Equal area property of narrow resonances induced in coupled Doppler-broadened systems by intense laser radiation*

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It is well known that the *spectral line shape* of the narrow change signal induced at one Doppler-broadened transition by an intense laser field resonating with a second one can be extremely complex and nonclassical in form, the exact details depending on such factors as the direction of observation relative to the propagation direction of the laser field and its spatial form (traveling wave or standing wave). Nevertheless, the *area* under the change signal line shape, i.e. its frequency-integrated intensity, is always the same for a given laser field, and is independent of the coupling between laser and change-signal fields. A proof of this *equal area property*, based on quantum-mechanical transition-rate theory, is given for a three-level system composed of degenerate or near-degenerate states. The resulting expression is also applicable to line-shape calculations for experiments which study resonance fluorescence induced by intense laser or resonance radiation or other forms of excitation.

In a recently introduced spectroscopic technique called laser-induced line narrowing,[1] one observes the radiation arising from a Doppler-broadened transition as influenced by an intense laser field resonating with a coupled transition[2] (Fig. 1). Viewed along the axis of the laser field, a narrow resonant change signal appears superimposed on the Doppler profile. This change signal can be studied either in spontaneous emission or by probing with a weak tunable monochromatic field, and it was shown in Ref. 1 that the spectral line shape observed in the two cases is the same.

The overall behavior of the change signal can be understood by noting that the applied laser field induces changes in the population of the common level over a narrow range of velocities, and this change in the velocity distribution manifests itself in the radiation at the coupled transition. In many cases, however, the spectral line shape of the change signal is not classical in form and cannot be accounted for on the basis of population changes alone.[3-8] For instance, the line shape of the change signal viewed in the propagation direction of the laser field can be significantly different from that viewed in the reverse direction. Furthermore, under certain conditions the change signal splits into two. In other cases one portion of the line shape may be in the absorption phase while another portion is in the emission phase. In fact, even if the level populations of the laser transition are equal, so that no net population change can be induced by the laser field, a change signal at the coupled transition can still occur, an effect certainly not explainable on the basis of population changes. These nonclassical effects occur because of the coherent nature of the interaction between the laser field and the radiation at the coupled transition, which gives rise to Raman-type transitions[2] in which atoms produced in one level can make coherent transitions to the other levels without loss of phase memory.

Nevertheless, in spite of the complex shape of the change signals, the area under the change signal line shape, i.e. its frequency-integrated intensity, is always the same as that predicted on the basis of population changes.[2] We refer to this result as the *equal area property*. This feature is not restricted to inhomogeneously broadened transitions. In fact, the equal area property was first pointed out in an analysis of a homogeneously broadened three-level system.[9] In this paper we shall show that the equal area property follows from general quantum-mechanical considerations, that it holds regardless of whether the transition is homogeneously or inhomogeneously broadened, and that it is independent of whether the radiation fields are in the form of travelling waves or standing waves.

The proof is based on the derivation of an expression for the frequency-integrated spontaneous

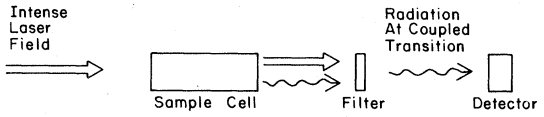

FIG. 1. Schematic experimental arrangement for observing laser-induced line-narrowing change signals. The change signals can be studied either in spontaneous emission or by probing with a weak monochromatic field.

emission intensity occurring at one transition in the presence of an intense laser field resonating with a coupled transition. This expression is also important in studies of resonance fluorescence induced by intense laser radiation, as in the standing-wave saturation technique,[10] in level crossing studies,[11,12] and in quantum-beat experiments.[13,14] In the standing-wave saturation technique one studies the narrow resonance produced when an intense *standing-wave* laser field is tuned through the central portion of a Doppler-broadened transition, as manifested in the spontaneous emission occurring at a coupled transition.[10] In level-crossing experiments one observes the fluorescence change signal emitted by a transition pumped by an intense cw travelling-wave laser field or by resonance radiation as a function of Stark or Zeeman tuning of the sample being studied.[11,12] In the quantum-beat effect one studies the modulation in the decay curve of the fluorescence emitted from near-degenerate coupled transitions excited by a short pulse.[13,14] In analyzing these effects, it is important to consider the degeneracy (or near degeneracy) of the energy levels involved.

