Theoretical aspects and experimental consequences of multiple photon-dressed discrete states in the atomic or molecular continuum

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The creation of multiple photon-dressed discrete states in the continuum by coupling N levels to a single resonance (atomic or molecular) is discussed. The conditions for the appearance of such states are established along with the experimental consequences that would reveal their presence. These include time-resolved ionization measurements as well as analysis of ionization yield as a function of frequencies.

I. INTRODUCTION

This paper concerns coupling of a single resonance to N levels of an atomic (or molecular) system by external electromagnetic fields and is to be considered as an extension and completion of our previous communication.[1] Previously, a two-level system with a resonance was considered. Here we generalize the analysis to a system with N bound states and a resonance and show that under proper conditions there exists N photon-dressed discrete states embedded in the dressed continuum. We also discuss the significance of these states, their measurability, and effects that they produce under normal spectroscopic conditions.

Before proceeding on to the subject matter, a short preamble to this problem will not be out of context. In quantum mechanics the subject of creating a bound state in the continuum part of a spectrum has a long history.[2–5] It has been shown how model Hamiltonians may be constructed which support a discrete state in the continuum. While apparently of mathematical interest, their utility for physical systems has been speculated for some time. Also, mathematical literature has been replete with the problem of the existence of bound states in the continuum.[6] Our aim in this paper is not to address this issue directly. We simply show that under very general conditions dressing of isolated systems by photons may result in generation (to a very good approximation) of such discrete states. The key issue is to find these conditions, since as is well known from any consideration of the theory of photoionization or photodissociation, we have usually only irreversible processes. If one takes a single bound state coupled to a continuum by a matrix element which slowly varies in energy (the usual condition for real systems), it is hard to produce anything other than the almost Lorentzian dissolution of the bound state in the diagonalized (dressed) continuum. In the time domain, this means that the population of the initial bound state decays exponentially. However, if the continuum contains a resonance (due to autoionization or predissociation) the diagonalization produces new structures, which could be in principle detected looking at the energy distribution of electrons or fragments, for fixed photon energy (the distribution is expected to be non-Lorentzian), or at the time dependence of the depletion of the initial state.

In the next section, we treat the general problem of N levels interacting with the continuum containing a resonance with N different frequencies. We show that, in general, it is possible to create N distinct photon-dressed discrete states for such a system. The conditions under which this happens will be discussed along with the general demonstration.

We go on to consider some special cases. Firstly, we consider the radiative coupling of a bound state to a resonance; secondly, the coupling by two electromagnetic (e.m.) fields of a pair of bound states to a flat (i.e., structureless) continuum; and finally, a resonance coupled to two bound states by two e.m. fields.

Thus our treatment unifies and generalizes all these above-mentioned particular examples which have been subject to previous investigations by various authors.[7–17] The characteristic which unifies all these processes is the interaction of overlapping resonances created by the e.m. fields and/or pre-existing ones.

In the succeeding section we consider coupling of two bound states with a resonance. There have been few works along these lines.[12] We systematize the previous findings here, pointing out in essence two major ones. Firstly, we determine the loci of the curves in the plane of the two detunings on which at least one discrete state lies. Secondly, we show that, where these curves intersect, one finds the maximal number (namely two here) of discrete states. The energies of these states are explicitly obtained in terms of the atomic parameters. The consequences that follow are analyzed, namely, the possibility of quantum beat spectroscopy. While a single discrete state leads to partial freezing of the flow into the continuum, the generation of more than one of these states leads to the oscillation of this persistence, i.e., we have an ionization (or

dissociation) yield which will show beat structure as a function of the pulse length of the external fields. The case of a three-level scheme, since it allows one to have an analytic expression for the beat frequency, permits one also to investigate how this beat frequency depends on both internal (atomic or molecular) as well as external (field strengths and frequencies) parameters. We give some numerical examples of these beats and discuss their significance. The last section is reserved for some concluding remarks.

II. N LEVELS COUPLED TO A RESONANCE

In this section we consider the problem of N bound states $|1\rangle, |2\rangle, \ldots, |N\rangle$ coupled by N lasers $\omega_1, \ldots, \omega_N$ to the same resonance $|a\rangle$ (see Fig. 1). We treat the fields in a second quantized form and perform a resonant approximation, which consists in reducing the full space, i.e., the tensorial product of atomic (molecular) and photon states, to a resonant subspace in which the dynamics takes place predominantly. This is done as follows. Suppose our system is at time $t=0$ in the product state $|1;n_1\omega_1, \ldots, n_N\omega_N\rangle$ (the notation is contracted for simplicity). When the field-matter interaction is switched on only the energy-conserved one-photon transitions to $|j;n_1\omega_1, \ldots, (n_j-1)\omega_j, \ldots, n_N\omega_N\rangle \equiv |j\rangle$ are allowed. The Hamiltonian can then be written in the following spectral way (neglecting also continuum-continuum transitions and spontaneous emission):

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

where

$$H_0 = \sum_j \varepsilon_j |j\rangle\langle j| + \sum_\alpha \int dE\, E |E,\alpha\rangle\langle E,\alpha| \,, \tag{2}$$

$$V = V^r + V^i \,, \tag{3}$$

and

$$V^i = \sum_\alpha |a\rangle\langle E,\alpha| V^i_{aE\alpha} + \text{H.c.} \,,$$
$$V^r = \sum_j V^r_j, \quad V^r_j = |a\rangle\langle j| (V^r_j)_{aj} + \text{H.c.} \tag{4}$$

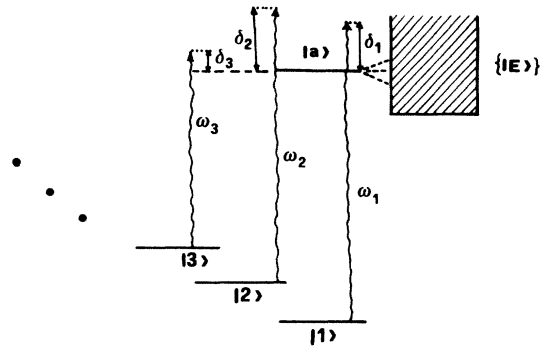

FIG. 1. N bound levels coupled to a resonance by N frequencies.

