Laser-induced bound and metastable states in bound-continuum systems

Kai-Shue Lam
Department of Physics, California State Polytechnic University, Pomona, California 91768

Thomas F. George
Department of Chemistry and Department of Physics, State University of New York at Buffalo, Buffalo, New York 14260
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A monochromatic laser is shown to be capable of inducing a bound state and multiple metastable states not originally present in the field-free spectrum of a bound-continuum system. For the bound state to exist, the field intensity must be larger than a certain frequency-dependent critical value. The widths of the metastable states are found to be in general proportional to renormalized field intensities rather than the physical field intensity. The energies can be classified as being in the metastable or unstable regimes, depending on whether the observed widths are smaller or larger than the corresponding "bare" widths predicted by perturbation theory. The relation of the Fano or laser-induced "autoionization" states to the scattering states is also demonstrated.

I. INTRODUCTION

Systems involving interactions between bound and continuum states present many interesting features in their spectral properties. The classic example is that of configuration interaction in atomic autoionization,[1] whose spectra exhibit characteristic asymmetric peaks. Usually, the bound-continuum coupling is considered to be completely system dependent, so that the spectral structure is fixed once the system is specified. The introduction of a laser, however, changes the situation drastically: The radiative bound-continuum coupling can readily be manipulated by varying the laser characteristics so as to produce many novel spectral features in a given system. Very interesting studies of this situation have recently been carried out on the processes of laser-induced autoionization[2–7] and photodetachment of electrons from negative ions.[8–11] In the former group of studies it is found that many details of the radiative interaction, such as radiative decay of the unperturbed continuum, phase fluctuation and relaxation of the atomic system due to both atomic collisions and laser-phase fluctuations, and even weak couplings between the electron and photon continua, can lead to very distinctive features in both the electron (autoionization) and photon (fluorescence) spectra. In the second group of studies, results of particular importance are the possibility of population trapping (formation of bound states) due to the interference between continuum-continuum transitions,[8,9] and the nonexponential time-decay due to threshold effects when the laser is tuned near the continuum threshold.[10]

In most of these studies, the coupling mechanisms between the bound states and different continua are quite involved. In addition to radiative bound-bound couplings, there are both the field-free configuration interaction and radiative bound-continuum couplings. In some cases, continuum-continuum couplings, both of the electron-electron and electron-photon types, are also treated. While such coupling schemes may be typical in many systems, they are perhaps not the most suitable for studying the effects due solely to the radiative bound-continuum coupling.

In this paper, we make use of the simplest possible model, discussed in Sec. II, to focus exclusively on this coupling and present a systematic study of the spectral problem associated with it. This case already brings into focus several interesting basic features in the bound-continuum spectrum which have largely been ignored previously. The generality of the model also makes it suitable for the investigation of a variety of other problems involving charge-transfer in molecular systems, such as gas-phase collisional ionization (Penning and associative) and neutralization (negative-ion formation) of impact ions in surface scattering.

An implicit assumption of many previous works is that the spectral range is invariant with respect to the "rediagonalization" imposed by the bound-continuum coupling. Thus, if the unperturbed spectrum consists of a discrete state embedded in a continuum, the exact spectrum is assumed to be just the continuum. In Sec. III this is found to be untrue if the coupling strength is larger than a certain frequency-dependent critical value. When this condition is fulfilled, a true bound state appears whose energy is lower than the threshold of the continuum. Its exact location depends critically on both the field strength and frequency of the laser. This fact makes the laser an ideal tool for inducing a "tunable" bound state originally not present in the field-free system.

The existence of a metastable state with energy centered near the energy of the unperturbed discrete state and a width directly proportional to the coupling strength is another frequently-exploited implicit assumption. In Secs. IV and V we show that when the coupling strength is large and strongly energy dependent, this assumption does not hold. Indeed, even when there is only one unperturbed discrete state, the laser may induce more than one metastable state whose energy levels bear no simple relationship to the energy of the unperturbed discrete state

[Eq. (37)]. This comes about because of the special properties of a principal-value Cauchy integral [Eq. (34)] usually taken to represent the line shift and conventionally discarded. In the case of strong radiative coupling, this quantity turns out to be of paramount importance. In Sec. IV a specific model expression for the coupling [Eq. (44)] is used in the calculation of this integral to demonstrate explicitly its effect on the energies of the metastable states (Figs. 3 and 4). Just as in the case of the bound state, the laser-induced metastable states are also found to be tunable by variation of the laser characteristics, but in a more subtle way than first expected. The principal-value integral also critically affects the widths of the metastable states through the requirement of coupling-constant renormalization when the field strength is large. Thus it is shown in Sec. V that the physically observed widths are not directly proportional to the physical field intensity but to a renormalized field intensity which may be larger or smaller than the physical one, depending on the energy of the metastable state. For a given physical field strength and frequency, we show that the energy range of the continuum may be divided into three regimes: the metastable regime, where the renormalized intensity is smaller than the physical one (line narrowing); the unstable regime, where the renormalized intensity is larger than the physical one (line broadening); and the unphysical regime, where the renormalized intensity is negative. These results point to the fact that in addition to the energy levels, the lifetimes of the metastable states can also be "tuned" by the laser.

In Sec. VI we apply the foregoing results to a derivation of an expression for the dissociation or recombination spectrum [Eq. (68)]. It is shown exactly how the resonance peaks of the metastable states appear and how they are influenced by the nonresonant background. Of special interest is the fact that in the case where a true bound state is present, an extra term [the first term inside the large parentheses in Eq. (68)] is added into the nonresonant background which is especially important near the dissociation threshold [Fig. 4(b)].

In considering the bound and metastable states we have found it necessary to introduce the scattering states, which are continuum eigenstates of the total Hamiltonian of our model. Not only are these states extremely important within the structure of the theory, as shown in Sec. IV, but they are useful in the direct calculation of the S matrix from which information on both bound and metastable states can be drawn. They are also the basic ingredients out of which eigenstates satisfying various boundary conditions can be constructed. In particular, the Fano states, which lead to the asymmetric profiles mentioned at the beginning of this section, are just standing-wave combinations of the outgoing and incoming scattering states. This is demonstrated in Sec. VII.

II. THE MODEL

Our analytical work on the metastable states is based on the Lee model of unstable particles.[12,13] Although this model was originally introduced to treat the problem of the renormalizability of field theories,[14] in several variant forms it has become the prototype model of many bound-continuum problems in diverse fields of physics.[15] In the present work we adapt it to the radiative coupling case and provide a derivational and interpretive framework which is unique to that coupling.

We consider the simplest model capable of describing the laser-induced bound state and metastable states. Fig. 1 depicts schematically the energy spectrum of the model. The single discrete state, of energy ϵ_d, is completely outside the continuum band; and the energy gap Δ, separating ϵ_d and the continuum (dissociation) threshold μ, is taken to be sufficiently large such that there can be no field-free bound-continuum interaction. A radiative bound-continuum interaction, however, can be induced by a laser with frequency $\omega \gtrsim \Delta$ ($\hbar$ set equal to 1). We can thus write the Hamiltonian as

$$H = H_0 + H' , \tag{1}$$

where

$$H_0 = \epsilon_d c_d^\dagger c_d + \sum_\epsilon \epsilon c_\epsilon^\dagger c_\epsilon + \omega a_\omega^\dagger a_\omega , \tag{2}$$

and

$$H' = \sum_\epsilon (a_\omega^\dagger + a_\omega)(V_{\epsilon\omega} c_d^\dagger c_\epsilon + V_{\epsilon\omega}^* c_d c_\epsilon^\dagger) . \tag{3}$$

c_d ($c_d^\dagger$), c_ϵ ($c_\epsilon^\dagger$), and a_ω ($a_\omega^\dagger$) are the destruction (creation) operators for the discrete state, the continuum state of energy ϵ, and the photon state of frequency ω, respectively; and $V_{\epsilon\omega}$ is the radiative bound-continuum coupling strength for energy ϵ. In addition to the simplifying approximation of a single-mode field, the spin of the photon (leading to polarization effects) has also been neglected.

