## Electromagnetically induced transparency with structured multicontinua

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We explore theoretically the consequence of the presence of structured multiple continua on the electromagnetically induced transparency (EIT) effect with pulsed lasers. The theory is developed in a rigorous manner using the overlapping resonances perspective. As one consequence of the theory we show that due to the structure in the continuum (which can also be induced optically) the EIT line shapes become asymmetric for a strong coupling piulse.

DOI: 10.1103/PhysRevA.75.013424

PACS number(s): 32.80.Qk, 42.50.Gy

## I. INTRODUCTION

Electromagnetically induced transparency (EIT) ([1-9]) is one of the prime examples for the use of quantum interferences in changing the optical properties of material systems. In the original EIT scenario [1,2], one allows two light beams to interact with a three-level material system so as to minimize, and often completely eliminate, the absorption of the same light beams as they pass through a thick sample.

The EIT phenomenon is intimately related to the formation of *resonances*—the broadening of spectral lines due to the interaction of bound states with a (radiative or nonradiative) continuum of states. When the broadening becomes comparable to the spacing between the lines, the resonances are said to *overlap*. This property can be controlled optically due to the (Autler Townes [10]) splitting of *dressed states*, which depends on the intensity of the light field used to induce the Autler Townes (AT) splitting. Once the relative positions of the various levels are known, there is a wellestablished theory [11–15] for dealing with interferences between overlapping resonances.

It was realized early on [14], following the analysis of Fano [11], that the interference between overlapping resonances can give rise to "*dark states*." Such dark states, which are characterized by the vanishing of photoabsorption, were found experimentally a few years later [16,17]. They feature very highly in many applications in coherent optics and in particular in "lasing without inversion" [1,18,19] and adiabatic passage phenomena [5,20].

The emergence of dark states as a result of overlapping resonances was at the heart of the original EIT idea of Harris *et al.* [1,2]. However, only limited use of the general theory of overlapping resonances has been subsequently made. In particular, the Wigner Weisskopf approximation [21], according to which the resonance width is independent of the energy ("unstructured continuum") was invariably assumed. The widths of the various levels were thus represented [7] as constant imaginary additions to the Hamiltonian matrix elements, or, in the density-matrix description of the process, as decay and dephasing *rates*.

An immediate consequence of this approximation is the neglect of the resonance *level shifts*. The reason is that the shifts, which are given as the Hilbert transform of the level widths, vanish for a constant function. Past treatments of EIT have also neglected multichannel effects, arising when each level is coupled to a multiplicity of continua.

The above are important effects because structure and multiplicity of continua abound in real (molecular) systems [22]. One finds in such systems that levels decay indirectly due to the existence of "tierlike" coupling schemes where levels belonging to a given vibrational mode ("tier") are coupled to levels belonging to just one other tier, which are coupled to yet another limited set of levels, and so on, until one reaches the continuum tiers. The tierlike scheme thus leads to highly "structured" continua, which are in addition, often coupled to one another, leading to "multichannel" scattering [13] and dissociation [23].

In this paper we present a more complete theory of EIT, based on the overlapping resonances perspective, in which structured multiple continua are considered. One of the outcomes of our theory is that the existence of structures (which can also be induced optically—as in the "laser-induced continuum structure" (LICS) effect [24]) may change the EIT line shapes and renders them asymmetric.

## II. ELEMENTS OF THE THEORY OF MULTICHANNEL OVERLAPPING RESONANCES

We start by briefly reviewing elements of "partitioning" theory [11-15,25] used in the remainder of this paper to treat the interference between overlapping resonances. The physical situation we address is illustrated in Fig. 1 in which (as explained in Sec. III below) two overlapping resonances are created by optically splitting a single resonance. Assuming that we have a situation in which bound states interact with continuum states, we define, according to this approach, two hermitian projection operators Q and P, satisfying the equalities

$$Q^{\dagger} = Q, \quad P^{\dagger} = P, \quad QQ = Q, \quad PP = P,$$
$$PQ = QP = 0, P + Q = I, \tag{1}$$

where I is the identity operator. The Q and P operators are chosen to project out the subspaces spanned by the bound states and the continuum states, respectively. Further, as Eq. (1) indicates, they are orthogonal, e.g., they may project onto the vacuum photon states and the one-photon states, or any two subspaces known to be orthogonal.

The full scattering incoming states  $|E, \mathbf{n}^-\rangle$ , are eigenstates of the Schrödinger equation



FIG. 1. A ground state  $|E_1\rangle$  is excited by a weak laser pulse  $\varepsilon_1$  to a resonance state  $|E_0\rangle \in Q$  decaying radiatively or nonradiatively to space *P*. The  $|E_0\rangle$  state is coupled optically to a third state  $|E_2\rangle$  by a strong guiding field  $\varepsilon_2$  and undergoes as a result Autler-Townes splitting. As a result of the splitting and the decay, an EIT "hole" is formed at  $E=E_0$ .

$$[E - i\epsilon - H]|E, \mathbf{n}^{-}\rangle = 0, \qquad (2)$$

where  $-i\epsilon$  gives rise to "incoming" boundary conditions [13,26]. Using the orthogonality of *P* and *Q*, we obtain two coupled equations,

$$[E - i\epsilon - PHP]P|E, \mathbf{n}^{-}\rangle = PHQ|E, \mathbf{n}^{-}\rangle, \qquad (3)$$

$$[E - i\epsilon - QHQ]Q|E, \mathbf{n}^{-}\rangle = QHP|E, \mathbf{n}^{-}\rangle.$$
(4)