The continuum states have been labeled by the energy and a further quantum number (or set of quantum numbers) α.

Here V^r_j is the radiation coupling due to the field ω_j whereas V^i is the internal coupling responsible for the decay of state $|a\rangle$ (autoionization, predissociation). The next step consists in projecting out the continuum. This is done by using the projection operators

$$P = \sum_j |j\rangle\langle j| \,, \tag{5}$$

$$Q = 1 - P = \sum_\alpha Q^\alpha \,, \tag{6}$$

$$Q^\alpha = \int |E,\alpha\rangle\langle E,\alpha|\, dE \,. \tag{7}$$

The effective Hamiltonian in the P subspace is then

$$H^{\text{eff}} = PHP + PHQ(E - QHQ)^{-1}QHP \,. \tag{8}$$

Taking into account Eqs. (2)–(7), Eq. (4) can be rewritten as

$$H^{\text{eff}} = PH_0P + PH_0Q(E - QHQ)^{-1}QH_0P \,. \tag{9}$$

The above effective Hamiltonian has the following representation in the basis of the N bound states plus $|a\rangle$:

$$H^{\text{eff}} \equiv \begin{pmatrix} -i\gamma_1+\delta_1 & -\pi V^r_{1E}V^r_{E2}(q_{12}+i) & \cdots & -\pi V_{1E}V_{EN}(q_{1N}+i) & -\pi V_{1E}V_{Ea}(q_1+i) \\ & -i\gamma_2+\delta_2 & \cdots & -\pi V_{2E}V_{EN}(q_{2N}+i) & -\pi V_{2E}V_{Ea}(q_2+i) \\ & & \cdots & \cdots & \cdots \\ & & & -i\gamma_N+\delta_N & -\pi V_{NE}V_{Ea}(q_N+i) \\ & & & & -i\gamma_a \end{pmatrix} . \tag{10}$$

Let us explain the notation used after noting that assuming that the Hamiltonian in Eq. (1) has real matrix elements (which may be done without loss of generality) the above effective Hamiltonian is a complex symmetric matrix. In writing Eq. (10) the zero of energy has been chosen to be $\varepsilon_a + (n_1-1)\omega_1 + n_2\omega_2 + \cdots + n_N\omega_N$.

Furthermore, the field-dependent widths γ_j are defined as usual

$$\gamma_j = \pi V^2_{jE} = \pi \sum_\alpha V^2_{jE\alpha} \,. \tag{11}$$

The corresponding shifts are

$$\sigma_j = PP \sum_\alpha \int \frac{V^2_{jE'\alpha}}{E-E'} dE' \ . \tag{12}$$

The latter do not appear explicitly in Eq. (8) being incorporated in the generalized detunings

$$\delta_j = \varepsilon_j + \sigma_j + \omega_j - \varepsilon_a \quad (j=1,\ldots,N) \ . \tag{13}$$

The width γ_a is field independent being due to the internal perturbation

$$\gamma_a = \pi V^2_{aE} = \pi \sum_\alpha V^2_{aE\alpha} \ . \tag{14}$$

The corresponding shift is contained in the energy ε_a.

The off-diagonal matrix elements of H^{eff} coming from the second term in the right-hand side (rhs) of Eq. (8) play a crucial role in what follows. From Eq. (8) one has

$$(H^{\text{eff}})_{jk} = \sum_\alpha \int \frac{V^r_{jE'\alpha} V^r_{E'\alpha k}}{E-E'} dE'$$

$$= -\pi V^r_{jE} V^r_{Ek} (q_{jk} + i) \ , \tag{15}$$

$$(H^{\text{eff}})_{ja} = \sum_\alpha \int \frac{V^r_{jE'\alpha} V^i_{E'\alpha a}}{E-E'} dE'$$

$$= -\pi V^r_{jE} V^i_{Ea} (q_j + i) \ , \tag{16}$$

where

$$q_{jk} = -PP \int \frac{V^r_{jE'} V^r_{E'k}}{E-E'} dE' (\pi V^r_{jE} V^r_{Ek})^{-1} \ , \tag{17}$$

$$q_j = \left[-PP \int \frac{V^r_{jE'} V^i_{E'a}}{E-E'} dE' + V^r_{ja} \right] (\pi V^r_{jE} V^i_{Ea})^{-1} \ , \tag{18}$$

and

$$V^r_{jE} V^r_{Ek} \equiv \sum_\alpha V^r_{jE\alpha} V^r_{E\alpha k} \ , \tag{19}$$

$$V^r_{jE} V^i_{Ea} \equiv \sum_\alpha V^r_{jE\alpha} V^i_{E\alpha a} \quad (j,k=1,\ldots,N) \ . \tag{20}$$

The effective Hamiltonian in Eq. (7) is, in principle, energy dependent. Here, however, we suppose that the bound-continuum matrix elements $V^r_{jE\alpha}, V^i_{aE\alpha}$ are nearly constant in the energy range of interest and consider the effective Hamiltonian as fixed (this is a generalized pole approximation).