The eigenstates $\{ | n_d, n_\epsilon, n_\omega \rangle \}$ of H_0 form a complete set. Since $n_d + n_\epsilon$ is a constant of motion, we choose to work in the sector of the entire Hilbert space in which $n_d + n_\epsilon = 1$, with the completeness relation

$$\sum_n \left[| d_n \rangle \langle d_n | + \sum_\epsilon | \epsilon_n \rangle \langle \epsilon_n | \right] = 1 , \tag{4}$$

where

$$| d_n \rangle \equiv | 1,0,n \rangle , \tag{5}$$

$$| \epsilon_n \rangle \equiv | 0,1,n \rangle , \tag{6}$$

and n is any positive integer or zero. In this representa-

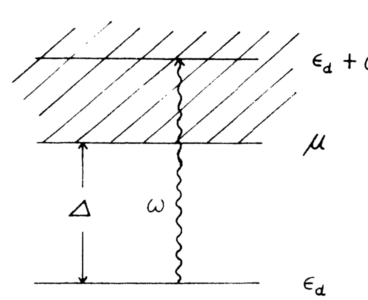


FIG. 1. Schematic energy-level diagram for a model bound-continuum system interacting with an external radiation field.

tion all matrix elements of the form $\langle d_n | H | \epsilon_{n\pm 1}\rangle$ are nonvanishing. Thus the entire sector of the Hilbert space spanned by $\{ |d_n\rangle, |\epsilon_n\rangle \}$ is in principle required for the description of the radiative interaction. This sector, however, separates still further into an infinity of subspaces, each invariant under H, if the rotating-wave approximation (RWA) is made. The RWA simply states that all antiresonant processes are forbidden. With respect to the energy spectrum of Fig. 1, this means that

$$\langle \epsilon_n | H | d_{n-1} \rangle = 0 \qquad (7)$$

for all n. Each irreducible subspace can then be labeled by a fixed n and has a completeness relation of the form

$$| d_{n+1} \rangle \langle d_{n+1} | + \sum_{\epsilon} | \epsilon_n \rangle \langle \epsilon_n | = 1 \; . \qquad (8)$$

From now we shall assume the validity of the RWA and work exclusively in the subspace where $n = N$ is fixed and $n_d + n_\epsilon = 1$. N will be taken to the number of photons in the external field and gives a measure of the latter's intensity.

As pointed out in the Introduction, two examples to which this model can be applied, are the problems of gas-phase collisional-ionization and charge transfer in ion-surface scattering. In the first case, typical processes are of the type

$$A^* + B + \omega \rightarrow \left. \begin{matrix} A + B^\dagger \\ (AB)^\dagger \end{matrix} \right\} + e^- \; .$$

Using our model, $|d_{n+1}\rangle$ would be the discrete state corresponding to the molecular-field complex $A^* + B + (n+1)\omega$ and $|\epsilon_n\rangle$ the electronic continuum state with energy $\epsilon + n\omega$ above the ionization threshold corresponding to the ion-complex $A + B^\dagger$ or $(AB)^\dagger$. In the second case, $|d_{n+1}\rangle$ may be the field-dressed valence state of the impact ion, and $|\epsilon_n\rangle$ the corresponding field-dressed electronic band-state of the metallic surface. In the present work, however, our model has not been adapted for the treatment of the detailed dynamics of the nuclear motion in these processes.

III. THE BOUND STATE

We proceed to solve the eigenvalue problem for H in the subspace specified following Eq. (8). Let $|E(N)\rangle$ be an eigenstate of H:

$$H | E(N) \rangle = E | E(N) \rangle \; . \qquad (9)$$

It follows from Eq. (8) that $|E(N)\rangle$ can be expanded as

$$| E(N) \rangle = \beta_E | d_{N+1} \rangle + \sum_{\epsilon} \chi_{E\epsilon} | \epsilon_N \rangle \; . \qquad (10)$$

It is then straightforward to apply Eqs. (1), (2), (3), and (10) to Eq. (9) and obtain

$$E\beta_E | d_{N+1} \rangle + \sum_{\epsilon} \chi_{E\epsilon} E | \epsilon_N \rangle = \beta_E \left[[\epsilon_d + (N+1)\omega] | d_{n+1} \rangle + \sqrt{N+1} \sum_{\epsilon} V^*_{\epsilon\omega} | \epsilon_N \rangle \right] + \sum_{\epsilon} \chi_{E\epsilon} [(\epsilon + N\omega) | \epsilon_N \rangle + \sqrt{N+1} V_{\epsilon\omega} | d_{N+1} \rangle] \; . \qquad (11)$$

Multiplying by $\langle d_{N+1}|$ and $\langle \epsilon'_N |$ leads, respectively, to the equations

$$E\beta_E = \beta_E [\epsilon_d + (N+1)\omega] + \sum_{\epsilon} \chi_{E\epsilon} \sqrt{N+1} V_{\epsilon\omega} \qquad (12)$$

and

$$\chi_{E\epsilon'} E = \beta_E \sqrt{N+1} V^*_{\epsilon'\omega} + \chi_{E\epsilon'} (\epsilon' + N\omega) \; , \qquad (13)$$

from which β_E and $\chi_{E\epsilon}$ can be eliminated to obtain

$$E - [\epsilon_d + (N+1)\omega] + \sum_{\epsilon} \frac{(N+1) | V_{\epsilon\omega} |^2}{\epsilon + N\omega - E} = 0 \; . \qquad (14)$$

This is the eigenvalue equation for H.

Defining an energy scale such that $N\omega = 0$ and converting the sum in the last equation to an integral, we have

$$E - (\epsilon_d + \omega) + g^2 \int_\mu^\infty d\epsilon \frac{\rho(\epsilon) | V_{\epsilon\omega} |^2}{\epsilon - E} = 0 \; , \qquad (15)$$

where $g^2 \equiv N + 1$ designates a dimensionless coupling strength that is proportional to the intensity of the external field, and $\rho(\epsilon)$ is a density of states appropriate to the continuum. The possible existence of a bound state can now be inferred from Eq. (15). Such a state exists if this equation has a real solution $E = \epsilon_b < \mu$. Since

$$\phi(E) \equiv g^2 \int_\mu^\infty d\epsilon \frac{\rho(\epsilon) | V_{\epsilon\omega} |^2}{\epsilon - E} \qquad (16)$$

is a monotonically increasing function of E for $E < \mu$, we infer that there exists one and only one bound state for Eq. (15) if

$$\phi(\mu) > \omega - \Delta \; . \qquad (17)$$

Thus no bound state can exist if the coupling strength falls below the critical value given by

$$g^2_{\text{crit}} = (\omega - \Delta) \Big/ \int_\mu^\infty d\epsilon \frac{\rho(\epsilon) | V_{\epsilon\omega} |^2}{\epsilon - \mu} \; . \qquad (18)$$

It is also clear that the position of the bound state, ϵ_b, shifts down as g^2 is increased and/or ω is decreased, provided $\omega \gtrsim \Delta$. We are thus led to the following interesting conclusions: As the intensity of the external field is increased, the latter has the effect of "pulling down" the energy level of the induced bound state, while, with a tunable laser, the same result can be achieved by lowering the field frequency. Hence, there exists the possibility of "fine-tuning" the energy of the laser-induced bound state by varying the field strength and frequency independently or simultaneously.

An explicit expression for the bound state can be given as follows:

$$|b_N\rangle=\beta_b\left[|d_{N+1}\rangle-g\int_\mu^\infty d\epsilon\frac{\rho(\epsilon)V^*_{\epsilon\omega}}{\epsilon-\epsilon_b}|\epsilon_N\rangle\right], \quad (19)$$

where

$$\beta_b=\left[1+g^2\int_\mu^\infty d\epsilon\frac{\rho(\epsilon)|V_{\epsilon\omega}|^2}{(\epsilon-\epsilon_b)^2}\right]^{-1/2} \quad (20)$$

and $\epsilon_b<\mu$. This result can be easily obtained from Eqs. (10) and (13) and the normalization requirement

$$\langle b_N|b_N\rangle=1\ . \quad (21)$$

Thus a system initially in the state $|d_{N+1}\rangle$ has a probability

$$P_{bd}=|\langle b_N|d_{N+1},t\rangle|^2=\beta_b^2 \quad (22)$$

of being in the tunable bound state $|b_N\rangle$ at later times. We also see that the probability of bound-state formation from a continuum state $|\epsilon_N\rangle$, $P_{b\epsilon}$, is given by

$$P_{b\epsilon}=|\langle b_N|\epsilon_N,t\rangle|^2=\frac{\beta_b^2g^2|V_{\epsilon\omega}|^2}{(\epsilon-\epsilon_b)^2}\ . \quad (23)$$