We define two basis sets,  $|E, \mathbf{n}_1^-\rangle$  and  $|\phi_s\rangle$ , which are the solutions of the *homogeneous* (decoupled) parts of Eqs. (3) and (4). That is,

$$[E - i\epsilon - PHP]|E, \mathbf{n}_1\rangle = 0, \qquad (5)$$

$$[E_s - QHQ] |\phi_s\rangle = 0. \tag{6}$$

Implicit in Eqs. (5) and (6) is that  $|E, \mathbf{n}_1^-\rangle \in P$  and  $|\phi_s\rangle \in Q$  and as such they are orthogonal to one another. We, in fact, assume that each basis set spans the entire subspace to which it belongs, hence we can write an explicit representation of Q and P as

$$Q = \sum_{s} |\phi_{s}\rangle \langle \phi_{s}|, \quad P = \sum_{\mathbf{n}} \int dE |E, \mathbf{n}_{1}^{-}\rangle \langle E, \mathbf{n}_{1}^{-}|.$$
(7)

Using Eq. (7) we can therefore write

$$|E,\mathbf{n}^{-}\rangle = [P+Q]|E,\mathbf{n}^{-}\rangle$$

in terms of Q and P as

$$|E,\mathbf{n}^{-}\rangle = \sum_{s} |\phi_{s}\rangle\langle\phi_{s}|E,\mathbf{n}^{-}\rangle + \sum_{\mathbf{n}'} \int dE'|E',\mathbf{n}_{1}'^{-}\rangle$$
$$\times\langle E',\mathbf{n}_{1}'^{-}|E,\mathbf{n}^{-}\rangle.$$
(8)

We now solve for  $P|E, \mathbf{n}^{-}\rangle$  by writing it as a sum of the

homogeneous solution of Eq. (5) and a particular solution of Eq. (3) obtained by inverting  $[E-i\epsilon-PHP]$ ,

$$P|E,\mathbf{n}^{-}\rangle = P|E,\mathbf{n}_{1}^{-}\rangle + [E - i\epsilon - PHP]^{-1}PHQ|E,\mathbf{n}^{-}\rangle.$$
(9)

Substituting this solution into Eq. (4) we obtain that

$$Q|E,\mathbf{n}^{-}\rangle = [E - i\epsilon - Q\mathcal{H}Q]^{-1}QHP|E,\mathbf{n}_{1}^{-}\rangle, \qquad (10)$$

where

$$QHQ \equiv QHQ + QHP[E - i\epsilon - PHP]^{-1}PHQ.$$
(11)

An explicit representation of Eq. (10) is obtained by using the well-known identity,

$$[E - i\epsilon - PHP]^{-1} = \mathsf{P}_{v}[E - PHP]^{-1} + i\pi\delta(E - PHP),$$
(12)

with  $P_v$  denoting a Cauchy principal-value integral, as

$$QHQ = QHQ + QHPP_{v}[E - PHP]^{-1}PHQ + i\pi QHP\delta(E - PHP)PHQ.$$
(13)

Given Eqs. (9) and (13) we can express, via Eq. (8), the full scattering wave function  $|E, \mathbf{n}^-\rangle$  in terms of  $|\phi_s\rangle$  and  $|E, \mathbf{n}_1^-\rangle$ .

# III. ELECTROMAGNETICALLY INDUCED TRANSPARENCY

We consider the situation illustrated in Fig. 1 in which a ground state  $|E_1\rangle$ , a resonance state  $|E_0\rangle$  (decaying radiatively or nonradiatively) coupled optically to a third state  $|E_2\rangle$  (which can also decay radiatively or nonradiatively) by a strong guiding field

$$\mathbf{E}_{2}(t) \equiv \hat{\boldsymbol{\varepsilon}}_{2} \boldsymbol{\varepsilon}_{2}(t) = \hat{\boldsymbol{\varepsilon}}_{2} R_{e} \mathcal{E}_{2}(t) \exp(-i\omega_{2}t), \quad (14)$$

where  $\hat{\boldsymbol{\varepsilon}}_2$  is the polarization direction vector. We probe the system by a weaker laser pulse

$$\mathbf{E}_{1}(t) \equiv \hat{\boldsymbol{\varepsilon}}_{1} \boldsymbol{\varepsilon}_{1}(t) = \hat{\boldsymbol{\varepsilon}}_{1} R_{e} \mathcal{E}_{1}(t) \exp(-i\omega_{1}t), \quad (15)$$

satisfying the  $\mathcal{E}_1(t) \ll \mathcal{E}_2(t)$  condition. Accordingly, we can treat  $\varepsilon_1(t)$  as a perturbation and obtain first the adiabatic eigenstates resulting from the  $\varepsilon_2(t)$ -induced interaction between the  $|E_0\rangle$  and  $|E_2\rangle$  states.

Therefore, temporarily neglecting  $\varepsilon_1(t)$ , we can expand the system wave function in just two states,

$$|\Psi(t)\rangle = b_0(t)|E_0\rangle \exp(-iE_0t) + b_2(t)|E_2\rangle \exp(-iE_2t),$$
 (16)

where a.u.  $(\hbar = 1)$  are used here and throughout this paper. Using the expansion of Eq. (16) we obtain from the timedependent Schrödinger equation

$$i\frac{\partial\Psi(t)}{\partial t} = H\Psi(t) = [H_M + H_{MR}]\Psi(t), \qquad (17)$$

where  $H_M$  is the material Hamiltonian and  $H_{MR}$  is the matterradiation interaction, given in the dipole approximation as

$$H_{MR} = -\vec{\mu} \cdot \mathbf{E}(t), \qquad (18)$$

the usual set of ordinary coupled differential equations for the  $\underline{b} \equiv (b_0, b_2)$  coefficients vector,

$$\frac{d}{dt}\underline{\mathbf{b}} = i\underline{\mathbf{H}} \cdot \underline{\mathbf{b}}(t), \tag{19}$$

where  $\underline{H}$  is given in the "rotating waves approximation" as

$$\underline{\mathbf{H}} = \begin{pmatrix} 0 & \Omega_2^*(t)e^{i\delta_2 t} \\ \Omega_2(t)e^{-i\delta_2 t} & 0 \end{pmatrix},$$
(20)

with the detuning and the Rabi frequency given, respectively as

$$\delta_2 \equiv \omega_2 - |E_0 - E_2|, \quad \Omega_2(t) \equiv \vec{\mu} \cdot \hat{\varepsilon}_2 \mathcal{E}_2(t).$$
(21)