We have now at our disposal a compact way of handling our problem of N levels radiatively coupled to a continuum supporting a resonance. Let us study the evolution operator for such a system. It can be partitioned as

$$U(t) = PUP + PUQ + QUP + QUQ \ . \tag{21}$$

Each term of the above decomposition can be calculated from the corresponding term of the resolvent:

$$G = (Z - H)^{-1}$$

and

$$U = \frac{i}{2\pi} \int_{-\infty+i\eta}^{+\infty+i\eta} G(E^+) e^{-iEt} dE \ ,$$

$$E^+ = E + i\eta, \quad \eta \to 0^+ \ . \tag{22}$$

Utilizing well-known results one has

$$PGP = [Z - PHP - PHQ(Z-QHQ)^{-1} QHP] \ , \tag{23}$$

$$PGQ = PGPPHQ(Z-QHQ)^{-1} \ , \tag{24}$$

$$QGP = (Z-QHQ)^{-1} QHPPGP \ , \tag{25}$$

$$QGQ = [Z - QHQ - QHP(Z-PHP)^{-1} PHQ] \ . \tag{26}$$

The first term in the rhs of Eq. (21) is needed for computing transition amplitudes between states which are bound in the absence of the fields.

The second term is needed for calculating the energy-resolved continuum spectrum starting from some bound state. Notice that the total flow of the population into the continuum can be calculated as 1 minus the sum of the population of states in the P subspace and this does not require evaluation of QGP.

The third and fourth terms in Eq. (21) are needed if one is interested in scattering situations (initial state in the continuum). In the present paper we only consider transitions between bound states and evaluate only PGP.

From Eqs. (21) and (23) it is clear that in order to compute the transition amplitudes starting from state $|1\rangle$ one must build up the matrix $PGP = (E - H^{\text{eff}})^{-1}$ and perform the integral in Eq. (22) (which can be done easily through the residue theorem). For example,

$$G_{j1} = g_{j1}(E)/f(E) \ , \tag{27}$$

where $f(E)$ and $g_{j1}(E)$ are respectively the determinant and the cofactor of the $j,1$ matrix element of the same matrix $(E - H^{\text{eff}})$. This means that $f(E)=0$ is the secular equation for H^{eff} so

$$f(E) = \prod_{\nu=1}^{N+1} (E - E_\nu) \ , \tag{28}$$

where E_ν are the complex eigenvalues. From Eq. (22):

$$U_{j1}(t) = \sum_{\nu=1}^{N+1} \left[g_{j1}(E_\nu) \exp(-iE_\nu t) \Big/ \prod_{\substack{\mu,\nu \\ \mu \neq \nu}} (E_\nu - E_\mu) \right] . \tag{29}$$

The above expression has been used for the calculations. Let us come to the investigation on the nature of the dressed resonances whose energies and widths are given respectively by the real and imaginary parts of E_ν $(\nu=1,2,\ldots,N+1)$. We are going to demonstrate that under certain conditions, discussed in the following, it is possible to vary the control parameters in order to produce N dressed resonances with zero widths, i.e., N true bound states embedded in the dressed continuum (in the following the intensities are kept fixed and the frequencies are considered as the variable parameters). To do this let us first reexamine the off-diagonal matrix elements of H^{eff}. From Eqs. (15) and (19) one has (assuming matrix elements of the full Hamiltonian to be real)

$$\mathrm{Im}(H_{jk}^{\mathrm{eff}})=-\pi\sum_{\alpha}V_{jE\alpha}^{r}V_{E\alpha k}^{r}\quad(j,k=1,\ldots,N)\tag{30}$$

(Im stands for the imaginary part). So from Cauchy inequality:

$$|\,\mathrm{Im}(H_{jk}^{\mathrm{eff}})\,|\le\left[\pi\sum_{\alpha}V_{jE\alpha}^{2}\pi\sum_{\alpha}V_{kE\alpha}^{2}\right]^{1/2}.\tag{31}$$

The same is true for $\mathrm{Im}(H_{ja}^{\mathrm{eff}})$. The sign in Eqs. (31) holds when $|j\rangle, |k\rangle$ (or $|j\rangle, |a\rangle$) are coupled only to one continuum, the same for both, i.e., when the sum over α in Eq. (31) reduces to only one term. This is true, for example, if $|j\rangle$ and $|k\rangle$ are S states of an atom and the radiation fields involved are linearly polarized. In fact, in this case, identifying α with the angular momentum quantum number l and m one has that only $l=1$, $m=0$ contribute. The requirements for creating N dressed bound states is that $|a\rangle$ and all the $|j\rangle$ are coupled to the same continuum. Let us now consider that this the case (maximum interference case). Taking into account that now

$$\mathrm{Im}(H_{jk}^{\mathrm{eff}})=\pi V_{jE}V_{Ek}\,,\tag{32}$$

$$\gamma_j=\pi V_{jE}^{2}\,,\tag{33}$$

$$\mathrm{Im}(H_{ja}^{\mathrm{eff}})=\pi V_{jE}V_{Ea}\,,\tag{34}$$

$$\gamma_a=\pi V_{Ea}^{2}\,.\tag{35}$$

The effective Hamiltonian for the maximum interference case we are considering can be written as $H^{\mathrm{eff}}=R-i\Gamma$, where R and Γ are now real symmetric matrices:

$$R\equiv\mathrm{Re}(H^{\mathrm{eff}})\,,\tag{36}$$

$$\Gamma\equiv-\mathrm{Im}(H^{\mathrm{eff}})=V\tilde{V}\,,\tag{37}$$

where

$$V=\pi^{1/2}\begin{pmatrix}V_{1E}\\V_{2E}\\\vdots\\V_{NE}\\V_{aE}\end{pmatrix},\tag{38}$$

and $\tilde{V}$ is the transpose of V (row vector).

We are now able to demonstrate the following theorem: for each given set of intensities of the N fields, it is always possible to find N generalized detunings $\delta_1,\ldots,\delta_N$ [Eq. (13)] in order to have N real eigenvalues of H^{eff}.

