Photon Spectrum in Laser-Induced Autoionization

Maciej Lewenstein[(a)] and Joseph W. Haus
Fachbereich Physik, Universität Essen, 4300 Essen 1, West Germany
and
Kazimierz Rzążewski
Institute for Theoretical Physics, Polish Academy of Sciences, PL-02-668 Warsaw, Poland
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A quantum electrodynamic treatment of an autoionizing atom excited by a laser field is presented. As the main result, the authors present the average number of photons emitted into a particular mode of the electromagnetic field. In the case of a symmetric autoionizing resonance, the spectrum presented here resembles the power spectrum in resonance fluorescence of a two-level atom. For an asymmetric resonance, however, this spectrum manifests the confluence of coherences. The total number of scattered photons has a maximum at the confluence intensity. The spectrum is asymmetric in this case.

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In this Letter we investigate the effect of spontaneous emission accompanying the autoionization process. Spontaneous emission to a third level has been previously discussed by Agarwal *et al.*[1] However, we consider spontaneous emission to the ground state. This has not been accounted for in earlier papers on autoionization. As our central result, we calculate the average number of scattered photons in a particular plane-wave mode. This property is another fundamental, quantum electrodynamic, strong-field effect and, as we shall show below, has similarities with the resonance-fluorescence power spectrum of the two-level atom.[2]

Recent work on laser-induced autoionization has revealed several interesting features of the strong-field spectra.[1,3-5] One remarkable feature of the laser-atom interaction is the confluence of coherences[3]; in the neighborhood of this point, the lifetime of the excited state increases dramatically and there is a sharp peak in the emitted-electron energy spectrum. This confluence, discovered in Ref. 3, is very difficult to observe because of poor spectral resolution of electrons. To overcome this obstacle, other authors introduced another laser beam and suggested a double-optical-resonance type experiment.[6] Furthermore, even with greatly improved spectral resolution, the width and height of the peaks are limited by dynamical disorder, such as, laser-phase fluctuations,[3b] inhomogeneous broadening,[7] and spontaneous emission.[1]

We consider the model of a bound state and continuum states coupled by an electromagnetic field:

$$H = H_A + H_F + H_{AF}, \tag{1}$$

with

$$H_A = -\hbar\omega_0 P_0 + \int_0^\infty d\omega\, \hbar\omega P_{\omega\omega},$$
$$H_F = \sum_\mu \int d^3k\, c\hbar k a_{\vec{k}\mu}^\dagger a_{\vec{k}\mu},$$
$$H_{AF} = \sum_\mu \int d^3k \int_0^\infty d\omega \hbar \vec{\Omega}(\omega)\cdot \epsilon_{\vec{k}\mu}\, g(k)(a_{k\mu}^\dagger D_\omega + \text{H.c.}),$$

where P_0 and $P_{\omega\omega}$ are the ground-state and continuum-state occupation operators, and D_ω, $D_\omega^\dagger$ are the corresponding polarization operators. $a_{\vec{k}\mu}^\dagger$, $a_{\vec{k}\mu}$ are the photon creation and annihilation operators for photons with momentum $\vec{k}$ and polarization μ. $g(k)$ is the form factor in the electric field operator.

As in Rzążewski and Eberly,[3] we use a two-Lorentzian generalization of the dipole moment $\vec{\Omega}(\omega)$:

$$\vec{\Omega}(\omega) = \frac{\vec{d}}{(4\pi\Gamma)^{1/2}}\; \frac{\Gamma}{\omega - i\Gamma} + \frac{1}{1-iq}\; \frac{\Gamma_1}{\omega + i\Gamma_1}, \tag{2}$$

where q is the Fano asymmetry parameter. Γ_1 is a parameter which is taken to infinity after the calculations have been performed; the squared amplitude of this function is called the Fano profile[8] in this limit.