We now transform H to a form that does not contain the highly oscillatory  $e^{-i\delta_2 t}$  terms, which might invalidate the adiabatic approximation. Multiplying Eq. (19) by a diagonal matrix,  $\exp(i\hat{\Delta}t/2)$ , with  $\hat{\Delta}$  being the diagonal detuning matrix,

$$\hat{\Delta} = \begin{pmatrix} -\delta_2 & 0\\ 0 & \delta_2 \end{pmatrix},\tag{22}$$

results in the following:

$$\exp(i\hat{\Delta}t/2)\frac{d}{dt}\mathbf{b} = i\exp(i\hat{\Delta}t/2)\cdot\mathbf{H}\exp(-i\hat{\Delta}t/2)\exp(i\hat{\Delta}t/2)\mathbf{b}(t).$$
(23)

We can eliminate the oscillatory terms by defining

$$\underline{\mathbf{c}} \equiv \exp(i\hat{\Delta}t/2)\underline{\mathbf{b}},\tag{24}$$

and obtain from Eq. (23) that,

$$\frac{d}{dt}\underline{\mathbf{c}} = i\underline{\mathbf{H}}' \cdot \underline{\mathbf{c}}(t), \qquad (25)$$

where

$$\underline{\mathbf{H}}' = \begin{pmatrix} -\delta_2/2 & \Omega_2^*(t) \\ \Omega_2(t) & \delta_2/2 \end{pmatrix}.$$
 (26)

Having removed the oscillatory  $\exp(\pm i \delta_2 t)$  terms, we now build adiabatic solutions by diagonalizing Eq. (26) using a  $2 \times 2$  unitary matrix,

$$\underline{\underline{V}} = \begin{pmatrix} \cos\theta & e^{-i\phi_2}\sin\theta \\ -e^{i\phi_2}\sin\theta & \cos\theta \end{pmatrix}.$$
 (27)

The corresponding diagonal eigenvalue matrix  $\hat{\lambda}$  is composed of the two roots,

$$\lambda_{1,2}(t) = \pm \lambda(t) = \pm \left[ \delta_2^2 / 4 + |\Omega_2(t)|^2 \right]^{1/2}.$$
 (28)

The  $\theta$  and  $\phi_2$  angles of U are given as

$$\tan \theta = \frac{|\Omega_2(t)|}{-[\delta_2^2/4 + |\Omega_2(t)|^2]^{1/2} + \delta_2/2},$$
 (29)

with  $\phi_2$  being the argument of  $\Omega_2$  [Eq. (21)].

Operating with  $\underline{U}^{\dagger}$  on Eq. (25) and defining  $\underline{a} \equiv (a_1, a_2) \equiv \underline{U}^{\dagger} \cdot \underline{c}$  we obtain by neglecting  $\underline{A} = \underline{U} \cdot d\underline{U}^{\dagger}/dt$ , the nonadiabatic coupling matrix, the adiabatic approximation for  $\underline{a}$ ,

$$\frac{d}{dt}\underline{\mathbf{a}} = i\hat{\boldsymbol{\lambda}}(t)\underline{\mathbf{a}}(t), \qquad (30)$$

whose solution is,

$$a_{1,2}(t) = \exp\left\{\pm i \int_{0}^{t} \lambda(t') dt' \right\} a_{1,2}(0)$$
  
=  $\exp\left\{\pm i \int_{0}^{t} \left[\delta_{2}^{2}/4 + |\Omega_{2}(t')|^{2}\right]^{1/2} dt' \right\} a_{1,2}(0).$  (31)

To gain insight into requirements for the validity of the adiabatic approximation we consider the case of  $\phi_2=0$  for which the nonadiabatic coupling matrix <u>A</u> is given as

$$\underline{\underline{A}} = \begin{pmatrix} 0 & \dot{\theta} \\ -\dot{\theta} & 0 \end{pmatrix}.$$
(32)

Thus, in order for adiabaticity to hold, the time derivative of the mixing angle must be small with respect to the gap between the eigenvalues,

$$\left|\dot{\theta}(t)\right| \ll \left|\lambda_2(t) - \lambda_1(t)\right|. \tag{33}$$

We now introduce the (weak)  $\varepsilon_1(t)$  pulse. Since  $|E_1\rangle$  is the initially populated state, in the absence of  $\varepsilon_1(t)$ , neither the  $|E_0\rangle$  or  $|E_2\rangle$  states, nor the  $|\lambda_1\rangle$  and  $|\lambda_2\rangle$  adiabatic states can ever be populated. Thus, in the absence of the  $\varepsilon_1(t)$  pulse, the only noticeable effect of the  $\varepsilon_2(t)$  pulse is to change the spectrum of the Hamiltonian.