Proof. First note that, due to Eq. (37), one has

$$\Gamma^{2}=V\tilde{V}V\tilde{V}=\beta V\tilde{V}=\beta\Gamma\,,\tag{39}$$

where β is a real number

$$\beta=\tilde{V}V\,.\tag{40}$$

Hence $(1/\beta)\Gamma$ is a projection matrix and as such it has one eigenvalue equal to 1 and the remaining N eigenvalues equal to 0. In order to prove the above theorem we have only to show that the N control parameters (i.e., the N frequencies) can be chosen in such a way that the real and imaginary part of H^{eff} commute, i.e.,

$$R\Gamma-\Gamma R=0\,.\tag{41}$$

In fact, in such case the eigenvalues of R and Γ give respectively the real and imaginary part of the eigenvalues of H^{eff} (Γ has N null eigenvalues). In order to show that Eq. (41) can be satisfied let us first rewrite it as

$$RV\tilde{V}-V\tilde{V}R=0\,.\tag{42}$$

Instead of tackling directly Eq. (41) we note that if V is an eigenvector of R, i.e.,

$$RV=\lambda V\tag{43}$$

(or $\tilde{V}\tilde{R}=\tilde{V}R=\lambda\tilde{V}$) then Eq. (41) is verified. Equation (43) gives rise to

$$\delta_1^{c}=\delta_j^{c}+\sum_{k\,(\neq1)}q_{1k}\gamma_k-\sum_{k\,(\neq j)}q_{jk}\gamma_k+(q_1-q_j)\gamma_a\qquad(j=2,\ldots,N)\,,\tag{44}$$

$$\delta_1^{c}=\sum_{k\,(\neq1)}q_{1k}\gamma_k+q_1\gamma_a-\sum_{k}q_k\gamma_k\,.\tag{45}$$

Hence we have N equations in the N unknown δ_i^{c}, which can always be solved. This completes the demonstration.

Equations (44) and (45) show also that the critical values of detunings at which N dressed bound states appear do not depend on the sign of the bound-continuum matrix elements.

For the case $N=1$, i.e., one bound state coupled to a resonance, Eq. (45) gives the following condition for δ_1:

$$\delta_1^{c}=q_1(\gamma_a-\gamma_1)\,.\tag{46}$$

For $N=2$ (i.e., two bound states coupled to a resonance by two lasers) one has, after a small amount of algebra, the following critical detunings occur:

$$\delta_1^{c}=q_1(\gamma_a-\gamma_1)-\gamma_2(q_2-q_{12})\,,\tag{47}$$

$$\delta_2^{c}=q_2(\gamma_a-\gamma_2)-\gamma_1(q_1-q_{12})\,.\tag{48}$$

The above condition was given in Ref. 1 with a different notation (the change necessary to recover the previous notation are the following: $a\to1$, $2\to3$, $\delta_1^{c}\to\delta_c$, $\delta_2^{c}\to\delta_c'$).

A. One bound state coupled to a resonance

The possibility of modifying a resonance through the photon coupling to a bound state has been discussed previously in the literature[1,7–9] so we discuss here only briefly this case. Let us first recall that, as mentioned in the Introduction, the 2×2 effective Hamiltonian has exactly the same form as that of two bound states coupled to a flat continuum by two fields.

It is easy to show that the photon-dressed bound state in which part of the population remains trapped is not merely a linear combination of the states which are bound in absence of the field, but involves the continuum as an important part. This is perhaps obvious for the one-field—one-resonance case, but establishes a significant

difference between the two-fields—no-resonance case and the well-known coherent trapping in a three-level Λ configuration, for which the trapping occurs in a linear combination of the two lower-lying bound states.[18]

B. Two bound states coupled to a resonance

Some results for this case have been already published.[1] The critical detunings at which, for fixed intensities, one creates two photon-dressed bound states embedded in the continuum are given in Eqs. (47) and (48). This corresponds to a point in the plane. One may study the possibility of creating just one bound state. This must be easier, and in fact an algebraic numerical study of the cubic secular equation, for the 3×3 effective Hamiltonian, shows that the locus of such points has dimension 1, i.e., it is a curve in the δ_1-δ_2 plane, as shown in Fig. 2.

The topological structure of such curves is quite interesting, being composed of three branches, two of which intersect in the critical point [Eqs. (47) and (48)].

Let us now examine the physical consequences of the above structure in the δ_1,δ_2 plane. Suppose we fix γ_1 and γ_3, and measure the ionization probability after a pulse of duration t as a function of t. The ionization probability (P_c) can be calculated as 1 minus the sum of populations of states $|1\rangle$ and $|2\rangle$:

$$P_c=1-P_1-P_2\ ,$$

and

$$P_1=|U_{11}(t)|^2\ , \tag{49}$$

$$P_2=|U_{21}(t)|^2\ , \tag{50}$$

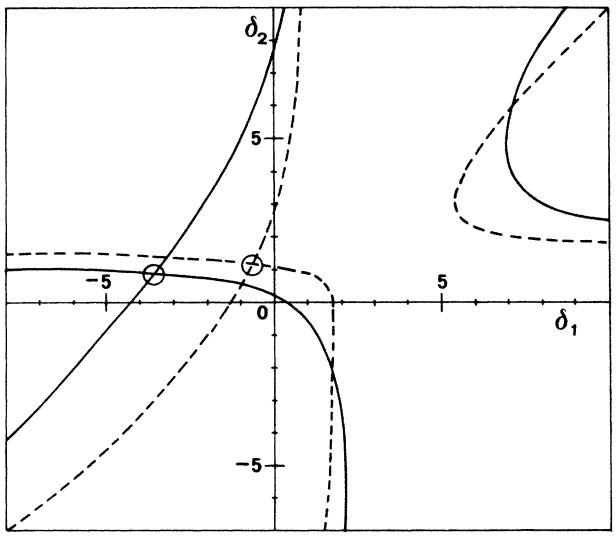

FIG. 2. Two three-branched curves in the plane of δ_1,δ_2 [see Eq. (13)] for which there exists at least one dressed discrete state. At the intersection of two branches (indicated by a circle) two discrete states coexist. Parameters are as follows: $\gamma_a=1.0$, $\gamma_2=0.6$, $q_1=2.0$, $q_2=4.0$. Solid line: $q_{12}=-5.0$, $\gamma_1=0.1$; dashed line: $q_{12}=1.0$, $\gamma_1=0.4$ (δ_1 and δ_2 are in units of γ_a). For further explanation see text.