A previous derivation[15] of an expression for laser-induced resonance fluorescence has been based on application of the correspondence principle to expressions for dipole radiation obtained from classical theory.[16] The resulting expression for the frequency-integrated emission signal depends, by virtue of the approach, only on the diagonal and near-diagonal density-matrix elements induced by the laser field. The off-diagonal elements, which give rise to Raman-type transitions as described above, are not accounted for in this treatment. The validity of this approach is not immediately obvious especially since, as explained above, the expression for the emission *line shape* from such a system does depend on the off-diagonal elements of the density matrix, and the line shape is very different from that predicted using classical arguments. The derivation presented here also provides a rigorous justification of the correspondence principle result for laser-induced resonance fluorescence.

The proof is obtained from quantum-mechanical transition rate theory.[17] Consider a three-level molecular system as shown in Fig. 2, where i, j, and k represent the close-lying states (degenerate or near-degenerate, e.g. M states) comprising levels 0, 1, and 2, respectively. The molecules are assumed to be at rest. (The results will be extended to moving molecules below.) Level 0, the common energy level, is radiatively coupled to levels 1 and 2. To be definite, level 0 is taken to lie above levels 1 and 2.[18] The energy of the mth sublevel ($m = i$, j, or k) is denoted by E_m and

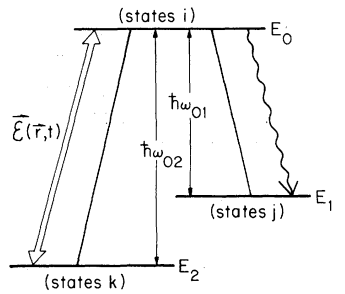

FIG. 2. Three-level system. Each level consists of a number of close-lying (degenerate or near-degenerate) states. An intense field $\vec{\mathcal{E}}(\vec{r},t)$ resonates with the 0-2 transition. The frequency-integrated spontaneous-emission intensity at the 0-1 transition is calculated.

$\hbar\omega_{mm'} = E_m - E_{m'}$ denotes the energy separation between states m and m'. Let $\vec{\mathcal{E}}(\vec{r}, t)$ represent a laser field at coordinates $(\vec{r}, t)$ of carrier frequency ω_L resonant with the 0-2 transition ($\omega_L \sim \omega_{02}$) but not with the 0-1 transition. The envelope of $\vec{\mathcal{E}}$ is taken to be a general function of space and time of arbitrary magnitude. We wish to calculate the fluorescence at the 0-1 transition emitted into a small solid angle, as influenced by the laser field resonating with the 0-2 transition. The problem is solved by considering the intense laser field to be classical and the spontaneous emission field to be quantized. A perturbation approach is adopted in which the laser field interacting with the molecules and the noninteracting background field together are considered to be the unperturbed system. The interaction of this background spontaneous emission field with the system is then treated as a small perturbation. The complete Hamiltonian is given by

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

where H_0 is the Hamiltonian of molecular system with stationary wave functions ϕ_m having energy eigenvalues E_m; H_f is the Hamiltonian of background radiation field with eigenstates $|n\rangle$ and energy eigenvalues $(n+\frac{1}{2})\hbar\omega$, with n being the photon occupation number and ω being close to ω_{01}; V is the interaction Hamiltonian coupling laser field to molecular system; and H' is the interaction Hamiltonian coupling background radiation field to molecular system.

The Schrödinger equation for the system is given by

$$(H_u + H')(\psi_u + \psi') = i\hbar \frac{\partial}{\partial t}(\psi_u + \psi'), \tag{2}$$

where

$$H_u = H_0 + H_f + V \tag{3}$$

is the Hamiltonian of the unperturbed system with wave function ψ_u and ψ' is the correction to the wave function caused by the perturbation H'.