Assuming that the adiabatic condition [Eq. (33)] indeed holds, the states seen by the  $\varepsilon_1(t)$  pulse with  $\varepsilon_2(t)$  on, are the adiabatic states  $|\lambda_1\rangle$  and  $|\lambda_2\rangle$ , rather than the  $|E_0\rangle$  and  $|E_2\rangle$  material states. Using the definition of a [Eqs. (24) and (27)], we can write the adiabatic states, using the identity  $b=e^{-i\hat{\Delta}t/2}/U \cdot a$ , as

$$\begin{aligned} |\lambda_{1}(t)\rangle &= e^{i\int_{0}^{t}\lambda(t')dt'+i\delta_{2}t/2-iE_{0}t} \\ &\times \{\cos \theta | E_{0}\rangle + \sin \theta e^{-i\phi_{2}(t)-i\omega_{2,0}t} | E_{2}\rangle \}, \\ |\lambda_{2}(t)\rangle &= e^{-i\int_{0}^{t}\lambda(t')dt'-i\delta_{2}t/2-iE_{0}t} \\ &\times \{-\sin \theta e^{i\phi_{2}(t)} | E_{0}\rangle + \cos \theta e^{-i\omega_{2,0}t} | E_{2}\rangle \}. \end{aligned}$$
(34)

Here  $|\lambda_1(t)\rangle$  and  $|\lambda_2(t)\rangle$  are obtained by setting either a

=(a<sub>1</sub> 0) or <u>a</u>=(0 a<sub>2</sub>) and  $\omega_{2,0} \equiv (E_2 - E_0)/\hbar$ . When  $\delta_2 = 0$  (i.e., when  $\omega_2$  is exactly resonant with the  $|E_2\rangle$  to  $|E_0\rangle$  transition), it follows from Eq. (29) that  $\theta = 3\pi/4$ . If, in addition, we assume that the pulse has no chirp (i.e., that the phase of  $\mathcal{E}_2(t)$ ,  $\phi_2(t)=0$ ), we have that

$$\begin{aligned} |\lambda_1(t)\rangle &= e^{i\int_0^t |\Omega_2(t')| dt' - iE_0 t} \{ |E_0\rangle - e^{-i\omega_{2,0} t} |E_2\rangle \} / \sqrt{2}, \\ |\lambda_2(t)\rangle &= e^{-i\int_0^t |\Omega_2(t')| dt' - iE_0 t} \{ |E_0\rangle + e^{-i\omega_{2,0} t} |E_2\rangle \} / \sqrt{2}. \end{aligned}$$
(35)

We see that the time evolution of the  $|E_0\rangle$  component of  $|\lambda_1(t)\rangle$  is governed by a "quasienergy" of  $E_0 - |\Omega_2(t)|$ , whereas the time evolution of the  $|E_0\rangle$  component of  $|\lambda_2(t)\rangle$  is governed by a "quasienergy" of  $E_0 + |\Omega_2(t)|$ . We say that

the two levels are "Autler-Townes" split by an amount equal to  $2|\Omega_2(t)|$ .

We now consider the broadening of the adiabatic levels due to the decay channels considered in Sec. II. When this broadening is comparable to, or in excess of the  $2|\Omega_2(t)|$ splitting (see Fig. 3) the switch on of the  $\varepsilon_1(t)$  pulse results in the simultaneous excitation of the two adiabatic eigenstates. The probability of a one-photon absorption to each scattering state is given in first-order perturbation theory as,

$$P_{\mathbf{n}}(E) = |2\pi\epsilon_1(\omega_{E,1})\mu_{1,\mathbf{n}}(E)|^2, \qquad (36)$$

where  $\mu_{1,n}(E)$  are bound-free dipole matrix elements between the ground (g) and excited (e) electronic states, given as

$$\boldsymbol{\mu}_{1,\mathbf{n}}(E) = \langle E_1 | \vec{\boldsymbol{\mu}}_{e,g} \hat{\boldsymbol{\varepsilon}} | E, \mathbf{n}^- \rangle. \tag{37}$$

 $\omega_{E,1}$  of Eq. (36) is the transition frequency between  $E_1$  and a continuum energy E,  $\omega_{E,1} = (E - E_1)/\hbar$ .  $\epsilon_1(\omega)$  is the temporal Fourier transform of the pulse,

$$\boldsymbol{\epsilon}_{1}(\boldsymbol{\omega}) \equiv \frac{1}{2\pi} \int dt \boldsymbol{\varepsilon}_{1}(t) \exp[-i\boldsymbol{\omega}(z/c-t)], \qquad (38)$$

where z is the direction of propagation of the light. Assuming for simplicity that no direct transitions to the continuum occur, i.e., that only the Q space of Eq. (7) is coupled radiatively to  $|E_1\rangle$ , we have that

$$\mu_{1,\mathbf{n}}(E) = \sum_{s} \langle E_1 | \vec{\mu}_{e,g} \hat{\boldsymbol{\varepsilon}} | \boldsymbol{\phi}_s \rangle \langle \boldsymbol{\phi}_s | E, \mathbf{n}^- \rangle.$$
(39)

If we now identify the  $|\phi_s\rangle$  bound states (which become resonances due to the interaction with the continuum) with the adiabatic states of Eq. (34), we can write that,

$$\boldsymbol{\mu}_{1,\mathbf{n}}(E) = \sum_{s=1,2} \langle E_1 | \vec{\boldsymbol{\mu}}_{e,g} \hat{\boldsymbol{\varepsilon}} | \boldsymbol{\lambda}_s \rangle \langle \boldsymbol{\lambda}_s | E, \mathbf{n}^- \rangle, \qquad (40)$$

where we have specialized the treatment to the interaction of just two resonances. Using Eq. (34) and the fact that  $\langle E_2 | \hat{\epsilon} \cdot \vec{\mu} | E_1 \rangle = 0$ , we have that

$$\mu_{1,\mathbf{n}}(E) = \langle E_1 | \vec{\mu}_{e,g} \hat{\boldsymbol{\varepsilon}} | E_0 \rangle e^{-iE_0 t} \Big\{ \cos \theta e^{i\delta_2 t/2 + i\int_0^t \lambda(t')dt'} \langle \lambda_1 | E, \mathbf{n}^- \rangle \\ -\sin \theta e^{-i\delta_2 t/2 - i\int_0^t \lambda(t')dt' + i\phi_2} \langle \lambda_2 | E, \mathbf{n}^- \rangle \Big\}.$$
(41)

