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Autoionizing states in strong laser fields

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A formalism is developed for the study of autoionizing states under the influence of strong lasers. It is cast in both a semiclassical as well as a fully quantum-mechanical form. A set of integrodifferential equations is derived and solutions are obtained for certain special cases. It is demonstrated that the strong field causes significant distortion of the line profile and that new effects appear in double optical resonance when autoionizing states are involved.

I. INTRODUCTION

For our purposes in this paper, an autoionizing resonance[1] is to be understood as a bound state involving the excitation of two electrons whose total excitation energy is above at least the first ionization threshold. As such it is bound only in some approximation that neglects its interaction with the continuum in which it is embedded. With this interaction included, the state becomes unstable against ionization with one electron being ejected. Since this is not the place for a comprehensive cataloguing of the literature, we simply cite some of the original experimental work of Garton and Codling,[2] Madden and Codling,[3] and the more recent work of Brown *et al.*[4] It is the observation of autoionizing resonances in photoabsorption that is relevant to this paper because our aim is the description of their behavior under strong light.

With the availability of strong tunable lasers, the first generalization of this work was undertaken by Armstrong, Wynne, and collaborators who in a series of papers[5,6] reported the observation of autoionizing resonances through two-photon excitation as well as the excitation of highly excited states through three-photon absorption in alkaline earth atoms. The major difference from the single-photon excitation is that the parity of a two-photon-excited autoionizing resonance is the same as that of the ground state owing to the well-known selection rule for two-photon absorption. Obviously these are autoionizing resonances that can not be reached via single-photon absorption. In a series of different experiments Gallagher, Cooke, and collaborators[7,8] have used two lasers to reach autoionizing states of Sr and Ba. Typically, they use two lasers in a two-photon excitation of a Sr Rydberg state as, for example, in the transition $5s^2 \rightarrow 5snd$. A third laser is then used to cause the transition $5snd \rightarrow 5p_jnd$ which leads to autoionization since the state $5p_jnd$ is above the continuum threshold.

Autoionizing resonances can also be excited by electron collision. In usual experiments of this type, the observation was limited to the decay of the resonance. But the intensity and resolution offered by lasers has made feasible a new type of experiment reported by Langendam *et al.*[9] It is the photoexcitation of an autoionizing resonance from a lower one which was created by electron attachment. In that experiment, by crossing an electron beam with a laser in the presence of Ne atoms, they formed a scattering resonance of Ne^- which by absorbing a laser photon was then excited to a higher resonance of Ne^-. Both of these resonances are not single states but belong to two distinct groups of states whose gross features had been suspected from earlier experiments in electron scattering. The higher resolution offered by the laser made possible the separate excitation of several pairs of transitions from one group to the other. Even though the laser intensity was by multiphoton standards low (about 10^6 W/cm^2), it was sufficient to excite by single-photon absorption a transition between two very short-lived states of the negative ion. So far this seems to be the only experiment in which two autoionizing states were connected via a photon transition. It is likely, however, that such transitions have played a role (perhaps an important role) in recent multiphoton ionization experiments[10] on alkaline earth atoms with lasers of much higher intensity. Although it is not clear yet what the detailed interpretation of these experiments will be, one of the central questions is: What happens to autoionizing resonances under strong laser fields?

From experience with bound-bound transitions, we already know that new effects appear when the strength of the induced transition approaches and exceeds the strength of other decay modes. These are usually referred to as saturation phenomena[11] and in bound-bound transitions they typically begin manifesting themselves when the rate of induced absorption (emission) becomes comparable to the spontaneous decay of the upper state. The equivalent situation in the case of an autoionizing state occurs when the induced transition begins to be-

come comparable to the autoionization rate. The problem is, however, expected to be more complicated owing to the interference effects that are an indispensable part of autoionizing decays. The purpose of this paper is to present the basic formulation and some general results on this question. As discussed in the following pages the problem can be treated at various levels of complexity. Our formal results are cast in fairly general terms so that they can be adapted to particular situations. We then discuss in some detail specific aspects that illustrate the new features due to the strong intensity. We have avoided presenting here complicated analytic solutions which in some cases are possible. They are postponed to follow-up papers which also deal with even more general questions. A brief exposition of the problem and a summary of some results has been given earlier by one of us.[12] Here we present a systematic exposition of the theory as well as results that were not contained in the earlier report.

There are many points of formal similarity between the problem formulated in Sec. III and multiphoton ionization in the version presented by Armstrong *et al.*[13] and further discussed by Beers and Armstrong.[14] In that work, the channel identified with autoionization is dependent on the radiation while here it is not. Thus the physical origin and nature of the effects analyzed here are different. More directly related to this work are results presented by Heller and Popov[15] which refer to the topic of Sec. V.

Recent activity[11,16] on saturation in resonant transitions under strong fields has focused on the effect of field correlations and bandwidth on ac Stark splitting and related phenomena. Despite our affection for that type of problems, we shall ignore that issue in the present context. Autoionizing states typically have widths from several to a few hundreds cm^{-1}. As a result, they will be much broader than most lasers that are apt to be employed in relevant experiments. In addition, it is the effect of the intensity that we wish to explore here which is best done with a field of bandwidth narrower than the autoionization width. Of course, questions of amplitude fluctuations become relevant even for narrow-bandwidth fields.[17] But the techniques for such problems are by now known and can be easily incorporated in the present formalism when needed. For the moment we choose to concentrate on the new aspect of this problem: The behavior of autoionizing states under strong electromagnetic fields.

II. FORMULATION

Traditionally, autoionizing resonances have been studied through their excitation either by single-photon absorption or by electron impact. There is a vast literature on the subject whose theoretical description can be and has been formulated in a number of ways. The formalism given by Fano[1] some time ago seems to be familiar to most, and especially to experimentalists. Thus we begin by formulating our problem first along lines that parallel those of Fano. Subsequently we also show how an equivalent formulation can be cast in terms of the resolvent operator whose use is fairly common in the study of multiphoton processes. Aside from matters of preference, different formalisms are often convenient for specific purposes such as, for example, the study of the effect of the pulse shape[18,19] of the laser or of its coherence properties. To begin with the simplest case, we consider the excitation of an autoionizing state by single-photon absorption. We want, however, to allow the radiation intensity to become very strong in the sense that it causes couplings stronger than autoionization. This implies that the interaction can not be treated by simply calculating the transition matrix element between the initial and autoionizing states. It is necessary that we treat exactly the coupling between these two states.

The system "atom plus field" is described by the Hamiltonian

$$H = H^0 + V + D , \tag{2.1}$$

where H^0 is part of the atomic Hamiltonian with a discrete and continuum set of (single-particle) states which are coupled through the (configuration) interaction V. The coupling to the radiation field is mediated by the interaction D. At this point we have in mind a semiclassical description in which the field is treated classically. The eigenstates of H^0 are known. Let $|g\rangle$ be the ground state, $|a\rangle$ an excited state, and $|c\rangle$ a set of continuum states, all eigenstates of H^0. Then we can write $H^0|g\rangle = E_g|g\rangle$, $H^0|a\rangle = E_a|a\rangle$, $H^0|c\rangle = E_c|c\rangle$, with E being the respective energies. We ignore, for the moment, other states of the atom but will include them later when we deal with more complicated interactions.

Although not necessary, it is useful to introduce the projection operators

$$P \equiv |g\rangle\langle g| \tag{2.2a}$$

and

$$Q \equiv |a\rangle\langle a| + \int dE_c |c\rangle\langle c| , \tag{2.2b}$$

which obey the relation

$$P + Q = 1 \tag{2.3}$$

as long as other states of the atom are ignored. Since $P+Q$ is the unity (identity) operator, we

have

$$H=(P+Q)H(P+Q)=PHP+PHQ+QHP+QHQ\,. \tag{2.4}$$

As usual, the eigenstates of H^0 are assumed orthonormal which has the consequence that $PQ=QP=0$ and $P^2=P$, $Q^2=Q$ the last two relations being inherent in the definition of a projection operator.

The state $|a\rangle$ whose energy lies above the continuum threshold is coupled to the states $|c\rangle$ through the potential V which however does not affect $|g\rangle$. On the other hand, D couples $|g\rangle$ with $|a\rangle$ as well as with $|c\rangle$. Diagonal matrix elements of both V and D are, as usual, assumed to vanish, without any essential loss of generality. The above relations can be summarized as follows:

$$\langle a|H|a\rangle=\langle a|QHQ|a\rangle=\langle a|H^0|a\rangle=E_a\,, \tag{2.5a}$$

$$\langle g|H|g\rangle=\langle g|PHP|g\rangle=\langle g|H^0|g\rangle=E_g\,, \tag{2.5b}$$

$$\langle c|H|c'\rangle=\langle c|QHQ|c'\rangle=\langle c|H^0|c'\rangle=E_c\,\delta(E_c-E_{c'})\,, \tag{2.5c}$$

$$\langle c|H|a\rangle=\langle c|QHQ|a\rangle=\langle c|V|a\rangle\equiv V_{ca}\,, \tag{2.5d}$$

$$\langle a|H|g\rangle=\langle a|QHP|g\rangle=\langle a|D|g\rangle\equiv D_{ag}\,, \tag{2.5e}$$

$$\langle c|H|g\rangle=\langle c|QHP|g\rangle=\langle c|D|g\rangle\equiv D_{cg}\,, \tag{2.5f}$$

$$\langle c|V|g\rangle=0 \text{ and } \langle c|V|c'\rangle=0\,. \tag{2.5g}$$

To connect this with Fano's treatment, we take the part QHQ of the total Hamiltonian and consider its eigenstates. Thus if we set

$$\tilde{H}\equiv QHQ\,, \tag{2.6}$$

the eigenstates

$$\tilde{H}|\Psi_{\tilde{E}}\rangle\equiv\tilde{E}\,|\Psi_{\tilde{E}}\rangle \tag{2.7}$$

are labeled by the continuous energy eigenvalue $\tilde{E}$. Equation (2.7) implies that we have solved exactly (at least formally) the coupling due to V thus obtaining the new eigenstates. At this point we can simply take the results of Fano[1] and refer the reader to his paper for the derivation. The eigenfunction $|\Psi_{\tilde{E}}\rangle$ can be written as

$$|\Psi_{\tilde{E}}\rangle=\frac{\sin\Delta(\tilde{E})}{\pi V_{\tilde{E}}}\,|\Phi_{\tilde{E}}\rangle-\cos\Delta(\tilde{E})|c\rangle\,, \tag{2.8}$$

where

$$|\Phi_{\tilde{E}}\rangle=|a\rangle+\mathrm{P}\int dE_c\,\frac{V_{ca}(E_c)}{\tilde{E}-E_c}\,|c\rangle\,, \tag{2.9}$$

and P denotes the principal value of the integral. The parameter Δ is defined by

$$\Delta=-\arctan\frac{\pi|V_{ca}(\tilde{E})|^2}{\tilde{E}-E_a-F(\tilde{E})} \tag{2.10}$$

and is a function of $\tilde{E}$. The matrix element $V_{ca}(\tilde{E})$ represents the (configuration) coupling of $|a\rangle$ to the continuum and is a smooth function of energy. We shall also use the abbreviated notation $V_{ca}(\tilde{E})\equiv V_{\tilde{E}}$. The quantity $F(\tilde{E})$ represents an energy-dependent shift of E_a. It can present additional complexity if it is strongly dependent on $\tilde{E}$. If that were the case it would probably mean that the continuum was not smooth and that additional bound states like $|a\rangle$ ought to be taken into account which is contrary to the model we wish to study here. Thus it is only a smooth dependence on $\tilde{E}$ that we can expect and for our purposes it is sufficient to replace for the moment $F(\tilde{E})$ by its value at $\tilde{E}=E_a$ and replace E_a by a shifted energy $\bar{E}_a$ given by

$$\bar{E}_a\equiv E_a+F(E_a)\,. \tag{2.11}$$

From Eqs. (2.8) and (2.9) we recognize that $|\Psi_{\tilde{E}}\rangle$ consists of a linear superposition of the bound state $|a\rangle$ and the whole continuum. The energy difference in the denominator of Eq. (2.10) suggests the well-known dispersive character of this superposition.

