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Theory and experimental consequences of generation of a pair of photon-dressed discrete states by external electromagnetic fields in the atomic or molecular continuum

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It is shown theoretically that double resonance through the continuum can be utilized to produce two photon-dressed discrete states embedded in the continuum. The significance of creating such a pair of states is discussed, along with its experimental consequences. Especially important are quantum beats of population, which offer novel spectroscopic opportunities.

The purpose of this Rapid Communication is to demonstrate that it is possible to create a *pair of photon-dressed bound states* in the continuum of atoms (or molecules) induced by external electromagnetic fields and then observe quantum beating between them. The importance of such generation of a pair of bound states lies not only on the often-raised question regarding the creation of bound states in the continuum, but equally on the prospect of an entirely new class of quantum beat spectroscopy,[1-6] enlarging from the beam-foil technique, single-photon beam or crossed electrons and photon beams to two-photon beams (either two lasers or a laser along with synchrotron radiation).

The point we wish to emphasize is the permanence of a bound state in the continuum of a system. The first effort in this direction is due to von Neumann and Wigner.[7] This pioneering work, which was to be corrected and extended,[8] has been a constructive approach to see under what conditions a bound state may be embedded in the continuum. (The literature is filled with models akin to that of Friedrichs,[9] in which a bound state embedded in the continuum becomes unstable due to the interaction with the continuum and dissolves into it. References 7 and 8, along with this contribution, are the antithesis of this phenomenon.) The creation of a bound state through the interaction of a discrete state with continuum by an external electromagnetic field was envisaged separately,[10] where the coupling of a discrete state to the continuum was taken to be the main parameter of the problem. Later, there had been a surge of interest in the possibility of modifying the structure of the continuum, where a structure was present, such as an autoionizing or a predissociating state.[11-14] Closely related are the efforts in which two fields are utilized to couple two bound states via the continuum, causing population trapping[15] and consequent harmonic generation,[16] as well as a double-resonance scheme of three levels interacting with both continuum and external electromagnetic fields.[17-19]

In many of these later works it was seen that a stable state can be formed in the continuum, induced by the external field and therefore present only when the interaction is on. Investigations have also been made on the stability of the discrete state in the presence of spontaneous emission.[20]

A two-level system interacting with two electromagnetic fields through a resonance (see Fig. 1) offers a further interesting feature, namely, the creation of two bound states in the dressed continuum. The way it comes about is seen transparently by examining the poles of the resolvent, which can be obtained as eigenvalues of the effective Hamiltonian obtained by projecting onto the discrete states:[17,18]

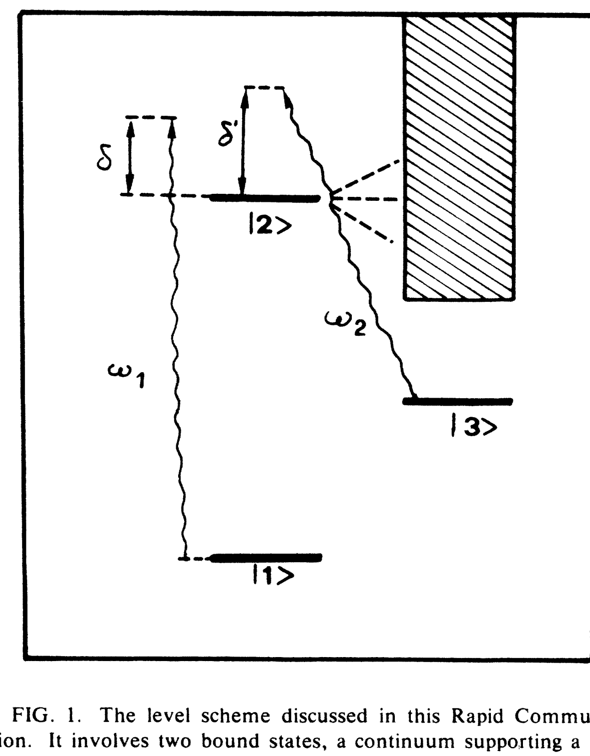

FIG. 1. The level scheme discussed in this Rapid Communication. It involves two bound states, a continuum supporting a resonance, and two frequencies ω_1 and ω_2. The broken lines from state $|2\rangle$ to the continuum signify internal coupling giving rise to autoionization or predissociation.