When  $\omega_2 = \omega_{2,0}$ , i.e., it is exactly on resonance, and  $\phi_2 = 0$ , this expression reduces to

$$\mu_{1,\mathbf{n}}(E) = \frac{1}{\sqrt{2}} e^{-iE_0 t} \langle E_1 | \vec{\mu}_{e,g} \hat{\boldsymbol{\varepsilon}} | E_0 \rangle \Big\{ \langle \lambda_1 | E, \mathbf{n}^- \rangle e^{i\int_0^t |\Omega_2(t')| dt'} \\ - \langle \lambda_2 | E, \mathbf{n}^- \rangle e^{-i\int_0^t |\Omega_2(t')| dt'} \Big\}.$$
(42)

Using Eq. (10) we can write an exact expression for the amplitude of observing the  $|\lambda_s\rangle$  states as

$$\langle \boldsymbol{\lambda}_{s} | \boldsymbol{E}, \mathbf{n}^{-} \rangle = \sum_{s'} \langle \boldsymbol{\lambda}_{s} | [\boldsymbol{E} - i\boldsymbol{\epsilon} - \boldsymbol{Q}\mathcal{H}\boldsymbol{Q}]^{-1} | \boldsymbol{\lambda}_{s'} \rangle \langle \boldsymbol{\lambda}_{s'} | \boldsymbol{H} | \boldsymbol{E}, \mathbf{n}_{1}^{-} \rangle.$$

$$(43)$$

Following Ref. [14] we can write the E-QHQ matrix of Eq. (13) in the two overlapping resonances case as

$$E - Q\mathcal{H}Q = \begin{pmatrix} E - E_0 - \lambda_1 - \Delta_{1,1} - i\Gamma_{1,1}/2 & -\Delta_{1,2} - i\Gamma_{1,2}/2 \\ -\Delta_{2,1} - i\Gamma_{2,1}/2 & E - E_0 - \lambda_2 - \Delta_{2,2} - i\Gamma_{2,2}/2 \end{pmatrix},$$
(44)

where

$$\Gamma_{s,s'}(E) = 2\pi \sum_{\mathbf{n}} \langle \lambda_s | H | E, \mathbf{n}_1^- \rangle \langle E, \mathbf{n}_1^- | H | \lambda_{s'} \rangle$$
(45)

and

$$\Delta_{s,s'}(E) = \mathsf{P}_{v} \int dE' \sum_{\mathbf{n}} \frac{\langle \lambda_{s} | H | E', \mathbf{n}_{1}^{-} \rangle \langle E', \mathbf{n}_{1}^{-} | H | \lambda_{s'} \rangle}{E - E'}.$$
(46)

We obtain that the inverse matrix is given as

$$[E - Q\mathcal{H}Q]^{-1} = \frac{1}{\mathcal{D}} \begin{pmatrix} E - E_0 - \lambda_2 - \Delta_{2,2} - i\Gamma_{2,2}/2 & \Delta_{1,2} + i\Gamma_{1,2}/2 \\ \Delta_{2,1} + i\Gamma_{2,1}/2 & E - E_0 - \lambda_1 - \Delta_{1,1} - i\Gamma_{1,1}/2 \end{pmatrix},$$
(47)

where

$$\mathcal{D} = (E - E_0 - \lambda_1 - \Delta_{1,1} - i\Gamma_{1,1}/2)(E - E_0 - \lambda_2 - \Delta_{2,2} - i\Gamma_{2,2}/2) - (\Delta_{1,2} + i\Gamma_{1,2}/2)(\Delta_{2,1} + i\Gamma_{2,1}/2).$$
(48)

Using Eq. (43) we obtain that

$$\langle \lambda_{1} | E, \mathbf{n}^{-} \rangle = \frac{1}{\mathcal{D}} [(E - E_{0} - \lambda_{2} - \Delta_{2,2} - i\Gamma_{2,2}/2) \langle \lambda_{1} | H | E, \mathbf{n}_{1}^{-} \rangle + (\Delta_{1,2} + i\Gamma_{1,2}/2) \langle \lambda_{2} | H | E, \mathbf{n}_{1}^{-} \rangle],$$

$$\langle \lambda_{2} | E, \mathbf{n}^{-} \rangle = \frac{1}{\mathcal{D}} [(\Delta_{2,1} + i\Gamma_{2,1}/2) \langle \lambda_{1} | H | E, \mathbf{n}_{1}^{-} \rangle + (E - E_{0} - \lambda_{1} - \Delta_{1,1} - i\Gamma_{1,1}/2) \langle \lambda_{2} | H | E, \mathbf{n}_{1}^{-} \rangle].$$
(49)

It follows from Eq. (34) that

$$\langle \lambda_1 | H | E, \mathbf{n}_1^- \rangle = e^{-i \int_0^t \lambda(t') dt' - i \delta_2 t/2 + i E_0 t} \\ \times \{ \cos \theta V_{0,\mathbf{n}} + \sin \theta e^{i \phi_2 + i \omega_{2,0} t} V_{2,\mathbf{n}} \},$$

$$\begin{aligned} \langle \boldsymbol{\lambda}_2 | \boldsymbol{H} | \boldsymbol{E}, \mathbf{n}_1^- \rangle &= e^{i f_0^t \boldsymbol{\lambda}(t') dt' + i \delta_2 t/2 + i E_0 t} \\ &\times \{ -\sin \theta e^{-i \phi_2} V_{0,\mathbf{n}} + \cos \theta e^{i \omega_{2,0} t} V_{2,\mathbf{n}} \}, \end{aligned}$$
(50)

where

$$V_{0,\mathbf{n}}(E) \equiv \langle E_0 | H | E, \mathbf{n}_1^- \rangle, \quad V_{2,\mathbf{n}}(E) \equiv \langle E_2 | H | E, \mathbf{n}_1^- \rangle.$$
(51)

