COHERENT AND INCOHERENT EFFECTS IN LASER-INDUCED AUTOIONIZATION

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I. INTRODUCTION

Study of the autoionizing states of the atom and their interaction with the radiation field is becoming increasingly important both from the theoretical and the applied point of view. On the applied side it provides a possible way of producing VUV radiation\(^1\)–\(^3\) and thus under suitable pumping conditions, laser action in VUV, using the autoionizing states, is possible. The interaction also results in very interesting fundamental physical effects,\(^4\) which again could lead to the very efficient VUV devices. Normally the interaction of the autoionizing states\(^5\) with laser fields is studied by ignoring all incoherent effects. However, one would expect that any source of incoherence such as spontaneous emission, recombination of electrons and ions, collisions, etc., would affect all the basic phenomena that can arise in the interaction of autoionizing states with fields, particularly if resonances are involved. We have undertaken a systematic study of some of the incoherent effects in laser-induced autoionization. I will primarily deal with the effects of spontaneous emission and recombination. This lecture is based on the work carried out with several physicists — S. Haan, K. Burnett, J. Cooper, D. Agassi and P. Anantha Lakshmi.

II. BASIC MODEL, DYNAMICAL EQUATIONS AND THEIR SOLUTION

We consider the simplified model of Fig. 1, which is self explanatory. The autoionizing state \(|a\rangle\) can decay to the unperturbed continuum of states \(|e\rangle\) and to the state \(|i\rangle\) by spontaneous emission. The continuum of the electronic states also decays to \(|i\rangle\)
Fig. 1. Schematic diagram of the energy levels with various radiative and configuration interactions.

by recombination. After making the rotating wave approximation and the transformation to the rotating frame, the atom-laser field interaction can be written in the form

\[ H = \int (\varepsilon - \omega_0) |\varepsilon \rangle \langle \varepsilon | d\varepsilon + \int \frac{V_{\varepsilon i}}{\varepsilon} |\varepsilon \rangle \langle \varepsilon i | + H.C. \]  \hspace{1cm} (2.1)

The state $|\varepsilon \rangle$ in (2.1) is the Fano state obtained by the diagonalization of the interaction between the unperturbed continuum and the state $|a\rangle$:

\[ |\varepsilon \rangle = b(\varepsilon, a)|a\rangle + \int b(\varepsilon, \varepsilon')|\varepsilon'\rangle d\varepsilon' , \hspace{1cm} (2.2)\]

where

\[ b(\varepsilon, a) = \frac{\sin \Delta}{\pi V_\varepsilon} , \hspace{1cm} b(\varepsilon, \varepsilon') = \frac{V_{\varepsilon', \varepsilon}}{(\varepsilon - \varepsilon')\pi V_\varepsilon} - \cos \Delta \delta(\varepsilon - \varepsilon') , \hspace{1cm} (2.2)\]

\[ \tan \Delta = -\pi \left| V_\varepsilon \right|^2/(\varepsilon - \varepsilon_a - F(\varepsilon)) , \hspace{1cm} \Gamma = 2\pi \left| V_\varepsilon \right|^2 . \hspace{1cm} (2.3)\]

Here $\Gamma$ gives the autoionization rate and $F(\varepsilon)$ is the shift due to the configuration interaction. Generally $F(\varepsilon)$ is small and hence we will ignore it.

We next consider the interaction of the atomic system with the vacuum of the radiation field to account for the spontaneous emission and the recombination effects. Using the standard methods and making the Born and Markov approximations on this interaction, we find the dynamical equation for the atomic system

\[ \frac{\partial \rho}{\partial t} = -i[H, \rho] - \frac{\gamma_0}{2} (A^+ A \rho - 2A \rho A^+ + \rho A^+ A) . \hspace{1cm} (2.4)\]
where the operator $A$ plays the role of the dipole moment operator connecting the Fano continuum and $|i\rangle$

$$A = \int d\varepsilon |i\rangle \langle \varepsilon | B_{\varepsilon a} \quad B_{\varepsilon a} = \langle a | \varepsilon \rangle \left\{ 1 + \frac{2(\varepsilon - \varepsilon_a)}{\Gamma q} \right\}$$  \hspace{1cm} (2.5)$$

and where $q$ is Fano's asymmetry parameter.