The usual weak-field photoexcitation of the autoionizing state is calculated by means of the transition matrix element $\langle\Psi_{\tilde{E}}|D|g\rangle$ which is employed in the standard expression for the transition probability per unit time. Using previous equations we have

$$\langle\Psi_{\tilde{E}}|D|g\rangle=\frac{\tilde{D}_{Eg}}{\pi V_{\tilde{E}}}\sin\Delta-D_{cg}\cos\Delta\,, \tag{2.12a}$$

where

$$\tilde{D}_{\tilde{E}g}\equiv\langle\Phi_{\tilde{E}}|D|g\rangle\,. \tag{2.12b}$$

The interaction D has the usual form

$$D=\vec{\mu}\cdot\vec{\epsilon}\,\mathcal{E}(t) \tag{2.13}$$

with $\vec{\mu}=-e\,\vec{r}$ being the dipole operator of the atom, $\vec{\epsilon}$ the polarization vector of the externally applied electromagnetic field, and $\mathcal{E}(t)$ its amplitude. We need be concerned only with monochromatic fields in this paper. Thus we take

$$\mathcal{E}(t)=\mathcal{E}_0\,e^{i\omega t}+\mathcal{E}_0^*\,e^{-i\omega t}\,, \tag{2.14}$$

where $\mathcal{E}_0$ is now the constant amplitude of the electric field. The dipole approximation implied in Eq. (2.13) is not necessary and can be easily changed at any stage in the calculation. If one were to calculate the transition probability from $|g\rangle$ to $|a\rangle$ as a function of ω the resulting curve would be the well-known asymmetric profile with width Γ given by

$$\Gamma=2\pi|V_{\tilde{E}}|^2\,. \tag{2.15}$$

How the profile arises becomes more evident if,

by combining previous equations, we write $\langle\Psi_{\tilde{E}}|D|g\rangle$ as

$$\langle\Psi_{\tilde{E}}|D|g\rangle=\frac{\tilde{D}_{\tilde{E}g}}{\pi V_{\tilde{E}}^{*}}\frac{\frac{1}{2}\Gamma}{[E-\overline{E}_a)^2+\frac{1}{4}\Gamma^2]^{1/2}}$$

$$+D_{cg}(\tilde{E})\frac{\tilde{E}-\overline{E}_a}{[(\tilde{E}-\overline{E}_a)^2+\frac{1}{4}\Gamma^2]^{1/2}}, \qquad (2.16)$$

where $\tilde{E}$ is to be understood as $E_g+\hbar\omega$. Obviously, taking $|\langle\Psi_{\tilde{E}}|D|g\rangle|^2$ we will have an asymmetric profile as the photon energy varies around the energy difference $\overline{E}_a-E_g$. The asymmetry originates from the second term of Eq. (2.16) since it changes sign from one side of the resonance to the other. It is often said that the asymmetry results from the interference between the direct transition to the continuum ($\tilde{D}_{cg}$) and the transition to the autoionizing state ($\tilde{D}_{\tilde{E}g}$). It must of course be kept in mind that the continuum is also involved in the matrix element $\tilde{D}_{\tilde{E}g}$ [see Eqs. (2.12b) and (2.9)] through the configuration interaction.

It will be noticed that the width Γ as written above is energy dependent. This dependence is directly related to the energy dependence of the matrix element V_{ca}. Matrix elements such as this, as well as D_{cg}, depend on the energy E_c of the continuum state $|c\rangle$. As we have seen they occur in the previous equations either evaluated at $E_c=\tilde{E}$ or integrated over E_c. Having in mind the remarks made earlier in connection with $F(\tilde{E})$, we will often assume these energy dependences to be sufficiently weak—compared to the variation implied by the resonance denominator—to enable us to neglect this energy dependence and take those matrix elements evaluated at $\tilde{E}=E_a$.

Another quantity that is an important parameter in this formalism is the so-called q value defined by

$$q(\tilde{E})\equiv\frac{\langle\Phi_{\tilde{E}}|D|g\rangle}{\pi V_{\tilde{E}}^{*}\langle c|D|g\rangle_{\tilde{E}}}=\frac{\tilde{D}_{\tilde{E}g}}{\pi V_{\tilde{E}}^{*}D_{cg}(\tilde{E})}. \qquad (2.17)$$

It is a measure of the importance of the direct transition from $|g\rangle$ to the continuum compared to the transition via the autoionizing state. High q means weak direct transition and hence symmetric line shape since the interference is minimized. Again q depends on $\tilde{E}$ but we may sometimes want to simply take its value at $\tilde{E}=E_a$.

There is an energy $\tilde{E}_0$ for which $\langle\Psi_{\tilde{E}_0}|D|g\rangle=0$. From Eqs. (2.16) and (2.9) it is evident that this occurs when

$$\frac{\sin\Delta_0}{\cos\Delta_0}=\frac{\pi V_{\tilde{E}_0}^{*}\langle c|D|g\rangle_{\tilde{E}_0}}{\langle\Phi_{\tilde{E}_0}|D|g\rangle_{\tilde{E}_0}}, \qquad (2.18)$$

where the subscript 0 indicates that the quantity is evaluated at $\tilde{E}=\tilde{E}_0$. If in addition one introduces

$$\epsilon\equiv\frac{\tilde{E}-\overline{E}_a}{\frac{1}{2}\Gamma(\tilde{E})}=-\cot\Delta \qquad (2.19)$$

and the quantity

$$R^2\equiv\frac{|\langle\Psi_{\tilde{E}}|D|g\rangle|^2}{|\langle c|D|g\rangle_{\tilde{E}}|^2}, \qquad (2.20)$$

which is an expression for the line profile, in terms of ϵ and q, R^2 becomes

$$R^2=\frac{(q+\epsilon)^2}{1+\epsilon^2}=1+\frac{q^2-1+2q\epsilon}{1+\epsilon^2}. \qquad (2.21)$$

These are equations that are often quoted in papers dealing with studies of autoionizing states. Here they serve as points of departure for the study of the more general problem that is our main concern.

III. STRONG COUPLING OF AN AUTOIONIZING RESONANCE TO A BOUND STATE

A. General case

If the electromagnetic field that couples $|g\rangle$ to $|a\rangle$ becomes sufficiently strong, the transition is not necessarily describable in terms of a transition probability per unit time. To provide a quantitative description for that case we must consider the evolution of the system in some detail.

At $t=0$ the atom is in $|g\rangle$. At any later time its wave function can be written as

$$\Psi(t)=U_g(t)|g\rangle+\int d\tilde{E}'U_{\tilde{E}'}(t)|\Psi_{\tilde{E}'}\rangle. \qquad (3.1)$$

The time evolution of $\Psi(t)$ is determined by the Schrödinger equation

$$\frac{\partial}{\partial t}\Psi(t)=-\frac{i}{\hbar}H\Psi(t), \qquad (3.2)$$

under the initial condition $\Psi(0)=|g\rangle$. Substitution of Eq. (3.1) into (3.2) gives

$$\frac{d}{dt}U_g(t)|g\rangle+\int d\tilde{E}'\frac{d}{dt}U_{\tilde{E}'}(t)|\Psi_{\tilde{E}'}\rangle$$

$$=-\frac{i}{\hbar}HU_g(t)|g\rangle-\frac{i}{\hbar}\int d\tilde{E}'HU_{\tilde{E}'}(t)|\Psi_{\tilde{E}'}\rangle. \qquad (3.3)$$

From this we obtain differential equations for the time-dependent expansion coefficients $U(t)$ in the standard fashion. We take the inner product of this equation first with $\langle g|$ and then with $\langle\Psi_{\tilde{E}}|$. In doing so, the form for H given in Eq. (2.4) and the definition of $|\Psi_{\tilde{E}}\rangle$ must be taken into ac-

count and Eqs. (2.5) will determine which terms survive. Thus it is only the term QHP that has nonvanishing matrix elements between $|g\rangle$ and $\langle\Psi_{\tilde{E}}|$, and of course the term PHQ between $\langle g|$ and $|\Psi_{\tilde{E}}\rangle$. The resulting equations are

$$\frac{d}{dt}U_g(t) = -\frac{i}{\hbar}E_g U_g(t) - \frac{i}{\hbar}\int d\tilde{E}\langle g|PHQ|\Psi_{\tilde{E}'}\rangle U_{\tilde{E}'}(t) , \tag{3.4a}$$

$$\frac{d}{dt}U_E(t) = -\frac{i}{\hbar}\tilde{E}U_{\tilde{E}}(t) - \frac{i}{\hbar}\langle\Psi_{\tilde{E}}|QHP|g\rangle U_g(t) , \tag{3.4b}$$

where an expression for $\langle\Psi_{\tilde{E}}|QHP|g\rangle$ is given by Eq. (2.12a). To compress the notation, we introduce $\omega_g \equiv E_g/\hbar$, $\tilde{\omega} \equiv \tilde{E}/\hbar$, and $U_{\tilde{\omega}} \equiv \hbar^{1/2}U_{\tilde{E}}$. The matrix elements of D are matrix elements of $(\vec{\mu}\cdot\vec{\epsilon})$ $(\mathcal{E}_0 e^{i\omega t} + \mathcal{E}_0^* e^{-i\omega t})$ which involve time-dependent factors. Because of their time-dependent factors, the equations are put into a more convenient form if we introduce new (transformed) expansion coefficients $v(t)$ defined by

$$v_g(t) \equiv U_g(t)e^{i\omega_g t}$$

and

$$v_{\tilde{\omega}}(t) \equiv U_{\tilde{\omega}}(t)e^{i\tilde{\omega}t} . \tag{3.5}$$

Substituting into Eqs. (3.4) and combining the time-dependent exponentials we find that they occur in the forms $e^{[i\omega \pm i(\tilde{\omega}-\omega_g)]t}$. Although $\tilde{\omega}$ is a continuous variable, we know that it appears in functions peaked at $\tilde{\omega} \simeq \omega_a$, because we do have a bound state embedded in the continuum. These peaked functions are represented by $\sin\Delta$ and $\cos\Delta$ [see Eqs. (2.12) and (2.16)]. This means that the exponent $[\omega + (\tilde{\omega} - \omega_g)]t$ leads to terms antiresonant with the photon frequency, while $[\omega - (\tilde{\omega} - \omega_g)]t$ leads to resonant terms. It is understood of course that the photon frequency is to be tuned around the resonant transition frequency $(\omega_a - \omega_g)$, where $\omega_a \equiv E_a/\hbar$. We can therefore neglect the antiresonant terms. This approximation is nothing else but the so-called "rotating wave approximation" and its implications have been well studied and understood in the context of bound-bound transitions. As long as the detunings $\omega - (\omega_a - \omega_g)$ are small compared to ω, there is no reson to be concerned about this approximation. The resulting equations for the v's are

$$\frac{d}{dt}v_g(t) = -i\int d\tilde{\omega}\, M_{\tilde{\omega}g}^* \mathcal{E}_0 e^{i(\omega-\tilde{\omega}+\omega_g)t} v_{\tilde{\omega}}(t) , \tag{3.6a}$$

$$\frac{d}{dt}v_{\tilde{\omega}}(t) = -iM_{\tilde{\omega}g}\mathcal{E}_0^* e^{-i(\omega-\tilde{\omega}+\omega_g)t} v_g(t) , \tag{3.6b}$$

$$M_{\tilde{\omega}g} \equiv \langle\Psi_{\tilde{E}}|\mu|g\rangle\hbar^{-1/2} = \hbar^{-1/2}\left(\frac{\vec{\mu}_{\tilde{E}g}}{V_{\tilde{E}}^*}\sin\Delta - \mu_{cg}\cos\Delta\right) , \tag{3.7a}$$

and

$$\tilde{\mu}_{\tilde{E}g} \equiv \langle\Phi_{\tilde{E}}|\mu|g\rangle . \tag{3.7b}$$

By μ we shall, from here on, understand $\vec{\mu}\cdot\vec{\epsilon}$, the projection of $\vec{\mu}$ on the polarization vector of the radiation. The relation between $U_{\tilde{\omega}}$ and $U_{\tilde{E}}$ is such that $\int d\tilde{\omega}|U_{\tilde{\omega}}|^2 = \int d\tilde{E}|U_{\tilde{E}}|^2$. The quantity $M_{\tilde{\omega}g}$ is a dipole matrix element between the ground state and the state $|\Psi_{\tilde{E}}\rangle$ which contains $|a\rangle$ as well as the continuum $|c\rangle$. But μ_{cg} is the usual bound-free dipole matrix element that appears in photoionization. If the continuum were "switched off", $M_{\tilde{\omega}g}$ would simply reduce to the matrix element μ_{ag} between two bound states; a point to which we return later on. Since in Eq. (3.6a) an integration over all $\tilde{\omega}$ is involved, the reader may become concerned about the validity of the rotating wave approximation. Its validity remains unaffected because of the peaked nature of $M_{\tilde{\omega}g}$ as a function of $\tilde{\omega}$. Incidentally, the rotating wave approximation should have also been mentioned in the discussion of Sec. II as it is inevitably implicit in any formulation of photoabsorption in terms of a transition probability per unit time. We postponed its discussion on purpose, however, so that it could be seen more clearly emerging from the formalism.