$$H^{\mathrm{eff}}=\begin{vmatrix} -i\gamma_1+\delta & -(\gamma_1\gamma_2)^{1/2}(q_1+i) & -(\gamma_1\gamma_3)^{1/2}(q_{13}+i) \\ -(\gamma_1\gamma_2)^{1/2}(q_1+i) & -i\gamma_2 & -(\gamma_2\gamma_3)^{1/2}(q_3+i) \\ -(\gamma_1\gamma_3)^{1/2}(q_{13}+i) & -(\gamma_2\gamma_3)^{1/2}(q_3+i) & -i\gamma_3+\delta' \end{vmatrix} \ . \tag{1}$$

The shifts are contained in the detunings (see also Fig. 1). Shifts and widths induced by the fields are defined as usual.[17]

We shall focus exclusively on the parameters of the external fields (the two detunings and the two field strengths) as variables of the problem; therefore, what follows will hold *universally* for all atoms and molecules which have resonances. Let us keep the field strengths arbitrary. The generalized Fano parameters q_1 and q_3 are the ones that one would normally need to parametrize photoionization data from the atomic states corresponding to the ground and excited state of any atom, respectively. The introduction of q_{13} is necessitated by the fact that there is in general a real part in the interaction between the states $|1\rangle$ and $|3\rangle$ intermediated by the continuum.

Diagonalization of the matrix given in (1) produces the dressed resonances of the system as well as the eigenvalues. Given an atom or a molecule, we have four laser variables at our disposal, the two detunings and the two field strengths. We look for a *real* eigenvalue (for arbitrary γ_1 and γ_3) by varying δ and δ'. Typical results are shown in Fig. 2, which shows two of such three-branched curves.

For fixed values of all the atomic parameters and field strengths, a real eigenvalue is obtained only on the points of the three-branched curves given in Fig. 2. To make it clear that the topology is very sensitive to the exact values of all the quantities involved, two (three-branched) curves for two different values of q_{13} are shown. It should, however, be clear that for any two levels of a given atom and laser frequencies q_{13} is fixed.[17,18]

The crossing point of two branches corresponds to two bound states. This can be further checked by numerical diagonalization of H^{eff} [Eq. (1)]. Since the imaginary part of H^{eff} [Eq. (1)] has two zero eigenvalues, one finds that there are two real eigenvalues only when

$$\delta=\delta_c=q_1(\gamma_2-\gamma_1)-\gamma_3(q_3-q_{13}) \ ,$$
$$\delta'=\delta_c'=q_3(\gamma_2-\gamma_3)-\gamma_1(q_1-q_{13}) \ . \tag{2}$$

This is precisely where the crossing point lies. The two real eigenvalues at this crossing point are ($\Gamma\equiv\gamma_1+\gamma_2+\gamma_3$)

$$\begin{aligned} E_\pm = \pm\frac{1}{2}\Bigg\{ & -\frac{4}{\Gamma}(\delta_c'-2\delta_c)[\delta_c(\gamma_1+\gamma_3)-(\delta_c'-\delta_c)(\gamma_1+\gamma_2)-2\gamma_1\gamma_2q_1-2\gamma_1\gamma_3q_{13}-2\gamma_2\gamma_3q_3] \\ & -4[\delta_c(\delta_c-\delta_c')-\gamma_2\gamma_3q_3^2-\gamma_1\gamma_3q_{13}^2-\gamma_1\gamma_2q_1^2] \\ & -\frac{3}{\Gamma^2}[\delta_c(\gamma_1+\gamma_3)+(\delta_c-\delta_c')(\gamma_1+\gamma_2)-2\gamma_1\gamma_2q_1-2\gamma_1\gamma_3q_{13}-2\gamma_2\gamma_3q_3]\Bigg\}^{1/2} \\ & -\frac{1}{2\Gamma}[\delta_c(\gamma_1+\gamma_3)+(\delta_c-\delta_c')(\gamma_1+\gamma_2)-2\gamma_1\gamma_2q_1-2\gamma_1\gamma_3q_{13}-2\gamma_2\gamma_3q_3] \ . \end{aligned} \tag{3}$$

A remarkable aspect of this crossing of curves signifying the generation of two dressed bound states is that no stringent condition is imposed either on the atomic variables or the external field strengths. One can perform an experiment at the required detunings [Eq. (2)], say δ_c, δ_c' commensurate with the rest of the parameters. The obvious analogy is that of coherence trapping in the Λ configuration of three-level systems,[21,22] where specific linear combination of states may be decoupled from the field by choosing the appropriate detunings.