Consider a molecule at position $\vec{r}$ and time t, produced in molecular state m at $t=t_0$ with no photons present in the background radiation field. The unperturbed wave function may be expanded in the form

$$\psi_u(\vec{r},t;t_0,m)=\sum_{m'=i,j,k} A_{m'}(\vec{r},t;t_0,m)\phi_{m'}|0\rangle\exp\left(-\frac{i}{\hbar}(E_{m'}+\tfrac{1}{2}\hbar\omega)(t-t_0)\right), \tag{4a}$$

where the A_m's, the time-varying probability amplitudes of the unperturbed system, are subject to the initial conditions

$$A_{m'}(\vec{r},t_0;t_0,m)=\delta(m,m'). \tag{4b}$$

At a later time the system may make a transition into state $|n\rangle$ of the background radiation field. The perturbation wave function will be of the form

$$\psi'(\vec{r},t;t_0,m)=\sum_n\sum_{m'} a_{m'n}(\vec{r},t;t_0,m)\phi_{m'}|n\rangle\ \exp\{-(i/\hbar)[E_{m'}+(n+\tfrac{1}{2})\hbar\omega](t-t_0)\}, \tag{5a}$$

where the $a_{m'n}$'s, the time-varying probability amplitudes of the perturbed system, are subject to initial conditions

$$a_{m'n}(\vec{r},t_0;t_0,m)=0. \tag{5b}$$

To lowest order in the perturbation Eq. (2) reduces to

$$H_u\psi'+H'\psi_u=i\hbar\frac{\partial}{\partial t}\psi'. \tag{6}$$

Substituting Eqs. (3)–(5) into Eq. (6), multiplying on the left side by $\langle 1|\phi_j^*$, and integrating over space in the usual manner, one obtains[19]

$$\dot{a}_{j1}=-\frac{i}{\hbar}\sum_i H'^*_{ji}A_i\,e^{-i(\omega_{ij}-\omega)(t-t_0)}, \tag{7}$$

where

$$H'_{ji}=\int\langle 1|\phi_j^*(\vec{R})H'\phi_i(\vec{R})|0\rangle\,d^3R. \tag{8}$$

Accordingly, the perturbation probability amplitudes a_{j1} may be expressed in terms of the probability amplitudes of the unperturbed system, which are assumed known. Note, however, that Eq. (7) cannot be integrated explicitly, since A_i is a time-varying function.[20]

The probability of a molecule produced in level m at time t_0 being found in any one of the states of level 1 at time $t>t_0$ accompanied by emission of a photon at frequency ω is given by

$$\sum_j\int|a_{j1}(\vec{r},t;t_0,m)|^2\rho(\omega)\,d\omega, \tag{9}$$

where $\sum_j$ indicates sum over all the states of level 1 and $\rho(\omega)$ is the density of final photon states of energy $\hbar\omega$ in volume V emitted into solid angle $d\Omega$, given by

$$\rho(\omega)=\omega^2\,d\Omega\,V/8\pi^3c^3. \tag{10}$$

The corresponding transition rate per atom is given by

$$W(\vec{r},t;t_0,m)=\frac{d}{dt}\sum_j\int|a_{j1}|^2\rho(\omega)\,d\omega$$

$$=2\mathrm{Re}\sum_j\int\dot{a}_{j1}a_{j1}^*\rho(\omega)\,d\omega, \tag{11}$$

and the corresponding emission intensity is $\hbar\omega W$. The net intensity of emission is comprised of contributions from molecules produced at all initial times t_0 in all possible states m. The rate of formation of molecules in level m is given by $n_m\gamma_m$, where n_m is the density of molecules in state m in the absence of the laser field and γ_m is the net rate of decay of population in level m due to radiative and collisional processes.[21] Therefore, the net intensity emitted at frequency ω into solid angle $d\Omega$, $I(\vec{r},t,d\Omega)$, is obtained by multiplying $\hbar\omega W$ by the rate of formation of molecules in state m, integrating over all possible initial times, and summing over initial states:

$$I(\vec{r},t,d\Omega)=\hbar\omega\sum_m n_m\gamma_m\int_{-\infty}^{t}W(\vec{r},t;t_0,m)\,dt_0. \tag{12}$$

This is the desired quantity.