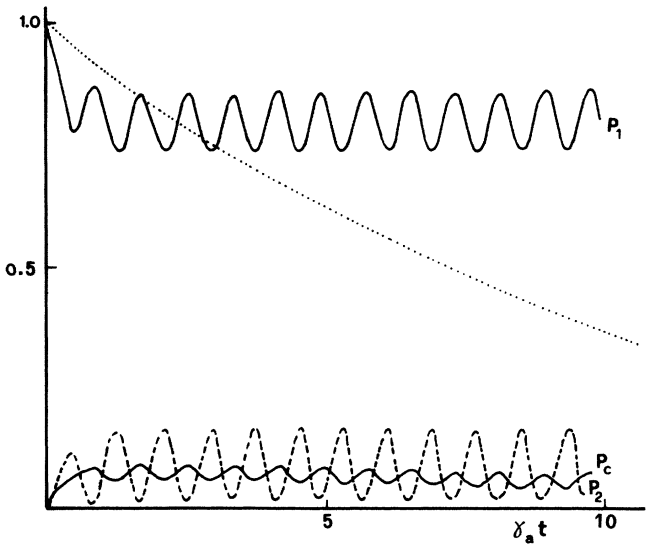

FIG. 3. Time dependence of population of state $|1\rangle(P_1)$, state $|2\rangle(P_2)$ and of the continuum ($P_c=1-P_1-P_2$) when δ_1 and δ_2 are at the intersection of the solid lines in Fig. 2 (indicated by a circle). Dashed line shows P_1 when just the field ω_1 is present. Note the persistent oscillations created by the two photon-dressed discrete states. Here parameters are as for the solid line of Fig. 2.

[see Eq. (29)].

Recalling that E_1,E_2,E_3 are complex eigenvalues of H^{eff} and that they are functions of δ_1 and δ_2 (for fixed intensities and given q's). For almost all δ_1,δ_2 the three E's are complex and P_c, after a transient regime, goes to 1 as t increases, since, as is clear from Eq. (29), P_1 and P_3 go to 0. This means that in almost all cases one has saturation after a sufficient time.[1]

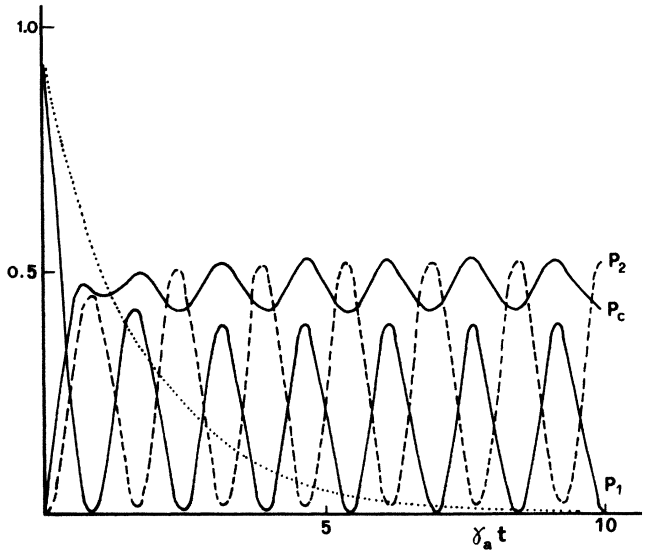

FIG. 4. Same as in Fig. 3 for the set of parameters which refer to the dashed line in Fig. 2.

Now suppose we choose δ_1 and δ_2 so that they are on the curve shown in Fig. 2. In such a case one of the three eigenvalues E_1, E_2, E_3 is real so that as $t \to \infty$ P_1 and P_2 become constant. Hence, in this case, after the transient one must observe that P_c goes to a constant value, different from 1.[1] The nonsaturation effect is a direct consequence of the creation of a true photon-dressed discrete state in which part of the population remains trapped when the fields are on.

The third possibility is that we fix δ_1 and δ_2 so that $\delta_1=\delta_1^c$ and $\delta_2=\delta_2^c$. In this case two terms survive in P_1 and P_2 giving rise to persistent oscillations in P_c as well as in P_1 and P_2. This typical quantum beat structure (see Figs. 3 and 4) is the sign that now two photon-dressed discrete states are present in the spectrum of the full Hamiltonian, and that both are contained in the nonstationary state we have prepared by the double-resonance irradiation. The beat frequency corresponds to the energy difference between such states. From perusal of Fig. 2, we see that under the conditions $\delta_1=\delta_1^c$, $\delta_2=\delta_2^c$, two discrete states are formed in the continuum in the presence of external electromagnetic fields. These two real eigenvalues are also easy to obtain analytically. They are given by