In our case, by choosing to do an experiment at δ_c, δ_c' we can bring the question of generation of stable states to the domain of usual spectroscopy such as double-resonance spectroscopy[23] and need not concern ourselves with the question of high field strengths at all.

If the system is in state $|1\rangle$ initially (see Fig. 1) and no bound states are created, the populations of states $|1\rangle$ and $|3\rangle$ decay to zero, due to the flow of the entire population into the continuum [Fig. 3(a)]. With a single bound state created, ionization is partially "frozen" and a part of the population remains indefinitely in the discrete part of the field-free spectrum [Fig. 3(b)]. With the generation of two bound states, we expect that the ionization will also freeze, but will additionally show oscillations. This is explicitly seen

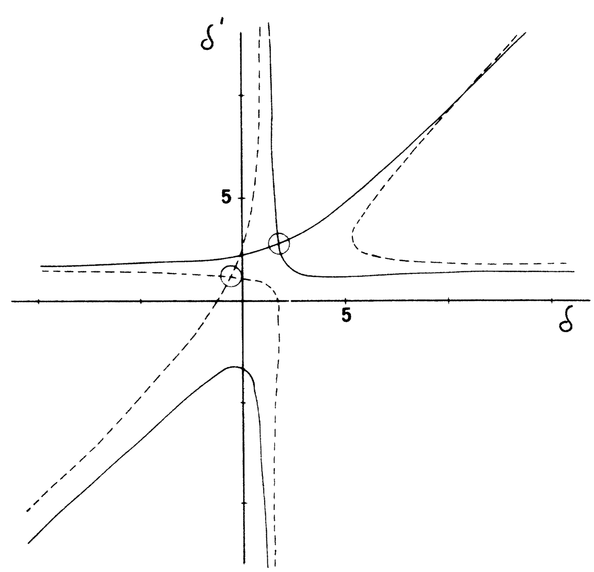

FIG. 2. The locus of points in the δ, δ' plane, in which there is a real eigenvalue (at least). Here $\gamma_1=0.4$, $\gamma_3=0.6$ (δ, δ', γ_1, and γ_3 are all in units of γ_2), $q_1=2$, and $q_3=4$. The two three-branched curves differ for the value of the parameter q_{13} (see the text). Broken line: $q_{13}=1$; full line: $q_{13}=5$.