We calculate the total number of photons emitted to the electromagnetic field modes, rather than a standard power spectrum, because we are treating a transient phenomenon here. The power spectrum tends to 0 as $t\to\infty$ in our case. It is interesting to note that inspite of its being transient, we can study the process in some detail without computing time-dependent quantities. This spectrum of scattered photons, averaged over solid angles, Ω_k, is given in the long-time limit by

$$S(kc) = \lim_{t\to\infty} k^2 \sum_\mu \int d\Omega_k \langle 0 | a_{s,\vec{k}\mu}^\dagger(t) a_{s,\vec{k},\mu}(t) | 0\rangle, \tag{3}$$

where the initial state of the system $|0\rangle$ requires that the atom be in the ground state and the electromagnetic field be in a coherent state describing a linearly polarized monochromatic electric field at frequency ω_L, and having an amplitude $\Omega_0 = \vec{d}\cdot\vec{E}/\hbar$, which we refer to as the Rabi frequency.

The calculation of $S(kc)$ requires the solution of integro-differential equations for the coupled atom-field moments. The equations form a closed hierarchy of coupled equations with use of the Born and Markov approximations.[9] The resulting equations can be solved exactly; the method of solution, too lengthy for this Letter, is similar to that used in Ref. 3b. The limit $\Gamma_1\to\infty$ is taken after all integrals have been carried out; the ionization threshold effects can be safely neglected and frequency integrals over the entire real axis are performed.[10,11]

Consider first the case of a symmetric Fano profile ($q=\infty$). The normalized photon spectrum is given by

$$S(kc) = \pi^{-1}\,\mathrm{Re}\, G(z)\left(z+\Gamma+\Gamma_s+i\Delta+\frac{\Omega_0^2(z+\Gamma)}{2[z+2(\Gamma+\Gamma_s)](z+\Gamma+\Gamma_s+i\Delta)}\right)_{z=-i(kc-\omega_L)}, \tag{4}$$

$$G(z) = \frac{(z+\Gamma+\Gamma_s-i\Delta)[z+2(\Gamma+\Gamma_s)]}{z\{[z+2(\Gamma+\Gamma_s)][(z+\Gamma+\Gamma_s)^2+\Delta^2]+\frac{1}{2}\Omega_0^2(z+\Gamma+\Gamma_s)\}+\frac{1}{2}\Omega_0^2(z+2\Gamma)(z+\Gamma+\Gamma_s)}, \tag{5}$$

where the detuning is $\Delta=\omega_0-\omega_L$ and the spontaneous emission rate is $\Gamma_s = 2|\vec{d}|^2\omega_0^3/3c^3$. The denominator of $G(z)$ is a fourth-order polynomial; for $\Delta=0$ one of the roots cancels; for the case $\Omega_0 \ll \Gamma$ the resulting third-order polynomial has the roots

$$z = -(\Gamma+\Gamma_s),\quad -2(\Gamma+\Gamma_s),\quad -\Omega_0^2\Gamma/2(\Gamma+\Gamma_s)^2. \tag{6}$$

The maximum of the spectrum, $ck=\omega_L$, has the height

$$S(\omega_L) = 2(\Gamma+\Gamma_s)^2/\pi\Gamma\Omega_0^2. \tag{7}$$

The final root in Eq. (6) in this weak-coupling limit dominates the spectrum and the spectrum is power broadened. Note that the peak narrows as the spontaneous lifetime is shortened. This is so because the spontaneous emission increases the lifetime of the atom by returning it to the ground state.