$$E_{\pm}=\pm\frac{1}{2}\left[-\frac{4}{\Gamma}(\delta_2^c-2\delta_1^c)[\delta_1^c(\gamma_1+\gamma_2)-(\delta_2^c-\delta_1^c)(\gamma_1+\gamma_a)-2\gamma_1\gamma_a q_1-2\gamma_1\gamma_2 q_{12}-2\gamma_2\gamma_a q_2]\right.$$
$$-4[\delta_1^c(\delta_1^c-\delta_2^c)-\gamma_2\gamma_a q_2^2-\gamma_1\gamma_2 q_{12}^2-\gamma_1\gamma_a q_1^2]$$
$$\left.-\frac{3}{\Gamma^2}[\delta_1^c(\gamma_1+\gamma_2)+(\delta_1^c-\delta_2^c)(\gamma_1+\gamma_a)-2\gamma_1\gamma_a q_1-2\gamma_1\gamma_2 q_{12}-2\gamma_2\gamma_a q_2]\right]^{1/2}$$
$$-\frac{1}{2\Gamma}[\delta_1^c(\gamma_1+\gamma_2)+(\delta_1^c-\delta_2^c)(\gamma_1+\gamma_a)-2\gamma_1\gamma_a q_1-2\gamma_1\gamma_2 q_{12}-2\gamma_2\gamma_a q_2]\quad(\Gamma=\gamma_1+\gamma_2+\gamma_a)\,, \tag{51}$$

where the + and the − subscripts on E denotes the two eigenvalues. Evidently these two real quantities are functions of all the parameters that have been necessary to consider the two discrete levels and the structured interacting continuum along with the four independent parameters (the frequencies and the field strengths) of the extern .. ields. Even though the formulas above are explicit, their content is far from being transparent without some examples. This is due to the fact that the parameters enter in a nonlinear manner in the expressions. Some examples are shown in Figs. 5 and 6. In Fig. 5, γ_1 and γ_2

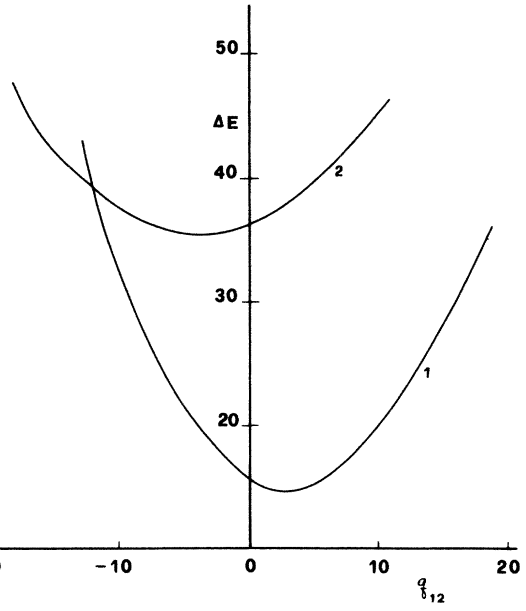

FIG. 5. Beat frequency as a function of the atomic parameter q_{12}. Both curves are calculated at $\gamma_1=\gamma_2=\gamma_a=1$. Curve 1 corresponds to $q_1=1$, $q_2=-9$, while curve 2 refers to $q_1=2$, $q_2=4$.

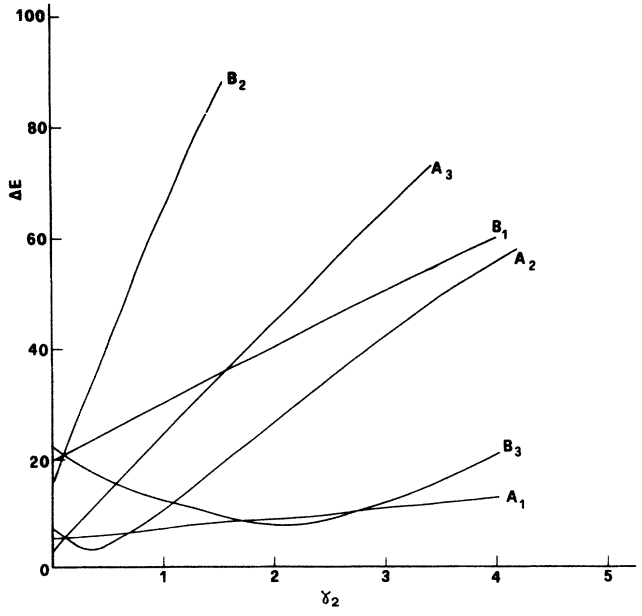

FIG. 6. Beat frequency as a function of γ_2 (see text) for a set of values of the atomic parameters. Curves A_1, A_2, and A_3 correspond to $q_{12}=3$, 12, and -6, respectively ($q_1=1$, $q_2=4$) while curves B_1, B_2, and B_3 refer to $q_{12}=-4$, $+14$, and -14, respectively ($q_1=1$, $q_2=-9$). All six curves have $\gamma_1=0.1$, $\gamma_2=1.0$.

have been fixed and the beat frequency has been plotted as a function of q_{12} with two sets of values for the pair (q_1,q_2). It is easy to see that in this case

$$\Delta E \equiv E_+ - E_- = \alpha_0 + \alpha_1 q_{12} + \alpha_2 q_{12}^2 , \tag{52}$$

where α_0, α_1, and α_2 are constants. The results are parabolas with minima at $q_{12} = -2\alpha_2/\alpha_1$ and

$$(\Delta E)_{\min} = \alpha_0 + 2\alpha_2(2\alpha_2^2/\alpha_1^2 - 1) , \tag{53}$$

where such minima might lie is shown in the two curves with $q_1 = 2.0, q_2 = 4.0$ and $q_1 = 1.0, q_2 = -9.0$. The reason that $|q_2| > |q_1|$ have been chosen as examples is due to the fact that the same resonance probed from the excited states in many cases tends to be more symmetric than from the ground states, for the atoms. From the shapes of these two curves, we see that the minimum of the beat frequency can be either for positive or negative values of q_{12}. These theoretical curves are of course nothing more than that. The experimental beat frequency (given q_1 and q_2, which are determined for independent measurements) would reveal the value of q_{12} which while theoretically calculable has, to our knowledge, no other experimental method of measurement. These universal parabolas show how the potential values of q_{12} restrict the values of the beat frequencies.