It may be noted that if the recombination effects associated with the unperturbed continuum were ignored, then $B_{\varepsilon a} = \langle a | \varepsilon \rangle$. In order to see this particular limit of our results, we will replace $q$ in the curly bracket of (2.5) by $q_F$. The limits $q_F \rightarrow q$ and $\infty$ would thus give results with and without recombination effects. Even though (2.4) is quite complex, its exact solution can be obtained subject to the arbitrary initial condition

$$\rho_{\alpha \beta}(0) = \rho_{\alpha \beta} = \psi_{\alpha}^*(\psi_{\beta} - \psi_{\beta}^*).$$  \hspace{1cm} (2.6)$$

The result for the Laplace transform of $\rho$ can be written in the form

$$\hat{\rho} = \hat{\rho}(1) + \frac{1 - z \text{Tr} \hat{\rho}(1)}{z \text{Tr} \hat{\rho}(2)} \hat{\rho}(2),$$  \hspace{1cm} (2.7)$$

$$\rho_{\alpha \beta}^{(1)}(t) = \psi_{\alpha}^{(1)(t)}(t), \quad \rho_{\alpha \beta}^{(2)}(0) = 1, \quad \rho_{\varepsilon \varepsilon}^{(2)}(0) = 0.$$

The time dependence of $\psi$ is given by the set of equations

$$\dot{\psi}_{\varepsilon 1} = -i\Delta_{\varepsilon 1} \psi_{\varepsilon 1} - iv_{\varepsilon 1} \psi_{\varepsilon 1} - \frac{\gamma_{\varepsilon 1}}{2} \int d\varepsilon B_{\varepsilon a} \psi_{\varepsilon 1} \langle a | \varepsilon \rangle, \quad \Delta_{\varepsilon 1} = \varepsilon_1 - \omega_{\varepsilon 1},$$

$$\dot{\psi}_{1} = -i \int v_{\varepsilon 1} \psi_{\varepsilon 1} d\varepsilon.$$  \hspace{1cm} (2.9)$$

Starting from (2.9), one can obtain for the Laplace transforms of $\psi$'s, an integral equation with separable kernel. Such an integral equation is exactly solvable. For example, one has

$$\hat{\Phi}(z) = \int B_{\varepsilon a} d\varepsilon \hat{\psi}_{\varepsilon}^{(2)}(z) = -iz m_2 \left( \frac{2}{\gamma_0} \right)^{1/2} \left\{ z(1 + m_{11})(1 + m_{22}) - zm_{12} m_{21} \right\}^{-1}$$

$$\hat{\psi}_{\varepsilon}^{(2)}(z) = -i(z + i\Delta_{\varepsilon})^{-1} \left\{ z(1 + m_{11})(1 + m_{22}) - zm_{12} m_{21} \right\}^{-1}$$

$$\times \left\{ v_{\varepsilon 1}(1 + m_{22}) - zm_{21} \left( \frac{\gamma_0}{2} \right)^{1/2} B_{\varepsilon a} \right\}.$$  \hspace{1cm} (2.10)$$
The matrix $m$ is the key quantity which determines the behavior of the various photoelectron and photoemission spectra. In particular, the time dependence of $\psi_\varepsilon$ will have the form $\alpha \exp(-i\Delta \varepsilon t) + \beta_1 \exp(z_1 t) + \beta_2 \exp(z_2 t)$, where $z_1$ are the two complex zeroes of $z \det(1 + m(z))$, assuming that the unperturbed continuum can be taken to be flat. These roots are given by