IV. THE SCATTERING AND METASTABLE STATES

To deduce the existence of metastable states, we look at the continuum-continuum transition specified by the scattering process $|\epsilon_N\rangle\to|\epsilon'_N\rangle$. For this purpose we introduce the scattering states $|E^\pm(N)\rangle$ which are exact continuum eigenstates of H:

$$H|E^\pm(N)\rangle=E|E^\pm(N)\rangle,\ \ \mu<E<\infty \quad (24)$$

$$|E^\pm(N)\rangle=|E_N\rangle+\sum_\epsilon\chi^\pm_{E\epsilon}|\epsilon_N\rangle+\beta^\pm_E|d_{N+1}\rangle\ , \quad (25)$$

where $|E^+(N)\rangle$ and $|E^-(N)\rangle$ stand for the outgoing and incoming states, respectively. The corresponding amplitudes $\chi^\pm_{E\epsilon}$ and $\beta^\pm_E$ can be determined unambiguously from Eq. (24):

$$\beta^\pm_E=\frac{gV_{E\omega}}{h(E\pm i\eta)}\ , \quad (26)$$

$$\chi^\pm_{E\epsilon}=\frac{g\beta^\pm_E V^*_{\epsilon\omega}}{E-\epsilon\pm i\eta}\ , \quad (27)$$

where

$$h(z)\equiv z-(\epsilon_d+\omega)+\phi(z)\ , \quad (28)$$

and $\phi(z)$, with z in general complex, is the Cauchy integral

$$\phi(z)\equiv g^2\int_\mu^\infty d\epsilon\frac{\rho(\epsilon)|V_{\epsilon\omega}|^2}{\epsilon-z} \quad (29)$$

[cf. Eq. (16)]. This function has a square-root branch structure with branch point at $\epsilon=\mu$ and a discontinuity of $2i\pi g^2\rho(\epsilon)|V_{\epsilon\omega}|^2$ across the branch cut (μ,∞). The double-valuedness differentiates between the outgoing and incoming scattering solutions. In Eq. (25), $|E_N\rangle$ represents the incident wave and $\sum_\epsilon\chi^\pm_{E\epsilon}|\epsilon_N\rangle$ the scattered wave. Given the completeness relation of Eq. (8), the following completeness relations hold for the scattering states:

$$\sum_E|E^\pm(N)\rangle\langle E^\pm(N)|=1\ ,\ \ g<g_{\text{crit}} \quad (30)$$

$$\sum_E|E^\pm(N)\rangle\langle E^\pm(N)|+|b_N\rangle\langle b_N|=1,\ \ g>g_{\text{crit}}\ . \quad (31)$$

This will be proved in the Appendix.

Using Eqs. (25)–(27), the S matrix for the scattering process $|\epsilon_N\rangle\to|\epsilon'_N\rangle$ can be written down directly:

$$\begin{aligned}S_{\epsilon'\epsilon}&=\delta_{\epsilon'\epsilon}-2\pi i\delta(\epsilon'-\epsilon)T_{\epsilon'\epsilon}\\&=\delta_{\epsilon'\epsilon}-2\pi i\delta(\epsilon'-\epsilon)\langle\epsilon'_N|H'|\epsilon^+_N\rangle\\&=\delta_{\epsilon'\epsilon}-2\pi i\delta(\epsilon'-\epsilon)\frac{g^2|V_{\epsilon\omega}|^2}{h(\epsilon+i\eta)}\\&=\delta_{\epsilon'\epsilon}\left[1-\frac{i\Gamma_0(\epsilon)}{\epsilon-(\epsilon_d+\omega)+g^2P(\epsilon)+i\Gamma_0(\epsilon)/2}\right],\end{aligned} \quad (32)$$

where

$$\Gamma_0(\epsilon)\equiv2\pi g^2\rho(\epsilon)|V_{\epsilon\omega}|^2 \quad (33)$$

and

$$P(\epsilon)\equiv\text{P}\int_\mu^\infty d\epsilon'\frac{\rho(\epsilon')|V_{\epsilon'\omega}|^2}{\epsilon'-\epsilon} \quad (34)$$

with P denoting the principal value of the integral. Thus

$$S_{\epsilon\epsilon}=e^{2i\delta(\epsilon)}\ , \quad (35)$$

where the scattering phase shift $\delta(\epsilon)$ is given by

$$\cot\delta(\epsilon)=-\frac{[\epsilon-(\epsilon_d+\omega)+g^2P(\epsilon)]}{\Gamma_0(\epsilon)/2}\ . \quad (36)$$

Since resonances occur at energies satisfying $\delta(\epsilon)=\pi/2$, the observed energies of the metastable states must satisfy approximately the equation[16]

$$\epsilon-(\epsilon_d+\omega)+g^2P(\epsilon)=0\ . \quad (37)$$

This equation is to be compared with Eq. (15) whose solution ($\epsilon=\epsilon_b<\mu$) for $g>g_{\text{crit}}$ is a pole of the S matrix $S_{\epsilon\epsilon}$ on the real axis and corresponds to a laser-induced bound state.

To study the roots of Eq. (37), we now choose a specific form for $\rho(\epsilon)|V_{\epsilon\omega}|^2$. The simplest choice for $\rho(\epsilon)$ is the free-electron energy density of states:

$$\rho(\epsilon)=\alpha\sqrt{\epsilon-\mu}\ , \quad (38)$$

where α is a constant. If

$$f(\epsilon)\equiv|V_{\epsilon\omega}|^2 \quad (39)$$

is then assumed to be an analytic function of ϵ which is real and single valued on the segment of the real axis (μ,∞), and has the property that

$$\rho(\epsilon)f(\epsilon)\to0\ \text{ as }\ |\epsilon|\to\infty\ ,$$

the principal-value integral in Eq. (34) can be evaluated readily by contour integration, without as yet specifying the form of $f(\epsilon)$. Thus,

$$P(E)\equiv \mathrm{P}\int_{\mu}^{\infty} d\epsilon \frac{\alpha\sqrt{\epsilon-\mu}f(\epsilon)}{\epsilon-E}$$
$$=\lim_{\lambda\to 0}\alpha\int_{\mu}^{\infty} d\epsilon \frac{\sqrt{\epsilon-\mu}f(\epsilon)(\epsilon-E)}{(\epsilon-E)^2+\lambda^2}$$
$$=\lim_{\lambda\to 0}\frac{\alpha}{2}\int_{C} d\epsilon \frac{(\epsilon-\mu)^{1/2}f(\epsilon)(\epsilon-E)}{(\epsilon-E)^2+\lambda^2},\quad E\ge\mu \qquad (40)$$

where the contour C is illustrated in Fig. 2. It follows from the residue theorem that

$$P(E)=\pi i\alpha\sum_{z_0}\frac{\mathrm{Res}[f(z_0)](z_0-\mu)^{1/2}}{z_0-E},\quad E\ge\mu \qquad (41)$$

where the sum is over the poles z_0 of $f(z)$, and $\mathrm{Res}[f(z_0)]$ denotes the residue of $f(z)$ at z_0. According to Schwarz's reflection principle, the poles of $f(z)$, if they exist at all, must exist in conjugate pairs. This guarantees that $P(E)$ as given by Eq. (41) is always real. We immediately note that $P(E)$ can be continued to the real values $E<\mu$ by

$$P(E)\equiv \int_{\mu}^{\infty} d\epsilon \frac{\alpha\sqrt{\epsilon-\mu}f(\epsilon)}{\epsilon-E}=\phi(E)/g^2,\quad E<\mu\ . \qquad (42)$$

This integral can also be obtained using the contour C and the residue theorem:

$$P(E)=\pi i\alpha\left[(E-\mu)^{1/2}f(E)+\sum_{z_0}\frac{\mathrm{Res}[f(z_0)](z_0-\mu)^{1/2}}{z_0-E}\right],\quad E<\mu\ . \qquad (43)$$

It is clear from Eqs. (41) and (43) that $P(E)$, as defined by Eqs. (40) and (42), is continuous at $E=\mu$.