Once q_1, q_2, and q_{12} are known, one may vary the beat frequency as a function of two external field strengths. These are quite nonlinear functions of γ_1 and γ_2. We have plotted a few examples in Fig. 6, where $\gamma_1 = 0.1$, a rather low value compared to the width scale of the resonance ($\gamma_a = 1.0$) for a range of γ_2 which goes from arbitrary small values to $\gamma_2 = 4.0$ thus covering a large zone in the field strength. The six curves correspond to the two sets, which in turn arise from the two curves of Fig. 5. The parameters that are used are all listed in the figure captions. Again these curves are shown as some possibly typical cases. Once q_{12} is fixed from measurement of the beat frequency at a given set of intensities, with the aid of curves as in Fig. 5 one can experimentally explore an analogous curve of Fig. 6. From the expression of ΔE which is

$$\Delta E = \left[-\frac{4}{\Gamma}(\delta_2^c - 2\delta_1^c)[\delta_1^c(\gamma_1+\gamma_2) - (\delta_2^c - \delta_1^c)(\gamma_1+\gamma_a) - 2\gamma_1\gamma_a q_1 - 2\gamma_1\gamma_2 q_{12} - 2\gamma_2\gamma_a q_2] \right.$$
$$-4[\delta_1^c(\delta_1^c - \delta_2^c) - \gamma_2\gamma_a q_2^2 - \gamma_1\gamma_2 q_{12}^2 - \gamma_1\gamma_a q_1^2]$$
$$\left. -\frac{3}{\Gamma^2}[\delta_1^c(\gamma_1+\gamma_2) + (\delta_1^c - \delta_2^c)(\gamma_1+\gamma_a) - 2\gamma_1\gamma_a q_1 - 2\gamma_1\gamma_2 q_{12} - 2\gamma_2\gamma_a q_2] \right]^{1/2} , \tag{54}$$

we expect strong nonlinearity in γ_2. How this translates into an experimental situation should be quite clear from these six curves. In a few cases, we see that the nonlinearity in the expression (54) is not in the zone of experimental interest, while for others this is the case. It is to be emphasized that given q_1, q_2, and q_{12}, one can experimentally verify the dependence of the beat frequency on the field strength by following a curve analogous to one of those depicted in Fig. 6.

There is another way of looking at the presence of bound states in the continuum. This consists in fixing one of the frequencies, say ω_2, i.e., δ_2, vary the other, i.e., ω_1 and measuring the ionization probability as a function of δ_1 for a given pulse length. For almost all δ_1 one has that the whole population flows into the continuum, since the three eigenvalues of H^{eff} are all complex. When one crosses the curves of Fig. 2, population flows into the continuum, since the three eigenvalues of H^{eff} are all complex. When one crosses the curves of Fig. 2, one bound state appears and some trapping occurs. This is clearly seen in Figs. 7 and 8 where the trapped population is plotted as a function of δ_1 for several values of δ_2. Every peak corresponds to the crossing of one of the branches of the curve in Fig. 2. When two peaks coalesce we have two

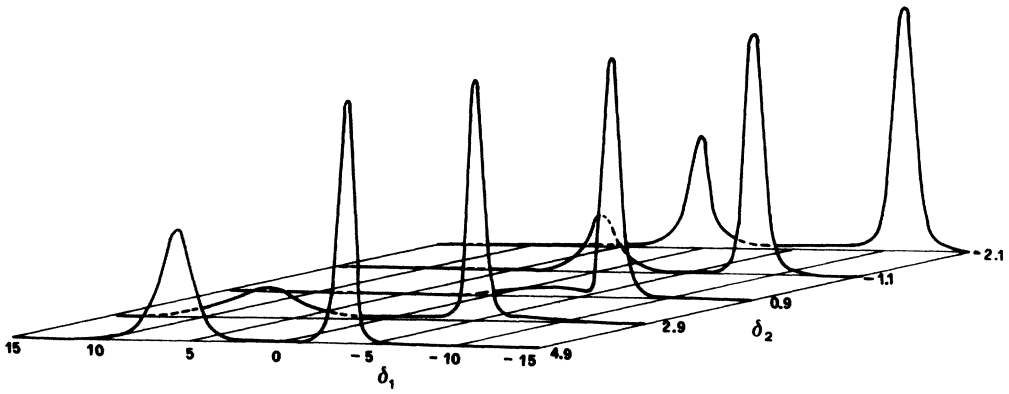

FIG. 7. Population trapped after a pulse whose length is 100 times the lifetime of the unperturbed resonance as a function of δ_1 for several values of δ_2 (see Fig. 1). Parameters are those of Fig. 2 (solid line). Note the merging of two peaks at the values of detunings which correspond to the point of intersection of solid lines in Fig. 2.

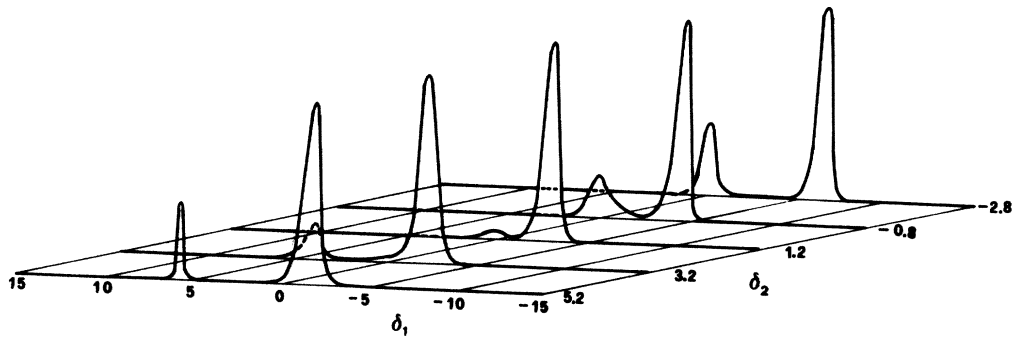

FIG. 8. Same as in Fig. 7 for the parameters of Fig. 2 (dashed line).

bound states simultaneously present and the trapped population increases. So, measuring the ionization yield one can experimentally test the structure of the δ_1,δ_2 plane (see Fig. 2).