We have now a set of two integrodifferential equations in the variables t and $\tilde{\omega}$. From their solution we must determine the amplitudes $U(t)$ in terms of which the probability of ionization is written as

$$P(T) = 1 - |U_g(T)|^2 - |U_a(T)|^2 , \tag{3.8a}$$

where T is the total time of interaction between atom and field. Depending on the particular experiment, T may be determined either by the duration of the laser pulse or the time it takes the atoms to traverse the light beam, whichever is smaller. Because we are dealing with strong field phenomena and/or short pulses, we must in general calculate a time-dependent probability which only in limiting cases may be expressible in terms of a transition probability per unit time that is time independent. The weak field limit is such a case. As expressed by Eq. (3.8a), the ionization probability is simply calculated as the number of atoms (or probability per atom) that are neither in state $|g\rangle$ nor in state $|a\rangle$ and are therefore in the continuum. An equivalent expression for $P(T)$ is

$$P(T) = \int dE_c |U_c(T)|^2 , \tag{3.8b}$$

where the integration is over the whole continuum. Although we have not so far written an expression for $U_a(t)$, because we are working with the states

$|\Psi_{\tilde{E}}\rangle$ which contain $|a\rangle$, it is easy to show that

$$U_a(t)=\int d\tilde{E}\,\frac{U_{\tilde{E}}(t)\sin\Delta}{\pi V_{\tilde{E}}}=\hbar^{1/2}\int d\tilde{\omega}\,\frac{U_{\tilde{\omega}}(t)\sin\Delta}{\pi V_{\tilde{E}}}\ , \tag{3.9}$$

which is evident if one refers to the definition of states $|\Phi_{\tilde{E}}\rangle$ and the expansion of $\Psi(t)$.

As expressed by Eqs. (3.8a) and (3.8b), $P(T)$ represents ionization at the end of the laser pulse. At that time, however, we have a population $|U_a(T)|^2$ in state $|a\rangle$. This population will in most cases decay into the continuum via the interaction V with a decay rate Γ. Only if radiative emission were significant, would it return to $|g\rangle$; a possibility that we may safely neglect in general. Therefore at a time $t>T$, the ionization probability $P(t)$ is given by

$$P(t)=1-|U_g(T)|^2-|U_a(T)|^2e^{-\Gamma(t-T)}. \tag{3.10a}$$

Depending now on how the ionization signal is collected, the last term in Eq. (3.10a) may or may not make a significant contribution. Perhaps for most realistic situations, we must take $\Gamma(t-T)\gg 1$ because all ions (or electrons) are collected. Thus we may calculate ionization as

$$P=1-|U_g(T)|^2. \tag{3.10b}$$

This may also seem to be more compatible with the standard understanding of autoionization where the state $|a\rangle$ is somewhat arbitrary in that it depends on what part of the atomic Hamiltonian is diagonalized exactly. The atom has no true bound states in that energy range, one could argue. Even with this arbitrariness, however, the fact is that we can have a "semibound" part of the state which, of course, decays eventually. Its decay rate depends on how the atomic Hamiltonian has been separated in the parts H^0 and V but it nonetheless represents a state. Needless to say, $|U_a|^2$ also depends on that separation. For $t\to\infty$, the ionization signal should not and does not depend on $|a\rangle$. It is, however, possible at least in principle, to determine ionization at finite t after the laser is off. Then $|U_a(t)|^2$ enters in the calculation. Recall that the choice of $|a\rangle$ also influences the Rabi frequency and hence $U_g(T)$. It is not strange therefore, that $|U_a(t)|^2$ appears in the equation for $P(t)$ for finite time t. Again, the actually observed signal does not depend on the choice of $|a\rangle$—if the problem is solved exactly—but it appears in the equation in much the same way that the particular representation affects the appearance of an equation. The salient point is that the interaction of autoionizing states with strong lasers affords the possibility of probing these time-dependent aspects. In fact, sometimes it is necessary to do so in order to understand the details of the interaction.

Returning now to Eqs. (3.6), we integrate (3.6b) formally, obtaining

$$v_{\tilde{\omega}}(t)=-i\int_0^t dt' M_{\tilde{\omega}g}\mathcal{E}_0^* e^{-i(\omega-\tilde{\omega}+\omega_g)t'}v_g(t')\ , \tag{3.11}$$

and then substitute this expression in (3.6a) with the result

$$\frac{d}{dt}v_g(t)=-\int d\tilde{\omega}\,|M_{\tilde{\omega}g}|^2|\mathcal{E}_0|^2\int dt' e^{-i(\omega-\tilde{\omega}+\omega_g)(t'-t)}v_g(t'). \tag{3.12}$$

This can be solved by Laplace transform. Defining

$$\int_0^\infty dt\,e^{-pt}v_g(t)\equiv u_g(p)\ , \tag{3.13}$$

and noting the initial condition $u_g(0)=1$, we obtain

$$u_g(p)=\frac{1}{p+\int\frac{d\tilde{\omega}\,|M_{\tilde{\omega}g}|^2|\mathcal{E}_0|^2}{p-i(\omega-\tilde{\omega}+\omega_g)}}=\frac{1}{p+S(p)}\ , \tag{3.14}$$

where we have defined

$$S(p)\equiv\int d\tilde{\omega}\frac{|M_{\tilde{\omega}g}|^2|\mathcal{E}_0|^2}{p-i(\omega-\tilde{\omega}+\omega_g)}\ . \tag{3.15}$$

In order to have a more direct correspondence with the resolvent operator formalism, which will be discussed later on, we introduce the transformation

$$p=-iz \tag{3.16}$$

and by direct substitution express $S(p)$ and $u_g(p)$ as functions of z. To obtain $v_g(t)$ we calculate the Laplace inversion integral on the complex z plane. It is known[20] that if we let $z=x+iy$, the inversion integral for $t>0$ can be obtained from

$$v_g(t)=\frac{-1}{2\pi}\int_{-\infty}^{+\infty}dx\,e^{-ixt}u_g^+(x)\ , \tag{3.17a}$$

where

$$u_g^+(x)=\lim_{\eta\to 0+}u_g(x+i\eta). \tag{3.17b}$$

The above inversion integral is identical to that obtained with the resolvent operator and the steps leading to it can be found in many references.[20]

An expression must be obtained now for $u_g^+(x)$. This involves a considerable amount of tedious algebra the main steps of which are shown in Appendix A. Here it will suffice to present the final expressions and discuss their physical significance. The expression for $u_g^+(x)$ is

$$u_g^+(x)=\frac{i}{x-s(x)+i\gamma(x)}\ , \tag{3.18}$$

where $s(x)$ and $\gamma(x)$ are the real and imaginary parts of $-iS(x)$ and are given by the equation

$$s(x)-i\gamma(x)=\frac{\pi}{\kappa}\frac{A_0+B_0X+C_0X^2}{X+i\kappa}-\frac{\pi}{\kappa}(B_0+C_0X+C_0\kappa N), \tag{3.19}$$

where the various parameters appearing above have emerged in the algebraic manipulation of $S(x)$ and are given by

$$A_0=\hbar^{-3}|\mathcal{E}_0|^2|\tilde{\mu}_{\tilde{E}g}|^2|V_{\tilde{E}}|^2\ , \tag{3.20a}$$

$$B_0=\hbar^{-2}|\mathcal{E}_0|^2 2\,\mathrm{Re}\,\frac{\tilde{\mu}^*_{\tilde{E}g}\tilde{\mu}_{cg}}{V_{\tilde{E}}}|V_{\tilde{E}}|^2\ , \tag{3.20b}$$

$$C_0=\hbar^{-1}|\mathcal{E}_0|^2|\mu_{cg}|^2\ , \tag{3.20c}$$

$$\kappa=\tfrac{1}{2}\Gamma/\hbar\ , \tag{3.20d}$$

and N is a pure number defined in Appendix A. Its magnitude depends on how fast (or slowly) μ_{cg} decreases as a function of the energy E_c. For the discussion here, we may consider it of order unity. In general, however, it will depend on the particular atom. Also, the parameters A, B, C are functions of $\tilde{E}=\hbar\tilde{\omega}$. The subscript 0 indicates that they are evaluated at $\tilde{\omega}=\omega_g+\omega$. Again this is acceptable because the structure in the continuum comes from the peaked functions discussed earlier, while the matrix elements themselves are slowly varying. This is not a necessary approximation in this formalism, but we adopt it since it facilitates greatly the discussion of the physical effects without any crucial loss of generality. If it ever were necessary, that slow energy dependence could be taken into account. The expression of Eq. (3.19) has been obtained by performing the integration over $\tilde{\omega}$ and X is an abbreviation defined by

$$X=x+\omega-(\bar{\omega}_a-\omega_g)\ . \tag{3.21}$$

It is the fact that $V_{\tilde{E}}$ and the matrix elements of μ are slowly varying functions of $\tilde{E}$ {compared to the Lorentzian $[(\tilde{E}-\bar{E}_a)^2+\frac{1}{4}\Gamma^2]^{-1}$} that enables us to perform the integrations over $\tilde{\omega}$ at this early stage, thus obtaining results that are independent of the particulars of the atom and reflect general properties of the process.

The time dependence of $v_g(t)$ is now obtained from the inversion integral

$$v_g(t)=-\frac{1}{2\pi i}\int_{-\infty}^{+\infty}dx\,\frac{e^{-ixt}}{x-s(x)+i\gamma(x)}\ , \tag{3.22}$$

where it must be noted that s and γ are not the standard shift and width because they depend on x in a way that is not negligible and contains in fact much of the physics of the problem. With a little more algebraic manipulation, Eq. (3.22) is put in a form that makes its meaning and connection with the weak-field case more evident. To this end, we introduce the quantity

$$\tilde{\Omega}^2\equiv\frac{\pi A_0}{\kappa}=\hbar^{-2}|\mathcal{E}_0|^2|\mu_{\tilde{E}g}|^2_{\tilde{E}=\bar{E}_a}\ , \tag{3.23}$$

which is identical to $\tilde{D}_{\tilde{E}g}$ of Eq. (2.12b), and the detuning

$$\delta\equiv\omega-(\bar{\omega}_a-\omega_g) \tag{3.24}$$

of the photon frequency ω from the exact center frequency $\bar{\omega}_a-\omega_g\equiv\bar{\omega}_{ag}$ of the autoionizing resonance. The inversion integral is now written as

$$v_g(t)=\frac{1}{2\pi i}\int_{-\infty}^{+\infty}dx\,\frac{(x+\delta+i\kappa)e^{-ixt}}{\Lambda(x)}\ , \tag{3.25a}$$

where

$$\Lambda(x)\equiv x(x+\delta+i\kappa)-\tilde{\Omega}^2\left(1+\frac{x+\delta}{q\kappa}\right)^2+\frac{\tilde{\Omega}^2}{q\kappa}\left(2+\frac{x+\delta+N\kappa}{q\kappa}\right)(x+\delta+i\kappa). \tag{3.25b}$$

Everything is now expressed in terms of the field-independent parameters q and $\kappa=\frac{1}{2}\Gamma/\hbar$ of the autoionizing resonance, and the field-dependent quantity $\tilde{\Omega}$. From its definition in Eq. (3.23), $\tilde{\Omega}$ will be recongnized as some generalized Rabi frequency. It would have been a usual Rabi frequency if the matrix element $\tilde{\mu}_{\tilde{E}g}$ coupled $|g\rangle$ with a bound state, in which case $\tilde{\Omega}$ would represent the frequency of induced transitions between $|g\rangle$ and the other bound state. Now, however, $\tilde{\mu}_{\tilde{E}g}$ couples $|g\rangle$ with the state $\Phi_{\tilde{E}}$ containing the bound state $|a\rangle$ as well as the continuum. Thus $\tilde{\Omega}$ does not represent simply transitions from $|g\rangle$ to $|a\rangle$ but also to $|c\rangle$ and in the calculation of $|\tilde{\mu}_{\tilde{E}g}|^2$ there is an interference between the two. This interference is of course mediated by the configuration interaction. As a result, when the field excites $|g\rangle$ to either $|a\rangle$ or $|c\rangle$, there are virtual transitions between $|a\rangle$ and $|c\rangle$. $\tilde{\Omega}$ is not the only quantity in which such interference is manifested. It is also to be found in the other terms of $\Lambda(x)$. But, that the interference is already present in the Rabi frequency points to a significant difference between the strong-field excitation of an autoionizing state from that of a bound state. With a little more algebra, Eq. (3.25b) can be put in the more compact form

$$\Lambda(x)=\left(x+\frac{\tilde{\Omega}^2}{q^2\kappa^2}(N\kappa+i\kappa)\right)(x+\delta+i\kappa)-\tilde{\Omega}^2\left(1-\frac{i}{q}\right)^2\ , \tag{3.26}$$

which makes easier the exploration of certain special cases for which analytic expressions can be obtained.