To lend concreteness to our model, we now adopt a specific form for $f(\epsilon)$:[17]

$$f(\epsilon)=\frac{\gamma^4}{(\epsilon-\epsilon_0)^2+\xi^2}\ , \qquad (44)$$

where γ, ϵ_0, and ξ are real constants, γ has the dimensions of energy, and $\epsilon_0>\mu$ may be taken to be an ω-dependent quantity. This form of $f(\epsilon)$ describes qualitatively the general situation that $|V_{\epsilon\omega}|^2$ peaks at some value $\epsilon_0(\omega)$ and is only significant over a certain range ξ within the continuum. Using Eq. (44) for $f(\epsilon)$, Eqs. (41) and (43) immediately lead to

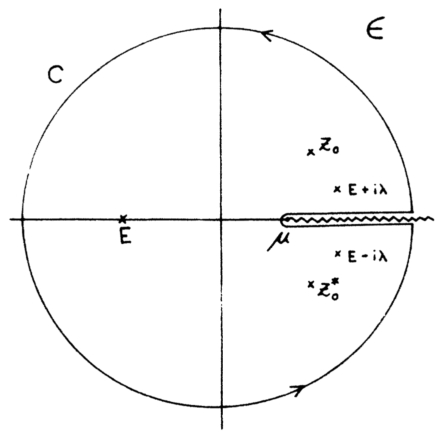


FIG. 2. Integration contour for $P(E)$ [Eq. (40)]. z_0^* and z_0 are poles of $f(\epsilon)$ [Eq. (39)].

$$P(E)=\begin{cases}P_>(E),\quad E\ge\mu & (45a)\\ P_>(E)+P_<(E),\quad E<\mu & (45b)\end{cases}$$

where

$$P_>(E)\equiv\frac{\beta\pi}{\xi}\frac{[(\epsilon_0-\mu)^2+\xi^2]^{1/4}[(\epsilon_0-E)\cos\theta/2+\xi\sin\theta/2]}{(\epsilon_0-E)^2+\xi^2}, \qquad (46)$$

$$P_<(E)\equiv\frac{-\beta\pi\sqrt{\mu-E}}{(E-\epsilon_0)^2+\xi^2} \qquad (47)$$

$$\beta\equiv\alpha\gamma^4 \qquad (48)$$

and

$$\theta\equiv\tan^{-1}[\xi/(\epsilon_0-\mu)],\quad 0<\theta<\pi/2\ . \qquad (49)$$

Using the forms in Eqs. (38) and (44) for $\rho(\epsilon)$ and $|V_{\epsilon\omega}|^2$ respectively, the integral

$$I\equiv g^2\int_{\mu}^{\infty} d\epsilon\frac{\rho(\epsilon)|V_{\epsilon\omega}|^2}{(\epsilon-\epsilon_b)^2}$$

in Eq. (20) can also be calculated using the contour C in Fig. 2 and the residue theorem:

$$I=\frac{g^2\beta\pi}{[(\epsilon_0-\epsilon_b)^2+\xi^2]}\left[\frac{1}{\sqrt{\mu-\epsilon_b}}\left[\frac{1}{2}-\frac{2(\mu-\epsilon_b)(\epsilon_0-\epsilon_b)}{(\epsilon_0-\epsilon_b)^2+\xi^2}\right]+\frac{[(\epsilon_0-\mu)^2+\xi^2]^{1/4}}{\xi[(\epsilon_0-\epsilon_b)^2+\xi^2]}\times\{\cos(\theta/2)[(\epsilon_0-\epsilon_b)^2-\xi^2]+2\xi(\epsilon_0-\epsilon_b)\sin(\theta/2)\}\right] \qquad (50)$$

with θ defined by Eq. (49). Finally, Eq. (44) also implies [cf Eq. (33)]

$$\Gamma_0(\epsilon)=\frac{2\pi g^2\beta\sqrt{\epsilon-\mu}}{(\epsilon-\epsilon_0)^2+\xi^2},\quad \epsilon\ge\mu\ . \qquad (51)$$

Figure 3 illustrates the graphical solution of Eq. (37) using the result of Eq. (45). The following parameters have been used: $\epsilon_d=1$; $\mu=2$; $\omega=5$; $\epsilon_0=\epsilon_d+\omega+0.5$; $\xi=1.5$; $g^2\beta=1$, 2, and 6. For $g^2\beta=1$ and 2, $g^2P(\mu)$

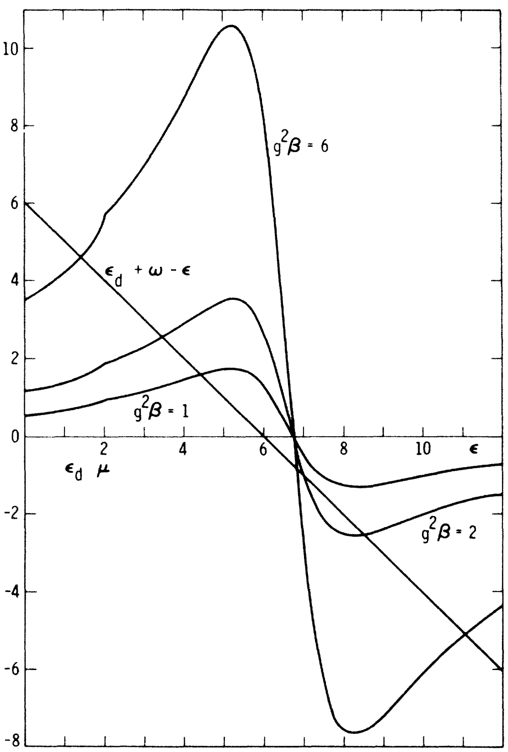


FIG. 3. Graphical solutions of Eq. (37). The curves are $g^2P(\epsilon)$ vs ϵ for three values of the coupling strength $g^2\beta$. $P(\epsilon)$ has been computed using Eqs. (45)–(47) and the parameters $\epsilon_d=1$, $\mu=2$, $\omega=5$, $\epsilon_0=\epsilon_d+\omega+0.5$, and $\xi=1.5$.

$=\phi(\mu)<\omega-\Delta$, and hence no bound state solutions ($E<\mu$) exist. For $g^2\beta=6$, however, $g^2P(\mu)>\omega-\Delta$, and a bound state exists at $\epsilon_b=1.4$. The critical coupling strength for the above parameters is $(g^2\beta)_{\rm crit}=4.21$ [Eq. (18)].

The most interesting result here is the possibility of multiple laser-induced metastable states. For $g^2\beta=1$ and 2, the smallest roots $>\mu$ have the property that their values decrease as g^2 is increased, or ω decreased, until, when Eq. (17) is satisfied, they become the bound-state roots of Eq. (15). These clearly correspond to metastable states. At this point it would be tempting to conclude that the other roots correspond to metastable states also. But whether this is so depends critically on the quantities $\Gamma_0(E_i)$ and $g^2(d/dE)P(E)\,|_{E=E_i}$ [from now on written as $g^2P'(E_i)$ where E_i is a root of Eq. (37)], as we shall see in the next section. All roots, however, are intimately related to both the field intensity and frequency. Hence, the energies of the metastable states can be fine tuned also, but they bear no simple relationship to $\epsilon_d+\omega$.

V. THE RENORMALIZED FIELD INTENSITY

In the usual perturbation treatment of resonance states, $P(\epsilon)$ and $\Gamma_0(\epsilon)$ are both regarded as small and weakly dependent on ϵ. In our model, there is then only one metastable state and it appears as an approximate pole, $\epsilon=\epsilon_0+\omega-g^2P-i\Gamma_0/2$, of the S matrix. The quantities $-g^2P$ and Γ_0 are interpreted as the shift and width of the state, respectively. The latter is simply proportional to g^2, or the physical field intensity.

This treatment becomes invalid when $P(\epsilon)$ and $\Gamma_0(\epsilon)$ vary significantly over a region ϵ of interest. Figure 3 illustrates such a case. As we have seen, the line centers of the metastable states are approximately given by solutions of Eq. (37) and bear no simple relationship to the "unperturbed" line center $\epsilon_d+\omega$. Moreover, the physical widths of these states have to be determined by a renormalization procedure.

Usually the object of this procedure is to "absorb" infinities which may appear in a field theory into redefined physical coupling constants in a consistent manner. In our model, an infinity may occur, for example, in the quantity $P(\epsilon)$. Due to the assumption following Eq. (39) on the analytical nature of $\Gamma_0(\epsilon)$, however, we have eliminated this possibility, although an identical procedure can be used to redefine the coupling constant g^2 so as to determine the physical widths.

