilde v_{\epsilon i}|^2}{\Gamma},\quad \delta_0=\frac{2}{\Gamma}[\omega-(\epsilon_a-\epsilon_f)]\ , \tag{12}$$

where $P(\delta_0)$ is the same polynomial that characterized the photoelectron spectra[3] $|S(\epsilon)|^2=2\Omega|\epsilon+q+i\gamma|^2/|P(\epsilon)|^2$, i.e.,

$$P(x)=x^2+x[-\alpha+i(\Omega+1+\gamma)]-\Omega(\gamma+q^2)-i(\alpha+\alpha\gamma-2\Omega q),\quad \alpha=\frac{2(\omega_l-\epsilon_a)}{\Gamma}\ . \tag{13}$$

Thus the photoelectron and photoemission spectra have a very similar structure except for the additional energy dependence in photoelectron spectra, which is significant near the Fano minimum $\epsilon=-q$. The structure of the photoemission spectra as computed from (12) is shown in Fig. 1. The narrow feature in the photoemission spectra near confluence which occurs at $\Omega=1+(\alpha/q)$ has a width of the order $\gamma(q+\alpha)/(2q+\alpha)$. It is clear that the photoemission spectra essentially provide us with a high-resolution method of probing the photoelectron spectra.

The total number N of photons emitted can be obtained by integrating (12) over all the frequencies, and the results are shown in Fig. 2. For small values of γ, the behavior is quite significant near conflu-

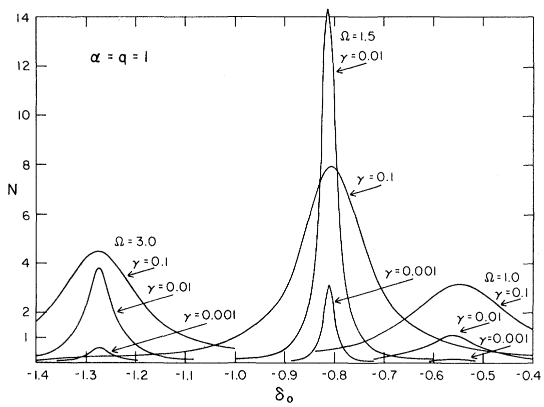

FIG. 1. Photoemission spectra $N(\delta_0)$ as a function of $\delta_0 \equiv 2[\omega - (\epsilon_a - \epsilon_f)]/\Gamma$ for various values of laser intensities Ω and the radiative linewidth γ.

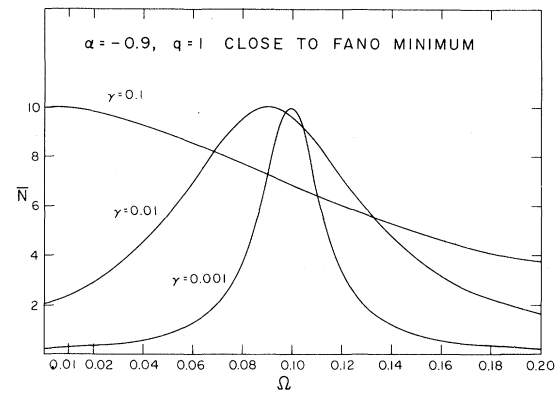

FIG. 3. Variation of $\bar{N}$ as a function of laser intensity (Rabi frequency Ω), when the laser is detuned close to Fano minimum, $\alpha = -0.9$, $q = 1$.

ence, i.e., for $\Omega = [1 + (\alpha/q)]$. The peak value for small γ is $q/(2q+\alpha)$. Thus the measurements of the total photon number will enable us to obtain rather easily the Fano asymmetry parameter q and also the width Γ of the autoionizing state (provided that the matrix element $|\tilde{v}_{\epsilon i}|$ is known) since a plot of peak position as a function of detuning is a straight line with an intercept on the detuning axis equal to Γ. Note that the total photoelectron emission [equal to $(1-N)$], will show a sharp dip near confluence ($\gamma \neq 0$).

In the foregoing analysis, we have assumed that the system decays to some final state $|f\rangle$ which is different from the state $|i\rangle$. An important question is now—what happens if the states $|f\rangle$ and $|i\rangle$ are identical? In such a case we have proved that in the long-time limit the results can be obtained from the results so far presented by using the scaling relations[11]

$$\bar{\rho}_{\epsilon\epsilon}(t \to \infty) = \frac{|S((\epsilon - \epsilon_a)/(\Gamma/2))|^2}{\int d\epsilon\, |S((\epsilon - \epsilon_a)/(\Gamma/2))|^2}\,, \quad (14)$$

$$\bar{N}(t \to \infty) = \frac{N}{1-N}\,. \quad (15)$$

Now, because of the factor $(1-N)^{-1}$ in (15), the structure of Fig. 2 sharpens further, as shown by dashed curves in Fig. 2. This is so because many more photons can be emitted as the system returns to the state $|i\rangle$ and then gets excited by the laser field and this process gets repeated. The effect of the normalization factor is very significant if the laser is tuned close to the Fano minimum, as shown in Fig. 3; the peak height for $\bar{N}$ being $q/(q+\alpha)$ since, in this case, basically the photoelectron channel is