Using Eq. (50) and neglecting the terms containing the highly oscillatory  $e^{\pm i\omega_{2,0}t}$  factors, we obtain that

$$\Gamma_{1,1} = \Gamma_0 \cos^2 \theta + \Gamma_2 \sin^2 \theta, \quad \Gamma_{2,2} = \Gamma_0 \sin^2 \theta + \Gamma_2 \cos^2 \theta,$$

$$\Gamma_{1,2} = (-\Gamma_0 + \Gamma_2)e^{-2if_0^i\lambda(t')dt' - i\delta_2 t + i\phi_2}\sin\theta\cos\theta, \quad (52)$$

where

$$\Gamma_{0}(E) \equiv 2\pi \sum_{\mathbf{n}} |\langle E, \mathbf{n}_{1}^{-} | H | E_{0} \rangle|^{2},$$
  

$$\Gamma_{2}(E) \equiv 2\pi \sum_{\mathbf{n}} |\langle E, \mathbf{n}_{1}^{-} | H | E_{2} \rangle|^{2}.$$
(53)

In the same manner

$$\Delta_{1,1} = \Delta_0 \cos^2 \theta + \Delta_2 \sin^2 \theta, \quad \Delta_{2,2} = \Delta_0 \sin^2 \theta + \Delta_2 \cos^2 \theta,$$

$$\Delta_{1,2} = (-\Delta_0 + \Delta_2)e^{-2i\int_0^t \lambda(t')dt' - i\delta_2 t + i\phi_2}\sin\theta\cos\theta, \quad (54)$$

where

$$\Delta_i(E) \equiv \left(\frac{1}{2\pi}\right) \mathsf{P}_v \int dE' \frac{\Gamma_i(E')}{E - E'}, \quad i = 0, 2.$$
 (55)

Hence from Eq. (49)

$$\langle \lambda_{1} | E, \mathbf{n}^{-} \rangle = \frac{e^{iE_{0}t}}{\mathcal{D}} \Big[ (E - E_{0} - \lambda_{2} - \Delta_{2,2} - i\Gamma_{2,2}/2) e^{-i\int_{0}^{t} \lambda(t')dt' - i\delta_{2}t/2} \{\cos \theta V_{0,\mathbf{n}} + \sin \theta e^{i\phi_{2} + i\omega_{2,0}t} V_{2,\mathbf{n}} \} \\ + (\Delta_{1,2} + i\Gamma_{1,2}/2) e^{i\int_{0}^{t} \lambda(t')dt' + i\delta_{2}t/2} \{-\sin \theta e^{-i\phi_{2}} V_{0,\mathbf{n}} + \cos \theta e^{i\omega_{2,0}t} V_{2,\mathbf{n}} \} \Big],$$

$$\langle \lambda_{2} | E, \mathbf{n}^{-} \rangle = \frac{e^{iE_{0}t}}{\mathcal{D}} \Big[ (\Delta_{2,1} + i\Gamma_{2,1}/2) e^{-i\int_{0}^{t} \lambda(t')dt' - i\delta_{2}t/2} \{\cos \theta V_{0,\mathbf{n}} + \sin \theta e^{i\phi_{2} + i\omega_{2,0}t} V_{2,\mathbf{n}} \} \\ + (E - E_{0} - \lambda_{1} - \Delta_{1,1} - i\Gamma_{1,1}/2) e^{i\int_{0}^{t} \lambda(t')dt' + i\delta_{2}t/2} \{-\sin \theta e^{-i\phi_{2}} V_{0,\mathbf{n}} + \cos \theta e^{i\omega_{2,0}t} V_{2,\mathbf{n}} \} \Big].$$

$$(56)$$

It follows from Eq. (41) that

$$\mu_{1,\mathbf{n}}(E) = \frac{\mu_{1,0}}{\mathcal{D}} \{\cos \theta [\{E - E_0 - \lambda_2 - (\Delta_0 + i\Gamma_0/2)\sin^2 \theta - (\Delta_2 + i\Gamma_2/2)\cos^2 \theta \} \{\cos \theta V_{0,\mathbf{n}} + \sin \theta e^{i\phi_2 + i\omega_{2,0}t} V_{2,\mathbf{n}} \} \\ + \{-\Delta_0 - i\Gamma_0/2 + \Delta_2 + i\Gamma_2/2\} e^{i\phi_2} \sin \theta \cos \theta \{-\sin \theta e^{-i\phi_2} V_{0,\mathbf{n}} + \cos \theta e^{i\omega_{2,0}t} V_{2,\mathbf{n}} \} ] \\ - \sin \theta e^{i\phi_2} [\{-\Delta_0 - i\Gamma_0/2 + \Delta_2 + i\Gamma_2/2\} e^{-i\phi_2} \sin \theta \cos \theta \{\cos \theta V_{0,\mathbf{n}} + \sin \theta e^{i\phi_2 + i\omega_{2,0}t} V_{2,\mathbf{n}} \} ] \\ + \{E - E_0 - \lambda_1 - (\Delta_0 + i\Gamma_0/2)\cos^2 \theta - (\Delta_2 + i\Gamma_2/2)\sin^2 \theta \} \{-\sin \theta e^{-i\phi_2} V_{0,\mathbf{n}} + \cos \theta e^{i\omega_{2,0}t} V_{2,\mathbf{n}} \} ] \}.$$
(57)

When we neglect the highly oscillatory  $e^{i\omega_{2,0}t}$  terms, we obtain

$$\mu_{1,\mathbf{n}}(E) = \frac{\mu_{1,0}V_{0,\mathbf{n}}}{\mathcal{D}} [E - E_0 + \lambda \cos 2\theta - (\Delta_2 + i\Gamma_2/2)].$$
(58)

Thus, if state  $|E_2\rangle$  is unstable, giving rise to the  $i\Gamma_2$  term, there is no real *E* for which the transition dipole matrix element vanishes.