The objective is to write the S matrix $S_{\epsilon\epsilon}$ [cf. Eq. (32)] in such a way that g^2 can be replaced by a renormalized coupling constant

$$g_R^2=Zg^2 \tag{52}$$

everywhere with Z denoting the renormalization constant (to be determined). We begin by making two "subtractions" at $\epsilon=E_i$, where E_i is a root of Eq. (37), on the quantity $1/(\epsilon'-\epsilon)$ appearing in $P(\epsilon)$ [Eq. (34)]:

$$\frac{1}{\epsilon'-\epsilon}=\left[\frac{1}{\epsilon'-\epsilon}-\frac{1}{\epsilon'-E_i}\right]+\frac{1}{\epsilon'-E_i}=\left[\frac{\epsilon-E_i}{(\epsilon'-\epsilon)(\epsilon'-E_i)}-\frac{\epsilon-E_i}{(\epsilon'-E_i)^2}\right]+\frac{\epsilon-E_i}{(\epsilon'-E_i)^2}+\frac{1}{\epsilon'-E_i}$$

$$=\frac{(\epsilon-E_i)^2}{(\epsilon'-E_i)^2(\epsilon'-\epsilon)}+\frac{\epsilon-E_i}{(\epsilon'-E_i)^2}+\frac{1}{\epsilon'-E_i}\ . \tag{53}$$

Then

$$\phi(\epsilon+i\eta)=(\epsilon-E_i)^2g^2A(\epsilon,E_i)+(\epsilon-E_i)g^2P'(E_i)+g^2P(E_i) \tag{54}$$

where

$$A(\epsilon,E_i)\equiv \mathrm{P}\int_\mu^\infty d\epsilon'\frac{\rho(\epsilon')\,|\,V_{\epsilon'\omega}\,|^2}{(\epsilon'-E_i)^2(\epsilon'-\epsilon)}\ . \tag{55}$$

Next, we use the fact

$$E_i-(\epsilon_d+\omega)+g^2P(E_i)=0 \tag{56}$$

to obtain

$$h(\epsilon+i\eta)=(\epsilon-E_i)[1+g^2P'(E_i)]+(\epsilon-E_i)^2g^2A+i\Gamma_0(\epsilon)/2 \tag{57}$$

$$=\frac{(\epsilon-E_i)}{Z_i}+(\epsilon-E_i)^2g^2A+i\Gamma_0(\epsilon)/2\ , \tag{58}$$

where

$$1/Z_i\equiv 1+g^2P(E_i) \tag{59}$$

defines the renormalization constant for the ith root of Eq. (37). Finally, Eq. (32) implies

$$S_{\epsilon\epsilon}=1-\frac{i2\pi Z_i g^2\rho(\epsilon)\,|\,V_{\epsilon\omega}\,|^2}{(\epsilon-E_i)+(\epsilon-E_i)^2Z_ig^2A+i\pi Z_ig^2\rho(\epsilon)\,|\,V_{\epsilon\omega}\,|^2}\ . \tag{60}$$

In this equation all factors of the "bare" coupling constant g^2 reappear as the renormalized coupling constant

$$g_{Ri}^2(g^2,\omega)\equiv Z_ig^2=g^2/[1+g^2P'(E_i(\omega,g^2))]\ , \tag{61}$$

where P' is considered a function of ω and g^2 through its dependence on E_i. Thus the renormalization procedure has been accomplished. In the neighborhood of $\epsilon=E_i$, we can drop the second-order factor $(\epsilon-E_i)^2Z_ig^2A$ and replace $\Gamma_0(\epsilon)$ by $\Gamma_0(E_i)$. The quantity

$$\Gamma_i(E_i)=\Gamma_0(E_i)/[1+g^2P'(E_i)] \tag{62}$$

can then be interpreted as the physical width of the metastable state at E_i, provided

$$g^2P'(E_i)>-1\ . \tag{63}$$

In addition, g_{Ri}^2 can be interpreted as a renormalized field intensity, which depends not only on the physical field intensity, but also on the field frequency.

It is interesting to note that, provided $g^2P'(E_i)>0$, the renormalized field intensity is always less than the physical intensity. Thus values of ϵ for which $P'(\epsilon)>0$ may be considered as the *metastable regime*: If an unstable state has energy E_i within this regime, its physical width is always smaller than its bare width. In this sense, a laser, in addition to creating a metastable state, may actually enhance the stability of that state. This may be observed through line narrowing in the spectrum as g^2P' is increased. For $-1<g^2P'<0$, Γ_i is still positive but $g_R^2>g^2$. The range of ϵ satisfying this inequality may be called the *unstable regime,* as Γ_i can become extremely large when $g^2P'\gtrsim -1$. In this regime a laser destabilizes an unstable state that it creates, thus causing line broadening as $g^2P'\to -1$. Finally for $g^2P'<-1$, the roots of Eq. (37) do not correspond to any physically observed states at all, since Γ_i is negative. Hence we may label the range of ϵ satisfying this condition the *unphysical regime.* With respect to Fig. 3, for example, the first roots $>\mu$ are in the metastable regime.

VI. DISSOCIATION AND RECOMBINATION

Laser-induced dissociation in our model is the process $|\,d_{N+1}\rangle\to|\,\epsilon_N\rangle$, accompanied by the absorption of a photon. The reverse process is that of recombination, $|\,\epsilon_N\rangle\to|\,d_{N+1}\rangle$, accompanied by stimulated emission of a photon. By the principle of microreversibility, the probabilities for these processes are identical. The second process is to be distinguished from the related process $|\,\epsilon_N\rangle\to|\,b_N\rangle$, which is also recombination but does not involve the emission or absorption of photons, and for which the final state $|\,b_N\rangle$ only exists when the laser is kept on. The probability for this last process has been given by Eq. (23).

The metastable states play a crucial role in dissociation (and recombination), and their properties for field-free processes, such as electron-molecule scattering, have been dealt with exhaustively using the analytic properties of the associated S matrices.[18] In perturbation theory, $|\,d_{N+1}\rangle$ goes over into the metastable state with the approximate energy $\epsilon_m=\epsilon_d+\omega-g^2P$, and we can think of this state as the one which decays into $|\,\epsilon_N\rangle$, with a lifetime $1/\Gamma_0$. Hence $P_{\epsilon d}$, the dissociation probability, is expected to peak only at $\epsilon=\epsilon_m$. As seen in the last section, this treatment fails when renormalization becomes important. Multiple metastable states may arise whose energies are quite distinct from ϵ_m. Furthermore, the lifetimes of these states may also differ significantly from $1/\Gamma_0$. In this case, the dissociation profile of $P_{\epsilon d}$ would be expected to exhibit multiple peaks with quite different strengths and widths. When $g>g_{\rm crit}$, a bound state also appears which may or may not be accompanied by metastable states. This situation further complicates the structure of $P_{\epsilon d}$. When there are no metastable states, one would only have a nonresonant background due to both the bound and scattering states. When the bound state exists together with one or more metastable states, resonance peaks superimposed on the nonresonant background would result. All these features of $P_{\epsilon d}$ make it a most sensitive probe into the effects of a laser on a bound-continuum system. Since the question of whether the bound state or metastable states exist depends on the field intensity and frequency, another way of viewing the situation is that by varying the laser characteristics one can induce vastly different kinds of dissociation spectra.

We shall now derive an expression for

$$P_{\epsilon d}(t)=|\,\langle\epsilon_N\,|\,d_{N+1}(t)\rangle\,|^2\ , \tag{64}$$

where $|d_{N+1}(t)\rangle$ denotes the time-evolved state which at $t=0$ is given by $|d_{N+1}\rangle$. This is most easily done by expanding $|d_{N+1}\rangle$ in items of the complete set of eigenstates of H, $\{|E^-(N)\rangle, |b_N\rangle\}$ [cf. Eq. (31)]. In doing so, we assume the general case where a bound state is present. The reason for using the incoming states will become apparent below. Using Eqs. (19), (25)–(27), and (31), we have

$$|d_{N+1}(t)\rangle = \sum_E e^{-iEt}|E^-(N)\rangle\langle E^-(N)|d_{N+1}\rangle + e^{i\epsilon_b t}|b_N\rangle\langle b_N|d_{N+1}\rangle$$
$$= \sum_E (\beta_E^-)^* e^{-iEt}|E^-(N)\rangle + \beta_b e^{-i\epsilon_b t}|b_N\rangle \,, \tag{65}$$

where β_E^- and β_b are given by Eqs. (26) and (20), respectively. Expanding $|E^-(N)\rangle$ and $|b_N\rangle$ again in terms of $\{|d_{N+1}\rangle, |\epsilon_N\rangle\}$ using Eqs. (25) and (19), Eq. (65) yields

$$\langle \epsilon_N | d_{N+1}(t)\rangle = (\beta_\epsilon^-)^* e^{-i\epsilon t} - \frac{g\beta_b^2 V_{\epsilon\omega}^* e^{-i\epsilon_b t}}{\epsilon-\epsilon_b} - gV_{\epsilon\omega}^* \sum_E \frac{e^{-iEt}|\beta_E^-|^2}{E-\epsilon-i\eta} \,. \tag{66}$$