If we call $x_\pm$ the roots of $\Lambda(x)$, then $\Lambda(x)=(x-x_+)(x-x_-)$ and a formal expression for the inversion integral is

$$v_g(t)=\frac{1}{x_+-x_-}[(x_++\delta+i\kappa)e^{-ix_+t} - (x_-+\delta+i\kappa)e^{-ix_-t}] . \quad (3.27)$$

If we were to substitute in this equation the expressions for $x_\pm$, the resulting expression would be too complicated to be of inspectional usefulness. Subsequently, we will examine some of its special cases which lend themselves to easier analytic exploration.

B. Special cases

Before proceeding with the exploration of the general result represented by Eq. (3.26), it is useful to consider certain special cases corresponding to limiting values of the parameters. The intention here is to show how our general equation reduces to two known results: The weak-field excitation of an autoionizing resonance and the strong-field coupling of two bound states.

The weak-field limit here corresponds to $\tilde{\Omega}\ll\kappa$. In exploring this case, we neglect the term $\tilde{\Omega}^2N/q^2\kappa$ from Eq. (3.26) because it represents a radiation-induced ac Stark shift. And this shift is absent from the usual treatment[1] of the photoexcitation of an autoionizing resonance since it is not important in that case. Then we need the roots of the equation

$$x^2+[\delta+i(1+\beta^2)\kappa]x-[x^2-i\beta^2\kappa(\delta+i\kappa)]=0, \quad (3.28a)$$

where we have introduced the abbreviations

$$\beta^2=\frac{\tilde{\Omega}^2}{q^2\kappa^2}$$

and

$$\alpha^2=\tilde{\Omega}^2\left(1-\frac{i}{q}\right)^2 . \quad (3.28b)$$

The roots are

$$x_\pm=-\tfrac{1}{2}[\delta+i(1+\beta^2)\kappa] \times\left[1\mp\left(1+4\frac{\alpha^2-i\beta^2\kappa(\delta+i\kappa)}{[\delta+i(1+\beta^2)\kappa]^2}\right)^{1/2}\right] . \quad (3.29)$$

Because of the weak-field assumption, we have $\beta^2\ll1$ and also $\alpha^2\ll\kappa^2$. As a result, we can take $\delta+i(1+\beta^2)\kappa\simeq\delta+i\kappa$ and also expand the square root in a Taylor series keeping only the term linear in the small quantity. Then the roots become

$$x_+=\frac{\alpha^2}{\delta+i\kappa}-i\beta^2\kappa \quad (3.30a)$$

and

$$x_-=-(\delta+i\kappa)-\frac{\alpha^2}{\delta+i\kappa}+i\beta^2\kappa . \quad (3.30b)$$

The inversion integral leads to

$$v_g(t)=\frac{1}{x_+-x_-}[(x_++\delta+i\kappa)e^{-ix_+t} - (x_-+\delta+i\kappa)e^{-ix_-t}] . \quad (3.31)$$

Because of the weak-field situation, we have $|x_+|\ll|x_-|$. In that limit, therefore, only the exponential e^{-ix_+t} matters because the other decays very fast. It is then a simple algebraic matter to show that the rate of decay of $|v_g(t)|^2$ is equal to $2\,\mathrm{Im}x_+$. This is shown most directly by calculating $[(d/dt)|v_g(t)|^2]_{t=0}$. From Eq. (3.20a) we obtain

$$-\mathrm{Im}x_+=\frac{\tilde{\Omega}^2}{q^2}\left(\frac{\kappa}{\kappa^2}+\frac{\kappa(q^2-1)+2q\delta}{\delta^2+\kappa^2}\right), \quad (3.32)$$

and upon using the notation $\delta=\epsilon\kappa$ of Sec. II we rewrite this as

$$-\frac{2\,\mathrm{Im}x_+}{2\pi}=\frac{1}{\hbar}|\langle c|D|g\rangle|^2\left(1+\frac{q^2-1+2q\epsilon}{1+\epsilon^2}\right), \quad (3.33)$$

which is the quantity $(1/\hbar)|\langle\Psi_E|D|g\rangle|^2$ as obtained by Fano and shown in Eq. (2.20).

The strong-field limit of the coupling between two bound states is obtained by letting $q\to\infty$ in Eq. (3.26) which then reduces to

$$x(x+\delta+i\kappa)-\tilde{\Omega}^2=0. \quad (3.34)$$

Its roots are

$$x_\pm=-\tfrac{1}{2}(\delta+i\kappa)\pm\tfrac{1}{2}[(\delta+i\kappa)^2+4\tilde{\Omega}^2]^{1/2} , \quad (3.35a)$$

and in the strong-field limit (that is when $|\delta+i\kappa|\ll\tilde{\Omega}^2$) they become

$$x_\pm=-\tfrac{1}{2}(\delta+i\kappa)\pm\tilde{\Omega} . \quad (3.35b)$$

The time development of $v_g(t)$ is then of the form

$$v_g(t)=\frac{1}{2\tilde{\Omega}}\left[[\tfrac{1}{2}(\delta+i\kappa)+\tilde{\Omega}]\exp-i\left(\tilde{\Omega}-\frac{\delta}{2}\right)t - [\tfrac{1}{2}(\delta+i\kappa)-\tilde{\Omega}]\exp i\left(\tilde{\Omega}+\frac{\delta}{2}\right)t\right], \quad (3.36)$$

which exhibits the typical oscillatory behavior of the the amplitude of a state $|g\rangle$ strongly coupled to another bound state $|a\rangle$. As usual,[11,17] the oscillation occurs at two different frequencies separated by $2\tilde{\Omega}$, the Rabi frequency. That $v_g(t)$ is in addition found to decay with half-width $\kappa/2$

is, strictly speaking, inconsistent with having taken the limit $q \to \infty$. Because we should then have set the coupling to the continuum equal to zero everywhere. Not having done so, Eq. (3.36) represents the solution for a ground state $|g\rangle$ strongly coupled to another bound state $|a\rangle$ which decays to other states via a coupling independent of the field. Moreover, these channels of decay should not interfere with the channel $|g\rangle \leftrightarrow |a\rangle$ if this equation is to be applicable.

We have thus seen that Eq. (3.26) is a generalization of two well-known types of interaction of atomic states with radiation. It is perhaps worth recalling at this point that autoionization does more than just cause the decay of $|a\rangle$ into the continuum. If that was all, the equation for the roots would look like Eq. (3.34). As we have seen, however, the interference aspects of autoionization affect not only the Rabi frequency but also introduce an additional width-shift term, namely, $(\tilde{\Omega}^2/q^2\kappa^2)(N\kappa + i\kappa)$, which as we shall see subsequently causes significant changes from what one would have expected on the basis of the two limiting cases.

IV. FORMULATION IN TERMS OF THE RESOLVENT OPERATOR

This section is devoted to a somewhat different formulation of the problem employing the resolvent operator[20] and treating the field quantum mechanically. The total Hamiltonian is again written as

$$H = H^0 + V + D \,, \tag{4.1a}$$

where

$$H^0 = H^A + H^R \,, \tag{4.1b}$$

which now contains two parts: The atomic part H^A whose eigenstates are the previously defined $|g\rangle$, $|a\rangle$, and $\{|c\rangle\}$, and the radiation part H^R with eigenstates the usual photon-number states $|n\rangle$. Only one mode of the field is assumed to be occupied, its frequency being ω. The initial state of the system is $|g; n\rangle$. The states connected to it via the absorption or emission of a photon are $|a; n-1\rangle$ and $\{|c; n-1\rangle\}$. For simplicity, we will use the labels g, a, and c for these states. This is equivalent to the rotating wave approximation of Sec. III B. The atomic states $|a\rangle$ and $\{|c\rangle\}$ are as before coupled to each other through configuration interaction.

In contrast to Sec. III A, H is time independent because it contains the Hamiltonian of the radiation as well. As a consequence, the time evolution of the wave function is given by

$$\Psi(t) = \exp\left(-i\frac{H}{\hbar}t\right)\Psi(0) \equiv U(t)\Psi(0) = U(t)|g;n\rangle \,, \tag{4.2}$$

thus defining the time-evolution operator $U(t)$ which can be expressed in terms of the resolvent operator

$$G(z) \equiv \frac{1}{z-H} = \frac{1}{z-H^0-V-D} \tag{4.3}$$

through the inversion integral

$$U(t) = -\frac{1}{2\pi i}\int_{-\infty}^{+\infty} dx\, e^{-ixt} G^+(x) \,, \tag{4.4}$$

whose meaning is identical to that of Eqs. (3.17). Taking into account the states of interest in this problem, $\Psi(t)$ is written in terms of the matrix elements of $U(t)$

$$\Psi(t) = U_{gg}|g;n\rangle + U_{ag}|a;n-1\rangle + \int dE_c U_{cg}|c;n-1\rangle \,, \tag{4.5}$$

and the matrix elements of $U(t)$ are obtained from the corresponding matrix elements of G through Eq. (4.4). The problem is thus reduced, as usual, to the calculation of matrix elements of $G(x)$. From Eq. (4.3) we have $(z-H)G = 1$, which leads to

$$(z-H)(P+Q)GP = P \,, \tag{4.6}$$

where P and Q are the projection operators defined in Sec. II. Multiplying this equation from the left by P or by Q, and making use of the properties of P and Q, we obtain the equations

$$(z - PHP)(PGP) - (PHQ)(QGP) = P \,, \tag{4.7a}$$

$$(z - QHQ)(QGP) - (QHP)(PGP) = 0 \,. \tag{4.7b}$$

The desired matrix element G_{gg} is now obtained from $\langle g|PGP|g\rangle$ while G_{ag} and G_{cg} from $\langle a|QGP|g\rangle$ and $\langle c|QGP|g\rangle$, respectively. For the first matrix element, we use Eq. (4.7a) and for the other two, Eq. (4.7b). In the process, we use Eqs. (2.5). The resulting equations are

$$(z - E'_g)G_{gg} - D_{ga}G_{ag} - \int dE'_c D_{gc}G_{cg} = 1 \,, \tag{4.8a}$$

$$-D_{ag}G_{gg} + (z - E'_a)G_{ag} - \int dE'_c V_{ac}G_{cg} = 0 \,, \tag{4.8b}$$

$$-D_{cg}G_{gg} - V_{ca}G_{ag} + (z - E'_c)G_{cg} = 0 \,. \tag{4.8c}$$

Since H^0 is here somewhat different than in Secs. III A and III B the energy E'_g contains the energy $n\hbar\omega$ of n photons while E'_a and E'_c contain the energy $(n-1)\hbar$. The end result as usual involves only energy differences. Thus we can take, without any further approximation,

$$E_g' = E_g + \hbar\omega, \quad E_a' = E_a$$

and

$$E_c' = E_c . \tag{4.9}$$

The matrix element G_{cg} can be eliminated by solving Eq. (4.8c), obtaining

$$G_{cg} = \frac{1}{z-\omega_c}\,(D_{cg}G_{gg} + V_{ca}G_{ag}) \tag{4.10}$$

and substituting into Eqs. (4.8a) and (4.8b) with the result

$$\left(z - E_g - \hbar\omega - \int dE_c \frac{|D_{cg}|^2}{z-E_c}\right)G_{gg} - \left(D_{ga} + \int dE_c \frac{D_{gc}V_{ca}}{z-E_c}\right)G_{ag} = 1 , \tag{4.11a}$$

$$-\left(D_{ag} + \int dE_c \frac{V_{ac}D_{cg}}{z-E_c}\right)G_{gg} + \left(z - E_a - \int dE_c \frac{|V_{ac}|^2}{z-E_c}\right)G_{ag} = 0 . \tag{4.11b}$$