In the numerical examples provided above on the consequences of multiple photon-dressed states in the atomic (molecular) continuum, the purely atomic parameters are q_i and q_{iN}. From perusal of the published results of autoionizing states from the ground states of atoms, q_1 is not difficult to obtain. The other q_i's are less well known. However, there has been notable experimental progress in this direction. On the other hand, q_{iN}'s are to be deduced from experimental results of double resonance through the continuum or from *ab initio* calculations. There are, to our knowledge, no accurate values of q_{iN}'s in the literature. The range of q_{12} that we have utilized to illustrate the theory has been based on our own estimate.

III. CONCLUSION

We have shown that coupling bound levels to a continuum may generate dressed discrete states. This result, while obtained after simplifying the problem through some approximations (neglecting of spontaneous emission, free-free transitions, and resonant approximation), has sufficient generality to produce a variety of experimental consequences, some of which have been investigated in this paper. It is particularly encouraging that the field intensities required can be maintained in the spectroscopic domain. One may note that a few affine experiments have already produced extremely interesting results.[19–22]

We have calculated time-resolved ionization (dissociation) probability and frequency-resolved ionization (dissociation) yields, bringing out the features due to creation of photon-dressed bound states. Various ancillary questions may still be examined but almost all of these follow in a straightforward manner from our treatment. Future work in this regard should perhaps be directed to specific atomic or molecular systems. Some effort in this direction with *ab initio* calculations is now in progress.

Note added. After this paper was written, we received a copy of a work by E. Kyrola[23] where N levels are coupled to a resonance. In that paper the technique of Laplace transform has been utilized to derive the photoelectron spectrum, an observable not considered by us.

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[1]A. Lami and N. K. Rahman, Phys. Rev. A **33**, 782 (1986). See also: (a) A. Lami and N. K. Rahman, in *Fundamentals of Lasar Interactions,* edited by F. Ehlotzky (Springer, Berlin, 1985); (b) N. K. Rahman and A. Lami, in *Advances in Laser Science,* edited by W. C. Stwalley and M. Lapp (AIP, New York, 1986), Vol. 1.

[2]J. von Neumann and E. Wigner, Phys. Z. **30**, 465 (1929).

[3]F. H. Stillinger and D. R. Herrick, Phys. Rev. A **11**, 446 (1975); J. Chem. Phys. **62**, 4360 (1975).

[4]L. Fonda and R. G. Newton, Ann. Phys. (N.Y.) **10**, 490 (1960).

[5]W. Friedrich and D. Wintgen, Phys. Rev. A **31**, 3964 (1985).

[6]See, e.g., K. O. Friedrichs, Comments Pure Appl. Math. **1**, 361 (1948).

[7]P. Lambropoulos and P. Zoller, Phys. Rev. A **24**, 379 (1981).

[8]K. Rzazewski and J. H. Eberly, Phys. Rev. Lett. **47**, 408 (1981); Phys. Rev. A **27**, 2026 (1983).

[9]P. Knight, Comments At. Mol. Phys. **15**, 193 (1984). This is the only review article in this area quoting almost all the references prior to that paper.

[10]G. S. Agarwal, S. L. Haan, K. Burnett, and J. Cooper, Phys. Rev. Lett. **48**, 1164 (1982).

[11]A. Lami and N. K. Rahman, in *Collisions and Half-Collisions with Lasers,* edited by N. K. Rahman and C. Guidotti (Harwood Academic, New York, 1984); Phys. Rev. A **26**, 3360 (1982); Opt. Commun. **43**, 383 (1982).

[12]Z. Deng and J. H. Eberly, J. Opt. Soc. Am. B **1**, 103 (1984); Z. Deng, *ibid.* **1**, 874 (1984).

[13]P. T. Greenland, J. Phys. B **15**, 3191 (1982).

[14]M. Crance and L. Armstrong, Jr., J. Phys. B **15**, 4637 (1982).

[15]K.-S. Lam and T. F. George, Phys. Rev. A **33**, 2491 (1986).

[16](a) A. D. Bandrauk and G. Turcotte, in *Collisions and Half-Collisions with Lasers,* edited by N. K. Rahman and C. Guidotti (Harwood Academic, New York, 1984); (b) A. D. Bandrauk, M. Giroux, and G. Turcotte, J. Phys. Chem. **89**, 4473 (1985).

[17]M. H. Nayfeh and W. Glab, Phys. Rev. A **25**, 1619 (1982). The usage of ionization quantum beats of this paper is not to be confused with the quantum beats from multiple photon-dressed discrete states. Transients in double resonance via continuum will in general have oscillatory behavior.

[18]E. Arimondo and J. Orriols, Lett. Nuovo Cimento **17**, 333 (1976).

[19]L. I. Pavlov, S. S. Dimov, D. I. Metchikov, G. M. Mileva, K. V. Stamenov, and G. B. Altschuler, Phys. Lett. **89A**, 441 (1982).

[20]S. S. Dimov, L. I. Pavlov, K. V. Stamenov, Y. I. Heller, and A. K. Popov, Appl. Phys. B **30**, 35 (1983).

[21]J.-Y. Liu, P. McNicholl, D. A. Armin, J. Ivri, T. Bergeman, and H. J. Metcalf, Phys. Rev. Lett. **55**, 189 (1985).

[22]E. B. Saloman, J. W. Cooper, and D. E. Kelleher, Phys. Rev. Lett. **55**, 193 (1985).

[23]E. Kyrola, J. Phys. B **19**, 1437 (1986).