$$z_{1,2} = -\frac{1}{2} \left( \varepsilon_\pm^* - \alpha \right), \quad \alpha = 2(\omega_a - \varepsilon_a)/\Gamma,$$

where $\varepsilon_\pm$ are the roots of the polynomial

$$P(\varepsilon) = \psi \varepsilon^2 + \left[ -\alpha \psi + \frac{2\gamma}{q_f} + i(\Omega + 1 + \gamma) \right] \varepsilon - \left[ \frac{2\alpha \gamma}{q_f} + \frac{\gamma^2}{q_f} \right] \psi \Omega + \frac{\gamma}{q_f} (1 - \frac{2q}{q_f}) + i\left[ 2q \Omega - \alpha(1 + \gamma) \right], \quad \psi = 2(\varepsilon - \varepsilon_a)/\Gamma.$$

We will now discuss the various consequences of (2.7).

### III. EFFECT OF SPONTANEOUS EMISSION ON FANO PROFILES

Fano profiles for the emitted photoelectrons can be obtained from our general expression by treating the interaction $v_{\varepsilon i}$ as weak so that it is sufficient to calculate $\lim_{t \rightarrow \infty} (dp_{\varepsilon i}/dt)$ to second order in $v_{\varepsilon i}$. The result is

$$S_F(\varepsilon) = \frac{(q + \varepsilon)^2 + \frac{2}{\Gamma}}{(1 + \frac{2}{\Gamma} + \psi^2(\varepsilon + \eta)^2)} + \frac{2\gamma}{q_f \psi \Gamma} \frac{(\varepsilon - \varepsilon_a)^2}{\Gamma}.$$

The results obtained thus far are quite general and no assumption regarding the structure of the unperturbed continuum has been made.
The behavior of $S\beta(\varepsilon)$ is given in Fig. 2 for various values of $\gamma = \gamma_0/T$. It is remarkable to note that if the recombination effects are ignored $q_f + \infty$, then $S\beta(\varepsilon)$ has no zero at $\varepsilon = -q$, whereas such a zero is restored if recombination is put in.

Fig. 2. Modified Fano profiles (after Ref. 7), $S\beta(\varepsilon)$ versus $\varepsilon$ for $q = 1$ and various $\gamma$. (a) The profile when recombination is neglected. The minimum at $\varepsilon = -q$ disappears, but the curves remain asymmetric. The crosses indicate the peaks of the curves for $\gamma = 0.1$ and 0.2. (b) The profile when recombination is included ($q_f = 1$). The minimum is maintained, but the asymmetry is quickly lost with increasing $\gamma$. The cross indicates the peak height for $\gamma = 0.1$. 
IV. PHOTOELECTRON SPECTRA

We next examine the photoelectron spectra $\rho_{\varepsilon\varepsilon}$ for arbitrary values of the field strengths, assuming that initially the atom was in the state $|i\rangle$. It then follows from (2.7) that

$$\hat{\rho}_{\varepsilon\varepsilon}(z) = \hat{\rho}^{(2)}_{\varepsilon\varepsilon}(z)/z \text{ Tr } \hat{\rho}^{(2)}(z), \quad (4.1)$$

$$\rho_{\varepsilon\varepsilon}(\omega) = \lim_{t \to \infty} \rho_{\varepsilon\varepsilon}(t) = \rho^{(2)}_{\varepsilon\varepsilon}(\omega)/\text{ Tr } \rho^{(2)}(\omega). \quad (4.2)$$

The time-dependent photoelectron spectra are extremely complex due to the denominator in (4.1) depending on the variable $z$; though these can be computed in terms of the zeroes $z_i$. The steady state situation is relatively simple. Since

$$\rho_{\varepsilon\varepsilon} = |\psi^{(2)}_{\varepsilon}(\omega)|^2,$$

we show in Fig. 3 the behavior of

$$|\psi^{(2)}_{\varepsilon}(\omega)|^2 \equiv S,$$

as a function of $\varepsilon$ for several values of the parameters $\gamma$ and $\Omega$. In order to give the main features of the spectra, we also ignore recombination effects. The case $\gamma = 0$ corresponds to the result of Rzazewski and Eberly. A small spontaneous emission decay