When the detuning 
$$\delta_2 = 0$$
,  $\cos 2\theta = 0$  and

$$\mu_{1,\mathbf{n}}(E) = \frac{1}{\mathcal{D}} \mu_{1,0} V_{0,\mathbf{n}} [E - E_0 - (\Delta_2 + i\Gamma_2/2)].$$
(59)

When we substitute the explicit form of  $\mathcal{D}$ , as given by Eq. (48), and the values of  $\Gamma_{i,j}$  as given by Eq. (52), we obtain that

$$\mathcal{D} = [E - E_0 - \lambda - \Delta_0 \cos^2 \theta - \Delta_2 \sin^2 \theta - i(\Gamma_0 \cos^2 \theta + \Gamma_2 \sin^2 \theta)/2] \times [E - E_0 + \lambda - \Delta_0 \sin^2 \theta - \Delta_2 \cos^2 \theta - i(\Gamma_0 \sin^2 \theta + \Gamma_2 \cos^2 \theta)/2] - [\Delta_0 - \Delta_2 + i(\Gamma_0 - \Gamma_2)/2]^2 \sin^2 \theta \cos^2 \theta.$$
(60)

When we irradiate exactly on resonance,  $\delta_2 = 0$ , and we have that  $\cos^2 \theta = \sin^2 \theta = 1/2$ . We obtain in that case that

$$\mathcal{D}(t) = \left[E - E_0 - (\Delta_0 + \Delta_2)/2 - i(\Gamma_0 + \Gamma_2)/4\right]^2 - \left[(\Delta_0 - \Delta_2)/2 + i(\Gamma_0 - \Gamma_2)/4\right]^2 - \lambda^2(t).$$
(61)

#### A. Unstructured continua

If we neglect the variation of  $\Gamma_{0,2}(E)$  with energy, and assume that  $E \ge 0$ , we have that the integrand defining  $\Delta_{0,2}$  is antisymmetric about *E* and is essentially zero at the integration limits, hence  $\Delta_i \approx 0$ . In that case

$$\mathcal{D}(t) = [E - E_0 - i(\Gamma_0 + \Gamma_2)/4]^2 + [\Gamma_0 - \Gamma_2]^2/16 - |\Omega_2(t)|^2$$
(62)

or

$$\mathcal{D}(t) = (E - E_0)^2 - i(E - E_0)(\Gamma_0 + \Gamma_2)/2 - \Gamma_0 \Gamma_2/4 - |\Omega_2(t)|^2.$$
(63)

Hence

(in a.u.) 0.04

.0 ⊈(E)

20.0 [E]

0.

-0.02

$$|\mathcal{D}(t)|^{2} = [(E - E_{0})^{2} - \Gamma_{0}\Gamma_{2}/4 - |\Omega_{2}(t)|^{2}]^{2} + [(E - E_{0})(\Gamma_{0} + \Gamma_{2})]^{2}/4.$$
(64)

The channel-specific probability of absorption of a photon of energy  $E-E_1$  from state  $|E_1\rangle$  is given, using Eqs. (59), (64), and (36) (assuming  $\Delta_2=0$ ) as

$$P_{\mathbf{n}}(E) = \frac{|2\pi\mu_{1,0}V_{0,\mathbf{n}}\boldsymbol{\epsilon}_{1}(\omega_{E,1})|^{2}[(E-E_{0})^{2}+\Gamma_{2}^{2}/4)]}{[(E-E_{0})^{2}-|\Omega_{2}(t)|^{2}-\Gamma_{0}\Gamma_{2}/4]^{2}+[(E-E_{0})(\Gamma_{0}+\Gamma_{2})/2]^{2}}.$$
(65)

P(E), the total probability for absorbing a photon of energy  $E-E_1$  from state  $|E_1\rangle$ , given as  $P=\sum nP_n(E)$ , is

$$P(E) = \frac{2\pi\Gamma_0|\mu_{1,0}\epsilon_1(\omega_{E,1})|^2[(E-E_0)^2 + \Gamma_2^2/4)]}{[(E-E_0)^2 - |\Omega_2(t)|^2 - \Gamma_0\Gamma_2/4]^2 + [(E-E_0)(\Gamma_0 + \Gamma_2)/2]^2},$$
(66)

where we have used Eq. (53), according to which,  $\Gamma_0 = 2\pi\Sigma_n |V_{0,n}|^2$ . We see that the basic form of the total photonabsorption probability remains essentially the same as in the single continuum case, with sums over channel-specific widths and shifts replacing the single-channel entities.



=0

## **B.** Structured continua

When the variation of  $\Gamma_{0,2}(E)$  with energy cannot be neglected we cannot assume that  $\Delta_{0,2}(E)$  vanish. In that case we need to compute  $\mathcal{D}$  according to Eq. (60). Assuming that  $\Gamma_{0,2}(E)$  can be parametrized, for example, as a sum of Lorentzian functions

$$\Gamma_i(E) = \sum_j \frac{A_{i,j} \gamma_{i,j}}{(E - e_{i,j})^2 + \gamma_{i,j}^2/4}, \quad i = 0, 2,$$
(67)

we have that

$$\Delta_{i}(E) = \frac{1}{2\pi} \mathsf{P}_{\nu} \int_{-\infty}^{\infty} dE' \frac{\Gamma_{i}(E')}{E - E'} = \frac{1}{2\pi} \mathsf{P}_{\nu} \int_{-\infty}^{\infty} dE' \sum_{j} \\ \times \frac{A_{i,j} \gamma_{i,j}}{(E' - e_{i,j} + i \gamma_{i,j}/2)(E' - e_{i,j} - i \gamma_{i,j}/2)(E - E')} \\ = \sum_{j} \frac{A_{i,j}(E - e_{i,j})}{(E - e_{i,j})^{2} + \gamma_{i,j}^{2}/4}.$$
(68)

An illustration of a typical case of structured continua of the dressed AT split pair of states is given in Fig. 2. We see