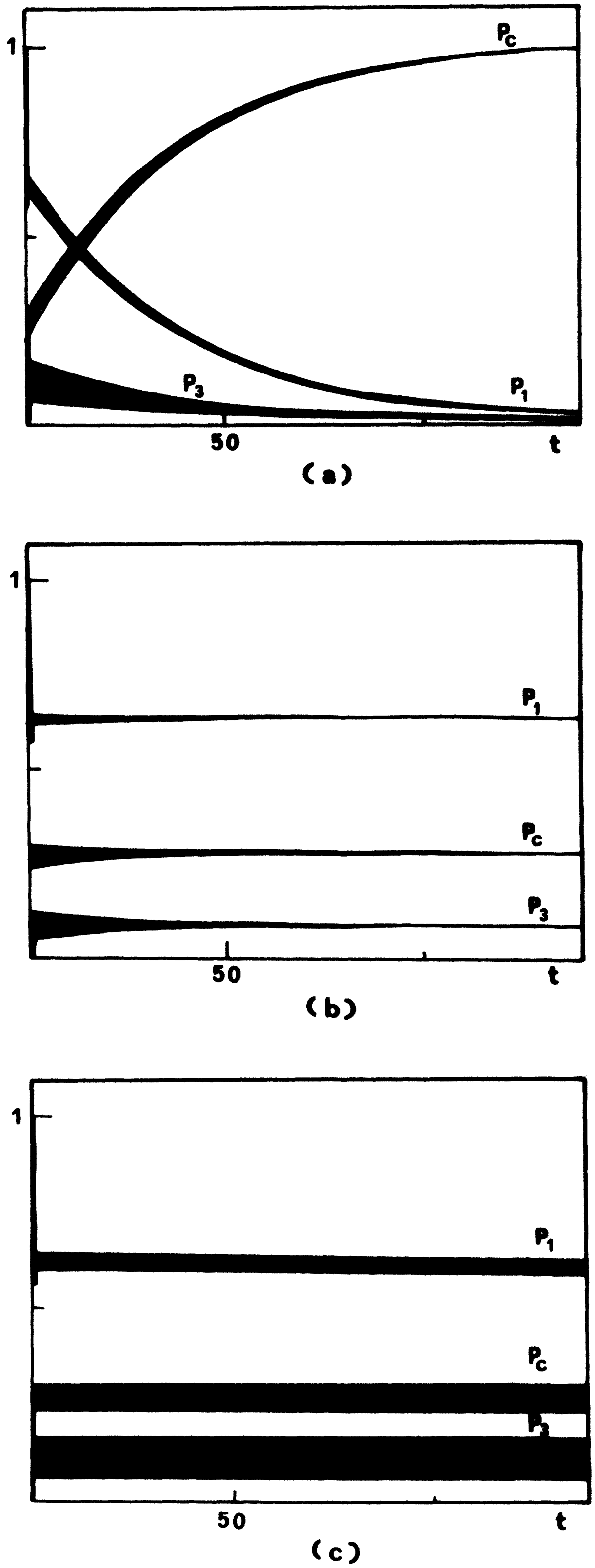

FIG. 3. (a), (b), and (c) show the populations of state $|1\rangle$ (P_1), of state $|3\rangle$ (P_3), and the continuum yield (P_c) as a function of time (in units of $1/\gamma_2$). The darkened areas are due to oscillations which are unresolved in the chosen time scale. γ_1, γ_3, q_1, and q_3 are as in Fig. 2 and $q_{13}=1$. (a) $\delta=-0.4$, $\delta'=1.1$; no dressed bound states are formed. (b) $\delta=-0.256$, $\delta'=1.964$; one dressed state is formed. (c) $\delta=-0.6$, $\delta'=1.2$; two dressed bound states are formed and the populations exhibit quantum beats between them.

in Fig. 3(c).

A detailed view of the populations of the two bound states $|1\rangle$ and $|3\rangle$, as well as that which has gone into the continuum is shown as function of time in Fig. 4. After an initial rise (fall) time, both the populations of $|1\rangle$ and $|3\rangle$ manifest clear evidence of oscillations, which is nothing but the quantum beating of the two field-generated bound states. Apart from the initial transients, these are pure oscillations with frequency equal to the difference of the two real eigenvalues, which in turn depends on the atom and the field parameters. (Figures 3 and 4 are two illustrations of such dependence.)

We next come to the specific systems and methods that may be applied to generate these discrete states and examine their quantum beats. The simplest manner to observe them is to examine the fluorescence of the upper level of the atom (molecule). The existence of the fluorescence will show the population of the initially empty state, while oscillations of the fluorescence will be the signature of the quantum beat of the field-generated discrete states. The other method may be the examination of the ionization (dissociation) yield, which will not show saturation, and the yield will oscillate with the duration (length of the pulse) of the external field.