The last term represents the overlap between the scattered-wave part of $|E^-(N)\rangle$ and $|\epsilon_N\rangle$. It thus vanishes as $t\to\infty$, since in the distant future the scattered part of an incoming scattering state vanishes. The mathematical statement of this fact is

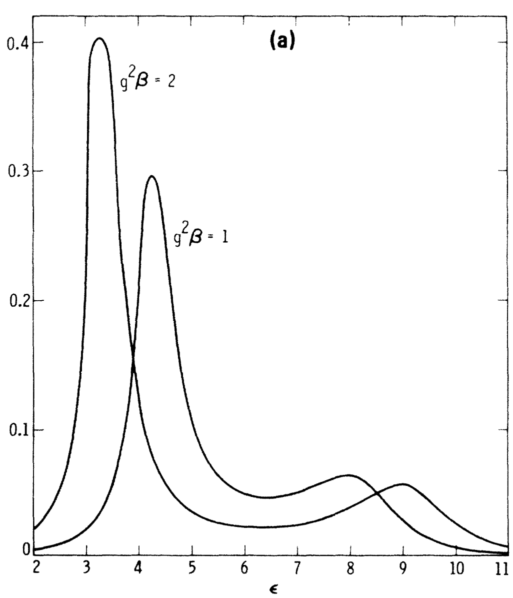
$$\lim_{t\to\infty} \frac{e^{i\omega t}}{\omega+i\eta} = 0 \,. \tag{67}$$

This equation also implies that the interference term due to the remaining two terms vanish, since ϵ is greater than μ and never equals ϵ_b. Thus we finally have

$$P_{\epsilon d} = g^2|V_{\epsilon\omega}|^2 \left\{ \frac{\beta_b^4}{(\epsilon-\epsilon_b)^2} + \frac{1}{[\epsilon-(\epsilon_d+\omega)+g^2P(\epsilon)]^2+\Gamma_0^2(\epsilon)/4} \right\} \,. \tag{68}$$

For the specific form of the $V_{\epsilon\omega}$ in Eq. (44), the integral I in Eq. (20) for β_b is given by Eq. (50). The first term in Eq. (68) represents the effects of the bound state and vanishes if $g<g_{\text{crit}}$. It contributes only to the nonresonant background since ϵ is never equal to ϵ_b and is most prominent near the dissociation threshold $\epsilon=\mu$. The second term is studied in detail in the last section. When metastable states exist it leads to resonance peaks. In the absence of metastable states it also contributes to the nonresonant background.

Figures 4(a) and 4(b) illustrate the profile $P_{\epsilon d}$ for the form of $V_{\epsilon\omega}$ given by Eq. (44) and the parameters used in Fig. 3. The narrow resonance peaks in Fig. 4(a) ($\epsilon\sim3.3$ and 4.3) are quite closely approximated by the solutions of Eq. (37) in the metastable regime (cf. Fig. 3) and stand in contrast to the broad ones in the unstable regime ($\epsilon\sim8$ and 9). It should be noted that in the latter regime, the resonance peaks are much less well approximated by the solutions to Eq. (37). In fact, for the case $g^2\beta=1$, no real



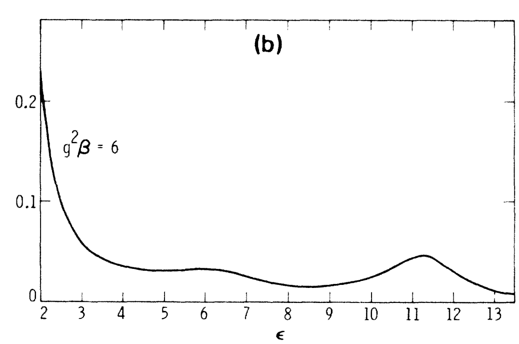


FIG. 4. The bound continuum profiles $\alpha P_{\epsilon d}$ [Eq. (68), α defined in Eq. (38)] for the coupling strengths $g^2\beta=1,2$ (a) and $g^2\beta=6$ (b), corresponding to the $g^2P(\epsilon)$ in Fig. 3.

solution exists at all in this regime, and the broad resonance occurs near the minimum of $\epsilon-(\epsilon_d+\omega)+g^2P(\epsilon)$ rather than at its zero (cf. Fig. 3). We also note the slight line narrowing in the metastable regime as the coupling constant $g^2\beta$ is increased.

Figure 4(b) illustrates clearly the threshold effect (for $\epsilon \gtrsim \mu$) due to the laser-induced bound state at $\epsilon_d=1.4$. This effect is totally absent in Fig. 4(a), which is for cases where no laser-induced bound state exists. Also, no narrow resonance peaks are present in the metastable regime, corresponding to the absence of a solution here to Eq. (37) (cf. Fig. 3). The broad resonance (at $\epsilon \sim 11.3$), however, persists in the unstable regime, along with the nonresonant background due to the first term in Eq. (68). This corresponds to the solution of Eq. (37) at $\epsilon \sim 11$ (Fig. 3). The other solution at $\epsilon \sim 6.9$ clearly lies in the unphysical regime ($g^2P'<-1$) and does not manifest itself as any resonance peak.

VII. FANO STATES

The laser-induced radiative coupling furnishes the equivalent of a "configuration interaction" between the discrete state $|d_{N+1}\rangle$ and the continuum states $|\epsilon_N\rangle$. As we have seen in the previous sections, this interaction is responsible for the formation of scattering states, a bound state when $g>g_{\text{crit}}$, and metastable states when suitable conditions on g^2 and ω are fulfilled. Under certain boundary conditions, stationary-wave eigenstates of H, as opposed to scattering states, are also important. These many manifest themselves as asymmetric peaks in absorption spectra, and in atomic and molecular physics they occur most commonly as autoionizing levels.[1] We shall now demonstrate that these so-called Fano states, $|E_F(N)\rangle$, are related to the scattering states introduced in Eq. (25) by

$$|E_F(N)\rangle=\frac{i}{2\sin\delta(E)}[\,|E^+(N)\rangle-|E^-(N)\rangle\,]\,, \tag{69}$$

where $\delta(E)$ is the scattering phase shift of Eq. (35).

We first note that, due to Eq. (24), $|E_F(N)\rangle$ is an eigenstate of H; and Eq. (69) implies that it is also a standing-wave state. Both $|E^+(N)\rangle$ and $|E^-(N)\rangle$ can be expanded in terms of $|d_{N+1}\rangle$ and $|\epsilon_N\rangle$ using Eq. (25). Thus, from Eqs. (26) and (27)

$$|E^+(N)\rangle-|E^-(N)\rangle=gV_{E\omega}\left[\left[\frac{1}{h(E+i\eta)}-\frac{1}{h(E-i\eta)}\right]|d_{N+1}\rangle +g\int_\mu^\infty d\epsilon\,\rho(\epsilon)V^*_{\epsilon\omega}\left[\frac{1}{h(E+i\eta)(E-\epsilon+i\eta)}-\frac{1}{h(E-i\eta)(E-\epsilon-i\eta)}\right]|\epsilon_n\rangle\right]. \tag{70}$$