Expressions for $G_{gg}(z)$ and $G_{ag}(z)$ are obtained by solving the system of these two algebraic equations. The resulting equations are

$$G_{gg} = 1/\Lambda'(z) \tag{4.12a}$$

and

$$G_{ag} = \frac{D_{ag} + \int dE_c \frac{V_{ac}D_{cg}}{z-E_c}}{\left(z - E_a - \int dE_c \frac{|V_{ac}|^2}{z-E_c}\right)\Lambda'(z)} , \tag{4.12b}$$

where

$$\Lambda'(z) = z - E_g - \hbar\omega - \int dE_c \frac{|D_{cg}|^2}{z-E_c} - \frac{\left(D_{ga} + \int dE_c \frac{D_{gc}V_{ca}}{z-E_c}\right)\left(D_{ag} + \int dE_c \frac{V_{ac}D_{ag}}{z-E_c}\right)}{z - E_a - \int dE_c \frac{|V_{ac}|^2}{z-E_c}} . \tag{4.13}$$

Under the assumption that the matrix elements under the integrals over E_c are slowly varying functions of E_c over the range of the resonance—an assumption that has also been employed in Sec. III B— z can be replaced by $E_g + \hbar\omega + i\epsilon$ (with $\epsilon \to 0+$) under the integrals. Thus using a well-known identity we take

$$\frac{1}{z-E_c} = \lim_{\epsilon\to 0+} \frac{1}{E_g + \hbar\omega - E_c + i\epsilon} = \mathrm{P}\int \frac{dE_c}{E_g + \hbar\omega - E_c} - i\pi\delta(E_g + \hbar\omega - E_c) , \tag{4.14}$$

often referred to as the "pole approximation." P denotes the principal value. Substitution into the integrals leads to shifts and widths. Thus we have

$$\int dE_c \frac{|D_{cg}|^2}{z-E_c} \simeq \mathrm{P}\int dE_c \frac{|D_{cg}|^2}{E_g + \hbar\omega - E_c} - i\pi\left|D_{cg}(E_g + \hbar\omega)\right|^2 \equiv S_g - i\tfrac{1}{2}\gamma_g , \tag{4.15}$$

$$\int dE_c \frac{|V_{ca}|^2}{E_g + \hbar\omega - E_c} \simeq \mathrm{P}\int dE_c \frac{|V_{cg}|^2}{E_g + \hbar\omega - E_c} - i\pi\left|V_{ca}(E_g + \hbar\omega)\right|^2 \equiv F_a - i\tfrac{1}{2}\Gamma_a , \tag{4.16}$$

$$D_{ag} + \int dE_c \frac{V_{ac}D_{cg}}{z-E_c} \simeq D_{ag} + \mathrm{P}\int dE_c \frac{V_{ac}D_{cg}}{E_g + \hbar\omega - E_c} - i\pi(V_{ac}D_{cg})_{E_g+\hbar\omega} \equiv \tilde{D}_{\tilde{E}g}(1 - i/q) . \tag{4.17}$$

The quantities S_g and γ_g introduced in Eq. (4.15), are, respectively, the shift and the ionization width of the state $|g\rangle$ due to direct transitions (virtual or real) into the continuum. In the formulation of Sec. III B, these quantities appeared in the form of the terms proportional to C_0. The other quantities appearing above, as for example, F_a, Γ_a, q, and $\tilde{D}_{\tilde{E}g}$, are identical to those defined in Secs. III A and III B. We will as before take these quantities as evaluated at $\tilde{E} = E'$. Note that the previous variable is the same as $E_g + \hbar\omega$. Using the above expressions it is now a matter of simple algebra to show that $\Lambda'(z)$ is identical to $\Lambda(x)$. All we need for this is to take

$$z - E_g - \hbar\omega = x , \tag{4.18}$$

which simply is a redefinition of z so as to be identical to the Laplace transform variable of Sec. III B. The translation defined by Eq. (4.18) is equivalent to the transformation of Eq. (3.5).

V. STRONG COUPLING OF TWO AUTOIONIZING RESONANCES

The next problem we want to formulate contains one additional autoionizing state but is considerably more complicated. It does, however, lead to a number of interesting new effects. Let $|b\rangle$ be a second bound state embedded in a continuum $|c_2\rangle$ and coupled to it through V. In general, $|a\rangle$ and $|b\rangle$ are assumed to be coupled to different continua. We will then label by $|c_1\rangle$ the continuum of $|a\rangle$. A second radiation field with n_2 photons and of frequency ω_2, near resonant with the transition $|a\rangle \to |b\rangle$ couples these two autoionizing resonances. It is

again assumed that a field of frequency ω_1 couples $|g\rangle$ to $|a\rangle$. Atoms can therefore be excited from $|g\rangle$ to $|a\rangle$ and from $|a\rangle$ to $|b\rangle$ which is assumed to lie above $|a\rangle$. Other excitations from the bound states to the various continua are also taking place as indicated in Fig. 1. In principle, one should keep all these couplings in the formalism since our purpose is to treat the problem for large field intensities. It may be possible in particular cases to neglect some of these channels. We begin, however, by considering the complete problem. Again, it can be done in either of the two formalisms discussed in the previous sections. We choose here the resolvent operator.

As in Sec. II, we take

$$P \equiv |g\rangle\langle g| , \tag{5.1a}$$

but Q is now somewhat different given by

$$Q \equiv |a\rangle\langle a| + |b\rangle\langle b| + \sum_{j=1}^{2} \int dE'_{c_j} |c_j\rangle\langle c_j| \tag{5.1b}$$

as it contains two bound states and two continua. The couplings between $|g\rangle$, $|a\rangle$, and the continuum $|c_1\rangle$ are determined by Eqs. (2.5). For the remaining couplings we have the equations

$$\langle b|H|b\rangle = \langle b|QHQ|b\rangle = \langle b|H^0|b\rangle = E'_b , \tag{5.2a}$$

$$\langle c_2|H|b\rangle = \langle c_2|QHQ|b\rangle = \langle c_2|V|b\rangle = V_{c_2 b} , \tag{5.2b}$$

$$\langle c_2|H|a\rangle = \langle c_2|QHQ|a\rangle = \langle c_2|D|a\rangle = D_{c_2 a} , \tag{5.2c}$$

$$\langle b|H|a\rangle = \langle b|QHQ|a\rangle = \langle b|D|a\rangle = D_{ba} , \tag{5.2d}$$

$$\langle c_1|H|b\rangle = \langle c_1|QHQ|b\rangle = \langle c_1|D|b\rangle = D_{c_1 b} , \tag{5.2e}$$

$$\langle c_2|H|c_1\rangle = \langle c_2|QHQ|c_1\rangle = \langle c_2|D|c_1\rangle = D_{c_2 c_1} , \tag{5.2f}$$

$$\langle c_2|H|c_2\rangle = \langle c_2|QHQ|c_2\rangle = \langle c_2|H^0|c'_2\rangle = E'_{c_2}\delta(E'_{c_2} - E'_{c'_2}) . \tag{5.2g}$$

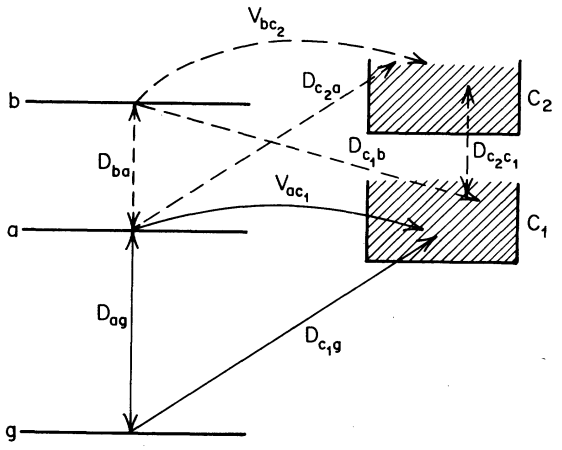

FIG. 1. Schematic representation of autoionizing states coupled to electromagnetic fields. D and V denote electromagnetic and configuration interactions, respectively. The shaded areas represent the continua to which the bound states a and b are coupled.

Since we are working with the resolvent operator, in the notation of Sec. IV, we must consider the states $|g;n_1,n_2\rangle$, $|a;n_1-1,n_2\rangle$, $|b;n_1-1,n_2-1\rangle$, $|c_1;n_1-1,n_2\rangle$, $|c_2;n_1-1,n_2-1\rangle$, with respective energies $E'_g = E_g + \hbar n_1\omega_1 + \hbar n_2\omega_2$, $E'_a = E_a + \hbar(n_1-1)\omega_1 + \hbar n_2\omega_2$, $E'_b = E_b + \hbar(n_1-1)\omega_1 + \hbar(n_2-1)\omega_2$, $E'_{c_1} = E_{c_1} + \hbar(n_1-1)\omega_1 + \hbar n_2\omega_2$, $E'_{c_2} = E_{c_2} + \hbar(n_1-1)\omega_1 + \hbar(n_2-1)\omega_2$, where as before the unprimed E denote atomic energies. Again, without further approximations we can take $E'_g = E_g + \hbar\omega_1 + \hbar\omega_2$, $E'_a = E_a + \hbar\omega_2$, $E'_{c_1} = E_{c_1} + \hbar\omega_2$, $E'_b = E_b$, and $E'_{c_2} = E_{c_2}$, because ultimately only differences between these energies will matter.

The total wave function now is of the form

$$\Psi(t) = U_{gg}(t)|g;n_1n_2\rangle + U_{ag}(t)|a;n_1-1;n_2\rangle + U_{bg}(t)|b;n_1-1;n_2-1\rangle + \int dE'_{c_1} U_{c_1 g}(t)|c_1;n_1-1;n_2\rangle + \int dE'_{c_2} U_{c_2 g}(t)|c_2;n_1-1;n_2-1\rangle . \tag{5.3}$$

As in Sec. IV, we obtain equations for the matrix elements of G corresponding to the above matrix elements of $U(t)$. This is accomplished by using Eqs. (4.7) in combination with Eqs. (5.2). The resulting equations are

$$(z - E'_g)G_{gg} - D_{ga}G_{ag} - \int dE'_{c_1} D_{gc_1} G_{c_1 g} = 1 , \tag{5.4a}$$

$$-D_{ag}G_{gg} + (z - E'_a)G_{ag} - \int dE'_{c_1} V_{ac_1} G_{c_1 g} - D_{ab}G_{bg} - \int dE'_{c_2} D_{ac_2} G_{c_2 g} = 0 , \tag{5.4b}$$

$$-D_{ba}G_{ag} + (z - E'_b)G_{bg} - \int dE'_{c_1} D_{bc_1} G_{c_1 g} - \int dE'_{c_2} V_{bc_2} G_{c_2 g} = 0 , \tag{5.4c}$$

$$-D_{c_1 g}G_{gg} - V_{c_1 a}G_{ag} - D_{c_1 b}G_{bg} + (z - E'_{c_1})G_{c_1 g} - \int dE'_{c_2} D_{c_1 c_2} G_{c_2 g} = 0 , \tag{5.4d}$$

$$-D_{c_2 a}G_{ag} - V_{c_2 b}G_{bg} + (z - E'_{c_2})G_{c_2 g} - \int dE'_{c_1} D_{c_2 c_1} G_{c_1 g} = 0 . \tag{5.4e}$$

This system of equations, although similar to that of Eqs. (4.8), contains an essential complication. The matrix elements $G_{c_1 g}$ and $G_{c_2 g}$ corresponding to the two continua are coupled through the laser fields. The coupling is represented by the last term in each of the last two equations. Its strength is characterized by the continuum-continuum (cc) matrix element $D_{c_1 c_2}$ which, in principle, will be proportional to the sum of both fields. If that coupling is very strong—in the sense that higher cc transitions must be included—the equations cannot be decoupled. In that regime of intensities (perhaps above 10^{14} W/cm^2) the whole model which is based on a finite number of bound states may become unreliable. There is, however, a range of intensities in which one can obtain solutions. We will postpone the discussion of more general solutions to another paper. Here we confine ourselves to the discussion of certain particular cases which illustrate the type of new effects to be expected from the coupling of autoionizing resonances with strong fields.