It is now necessary to obtain an explicit expression for W. Inserting Eq. (7) into Eq. (11) and regrouping the factors, one obtains

$$W=\frac{2}{\hbar^2}\mathrm{Re}\sum_{j,i,i'}A_i(\vec{r},t;t_0,m)e^{-i\omega_{ii'}(t-t_0)}g, \tag{13}$$

where

$$\mathcal{I} = \int_{t_0}^{t} dt' A_{i'}^{*}(\vec{r}, t; t_0, m) \int_0^{\infty} d\omega \rho(\omega) H'_{i'j}(\omega) H'_{ji}(\omega) e^{i(\omega - \omega_{i'j})(t-t')} . \tag{14}$$

(Note that $H_{ij} = H_{ji}^{*}$.) It is convenient to choose new variables of integration $\tau = t - t'$ and $\xi = \omega - \omega_{i'j}$. Then

$$\mathcal{I} = \int_0^{t-t_0} d\tau A_i^{*}(\vec{r}, t-\tau; t_0, m) \int_{-\omega_{i'j}}^{\infty} d\xi \rho(\xi + \omega_{i'j}) H'_{i'j}(\xi + \omega_{i'j}) H'_{ji}(\xi + \omega_{i'j}) e^{i\xi\tau} . \tag{15}$$

The ρ and the H''s are slowly varying functions and the time-varying phase factor $\exp(i\xi\tau)$ is dominant. The latter is a rapidly varying function of ξ and τ except near the origin of the ξ-τ plane. Accordingly, the slowly varying factors can be removed from the integrand and evaluated at $\xi = 0$. This gives

$$\mathcal{I} = \rho(\omega) H'_{i'j} H'_{ji}(\omega) \int_0^{t-t_0} d\tau A_{i'}^{*}(\vec{r}, t-\tau; t_0, m) \times \left[\frac{e^{i\nu\tau} - e^{-i\omega_{i'j}\tau}}{i\tau} \right] , \tag{16}$$

where $\mathcal{I}$ is to be evaluated at $\nu \to \infty$. The function in brackets is large near $\tau = 0$ but falls to zero for $\tau \gg 1/\omega$. Since A_i does not vary appreciably over this time,[20] it may be removed from the integral and evaluated at $\tau = 0$. This gives

$$\mathcal{I} = -i\zeta\rho H'_{i'j} H_{ji} A_{i'}^{*} , \tag{17}$$

with

$$\zeta = \int_{-\omega_{i'j}(t-t_0)}^{\nu(t-t_0)} \frac{e^{iu}}{u} du . \tag{18}$$

Note that for times $t - t_0 \gg 1/\omega$ the integration limits of ζ are large, and ζ approaches its asymptotic limit. Accordingly, we have

$$W = \frac{2\rho}{\hbar^2} \operatorname{Re} \sum_{j,i,i'} (-i\zeta) H'_{i'j} H'_{ji} A_i A_{i'}^{*} e^{-i\omega_{ii'}(t-t_0)} \tag{19}$$

$$= \frac{2\rho \operatorname{Im}\zeta}{\hbar^2} \sum_{j,i,i'} H'_{i'j} H'_{ji} A_i A_{i'}^{*} e^{-i\omega_{ii'}(t-t_0)} . \tag{20}$$

Since for $\omega(t - t_0) \gg 1$,

$$\operatorname{Im}\zeta = 2 \int_0^{\infty} \frac{\sin u}{u} du = \pi , \tag{21}$$

we obtain

$$W = \frac{2\pi\rho}{\hbar^2} \sum_{j,i,i'} H'_{i'j} H'_{ji} [A_i e^{-i\omega_i(t-t_0)}][A_{i'} e^{-i\omega_{i'}(t-t_0)}]^{*} . \tag{22}$$