Now it follows directly from Eqs. (28) and (36) that

$$\begin{aligned}\frac{1}{h(E+i\eta)}-\frac{1}{h(E-i\eta)}&=\frac{2i[-\Gamma_0(E)/2]}{[E-(\epsilon_d+\omega)+g^2P(E)]^2+\Gamma_0^2(E)/4}\\&=\frac{2i\sin\delta(E)}{\{[E-(\epsilon_d+\omega)+g^2P(E)]^2+\Gamma_0^2(E)/4\}^{1/2}}\end{aligned} \tag{71}$$

and

$$\begin{aligned}\frac{1}{h(E+i\eta)(E-\epsilon+i\eta)}-\frac{1}{h(E-i\eta)(E-\epsilon-i\eta)}&=\frac{-2i}{\{[E-(\epsilon_d+\omega)+g^2P(E)]^2+\Gamma_0^2(E)/4\}^{1/2}}\\&\quad\times\left[\sin\delta(E)\mathrm{P}\left[\frac{1}{\epsilon-E}\right]+\pi\cos\delta(E)\delta(E-\epsilon)\right],\end{aligned} \tag{72}$$

where $\delta(E-\epsilon)$ is the Dirac delta function. These equations substituted in Eq. (70) lead to the following form for $|E_F(N)\rangle$:

$$|E_F(N)\rangle=-\cos\delta(E)|E_N\rangle+\frac{gV_{E\omega}}{\Gamma_0(E)/2}\sin\delta(E)\left[|d_{N+1}\rangle+\mathrm{P}\int_\mu^\infty d\epsilon\frac{\rho(\epsilon)V^*_{\epsilon\omega}}{E-\epsilon}|\epsilon_N\rangle\right], \tag{73}$$

which is exactly the form given in Eq. (19) of Fano's paper.[1] The term in large parentheses has been called, by Fano, the modified discrete state. The orthonormality property of $|E_F(N)\rangle$ is also easily verified:

$$\begin{aligned}\langle E'_F(N)|E_F(N)\rangle&=\frac{1}{4\sin^2\delta(E)}[2\delta(E'-E)-\langle E'^+(N)|E^-(N)\rangle-\langle E'^-(N)|E^+(N)\rangle]\\&=\frac{\delta(E'-E)}{4\sin^2\delta(E)}(2-e^{-2i\delta(E)}-e^{2i\delta(E)})\\&=\delta(E'-E)\,.\end{aligned} \tag{74}$$

The second equality follows from the definition of the S matrix:

$$S(E)=\langle E^{-}(N)\,|\,E^{+}(N)\rangle \ . \tag{75}$$

VIII. SUMMARY AND DISCUSSION

Throughout the present work, the emphasis has been on the controllable nature of the radiative bound-continuum coupling and hence of the spectral structure. By varying both the frequency and the intensity of the laser, an unusual range of laser-induced effects can be achieved. The frequency mainly determines the position of the unperturbed discrete state while the intensity determines the coupling strength. First, we have seen that a point eigenvalue of the total Hamiltonian corresponding to a true bound state appears when the intensity is raised beyond a certain frequency-dependent critical value. Moreover, the energy of this bound state can be "tuned" by increasing the intensity further. Not only is the appearance of a new bound state of interest in itself, its presence also affects the nonresonant contribution to the dissociation spectrum, especially near the dissociation threshold. Through an examination of the continuum eigenstates (scattering states) and the S matrix, the structure of the metastable states is then revealed. Rather unexpectedly, it is found that even a monochromatic laser may induce more than one metastable state, with distinct energies not directly related to the energy of the unperturbed discrete state and the laser frequency. Moreover, the widths of the metastable states are found to be proportional to a renormalized intensity which may be larger or smaller than the physical field intensity. This interesting fact makes possible the broadening or narrowing, as well as a wide choice of the centers of the resonance peaks in the laser-induced dissociation spectrum as the laser characteristics are varied. The same conclusion also applies to the profiles generated by the Fano states, which are shown to be just coherent superpositions of the outgoing and incoming scattering states.

While the results derived in this paper are quite general, we must stress the specific limitations of our model. First, the only laser characteristics considered are the field intensity and frequency. Other factors such as pulse duration, bandwidth, polarization and coherence properties have been ignored. The rotating-wave approximation has also been used to delimit the size of the Hilbert space required for the treatment of radiative couplings. Finally, the field-free spectrum is assumed to be such that no field-free bound-continuum coupling is present. We expect that removal of one or more of the above restrictions would lead to very interesting physics in individual circumstances, but would generally leave the basic conclusions reached here substantially unchanged.

One interesting direction for futher investigations is the adaptation of the present model to the semiclassical treatment of collision problems involving atoms or ions. Examples are Penning or associative ionization[19] and atom and/or ion surface scattering.[20] The nuclear dynamics can be described by including explicit time dependences on the bound-continuum coupling $V_{\epsilon\omega}$ and the energy levels ϵ_d and μ.[21] This dynamics, of significance only within the duration of a collision period, has to be incorporated into the time development of the laser-induced bound, metastable or Fano states, and it is consequently expected to yield interesting structure in the wing regions of the dissociation or recombination profiles.

APPENDIX: PROOF OF THE COMPLETENESS RELATIONS (30) AND (31)

The proof given below follows closely that presented by Glaser and Källén[12] for a similar relation in the Lee model. We include it here for two reasons: first, for ease of reference and completeness; and second, to highlight the usefulness of the function-theoretic method in the demonstration of completeness relations. We begin by calculating the quantity on the left-hand side of Eq. (30). Making use of the expansion Eq. (25), we have

$$\begin{aligned}
\sum_E |E^{\pm}(N)\rangle\langle E^{\pm}(N)| = \sum_E \Bigg[& |E_N\rangle\langle E_N| + |d_{N+1}\rangle\langle d_{N+1}|\,|\beta_E^{\pm}|^2 + \sum_{\epsilon}\sum_{\epsilon'} (\chi_{E\epsilon'}^{\pm})^*\chi_{E\epsilon}^{\pm}\,|\epsilon_N\rangle\langle \epsilon'_N| \\
& + \beta_E^{\pm}\,|d_{N+1}\rangle\langle E_N| + (\beta_E^{\pm})^*\,|E_N\rangle\langle d_{N+1}| \\
& + \beta_E^{\pm}\sum_{\epsilon}(\chi_{E\epsilon}^{\pm})^*\,|d_{N+1}\rangle\langle\epsilon_N| + (\beta_E^{\pm})^*\sum_{E}\chi_{E\epsilon}^{\pm}\,|\epsilon_N\rangle\langle d_{N+1}| \\
& + \sum_{\epsilon}[\chi_{E\epsilon}^{\pm}\,|\epsilon_N\rangle\langle E_N| + (\chi_{E\epsilon}^{\pm})^*\,|E_N\rangle\langle\epsilon_N|\,] \Bigg] \\
= {} & |d_{N+1}\rangle\langle d_{N+1}| \sum_E |\beta_E^{\pm}|^2 + \sum_E |E_N\rangle\langle E_N| + |d_{N+1}\rangle \sum_E \left[\beta_E^{\pm} + \sum_{\epsilon}\beta_{\epsilon}^{\pm}(\chi_{\epsilon E}^{\pm})^*\right]\langle E_N| \\
& + \sum_E |E_N\rangle \left[(\beta_E^{\pm})^* + \sum_{\epsilon}(\beta_{\epsilon}^{\pm})^*\chi_{\epsilon E}^{\pm}\right]\langle d_{N+1}| \\
& + \sum_E\sum_{\epsilon} |E_N\rangle \left[\chi_{\epsilon E}^{\pm} + (\chi_{E\epsilon}^{\pm})^* + \sum_{\epsilon'}(\chi_{\epsilon'\epsilon}^{\pm})^*\chi_{\epsilon' E}^{\pm}\right]\langle\epsilon_N| \ .
\end{aligned} \tag{A1}$$

The sums may be converted to integrals by taking the continuum limit. Thus, using Eqs. (26), (28), (29), (33), and (34), we can write

$$\sum_E |\beta_E^{\pm}|^2 = g^2 \int_\mu^\infty dE \frac{\rho(E)\,|V_{E\omega}|^2}{|E-(\epsilon_d+\omega)+g^2P(E)\pm i\Gamma_0(E)/2|^2} = \frac{1}{\pi}\int_\mu^\infty dE \frac{\Gamma_0(E)/2}{[E-(\epsilon_d+\omega)+g^2P(E)]^2+\Gamma_0^2(E)/4}$$
$$= \frac{1}{\pi}\,\mathrm{Im}\int_\mu^\infty dE \frac{1}{h(E-i\eta)} = -\frac{1}{2\pi i}\int_{C'} dE \frac{1}{h(E)} , \tag{A2}$$

where C' is the contour shown in Fig. 2 minus the large circle. The last integral can easily be evaluated by considering the contour C itself (Fig. 2) and using the residue theorem. We have, evidently,

$$\int_{C'} dE \frac{1}{h(E)} = \int_C dE \frac{1}{h(E)} - \int_{C_R} dE \frac{1}{h(E)} , \tag{A3}$$

where C_R denotes the large circle with the radius $R \to \infty$. Since from Eqs. (28) and (29)

$$\lim_{|E|\to\infty} h(E) = E , \tag{A4}$$

it follows that

$$\int_{C_R} dE \frac{1}{h(E)} = 2\pi i . \tag{A5}$$

Now from the results of Sec. III, we see that when $g > g_{\text{crit}}$ there is a first-order zero of $h(E)$ at $E_b = \epsilon_b < \mu$ corresponding to the laser-induced bound state. On the other hand, it is readily seen [by looking at the imaginary part of $h(E)$ for complex E] that there can be no complex roots of $h(E)$ on the first sheet [see the remark following Eq. (29)]. These facts and the residue theorem, together with Eqs. (A2), (A3), and (A5), imply that

$$\sum_E |\beta_E^{\pm}|^2 = \begin{cases} 1, & g < g_{\text{crit}} \\ 1 - \dfrac{1}{h'(\epsilon_b)}, & g > g_{\text{crit}} \end{cases} \tag{A6}$$

where $h'(\epsilon_b)$ denotes the derivative of $h(E)$ evaluated at ϵ_b.