Depending on the particulars of the problem at hand, one may choose from a hierarchy of approximations that lead to a decoupling of $G_{c_1 g}$ and $G_{c_2 g}$ to various levels of accuracy. To be more specific, let us first note that by setting $D_{c_2 c_1} = 0$ in Eqs. (5.4d) and (5.4e) the two continua are decoupled. This, however, is more drastic than necessary as can be demonstrated by solving Eq. (5.4e) for $G_{c_2 g}$, substituting into Eq. (5.4d) and then solving it for $G_{c_1 g}$ thus obtaining

$$G_{c_2 g} = \frac{1}{z - E'_{c_2}} (D_{c_2 a}G_{ag} + V_{c_2 b}G_{bg}) + \frac{1}{z - E'_{c_2}} \int dE'_{c_1} D_{c_2 c_1} G_{c_1 g} , \tag{5.5}$$

$$G_{c_1 g} = \frac{1}{z - E'_{c_1}} (D_{c_1 g}G_{gg} + V_{c_1 a}G_{ag} + D_{c_1 b}G_{bg}) + \frac{1}{z - E'_{c}} \int dE'_{c_2} \frac{D_{c_1 c_2}}{z - E'_{c_2}} (D_{c_2 a}G_{ag} + V_{c_2 b}G_{bg}) + \frac{1}{z - E'_{c_1}} \int dE'_{c_2} \frac{D_{c_1 c_2}}{z - E'_{c_2}} \int dE'_{c_1} D_{c_2 c_1} G_{c_1 g} . \tag{5.6}$$

Now, as far as decoupling $G_{c_2 g}$ and $G_{c_1 g}$ is concerned, all we need to do is neglect the last term in Eq. (5.6). The result is not equivalent to having set $D_{c_2 c_1} = 0$ in Eqs. (5.4) because $D_{c_2 c_1}$ still appears in the second term of Eq. (5.6). Obviously, how important these various contributions are will in each case vary, depending on the strength of the $c_2 c_1$ matrix elements as compared to the other channels. Part of the interest in the processes discussed in this paper stems from the possibility they afford of exploring these questions.

As experience with multiphoton processes has shown, cc transitions even under strong lasers become important only above certain intensities. It is reasonable to expect therefore that in various intensity regimes one will be able to study effects that do or do not depend on $D_{c_2 c_1}$ in a significant way. In this paper we show results that do not depend on $D_{c_2 c_1}$ while in a follow-up paper we show how to incorporate its influence when necessary. For the moment we simply point out that the structure introduced in the continuum by the autoionizing resonances makes the solution of the general integral equation (5.6) easier (at least formally) than in the general case. In the remainder of this section we confine attention to the case of $D_{c_2 c_1} \to 0$. Thus from both Eq. (5.5) and (5.6) we retain only the first group of terms in their right-hand sides. We substitute then into Eqs. (5.4a)–(5.4c) and after some rearrangement and grouping of terms we arrive at the equations

$$\left(z + \frac{i}{2}\gamma_g\right)G_{gg} - \bar{D}_{ga}G_{ag} - \overline{D^{(2)}_{gb}}G_{bg} = 1, \tag{5.7a}$$

$$-\bar{D}_{ag}G_{gg} + \left(z + \delta_1 + \frac{i}{2}\gamma_a + \frac{i}{2}\Gamma_a\right)G_{ag} - \bar{D}_{ab}G_{bg} = 0, \tag{5.7b}$$

$$-\overline{D^{(2)}_{bg}}G_{gg} - \bar{D}_{ba}G_{ag} + \left(z + \delta_1 + \delta_2 + \frac{i}{2}\gamma_b + \frac{i}{2}\Gamma_b\right)G_{bg} = 0, \tag{5.7c}$$

where we have introduced the abbreviations

$$\bar{D}_{ga} = D_{ga} + \mathrm{P}\int dE'_{c_1} \frac{D_{gc_1}V_{c_1 a}}{E'_g - E'_{c_1}} - i\pi(D_{gc_1}V_{c_1 a})_{E'_g} , \tag{5.8a}$$

$$\overline{D^{(2)}_{gb}} = \mathrm{P}\int dE'_{c_1} \frac{D_{gc_1}D_{c_1 b}}{E'_g - E'_{c_1}} - i\pi(D_{gc_1}D_{c_1 b})_{E'_g} , \tag{5.8b}$$

$$\bar{D}_{ab} = D_{ab} + \mathrm{P}\int dE'_{c_1} \frac{V_{ac_1} D_{c_1 b}}{E'_g - E'_{c_1}} - i\pi (V_{ac_1} D_{c_1 b})_{E'_g}$$

$$+ \mathrm{P}\int dE'_{c_2} \frac{D_{ac_2} V_{c_2 b}}{E'_g - E'_{c_2}} - i\pi (D_{ac_2} V_{c_2 b})_{E'_g} . \qquad (5.8c)$$

To obtain $\bar{D}_{ag}$ from $\bar{D}_{ga}$, we simply transpose the indices in all matrix elements but do *not* take the complex conjugate of the whole expression. In an analogous way we obtain $\bar{D}_{ba}$ and $\bar{D}^{(2)}_{bg}$. The widths appearing in the above equations are defined by

$$\pi |D_{c_1 g}|^2_{E'_g} = \tfrac{1}{2}\gamma_g , \quad \mathrm{P}\int dE'_{c_1} \frac{|D_{c_1 g}|^2}{E'_g - E'_{c_1}} \equiv S_g , \qquad (5.9)$$

$$\pi |D_{c_2 a}|^2_{E'_g} \equiv \tfrac{1}{2}\gamma_a , \quad \mathrm{P}\int dE'_{c_2} \frac{|D_{c_2 a}|^2}{E'_g - E'_{c_2}} \equiv S_a , \qquad (5.10)$$

$$\pi |D_{c_1 b}|^2_{E'_g} \equiv \tfrac{1}{2}\gamma_b , \quad \mathrm{P}\int dE'_{c_1} \frac{|D_{c_1 b}|^2}{E'_g - E'_{c_1}} \equiv S_b , \qquad (5.11)$$

$$\pi |V_{c_1 a}|^2_{E'_g} \equiv \tfrac{1}{2}\Gamma_a , \quad \mathrm{P}\int dE'_{c_1} \frac{|V_{c_1 a}|^2}{E'_g - E'_{c_1}} \equiv F_a , \qquad (5.12)$$

$$\pi |V_{c_2 b}|^2_{E'_g} \equiv \tfrac{1}{2}\Gamma_b , \quad \mathrm{P}\int dE'_{c_2} \frac{|V_{c_2 b}|^2}{E'_g - E'_{c_2}} \equiv F_b , \qquad (5.13)$$

where the γ's and S's represent widths and shifts of the respective states induced by the field via coupling to one of the continua while the Γ's and F's are autoionization widths and shifts (see also Sec. II). The shifts do not appear explicitly in the equations (5.7) because they have been incorporated in the detunings which are defined by

$$\delta_1 \equiv \hbar\omega_1 - [(E_a + F_a + S_a) - (E_g + S_g)], \qquad (5.14a)$$

$$\delta_2 \equiv \hbar\omega_2 - [(E_b + F_b + S_b) - (E_a + F_a + S_a)]. \qquad (5.14b)$$

Since they contain the intensity-dependent shifts (which are proportional to the light intensity) these detunings must be understood as dynamic and not static detunings. They will change with changing intensity. As we shall see subsequently, some of the most interesting effects are associated with these shifts. In arriving at these equations, we have used the procedure of Sec. V with the same type of approximations. As in that section, the widths and shifts have been calculated at the energy E'_g a convenient but not necessary approximation.

The general solution of the system of Eqs. (5.7) leads to a third degree algebraic equation with respect to z. Thus analytic solutions for the general case, although possible, do not give much insight. They are, however, useful in special cases when, for example, some of the couplings (channels) are stronger than others. In all cases, the total ionization for an interaction time T is given by the equation

$$P(T) = 1 - |U_{gg}(T)|^2 - |U_{ag}(T)|^2 - |U_{bg}(T)|^2 , \qquad (5.15a)$$

while for times $t > T$, it is given by

$$P(t) = 1 - |U_{gg}(T)|^2 - |U_{ag}(T)|^2 e^{-\Gamma_a (t-T)} - |U_{bg}(T)|^2 e^{-\Gamma_b (t-T)} . \qquad (5.15b)$$

Again, in the limit of $\Gamma_a(t-T) \gg 1$ and $\Gamma_b(t-T) \gg 1$ the expression reduces to

$$P = 1 - |U_{gg}(T)|^2 . \qquad (5.15c)$$

If, of course, either $|a\rangle$ or $|b\rangle$ are not autoionizing, the corresponding Γ must be set equal to zero and the corresponding population will appear in Eq. (5.15c). An equivalent expression—a direct generalization of Eq. (3.9)—involving only the amplitudes of the continuum is given by

$$P(T) = \sum_{j=1}^{2} \int dE_{c_j} |U_{c_j g}(T)|^2 . \qquad (5.16)$$

To explore a case in which analytical expressions are feasible and useful, and also to show how these general equations reduce to more familiar results, we consider next the limit in which $|a\rangle$ is a bound state (or an extremely narrow autoionizing resonance) while $|b\rangle$ remains an autoionizing resonance. Under these conditions, we neglect the continuum c_1 completely. As a consequence $\bar{D}^{(2)}_{gb} = 0$ and also $\gamma_g = 0$, $s_g = 0$, $F_a = 0$, and $\Gamma_a = 0$ and $\gamma_b = 0$. The set of Eqs. (5.7) then becomes

$$z G_{gg} - \Omega_1 G_{ag} = 1 , \qquad (5.17a)$$

$$-\Omega_1 G_{gg} + \left(z + \delta_1 + \frac{i}{2}\gamma_a\right) G_{ag} - \tilde{\Omega}_2 \left(1 - \frac{i}{q_b}\right) G_{bg} = 0 , \qquad (5.17b)$$

$$-\tilde{\Omega}_2 \left(1 - \frac{i}{q_b}\right) G_{ag} + \left(z + \delta_1 + \delta_2 + \frac{i}{2}\Gamma_b\right) G_{bg} = 0 , \qquad (5.17c)$$

where in accordance with the notation of previous sections we have defined

$$\Omega_1 \equiv D_{ag} , \qquad (5.18a)$$

$$\bar{D}_{ba} = D_{ba} + \mathrm{P}\int dE'_{c_2} \frac{V_{bc_2} D_{c_2 a}}{E'_g - E'_{c_2}} - i\pi (V_{bc_2} D_{c_2 a})_{E'_g}$$

$$\equiv \tilde{D}_{\tilde{E}a} \left(1 - \frac{i}{q_b}\right) , \qquad (5.18b)$$

and

$$\tilde{D}_{\tilde{E}a} \equiv \tilde{\Omega}_2 . \qquad (5.18c)$$

Now Ω_1 is a conventional Rabi frequency between two bound states while $\tilde{\Omega}_2$ is an effective Rabi

frequency like the $\tilde{\Omega}$ of Sec. III. The q_b in Eq. (5.18b) is the q of autoionizing resonance b with a definition analogous to that of Eq. (2.17). The system of Eqs. (5.17) also leads to an algebraic equation of third degree but of a somewhat simpler form.

An even more restricted case of this system, for which analytic solutions can be obtained, corresponds to the so-called weak-probe approximation. This means that one of the fields is weak while the other is strong. If the weak field is tuned around the exact resonant frequency of its transition, the total ionization signal reflects (probes) the effect of the strong field on the transition it couples. We take here the first field as weak. It follows then that the first Rabi frequency Ω_1 is small in the sense $\Omega_1 \ll \gamma_a$, Γ_b, $\tilde{\Omega}_2$. This approximation allows us to obtain an analytic solution for the rate of total ionization given by

$$\frac{dP}{dt} = \Omega_1^2 \gamma_a \frac{(\delta_1+\delta_2+\frac{1}{2}\Gamma_b q_b)^2}{|f(\delta_1)|^2}, \tag{5.19a}$$

where

$$f(\delta_1) = \left(\delta_1+\frac{i}{2}\gamma_a\right)\left(\delta_1+\delta_2+\frac{i}{2}\Gamma_b\right)-\tilde{\Omega}_2^2\left(1-\frac{i}{q_b}\right)^2. \tag{5.19b}$$

It is because of the weak-probe approximation that a constant (time-independent) transition rate can be obtained for long times; in the sense that $\gamma_a T$, $\Gamma_b T$, $\Omega_2 T \gg 1$ and the transients have died out. Physically, the weak-probe situation corresponds to the excitation of very few atoms (per unit time) from $|g\rangle$ to $|a\rangle$. But once an atom is excited to $|a\rangle$ it is very quickly snatched away by the second (strong) field and its further development is determined by the particulars of the transition $|a\rangle \rightarrowtail |b\rangle$ and the continuum $|c_2\rangle$. The rate is determined by the weak (bottleneck) transition. One can investigate even more specialized cases corresponding to specific relations between the parameters of Eqs. (5.19). Such examples are discussed in Sec. VI.