Fig. 3. Spectra $|S(\varepsilon)|^2$ in units of $1/\pi T$ for $q = 1$, $\alpha = 1$, and for different values of field strengths $\Omega = 2\pi |\tilde{\nu}_{\varepsilon 1}|^2/T$ (after Ref. 6).
rate is found to have a drastic effect on the spectra for field strengths $\Omega \sim 2$ and for $q = 1$. The behavior can be understood from the zeroes of the polynomial $P(\varepsilon)$. From (2.10) it is clear that $S(\varepsilon)$ should show peaks at $\varepsilon = \varepsilon_\pm^*$. For $\gamma = 0$ and $\varepsilon = -q$, $P = 0$ at $\Omega = [1+(a/q)]$. Thus for $\Omega = [1+(a/q)]$, one of the roots lies exactly on the real axis at $\varepsilon = -q$ which is just the place where the Fano minimum occurs. Hence for $\Omega \sim 1 + a/q$, one will have a very sharp spike and the Fano minimum. This is due to the strong laser field interaction and this situation has been referred to as the "confluence of coherences" by Rzazewski and Eberly. A very small value of $\gamma$ gives a finite width to the spike resulting in considerable change in spike height.

We have investigated in great detail the behavior of the zeroes of $P(\varepsilon)$ and the results are shown in Fig. 4. Note that in the limit $\Omega \to 0$, the zeroes of $P(\varepsilon)$ are at $\varepsilon_+ = \alpha$, $\varepsilon_- = -(2\gamma/\psi_\alpha) - (i(1+\gamma)/\psi)$ and it is the zero at $\varepsilon_-$ that occurs in the Fano profiles (3.1).

![Diagram showing the motion of zeroes $\varepsilon_\pm$ of $P(\varepsilon)$ with increasing laser intensity $\Omega$ for $q = 1$, $\gamma = 0.10$ and $\alpha = 0.4$ and 1.0 with $q_\beta = 1$ (solid curves) and $q_\beta \to \infty$ (dashed curves) (after Ref. 7). The abscissae for the $\alpha = 0.4$ ($\alpha = 1.0$) curves are given at the bottom (top) of the graph. Recombination shifts the zeroes noticeably for weak fields, but is of little importance near the confluence.]

Fig. 4. Motion of the zeroes $\varepsilon_\pm$ of $P(\varepsilon)$ with increasing laser intensity $\Omega$ for $q = 1$, $\gamma = 0.10$ and $\alpha = 0.4$ and 1.0 with $q_\beta = 1$ (solid curves) and $q_\beta \to \infty$ (dashed curves) (after Ref. 7). The abscissae for the $\alpha = 0.4$ ($\alpha = 1.0$) curves are given at the bottom (top) of the graph. Recombination shifts the zeroes noticeably for weak fields, but is of little importance near the confluence.
V. PHOTOEMISSION SPECTRA

In this section we discuss various important features associated with the photons emitted from the autoionizing states. It can be shown that the average number of photons at the frequency \( \omega \) is related to the dipole-dipole correlation function by

\[
N(\delta) = \frac{\gamma_0}{2\pi} \int_0^t dt_1 \int_0^t dt_2 \langle A^+(t_1)A(t_2) \rangle e^{-i\delta(t_1-t_2)}, \quad \delta = \omega - \omega_0
\]

(5.1)

and that the total photon yield is given by

\[
N = \gamma_0 \int_0^t d\tau \langle A^+(\tau)A(\tau) \rangle .
\]

(5.2)

Using the definition of \( A \) and (2.7), the Laplace transform of \( N \) becomes

\[
\hat{N}(z) = \left( \frac{\gamma_0}{z} \right) \cdot \left( |\hat{\Phi}(t)|^2 / z \right) \text{Tr} \rho^{(2)}(z) ,
\]