We now consider the terms in Eq. (A1) involving $\chi_{\epsilon E}^{\pm}$. Using Eqs. (26) and (27), we have, recalling the derivation of Eq. (A2),

$$\sum_\epsilon \beta_\epsilon^{\pm}(\chi_{\epsilon E}^{\pm})^* = g\int_\mu^\infty d\epsilon \frac{\rho(\epsilon)\,|\beta_\epsilon^{\pm}|^2 V_{E\omega}}{\epsilon - E \mp i\eta} = \frac{gV_{E\omega}}{\pi}\int_\mu^\infty d\epsilon \left[\mathrm{Im}\frac{1}{h(\epsilon - i\eta)}\right]\frac{1}{\epsilon - E \mp i\eta}$$
$$= -\frac{gV_{E\omega}}{2\pi i}\int_{C'} d\epsilon \frac{1}{h(\epsilon)(\epsilon - E \mp i\eta)}$$
$$= -\frac{gV_{E\omega}}{2\pi i}\int_{C} d\epsilon \frac{1}{h(\epsilon)(\epsilon - E \mp i\eta)}$$
$$= \begin{cases} -\dfrac{gV_{E\omega}}{h(E\pm i\eta)}, & g < g_{\text{crit}} \\ -\dfrac{gV_{E\omega}}{h(E\pm i\eta)} + \dfrac{gV_{E\omega}}{h'(\epsilon_b)(E-\epsilon_b)}, & g > g_{\text{crit}} . \end{cases} \tag{A7}$$

The last equality has again been obtained by the residue theorem, and we note that $E \geq \mu$. Recalling the definition of $\beta_E^{\pm}$ given by Eq. (26), Eq. (A7) yields

$$\beta_E^{\pm} + \sum_\epsilon \beta_\epsilon^{\pm}(\chi_{\epsilon E}^{\pm})^* = \begin{cases} 0, & g < g_{\text{crit}} \\ \dfrac{gV_{E\omega}}{h'(\epsilon_b)(E-\epsilon_b)}, & g > g_{\text{crit}} . \end{cases} \tag{A8}$$

Note that the $(\pm)$ cases lead to the same result.

Next we calculate the last term in Eq. (A1). Proceeding as in Eq. (A7) and using the residue theorem yet another time, we have

$$\sum_{\epsilon'}(\chi^{\pm}_{\epsilon'\epsilon})^*\chi^{\pm}_{\epsilon' E}=\frac{g^2V_{\epsilon\omega}V^*_{E\omega}}{\pi}\int_\mu^\infty d\epsilon'\left[\mathrm{Im}\frac{1}{h(\epsilon'-i\eta)}\right]\frac{1}{(\epsilon'-\epsilon\mp i\eta)(\epsilon'-E\pm i\eta)}$$
$$=-\frac{g^2V_{\epsilon\omega}V^*_{E\omega}}{2\pi i}\int_C dz\frac{1}{h(z)(z-\epsilon\mp i\eta)(z-E\pm i\eta)}$$
$$=\begin{cases}-g^2V_{\epsilon\omega}V^*_{E\omega}H^{\pm}(\epsilon,E), & g<g_{\mathrm{crit}}\\ -g^2V_{\epsilon\omega}V^*_{E\omega}\left[H^{\pm}(\epsilon,E)+\dfrac{1}{(\epsilon-\epsilon_b)(E-\epsilon_b)h'(\epsilon_b)}\right], & g>g_{\mathrm{crit}}\end{cases} \tag{A9}$$

where

$$H^{\pm}(\epsilon,E)\equiv\frac{1}{h(\epsilon\pm i\eta)(\epsilon-E\pm i\eta)}+\frac{1}{h(E\mp i\eta)(E-\epsilon\mp i\eta)}\ . \tag{A10}$$

Thus, recalling the definition of $\chi^{\pm}_{E\epsilon}$ [Eq. (27)], we have

$$\chi^{\pm}_{\epsilon E}+(\chi^{\pm}_{E\epsilon})^*+\sum_{\epsilon'}(\chi^{\pm}_{\epsilon'\epsilon})^*\chi^{\pm}_{\epsilon' E}=\begin{cases}0, & g<g_{\mathrm{crit}}\\ -\dfrac{g^2V_{\epsilon\omega}V^*_{E\omega}}{h'(\epsilon_b)(\epsilon-\epsilon_b)(E-\epsilon_b)}, & g>g_{\mathrm{crit}}\ .\end{cases} \tag{A11}$$

Combining Eqs. (A6), (A8), and (A10) in Eq. (A1), we arrive at the result

$$\sum_E|E^{\pm}(N)\rangle\langle E^{\pm}(N)|=|d_{N+1}\rangle\langle d_{N+1}|+\sum_E|E_N\rangle\langle E_N|$$
$$=1,\quad g<g_{\mathrm{crit}} \tag{A12}$$

provided Eq. (8) holds. This is the result of Eq. (30). For the case $g>g_{\mathrm{crit}}$, we have

$$\sum_E|E^{\pm}(N)\rangle\langle E^{\pm}(N)|=\sum_E|E_N\rangle\langle E_N|+\left[1-\frac{1}{h'(\epsilon_b)}\right]|d_{N+1}\rangle\langle d_{N+1}|+|d_{N+1}\rangle\left[\sum_E\frac{gV_{E\omega}}{h'(\epsilon_b)(E-\epsilon_b)}\right]\langle E_N|$$
$$+\sum_E\left[|E_N\rangle\frac{gV^*_{E\omega}}{h'(\epsilon_b)(E-\epsilon_b)}\right]\langle d_{N+1}|-\sum_E\sum_\epsilon|E_N\rangle\frac{g^2V_{\epsilon\omega}V^*_{E\omega}}{h'(\epsilon_b)(\epsilon-\epsilon_b)(E-\epsilon_b)}\langle\epsilon_N|,\quad g>g_{\mathrm{crit}}\ . \tag{A13}$$

Now, from Eqs. (28) and (29), β_b as defined by Eq. (20) is actually given by

$$\beta_b=[h'(\epsilon_b)]^{-1/2}\ . \tag{A14}$$

Hence, Eq. (19) for the bound state can be written, on reverting from the integral to the sum,

$$|b_N\rangle=\frac{1}{[h'(\epsilon_b)]^{1/2}}\left[|d_{N+1}\rangle-\sum_E\frac{gV^*_{E\omega}}{E-\epsilon_b}|E_N\rangle\right]\ . \tag{A15}$$

This immediately yields

$$|b_N\rangle\langle b_N|=\frac{1}{h'(\epsilon_b)}\left[|d_{N+1}\rangle\langle d_{N+1}|-|d_{N+1}\rangle\sum_E\frac{gV_{E\omega}}{E-\epsilon_b}\langle E_N|-\sum_E|E_N\rangle\frac{gV^*_{E\omega}}{E-\epsilon_b}\langle d_{N+1}|\right.$$
$$\left.+\sum_E\sum_\epsilon|E_N\rangle\frac{g^2V_{\epsilon\omega}V^*_{E\omega}}{(\epsilon-\epsilon_b)(E-\epsilon_b)}\langle\epsilon_N|\right]\ . \tag{A16}$$

On adding this equation to Eq. (A13), we obtain

$$\sum_E|E^{\pm}(N)\rangle\langle E^{\pm}(N)|+|b_N\rangle\langle b_N|=\sum_E|E_N\rangle\langle E_N|+|d_{N+1}\rangle\langle d_{N+1}|$$
$$=1,\quad g>g_{\mathrm{crit}}\ , \tag{A17}$$

provided Eq. (8) holds. This is the result of Eq. (31).

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