For Δ, $\Omega_0 \gg \Gamma$, Γ_s, the photon spectrum has three peaks as shown in Fig. 1. The side peaks are displaced by $\beta = \pm(\Omega_0^2+\Delta^2)^{1/2}$ and have widths $\kappa = \Gamma+\frac{3}{2}\Gamma_s - \frac{1}{2}\Gamma_s\Delta^2/\beta^2$. The central peak has a width Γ. The ratio of the heights is

$$R = \frac{S(\omega_L)}{S(\omega_L\pm\beta)} = \frac{2[\frac{1}{2}\Gamma\Omega_0^2+2(\Gamma+\Gamma_s)\Delta^2][\Omega_0^2(2\Gamma+\Gamma_s)+2\Delta^2(\Gamma+\Gamma_s)]}{\Gamma(\Gamma+\Gamma_s)\Omega_0^4}. \tag{8}$$

In the limit $\Delta=0$, $\Gamma_s\ll\Gamma$, the ratio is 2. In Fig. 1, where $\Delta=0$, this spectrum exhibits the asymptotic properties discussed above for $\Omega_0\gg\Gamma$ for quite moderate values of Ω_0.

Our photon spectrum remains symmetric in $k-k_L$, even with detuning. There is a striking qualitative similarity of these results with the strong-field fluorescence spectrum of the two-level atom. However, the widths of our peaks are basically determined by the autoionization width Γ and *not* by the spontaneous decay width Γ_s (at least as long as $\Gamma_s\ll\Gamma$).

In this strong-field limit the peaks are reduced, see Fig. 1, as Γ_s is increased. This is because the transition rates are determined by the Rabi frequency which is much larger than Γ_s. So spontaneous emission does not prolong the lifetime of the atom.

For finite q completely new features occur. They are manifestations of those delicate interference phenomena discussed in earlier papers.[3] We do not present the rather lengthy formulas here; they shall be fully discussed in a forthcoming publication.[10]

We plot the photon spectrum for $q=5$, $\Delta=0$ in Fig. 2 for two values of Γ_s, as discussed in the figure caption. For small values of the Rabi frequency, only one peak is present. Increasing the Rabi frequency results in a splitting of the central peak into a three-peak spectrum again; however, the spectrum is asymmetric and the widths of the side peaks are broadened by further increase of the Rabi frequencies. The condition

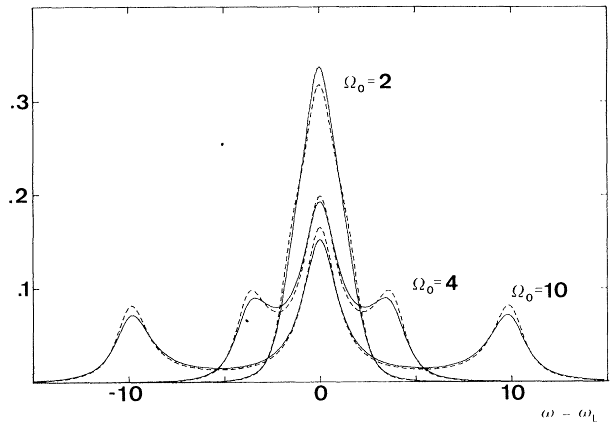

FIG. 1. The photon spectrum, Eq. (4), for $\Delta=0$ and $\Omega_0=2$, 4, and 10. The dashed lines are $\Gamma_s=0$ and the solid lines are $\Gamma_s=0.1$. All frequencies are scaled to Γ. Note that for a weak field the peak is enhanced, whereas for a strong field the peaks are reduced as discussed in the text ($\omega=ck$).

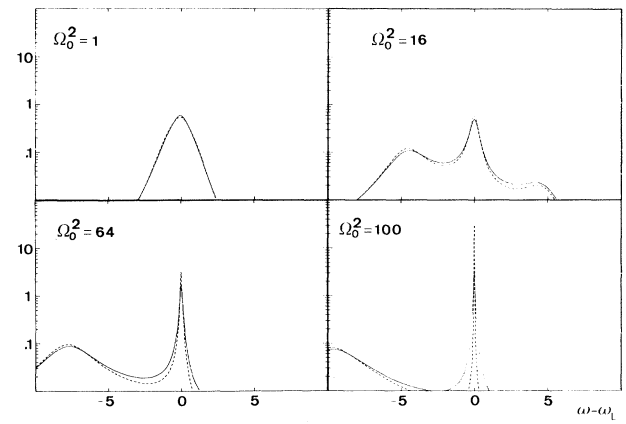

FIG. 2. A semilogarithmic plot of the photon spectrum for $q=5$, $\Delta=0$, and $\Omega_0{}^2=1$, 16, 64, and 100. The dashed curves are the results for $\Gamma_s=0.01$, while the solid curves represent $\Gamma_s=0.1$ ($\omega=ck$).