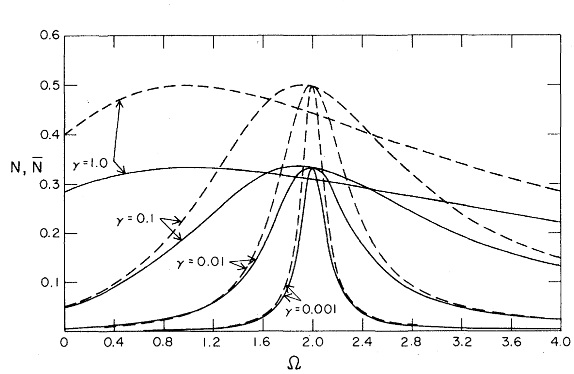

FIG. 2. Total number of photons N as a function of laser intensity for $\alpha = q = 1$. The dashed curves give the total number $\bar{N}$ [Eq. (15)] for the case in which the final state $|f\rangle$ is identical to the initial state $|i\rangle$.

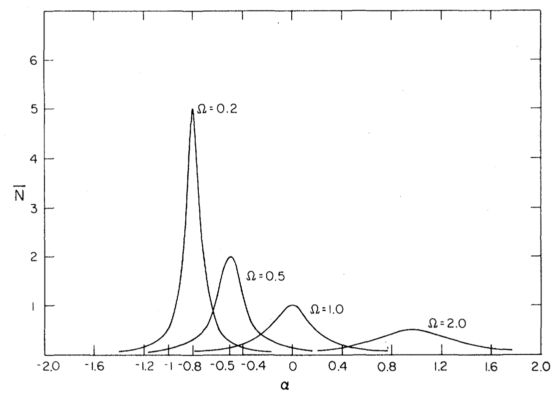

FIG. 4. Variation of the total number of photons as a function of laser detuning for $q = 1$, $\gamma = 0.01$, and for various values of Rabi frequency Ω.

closed and the system has no choice but to emit photons. Finally, Fig. 4 gives the variation of the total number of photons $\bar{N}$ as a function of laser detuning for various values of the laser intensity. This again has a sharp structure near confluence $\alpha = q(\Omega - 1)$ with peak height $\sim 1/\Omega$.

Thus, to conclude, the present study demonstrates how the confluence[2] of coherences at the Fano minimum in the laser induced autoionization is manifested in the photoemission spectra and how the photoemission spectra may be used with a very high degree of resolution as compared to the photoelectron spectra, in the study of the properties of autoionizing states.

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[1]For an earlier study of photoemission spectra in weak laser fields see, L. Armstrong, Jr., C. E. Theodosiou, and M. J. Wall, Phys. Rev. A 18, 2538 (1978); however, Dr. Armstrong (private communication) has informed us that a similar result is expected to hold good in strong fields; see also M. Crance and L. Armstrong (unpublished).
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[4]U. Fano, Phys. Rev. 124, 1866 (1961).
[5]For the use of quantum Langevin equations see, for example, M. Lax, Phys. Rev. 145, 110 (1966); I. R. Senitzky, *ibid.* 161, 165 (1967); and Ref. 6.
[6]G. S. Agarwal, in *Springer Tracts in Modern Physics*, edited by G. Höhler (Springer, Berlin, 1974), Vol. 70, p. 44.
[7]Compare Ref. 6, Sec. VII.
[8]Here we quote, for example, only the long-time behavior, though we have also calculated the time-dependent result.
[9]We have also verified the result (10) using an altogether different method based on resolvent formulation, which is well suited for handling single-photon situations and leads also to results where one does not have to make the Markov approximation for the radiation field. It may also be added that situations involving only a single spontaneously emitted photon can be treated by a variety of other methods (cf. Ref. 6), such as the Wigner-Weisskopf method; more sophisticated techniques like the master equations are crucial in the treatment of situations involving many spontaneously emitted photons.
[10]It should be noted that here we are calculating the number of photons in any given mode rather than its rate of change (as is usually the case in scattering problems). Note that in the photoelectron emission, all the population leaks out and nothing remains in the state $|i\rangle$ in the long-time limit. Questions regarding the pumping of the state $|i\rangle$ and the resulting photoelectron and photoemission spectra are planned to be examined elsewhere.
[11]The time-dependent results have a much more complicated structure, since the result in terms of Laplace transforms is $\hat{\rho}_{\epsilon\epsilon}(z) = [\hat{\rho}_{\epsilon\epsilon}(z)]/z \int d\epsilon\, \hat{\rho}_{\epsilon\epsilon}(z)$ and $\rho_{\epsilon\epsilon}(t) = |\psi_\epsilon(t)|^2$.