(5.3)

where \( \hat{\Phi}(z) \) is given by (2.10). The steady state expression for \( N \) is relatively simple

\[
N \xrightarrow{t \to \infty} \gamma_0 |\hat{\Phi}(0)|^2 / (\text{Tr} \rho^{(2)}(\infty)) .
\]

(5.4)

The expression for the spectrum can be obtained from (5.1) by using the solution (2.7) and the quantum regression theorem. We will not give the explicit form of the spectrum but present only some numerical results.

We first examine very briefly the photon spectra in the absence of any laser field but assuming that the atom has been excited to the autoionizing atom \( |a> \) at time \( t = 0 \) by some means. The spectrum of (pure) spontaneous emission then turns out to be

\[
N(\omega) = \frac{\gamma_0}{2\pi \psi^2} \frac{1}{4} / \left[ (\omega - \epsilon_a + \frac{\gamma_0}{\psi q_f})^2 + \frac{(\Gamma + \gamma_0)^2}{4\psi^2} \right]
\]

(5.5)

\[
\text{no recombination, } q_f \to \infty \quad \gamma_0 / 2\pi \frac{1}{(\omega - \epsilon_a)^2 + \left( \frac{\Gamma + \gamma_0}{2} \right)^2} .
\]

(5.6)

The total number of photons is

\[
N = \frac{\gamma_0}{\Gamma + \gamma_0} \cdot \frac{1}{2} \left( 1 + \frac{1}{q_f^2} \right)
\]

(5.7)
which can be compared with the total number $P_e$ of electrons

$$P_e = \frac{\Gamma}{\Gamma + \gamma_0} \cdot \frac{1}{\phi} \left( 1 + \frac{\gamma_0^2}{\Gamma^2 q_f^2} \right).$$

(5.8)

The branching ratios that follow from (5.7) and (5.8) agree with those given by Armstrong et al.\textsuperscript{13}

Various features of the photoemission from autoionizing states in strong fields are given in Figs. 5-8. The behavior of the total number of photons as a function of the applied field intensity is given in Fig. 5. For small values of $\gamma$ the peak in this curve occurs at $\Omega = 1 + (\alpha/q)$, which is precisely the field strength for confluence and is thus very significant. As discussed elsewhere\textsuperscript{6} this peak can be used to determine the Fano asymmetry parameter $q$. This figure also shows the relative importance of recombination effects as $\gamma_0$ is changed. The photon spectra are expected to have peaks at $\delta \equiv 2(\omega - \omega_L)/T = 0$, $\pm \text{Re}(\epsilon_+ - \epsilon_-)$. There are two peaks at $\delta = 0$ with different widths: $-2 \text{Im}(\epsilon_+)$, $-2 \text{Im}(\epsilon_-)$. The side peaks have a width $-\text{Im}(\epsilon_+ + \epsilon_-)$. The characteristics of the confluence are reflected by the presence of a very narrow spike at the laser

![Fig. 5. The total number of photons as a function of $\Omega$ for $q = 1$ and for various $\gamma$ with laser tuned close to Fano minimum ($\alpha = -q$). The dashed curves give results when recombination is ignored.](image-url)
Fig. 6. Spectrum for $\alpha = q = 1, Q = 3.0, \delta = 2(\omega - \omega_0)/T, \delta_0 = 2(\omega - \epsilon_a)/T$. The inset shows $N(\delta)$ for $\gamma = 0.1$ and the cross indicates spike height for $\gamma = 0.01$ (after Ref. 8).