Inserting Eq. (22) into Eq. (12), one obtains an expression for the net intensity emitted at the 0-1 transition into solid angle $d\Omega$:

$$I(\vec{r}, t, d\Omega) = (2\pi\omega\rho/\hbar) \sum_{j,i,i'} \bar{\rho}_{ii'}(\vec{r}, t) H'_{i'j} H'_{ji} , \tag{23}$$

where ρ, Eq. (10), is the density of final states and $\bar{\rho}_{ii'}$, the ii' element of the ensemble-averaged density matrix, is defined as[22]

$$\bar{\rho}_{ii'}(\vec{r}, t) = \sum_m n_m \gamma_m \int_{-\infty}^{t} dt_0 [A_i e^{-i\omega_i(t-t_0)}] \times [A_{i'} e^{-i\omega_{i'}(t-t_0)}]^{*} . \tag{24}$$

The quantity $\bar{\rho}_{mm'}$ obeys the well-known equation of motion[22]

$$\left(\frac{\partial}{\partial t} + \vec{v} \cdot \nabla \right) \bar{\rho}_{mm'} = \frac{i}{\hbar} [H_u, \bar{\rho}]_{mm'} - \gamma_{mm'} [\bar{\rho}_{mm'} - n_m \delta(m, m')] , \tag{25}$$

where H_u is the Hamiltonian of the unperturbed system, Eq. (13), and $\gamma_{mm'}$ is the decay constant associated with the mm' element of $\bar{\rho}$. The left-hand side of Eq. (25) is the total ("hydrodynamic") derivative of $\bar{\rho}_{mm'}$ and automatically takes into account the motion of an ensemble of molecules moving with velocity $\vec{v}$.

Equation (23) is the desired expression for the frequency-integrated spontaneous emission intensity.[23] Note that the $\bar{\rho}_{ii'}$ may be obtained directly from Eq. (25). This is generally much simpler and more convenient than calculating the amplitudes A_i using the Schrödinger equation. Also note that in the case of a Doppler-broadened system it is necessary to integrate $\bar{\rho}_{ii'}$ over the distribution of molecular velocities.

The above derivation of Eq. (23) assumed that the pump transition is distinct from the transition at which the emission is observed (three-level system). A similar calculation for the case where the spontaneous emission at the pump frequency itself is studied (two-level system) shows that Eq. (23) holds identically in that case also.

For electric dipole radiation a more explicit form of Eq. (23) may be given. In this case matrix elements of H' are given by[24]

$$H'_{ji} = i(2\pi\hbar\omega/V)^{1/2} \mu_{ji} , \tag{26}$$

where

$$\mu_{ji} = \vec{\mu}_{ji} \cdot \hat{\epsilon} , \tag{27}$$

with $\vec{\mu}_{ji}$ the electric dipole matrix element con-

necting state i of level 0 with state j of level 1, and $\hat{\epsilon}$ the polarization vector of the emitted photon. Specializing to a Doppler-broadened system, Eq. (23) then becomes

$$I(\vec{r}, t, d\Omega) = \frac{\omega^4 d\Omega}{2\pi c^3} \sum_{j,i,i'} \mu_{i'j}\mu_{ji}\langle\bar{\rho}_{ii'}\rangle, \qquad (28)$$

where the brackets indicate integration over velocity.

Equation (28) has a simple form. It is similar to the usual expression for spontaneous emission intensity as would occur in the absence of the laser field, except that the sums are taken over the near-diagonal elements of $\bar{\rho}$, as well as the diagonal elements. The diagonal contributions represent the changes in level population induced by the intense field. The near-diagonal contributions are interference terms, and are due to the phase coherence induced between closely spaced states by the intense laser field. Note, however, that the off-diagonal elements of $\bar{\rho}$ do not enter into this expression.

Equation (28) was obtained in Ref. 12 in a different manner, by explicitly calculating the line shape of emission induced at the 0-1 transition in the presence of an intense resonant cw monochromatic travelling-wave field, and then integrating over the line shape.[25] The line shape expression was found to be nonclassical, and was dependent on the off-diagonal matrix elements of $\bar{\rho}$, as well as diagonal and near-diagonal elements. Nevertheless, after frequency integration the expression simplified and reduced to Eq. (28). It should be noted that in the derivation given here the form of the A_m's (and therefore the $\bar{\rho}_{mm'}$'s) was left unspecified. Accordingly, the present result is general and holds for an applied field of arbitrary spatial and temporal form resonating with the 0-2 transition. This includes excitation either by intense monochromatic laser radiation or resonance radiation, continuous or pulsed, in the form of a travelling or standing wave. In fact, the result also holds for other forms of selective excitation, such as by a monoenergetic electron beam, since in the derivation nothing was assumed to restrict the form of V, the interaction Hamiltonian coupling the excitation source to the 0-2 transition.