VI. NUMERICAL RESULTS

Having established a formal framework for the coupling of autoionizing states to strong fields and explored some of its general predictions we turn now to the examination of certain quantitative predictions which provide illustrations of the effects to be expected. We begin by showing the effect of the time of interaction on the line profile of an autoionizing resonance. This is never discussed in standard treatments of autoionization because it does not matter. As long as the field is sufficiently weak, we have a time-independent transition probability per unit time and it is always assumed that the interaction time is sufficiently long for this transition probability to be meaningful; a condition always satisfied in traditional spectroscopy of photoionization. If, however, the light intensity is large or its duration short, the effect is important, as illustrated in Fig. 2. We have plotted total ionization as a function of detuning from resonance for a constant light intensity but different times of interaction. For this example, we have chosen a value $q = 5$ for the resonance and an intensity such that $\tilde{\Omega} = \Gamma$; i.e., the Rabi frequency $\tilde{\Omega}$ is equal to the autoionization width Γ. This certainly is not a weak but not a very strong intensity either. The five curves of the figure correspond to interaction times T ranging from $\Gamma T = 1$ to 20. These curves show that as long as $T \gtrsim 5\Gamma^{-1}$, the typical autoionization profile has developed with its distinct minimum and asymmetric shape. For small T, such as $T = \Gamma^{-1}$, the curve does not resemble at all the well known profile; it is flat. The detuning $\tilde{\Delta}$ is the dynamic detuning which includes the shift. This ac Stark shift also is something that is unimportant in weak fields but plays a significant role when the field becomes strong.

In the next example (Fig. 3) we take a long (in the above sense) time of interaction $T = 5\Gamma^{-1}$ and the same $q = 5$. Again we have plotted total ionization as a function of detuning from exact resonance for various values of intensity. The first curve ($\tilde{\Omega} = 0.25\Gamma$) corresponds to weak intensity while the last one ($\tilde{\Omega} = 10\Gamma$) to rather strong intensity for which the modified Rabi frequency is ten

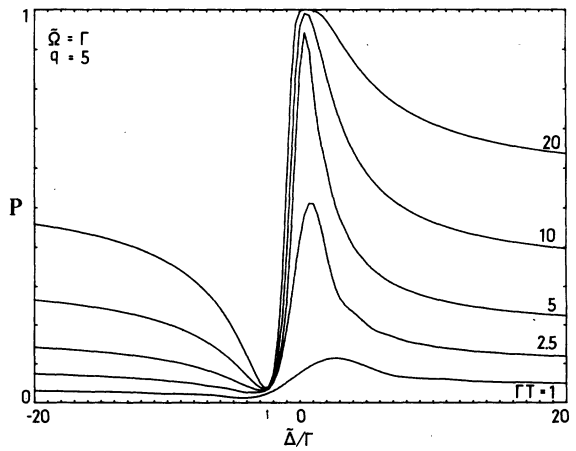

FIG. 2. Line shape of an autoionizing resonance as a function of the time of interaction between light and atom. Γ is the autoionization width, $\tilde{\Omega}$ the Rabi frequency, and $\tilde{\Delta}$ the dynamic detuning, as defined in the text. The arrow indicates the position of the field-free minimum.

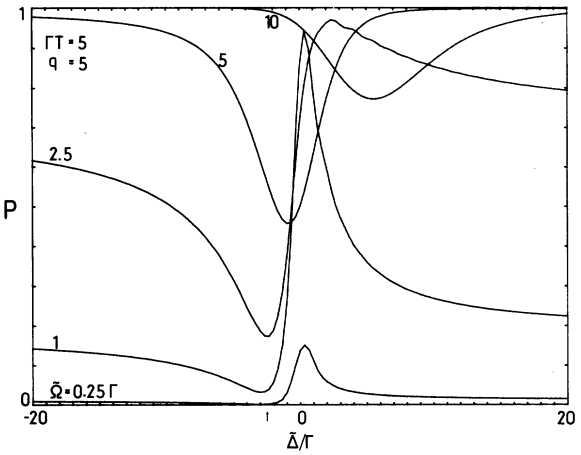

FIG. 3. Line shape of an autoionizing resonance as a function of field intensity. $\tilde{\Delta}$ is the dynamic detuning and the arrow indicates the position of the field-free minimum. The interaction time is $T = 5\Gamma^{-1}$.

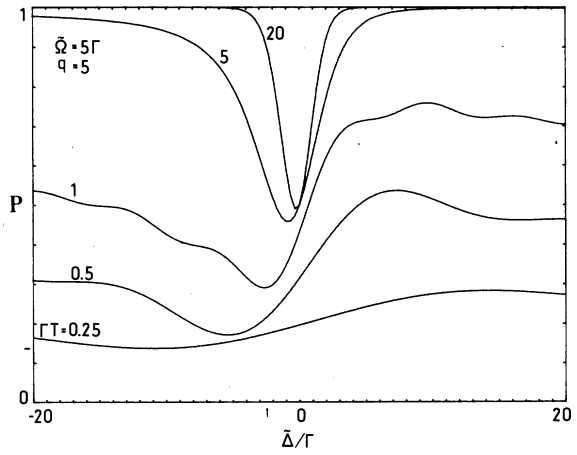

FIG. 4. Line shape of an autoionizing resonance as a function of interaction time for relatively strong field, i.e., $\tilde{\Omega} = 5\Gamma$. Arrow indicates the position of the field-free minimum.

times larger than the autoionization width. Here we see the first effect of the intensity. For $\tilde{\Omega} = \Gamma$ the line shape has the typical form, for $\tilde{\Omega} = 2.5\Gamma$ it still has a minimum and a maximum which however is now much less peaked, while for $\tilde{\Omega} = 5\Gamma$ the maximum no longer exists and the minimum is much shallower. And this trend continues for higher values of $\tilde{\Omega}$. This means that the probability of ionization tends to one, no matter what the detuning is. There is a region, however, where the probability increases much more slowly; namely, around the minimum. But the minimum does not remain in its weak-field position. It shifts by about 7Γ as $\tilde{\Omega}$ changes from 0.25Γ to 10Γ; that is as the field strength increases by a factor of 40. The existence of the minimum in the first place is the result of interference between the direct ionization and the indirect (via the configuration interaction) channels. The intensity modifies this interference because it affects some of the transitions but not the configuration interaction. Thus it changes the relative strengths. This shifting of the minimum will occur whether there is significant ac Stark shift of the resonance or not and must be viewed as a separate effect although both are present simultaneously.

Having seen how the intensity affects the line shape, we return once more to the effect of the time of interaction but this time combined with relatively strong field. In Fig. 4 we present the ionization profile for intensity such that $\tilde{\Omega} = 5\Gamma$ and as in Fig. 2 for times ranging from $\Gamma T = 0.25$ to 20. None of these curves now exhibits the typical profile. Even a moderately strong intensity has a profound effect on the line shape. The contrast is glaring when we compare one by one the curves of this figure with those of Fig. 2. The undulations evident especially in the curve for $T = 1$ are due to Rabi oscillations of the atom between states $|g\rangle$ and $|a\rangle$. Such undulations tend to be smoothed by the decay mechanisms (in this case ionization) of the process. It is only when a particular combination of interaction time and intensity conspire that they become evident on such curves. In other words, it is a particular relation between $\tilde{\Omega}$ and T that leads to these undulations. Typically, they will appear when T is equal to a few Γ^{-1}. But this should be viewed as a rough rule and not as an exact relation.

For weak fields, autoionization is proportional to the laser intensity. The process is then described by a transition probability per unit time linear in the light intensity. Thus the slope of the curve $\log P = f(I)$ (with P being the total ionization and I the photon flux) is one. The situation changes considerably when the field becomes strong. First of all, the process is not necessarily proportional to intensity. In addition, the resonant states $|g\rangle$ and $|a\rangle$ undergo ac Stark shifts which are proportional to the light intensity. The magnitudes of the shifts depend on the particular atom. But in any case, they increase linearly with light intensity and eventually become larger than Γ. It must also be kept in mind that the shift increases faster than $\tilde{\Omega}$ which is proportional to $I^{1/2}$. One of the many consequences of the shifts is that they alter the dependence of the total ionization on light intensity. We have already seen how the shifts enter in the detuning which thus becomes intensity dependent. If the laser frequency is at a certain detuning for weak inten-

sity, this detuning will change as the intensity rises. The atom may then shift either closer to or farther from resonance with the laser frequency. Mathematically, this means that the intensity appears in a complicated manner in the denominator which spoils the simple intensity dependence known from the weak-field situation. As we have already seen in previous sections, it is the difference between the shifts of $|g\rangle$ and $|a\rangle$ that matters and not the shifts of the individual states. Because this difference is what determines how much the frequency of the transition $|g\rangle \leftrightarrow |a\rangle$ changes. Obviously it is for detunings around $\Delta \simeq 0$ that the influence of the shifts on the intensity dependence will be the greatest because even small changes are then significant. These effects are shown in Fig. 5 where we have plotted $\partial 1gP/\partial 1gI$ as a function of detuning for four different situations, all however corresponding to an intensity such that $\Omega = \Gamma$ and $T = 5/\Gamma$. For large detunings, the slope approaches one, but around $\Delta = 0$ it undergoes significant changes and depending on the particular combination of parameters its value is seen to range from 2 to -2. It must be stressed here that these slopes correspond to this particular intensity. In general they will change with intensity since the function $P(I)$ is not necessarily represented by a simple power of I. Comparing Fig. 5(a) with Fig. 5(b) we see how the q value of the resonance affects the slope, while comparing 5(b) we see how the sign of the total shift affects the slopes. But even these comparisons give only a very small part of the whole picture. We have a multiparameter problem and a change of one of these parameters may significantly affect the relative effect of the others. For example, the change between 5(a) and 5(c) is less pronounced than the change between 5(b) and 5(d).

A plot that gives a pictorial sense of how ionization proceeds in time is given in Fig. 6. The total probability of ionization as a function of the time of interaction is plotted together with the occupation probability of states $|g\rangle$ and $|a\rangle$. These three probabilities add up to one [see Eq. (3.8)] and the oscillations in these curves reflect the (Rabi) oscillation of the atom between the resonantly coupled states $|g\rangle$ and $|a\rangle$. These oscillations are damped by ionization in a few ΓT's. This behavior is well known from the study of bound states coupled by an intense field.

Our final figure (7) has to do with the coupling of two autoionizing resonances by a strong field. We consider the case discussed in Sec. V and depicted in Fig. 1. Here we choose the first autoionizing resonance $|a\rangle$ very narrow and equivalent to a bound state. It is assumed coupled to the ground state $|g\rangle$ by a weak field while a second field, whose intensity we vary from weak to quite strong, couples $|a\rangle$ with a higher autoionizing resonance $|b\rangle$ which is characterized by $q = 5$ and an autoionization width Γ. The second field is assumed to be exactly resonant with the frequency of the transition $|a\rangle \leftrightarrow |b\rangle$. We then scan the frequency of the first (weak probe) field and plot total ionization as a function of its frequency ω, expressed in terms of its detuning Δ' from the resonance $|g\rangle \leftrightarrow |a\rangle$. When both fields are weak—as in the first curve of Fig. 7 where the Rabi frequency of the coupling $|a\rangle \leftrightarrow |b\rangle$ is $\Omega = 0.5\Gamma$—we have one peak at $\Delta' = 0$. This simply means that the atom can ionize only as long as the first photon has the energy to raise it from $|g\rangle$ to $|a\rangle$ from where it is then excited to $|b\rangle$ and to the continuum by the second resonant photon. The curve has

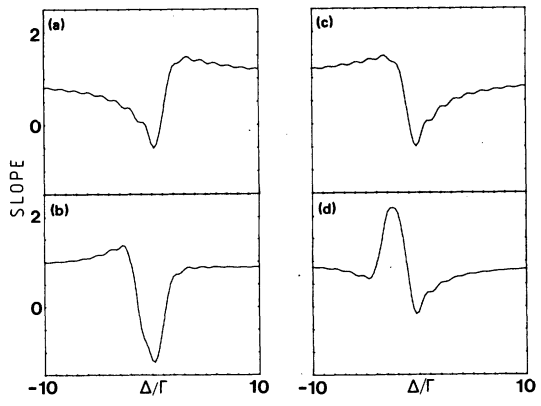

FIG. 5. The index of nonlinearity of ionization through an autoionizing resonance for a field such that $\tilde{\Omega} = \Gamma$. The interaction time is $T = 5\Gamma^{-1}$. The figure illustrates the effects of the shift. (a) $S = \Gamma$, $q = 10^6$; (b) $S = \Gamma$, $q = 5$; (c) $S = -\Gamma$, $q = 10^6$; (d) $S = -\Gamma$, $q = 5$.