FIG. 3. The formation of the EIT "hole" for unstructured continuum.  $\Gamma_0=0.05$ ,  $\Gamma_2=0$ , and  $\delta_2=0$ . Shown is the absorption line shape [P(E)], i.e., the absorption probability per unit energy, at three different times in the history of the guiding pulse  $[\varepsilon_2(t)]$ : (**x**)—at the peak of the guiding pulse t=0, ([]) as the pulse begins to wane, t=0.75, and (unmarked line)—at the tail of the pulse, t=1.5. A simple Gaussian pulse of the form  $\mathcal{E}_2(t)=\mathcal{E}_0 \exp(-t^2)$  was assumed.

that the shift function is *antisymmetric* about  $e_i$ , i=0,2. As a result, as shown e.g., in Figs. 7 and 8, the probability of absorption, given as the square of  $\mu_{1,n}(E)$  of Eq. (58), is no longer symmetric about the line center. Thus the appearance of asymmetry in the EIT absorption line shape is a hallmark of a structured continuum.

#### **IV. NUMERICAL EXAMPLES**

We present illustrative calculations on a three-level system of the type depicted in Fig. 1. We plot  $P_{\mathbf{n}}(E)$ , the absorption probability from the ground state  $(|E_1\rangle)$ , which is electric-dipole coupled to an upper state  $(|E_0\rangle)$ , that is in turn linked by a strong optical pulse to a third level  $(|E_2\rangle)$ . The



FIG. 4. The same as in Fig. 3 for a broadened  $|E_2\rangle$  ( $\Gamma_2=0.01$ ) with zero detuning ( $\delta_2=0$ ).





FIG. 5. The same as in Fig. 3 for a broadened  $|E_2\rangle$  ( $\Gamma_2=0.01$ ) with finite detuning ( $\delta_2=0.005$ ).

absorption line shapes are plotted as a function of the detuning from the  $E_1-E_0$  resonance at three different times in the history of the Gaussian pulse linking  $|E_0\rangle$  to  $|E_2\rangle$ , with t=0being the pulse maximum. The  $|E_0\rangle$  and  $|E_2\rangle$  states are coupled nonradiatively to some continuum channels representing the *P* space.

We first analyze the situation for *unstructured continua*. Figure 3 shows the situation when only the  $|E_0\rangle$  level (the one with the dipole-allowed transition to the ground state) is broadened ( $\Gamma_0=0.05$ ). We assume no detuning ( $\delta_2=0$ ) of the center of the strong pulse connecting the  $|E_0\rangle \leftrightarrow |E_2\rangle$  states. A *perfect* EIT dip is seen to arise. In contrast, Fig. 4 shows the zero-detuning ( $\delta_2=0$ ) situation when both levels ( $|E_0\rangle$  and  $|E_2\rangle$ ) are broadened ( $\Gamma_0=0.05$ ,  $\Gamma_2=0.01$ ). In this case, as clearly shown in Eqs. (65) and (66), the line shape does not dip to zero. Figure 5 pertains to the case when both levels ( $|E_0\rangle$  and  $|E_2\rangle$ ) are broadened ( $\Gamma_0=0.05$ ,  $\Gamma_2=0.01$ ) in the presence of detuning ( $\delta_2=0.005$ ). Here, the EIT does not dip to zero and the whole line shape is asymmetrically biased to the blue by an amount that depends on the intensity of the  $|E_0\rangle \leftrightarrow |E_2\rangle$  guiding field.



FIG. 6. EIT for *slightly* structured continuum ( $\gamma_0 = \gamma_2 = 0.5$ ) with no detuning ( $\delta_2 = 0$ ). The other parameters are as in Fig. 4.



FIG. 7. EIT for *highly* structured continuum ( $\gamma_0 = \gamma_2 = 0.05$ ) with no detuning ( $\delta_2 = 0$ ). The other parameters are as in Fig. 4.

We now turn our attention to the *structured continua* case. Figure 6 displays the absorption of two AT split levels for *slightly* structured continuum ( $\gamma_0 = \gamma_2 = 0.5$ ) with no detuning ( $\delta_2 = 0$ ). A slightly asymmetric line shape biased to the red is seen to arise. We next sharpen the continuum structure by letting  $\gamma_0 = \gamma_2 = 0.05$ , while applying the  $|E_0\rangle \leftrightarrow |E_2\rangle$  linking field *on resonance* ( $\delta_2 = 0$ ). As shown in Fig. 7, the line shapes are now highly *asymmetric*. This asymmetry is due, as shown in Fig. 2, to the asymmetry in the shift functions,  $\Delta_{0,2}(E)$ .

Finally, in Fig. 8 we show EIT for a *highly* structured continuum ( $\gamma_0 = \gamma_2 = 0.05$ ) with detuning ( $\delta_2 = 0.005$ ). As already shown in Fig. 5, the detuning introduces a bias to the blue, which counters the bias to the red introduced by the continuum structures. As a result, in comparison with Fig. 7, the line-shape asymmetry is somewhat reduced.



FIG. 8. EIT for *highly* structured continuum ( $\gamma_0 = \gamma_2 = 0.05$ ) with detuning ( $\delta_2 = 0.005$ ) at four values of the guiding pulse, with the uppermost trace corresponding to the peak of the guiding pulse. The lower energy peak was divided by a factor of 2 for clarity. The other parameters are as in Fig. 4.

#### **V. CONCLUSIONS**

We have presented a comprehensive theory of EIT in which both the structure and multiplicity of (coupled) continua are taken into account. The present treatment emphasizes the fact that EIT is a manifestation of interferences in the continuum. As such, it is a property of the way the full continuum eigenfunctions are convoluted with the matterradiation Hamiltonian and the initial bound states. The exact nature of this convolution depends on the type of spectroscopy used to probe the continuum; whether it is linear, or nonlinear, but the EIT line shapes, and especially the EIT dips, are properties of the continuum. They do not exist in purely bound state systems.

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