Fig. 7. Spectrum for same parameters as in Fig. 6 except $\gamma = 1.0$. The crosses indicate spike heights for smaller $\gamma$ (after Ref. 8).
Fig. 8. Spectrum for $\alpha = 0$, $\gamma = 0.1$, $q = 10$ and various laser intensities (after Ref. 8).

frequency ($\delta = 0$) since for $\Omega \sim 1 + (\alpha/q)$ and for small $\gamma$, the $\varepsilon_+$ has a negligibly small imaginary part. Near the confluence, the remaining three peaks are quite broad since $\text{Im} \varepsilon_+ \sim (2q+\alpha)/q$, $\text{Re}(\varepsilon_+ - \varepsilon_-) \sim -(2q+\alpha)$. The peaks at $\pm \text{Re}(\varepsilon_+ - \varepsilon_-)$ will be resolved only for large $q$. These general features are clearly seen in Figs. 6-8. The triplet structure of Fig. 8 is reminiscent of the Mollow triplet in the context of scattering from a strongly driven two-level atom. Notice also the prominent asymmetries of the spectra produced by scattering of radiation from autoionizing states.

VI. FOUR-WAVE MIXING PROFILES

The importance of the autoionizing states in producing VUV radiation using four-wave mixing has already been emphasized in the introduction. It is clear from the foregoing that the spontaneous emission from the autoionizing states would change the features of the four-wave profiles considerably. We present here the main results for the model shown in Fig. 9. The two-photon transition $|g\rangle \leftrightarrow |i\rangle$ is taken to be a weak one. Using techniques similar to those discussed above, one finds that the four-wave mixing is proportional to $|S(x)|^2$ where

$$S(x) = \sqrt{Q} q_1 \left(1 - \frac{1}{q_1} - \frac{1}{q_2} + \frac{ix}{q_1 q_2} \right)/D,$$

$$x = \frac{2}{F} (\epsilon_a - \omega_1 - 2\omega_2),$$

(6.1)
Fig. 9. Schematic diagram of the model used to calculate the four-wave mixing profiles.

\[ D \equiv \left( \frac{\gamma_1}{\Gamma} (x+\alpha) - i\omega q_1^2 \right) \left\{ \frac{\gamma_2}{\Gamma} \left( \frac{1}{q_1} - \frac{1}{q_2} \right)^2 + \left( 1 - \frac{i}{q_1} \right)^2 + \frac{(1+i\alpha)}{q_2^2} \right\} \\
+ (x+\alpha) \left[ (1+i\alpha) \left( 1 + \frac{\gamma_2}{\Gamma q_2} \right) + \frac{\gamma_2}{\Gamma} \left( 1 - \frac{i}{q_2} \right)^2 \right], \quad (6.2) \]

and where various symbols have obvious meanings. The effect of the spontaneous emission on the weak field profiles (\(|S(x)|^2 \propto \xi\)) is shown in Fig. 10. The profiles change considerably as \(\gamma_1\) and \(\gamma_2\) are changed. An important feature of Fig. 10 is the change in the peak positions with changes in \(\gamma_1\) and \(\gamma_2\). Curve 1 of Fig. 10 corresponds to the profiles calculated by Armstrong and Beers\(^2\) using different methods. Some of the characteristics of the strong field profiles are shown in Fig. 11. It should be noted that

\[ \lim_{q \to q_1} D \equiv \lim_{\gamma \to 0} P(-x) \quad (6.3) \]

and hence the four-wave mixing signal (6.1) will have a sharp spike at \(x = q_1\) corresponding to \(\xi = [1 + (\alpha/q_1)]\). Figure 11 indeed shows such spikes and the effect of spontaneous emission on the characteristics of the spike. The curves show that the dependence on \(\gamma_2\) is more dramatic than on \(\gamma_1\). Furthermore, for fixed \(\gamma_2\), an increase in \(\gamma_1\) leads to an increase in four-wave mixing signals. The relatively stronger dependence on \(\gamma_2\) can be understood by remembering that the calculations are done to the lowest order in the two-photon transition rate \(|g\rangle \leftrightarrow |1\rangle\). For large values of \(q\), the doublet structure of \(|S(x)|^2\) can be seen.
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REFERENCES