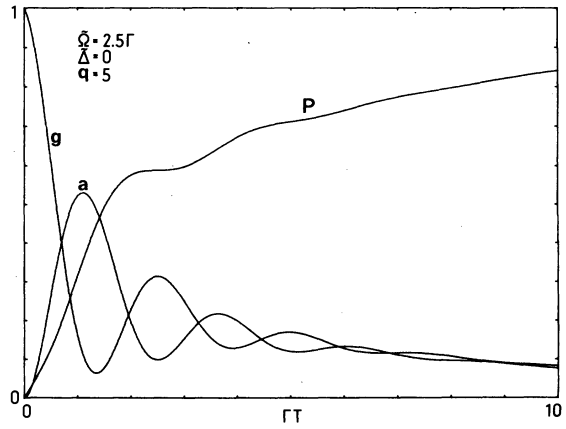

FIG. 6. Evolution in time of the populations of states g and a and of the photoionization signal for a resonant field ($\tilde{\Delta} = 0$) of strength such that $\tilde{\Omega} = 2.5\Gamma$.

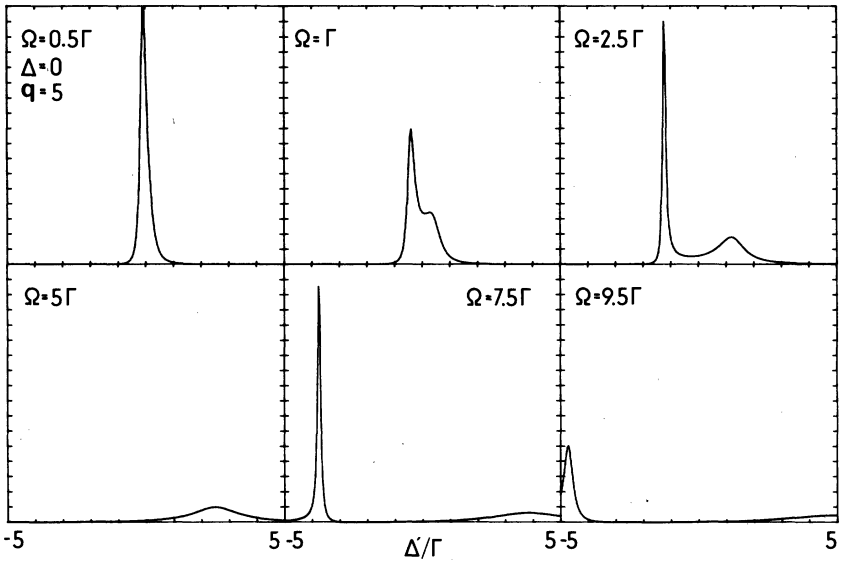

FIG. 7. ac Stark splitting of an autoionizing resonance in double optical resonance. The strong field is exactly on resonance ($\Delta=0$) and its strength is varied from $\Omega=0.5\Gamma$ to 9.5Γ. The detuning Δ' of the weak (probe) field is measured in units of autoionization width Γ. The calculation has been performed in the limit of a steady state. Stark shifts have been neglected.

width Γ because state $|b\rangle$ is broadened by autoionization. If the reader is concerned about the curve not being asymmetric, we note that it is Δ' and not Δ that is varied. By having fixed Δ at exact resonance with the $|a\rangle \rightarrow |b\rangle$ transition, we are at the center of the asymmetric profile which, for weak fields, would become apparent only if we scanned Δ and kept Δ' at resonance. The dynamics of the process is there nevertheless and is manifested even for Δ fixed at zero when the second field becomes strong. This is clearly seen in the third curve which is calculated for $\Omega=2.5\Gamma$. The transition from a single peak to a two-peak curve was to be expected since it reflects the ac Stark splitting of the transition $|a\rangle \rightarrow |b\rangle$ due to the strong field. We have a situation quite similar to ac Stark splitting in double optical resonance in a system with three bound states. As in that situation, the two peaks are separated by Ω the Rabi frequency. But the shape is now completely different; the two peaks are of unequal heights and widths. If state $|b\rangle$ simply decayed out of the space of the three coupled states with a rate Γ, the two peaks would have equal heights and widths $\Gamma/2$ as long as $\Delta=0$. Further increase of the intensity and hence of Ω would simply move them farther and farther apart but would not cause the asymmetry seeing in Fig. 7. Asymmetric peaks in that case[11,17] would occur only if $\Delta\neq 0$. The novel features we see here are due to the interference inherent in autoionization; the interference between direct ionization of $|a\rangle$ and ionization via $|b\rangle$ and its configuration interaction. How unusual this behavior is becomes more contrastingly evident in the fourth curve where for $\Omega=5\Gamma$ one of the peaks is absent, to reappear at higher intensities. From this sequence of curves we can piece together the underlying effect. Interference causes the two peaks to have unequal widths and as the intensity increases one of the peaks continues to narrow while the other continues to broaden. From a different viewpoint, this means that of the two dressed states created by the strong coupling one acquires a short and the other a long lifetime against ionization. At some critical intensity, the interference is just right for the width of the narrow state to become zero which means that it becomes stable against ionization. Thus we have the rather unexpected result of an autoionizing system becoming stabler with increasing laser intensity. Physically, this means that as the atom is brought to $|b\rangle$ by the second field, instead of autoionizing from there, because of the configuration coupling, it is brought back to $|a\rangle$ by the field thus lengthening its lifetime. The bound character of $|b\rangle$ is thus enhanced by the field. This of course is related to the fact that the strong field couples $|b\rangle$ to a bound state $|a\rangle$ whose autoionization rate we have assumed negligible. Without going into further detail in this paper, we simply point out that if we had kept the autoionization width of $|a\rangle$ and it was much smaller than that of $|b\rangle$ we would again have found one of

the components becoming narrower up to a certain intensity but the width would not go to zero. The last two graphs of Fig. 7 show both peaks becoming broader and broader as the intensity increases beyond the critical intensity $\Omega = 5\Gamma$. At the same time the centers of the peaks move farther apart as they are expected to do in any case of ac Stark splitting.

The calculations of this section have been performed under the assumption of a square pulse (suddenly switched on). As is well known from work in related contexts,[18,19] certain details will change if the pulse is switched on gradually, as in fact is the case in real experimental situations. We shall present an elaboration of this point in a follow-up paper.

VII. CONCLUDING REMARKS

Our main objective in this paper has been the study of the general features of autoionizing states coupled to strong laser fields. We have presented a fairly detailed description of the formalism because in subsequent papers we shall have to rely on it in order to discuss many new aspects of the problem. From the results presented in the previous sections, it is evident that the line shape undergoes severe distortion. When more than one autoionizing state is involved, the resulting phenomenon of ac Stark splitting exhibits behavior which is very different from the corresponding behavior of bound states. In fact, the novel features of this effect have many more unusual aspects as we shall show in our next paper.

Aside from questions pertaining to laser spectroscopy of autoionizing states, the problems formulated and solved in this paper are also relevant in other contexts. Most likely, they will eventually become of interest in the multiphoton ionization of alkaline earth atoms[10] when more detailed experimental information is obtained. The participation of autoionizing states in harmonic generation,[21] or more generally in wave mixing, is a problem that also will require further theoreticl understanding on a systematic basis. Equally important will be the investigation of the dynamics of lasing transitions via autoionizing resonances based on the phenomenon of radiative autoionization.[22-24] A scheme for generating coherent radiation through this process has recently been proposed.[23] If a laser is to operate via an autoionizing state the details of line-shape distortion because of saturation are very important in determining its characteristics. Finally, processes such as those depicted in Fig. 1 are related, albeit somewhat indirectly, to continuum-continuum transitions under strong fields.[25-27] This point is discussed in more detail in Ref. 12.

During the last five years or so, we have seen intense activity on phenomena resulting from the strong coupling of bound states with laser fields. Significant advances have been made in the theoretical understanding of the related effects[11,16,17] and experiments of increasing refinement are still underway. The analogous situation in relation to autoionizing states contains even larger variety and promises to prove even richer in new and interesting phenomena.

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APPENDIX A

In this Appendix, we give a brief account of the calculations that lead to Eq. (3.19). Beginning with Eq. (3.7a) and the definitions of $\sin\Delta$ and $\cos\Delta$ we write $|M_{\tilde{\omega}_g}|^2$ as

$$\delta\hbar^2 |M_{\tilde{\omega}_g}|^2 = \left[|\tilde{D}_{\tilde{E}g}|^2 |V_{\tilde{E}}|^2 + |D_{cg}|^2 (\tilde{E}-\overline{E}_a)^2 + |V_{\tilde{E}}|^2 2\,\mathrm{Re}\left(\frac{\tilde{D}_{\tilde{E}g} D_{cg}}{V_{\tilde{E}}}\right)(\tilde{E}-\overline{E}_a)\right] [(\tilde{E}-\overline{E}_a)^2 + \pi^2 |V_{\tilde{E}}|^4]^{-1}$$

$$\equiv y(\tilde{E})[(\tilde{E}-\overline{E}_a)^2 + \pi^2 |V_{\tilde{E}}|^4]^{-1}, \tag{A1}$$

which defines $y(\tilde{E})$. With this expression for $|M_{\tilde{\omega}_g}|^2$ the quantity $s(x) - i\gamma(x)$ appearing in Eq. (3.18) is written as

$$s(x) - i\gamma(x) = \mathrm{P}\int_{-\overline{\omega}_a}^{\infty} d\tilde{\omega}\, \frac{y(\tilde{\omega})}{(x-\tilde{\omega})[(\tilde{\omega}-\overline{\omega}_a)^2 + \kappa^2]} - i\pi \frac{y(x)}{(x-\overline{\omega}_a)^2 + \kappa^2}, \tag{A2}$$

where $\tilde{\omega} = \tilde{E}/\hbar$. If the matrix elements of D and V are assumed to be slowly varying (compared to the variation of the Lorentzian), $y(\tilde{\omega})$ becomes a polynomial of $(\tilde{\omega} - \overline{\omega}_a)$. Then the integral with the principal value can be calculated exactly using known expressions for definite integrals of this type. It is nevertheless important for the coefficient of $(\tilde{\omega} - \overline{\omega}_a)^2$ to go to zero sufficiently fast as $\tilde{\omega} \to \infty$. The term $|\tilde{D}_{\tilde{E}g}|^2 |V_{\tilde{E}}|^2$ also has similar

behavior for $\tilde{\omega} \to \infty$. Thus we write $y(\tilde{\omega})$ in the abbreviated form

$$y(\tilde{\omega}) = A(\tilde{\omega}) + B(\tilde{\omega})(\tilde{\omega} - \tilde{\omega}_a) + C(\tilde{\omega})(\tilde{\omega} - \tilde{\omega}_a)^2, \quad \text{(A3)}$$

where the definitions of A and B become obvious by comparing Eq. (A3) with Eq. (A1). In computing the integrals, the lower limit can be set equal to $-\infty$ in all of them except one which can be put in the dimensionless form

$$\int_{-\omega_a/\kappa}^{\infty} d\lambda \, \frac{\lambda b(\lambda)}{\lambda^2 + 1} \equiv N, \quad \text{(A4)}$$

where N is a pure number, $b(\lambda)$ is defined by $B(\tilde{\omega})/B_0$, B_0 by $B_0 = B(\tilde{\omega} = \omega_g + \omega)$, and λ by $\lambda = \tilde{\omega}/\kappa$. The meaning of the subscript 0 is the same as in Sec. III. The number N depends on how fast the bound-free matrix element μ_{cg} goes to zero as $E_c \to \infty$ and determines the magnitude of the laser-induced ac Stark shift of the state $|g\rangle$ due to its direct coupling with the continuum. For the rest of the integrals, it suffices to replace $A(\tilde{\omega})$ and $B(\tilde{\omega})$ by their values at $\tilde{\omega} = \omega_g + \omega$ thus obtaining the results shown in Eqs. (3.19) and (3.20).

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