In a nondegenerate system Eq. (28) becomes

$$I(\vec{r}, t, d\Omega) = (\omega^4 d\Omega/2\pi c^3)\langle\bar{\rho}_{00}\rangle|\mu_{01}|^2, \qquad (29)$$

where $\langle\bar{\rho}_{00}\rangle$ is the velocity-integrated population of level 0, as influenced by the laser field. This result indicates that in a nondegenerate system the frequency-integrated change signal intensity only depends on the change in population of the common level (0) induced by the laser field, independent of the coupling of the laser field with the radiation emitted at frequency ω. This is the statement of the equal-area property of the laser-induced line-narrowing effect.

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[17]See, for example, W. Heitler, *The Quantum Theory of Radiation* (Oxford U. P., Oxford, 1960). The usual derivations, however, are not applicable here, since the intense laser field causes the probability amplitudes of the unperturbed system (A_m's) to be time dependent.
[18]The proof follows similarly when level 0 is not uppermost.

[19]In obtaining Eq. (7) note that (i) matrix elements of V connecting to level 1 may be ignored since the laser field does not resonate with the 0-1 transition; (ii) in considering spontaneous emission at the 0-1 transition only matrix elements of H' connecting levels 0 and 1 need be retained.

[20]For continuous and/or intense laser fields, A_i varies at a rate $\sim\mu_{02}\mathcal{E}/\hbar$, where μ_{02} is the dipole moment matrix element of the 0-2 transition. In the case of transient excitation A_i may also vary $\sim\gamma_i$, the decay rate of state i. Both of these rates are much smaller than ω.

[21]A detailed discussion of the steps involved in obtaining the ensemble-averaged transition rate from the transition rate per atom is given in Sec. II A of Ref. 1.

[22]See, for example, B. J. Feldman and M. S. Feld, Phys. Rev. A 1, 1375 (1970), Sec. 2.

[23]The above derivation can easily be extended to the case of the emission (or absorption) signal induced by a weak tunable monochromatic probe field by considering n photons to be initially present in the background radiation field mode [cf. Eq. (4a)]. One then finds that the expression for the frequency-integrated gain coefficient, $\int\alpha(\omega)\,d\omega$, only depends on the diagonal and near-diagonal elements of $\bar{\rho}$, and not on the off-diagonal elements. For example, the stimulated emission counterpart of Eq. (28) is that

$$\int\alpha(\omega)\,d\omega=\frac{4\pi^2\omega}{\hbar c}\sum_{i,j}\left[\sum_{i'}\mu_{i'j}\mu_{ji}\langle\bar{\rho}_{ii'}\rangle-\sum_{j'}\mu_{j'i}\mu_{ij}\langle\bar{\rho}_{jj'}\rangle\right].$$

[24]W. Heitler, Ref. 17.

[25]Equation (28) of the present work appears as Eq. (44) of Ref. 12. Note that in the denominator of Eqs. (43) and (44) of Ref. 12, the factor $\hbar$ should be replaced by 2π. In Eq. (38) the factor $\frac{1}{2}$ should be deleted. Also, the coordinate systems in Fig. 4 of Ref. 12 were incorrectly labeled. Throughout the figure the polarization vector ϵ of the laser field should be oriented in the vertical direction, and the Stark or Zeeman field should point to the right. Then in Fig. 4(b) the z axis should be vertical, and in Fig. 4(c) the x' axis should be vertical, with the z' axis pointing to the right. The angles of the coordinate systems should be relabeled correspondingly. With the above changes the intensity distributions are correct as drawn. A complete list of the errata for Ref. 12 will appear elsewhere.