What kind of external field should be suitable for such experiments? We tentatively suggest synchrotron radiation and a laser. The synchrotron radiation has the right frequency range to couple a ground state to a resonance, while lasers will be suitable for coupling the excited state to the same resonance. Notice that experiments that involve both lasers and synchrotron are already state of the art.[24] The alternative is to use two different lasers with an appropriate atomic system. With the development of the lasers in the uv region, the prospects of such experiments are indeed on the rise. One may also expect to see variants of the scheme discussed in this Rapid Communication, in which one of the fields cause a multiphoton transition, or an alternate scheme involving a larger number of levels and field frequencies.[25] This will add a certain richness to the processes involved,

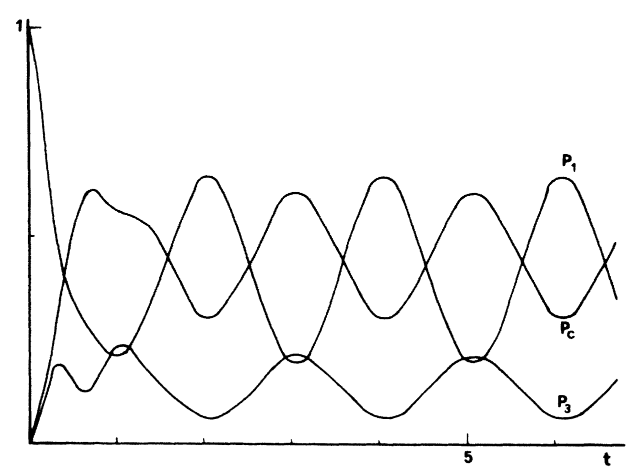

FIG. 4. Populations as functions of time when two bound states coexist. Here the time scale is enlarged and the oscillations (which persist indefinitely) are clearly resolved. The widths γ_1 and γ_3 are as in Figs. 2 and 3 and $q_1=2$, $q_3=4$, $q_{13}=5$.

keeping intact the central issue of this Rapid Communication.

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[1]S. Haroche, in *High-Resolution Laser Spectroscopy,* edited by K. Shimoda, Topics in Applied Physics, Vol. 13 (Springer, Heidelberg, 1976), p. 256.

[2]H. J. Andra, in *Progress in Atomic Spectroscopy, Part B,* edited by W. Hanle and H. Kleinpoppen, Physics of Atoms and Molecules (Plenum, New York, 1979), p. 829.

[3]R. L. Shoemaker and R. Brewer, Phys. Rev. Lett. **28**, 1430 (1972).

[4]P. J. O. Teubner, J. E. Furst, M. C. Tonkin, and S. J. Buckman, Phys. Rev. Lett. **46**, 1569 (1981).

[5]J. P. Heritage, T. K. Gustafsen, and C. H. Lin, Phys. Rev. Lett. **34**, 1299 (1975).

[6]W. Lange and J. Mlynek, Phys. Rev. Lett. **40**, 1373 (1978).

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

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

[9]K. O. Friedrichs, Commun. Pure. Appl. Math. **1**, 361 (1948).

[10]C. Cohen-Tanoudji and P. Avan, in *États Atomiques et Moléculaires Couplés a un Continuum: Atomes et Molecules Hautement Excités* (CNRS, Paris, 1977), p. 93.

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

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

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

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

[15]P. Knight, Comments At. Mol. Phys. **15**, 193 (1984). This review paper quotes most of the ancillary references in this area.

[16]L. I. Pavlov, S. S. Dimov, D. I. Metchikov, G. M. Mileva, K. V. Stamenov, and G. B. Altshuller, Phys. Lett. **89A**, 441 (1982); S. S. Dimov, L. I. Pavlov, K. V. Stamenov, Y. I. Heller, and A. K. Popov, Appl. Phys. B **30**, 35 (1983).

[17]A. Lami and N. K. Rahman, Phys. Rev. A **26**, 3360 (1982); Opt. Commun. **43**, 383 (1982).

[18]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), p. 373; in *Fundamentals of Laser Interactions,* edited by F. Ehlotzky (Springer, Heidelberg, 1985), p. 299.

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

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

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

[22]H. R. Gray, R. M. Whitley, and C. R. Stroud, Jr., Opt. Lett. **3**, 218 (978).

[23]K. Shimoda, in *Laser Spectroscopy of Atoms and Molecules,* edited by H. Walther (Springer, Berlin, 1976).

[24]F. J. Wuilleumier, in *Atomic and Molecular Physics Close to Ionization Thresholds in High Fields, Colloque International du CNRS* [J. Phys. (Paris) Colloq. **43**, Suppl. 11, C2-347 (1982)].

[25]A. Lami and N. K. Rahman (unpublished).