for the confluence is[3b]

$$\Omega_c{}^2=4\Gamma(1+q^2)(\Gamma-\Delta/q). \tag{9}$$

Near this value of the Rabi frequency, the spontaneous lifetime has the largest influence on the spectrum and determines the width of the central peak, as shown in Fig. 2 for $\Omega_0{}^2=100$. These results show that the structural features of the autoionizing resonance are present in the photon spectrum.

Generally, the total number of scattered photons per ionized atom is of the order Γ_s/Γ, so that for particular atomic systems good statistics are attainable. Again, an important exception is seen in the neighborhood of the confluence. In Fig. 3 we plot the integrated photon spectrum versus Rabi frequency for several values of detuning, $q=5$; this represents the total number of scattered photons. The curves show a dramatic enhancement in the total number of scattered photons at the confluence Ω_c, Eq. (9).

By appropriate choice of the detuning so that Ω_c is of order Γ, several orders of magnitude more photons should be emitted above the value Γ_s/Γ. When $\Omega_c=0$ the total number of photons emitted diverges as $\Omega_0\to 0$. The observation of this singularity requires measuring times of the order of the inverse power broadening (also, the effects of other fluctuations, e.g., laser-phase fluctuations, are dominant in this regime[10]). These times may be unrealistically long, whereas, for $\Omega_c\simeq\Gamma$ the basic emission time scale should be of the order $\Gamma_s{}^{-1}$. An experiment designed to count all the emitted photons would present an interesting test of the results presented above.

Note added.—After this manuscript was submitted the authors became aware of a recent publication by Agarwal *et al.*[12] Their photon spectra are calculated from the model of Ref. 1 and thus, there are principal differences between their results and ours. Our model fully accounts for correlations between successive photon events, whereas theirs does not include this physical process. Qualitative and quantitative differ-

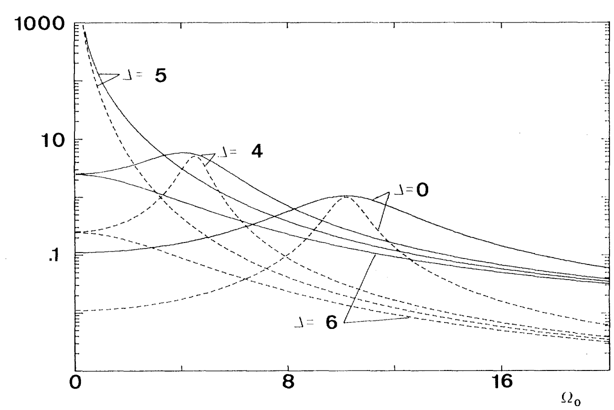

FIG. 3. A semilogarithmic plot of the total number of scattered photons vs the Rabi frequency for $q=5$ and four values of the detuning. The spontaneous emission rate $\Gamma_s=0.1$ is plotted as a solid line; $\Gamma_s=0.01$ is plotted as a dashed line. Near the confluence Ω_c, Eq. (9), there is an enhancement of the photon number. Note that the maximum is nearly independent of Γ_s!

ences between these two models will be discussed in a future comment.

[(a)]Permanent address: Institute for Theoretical Physics, Polish Academy of Sciences, Al. Lotników 32, PL-02-668 Warsaw, Poland.

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[10]Details of this calculation will appear in a future publication, M. Lewenstein, J. W. Haus, and K. Rzążewski, to be published.

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