Transit-time effects in power-broadened Doppler-free saturation resonances*

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A density-matrix calculation of the line shape for a Doppler-broadened two-level system subjected to a plane-standing-wave laser field with a Gaussian intensity profile is presented to fifth order in perturbation theory. The primary new result is that, for fixed pressure and intensity, the power-broadening contribution to the linewidth is at a minimum when the beam radius is approximately equal to the mean free path. In addition, an analytic solution for the linewidth in the collision-dominated regime is presented for general collision parameters.

I. INTRODUCTION

In recent years, the attainment of ultra-narrow Doppler-free saturation resonances[1-4] has made necessary the consideration of line-broadening effects due to the finite spatial extent of the laser field.[5,6] Several calculations which consider these effects on the Lamb dip have been reported. Transit-time effects in third-order perturbation theory were considered by Rautian and Shalagin.[7] In their paper the third-order line shape was numerically integrated; the line shape in the extreme transit-time regime was shown to be non-Lorentzian. Utilizing the results of Rautian and Shalagin, Baklanov *et al.*, using numerical techniques, have given the linewidth as function of the ratio of collision frequency to transit-time frequency.[8] In addition, they present closed-form expressions for the linewidth in the extreme collision dominated and transit-time dominated regimes.

Maeda and Shimoda have studied the effects of a Gaussian beam in a gas-laser cavity and find assymmetry in the Lamb dip and a shift in the center frequency.[9]

Bordé *et al.* have recently considered the transit-time problem for a Gaussian beam including the curvature of the phase front in a third-order perturbation calculation and find a shift in the line center. Hall and Bordé have verified this result experimentally.[10]

In the present paper, the line shape for a Doppler-broadened two-level system subjected to a plane-standing-wave laser field with a Gaussian intensity profile is given to fifth order in perturbation theory.[11] The fifth-order theory enables a determination of the transit-time corrections to a power-broadened line shape. As shown below, a general conclusion is that the power broadening contribution to the linewidth is significantly altered by the transit-time effects. In Sec. II, the exact line shape valid to fifth order in perturbation theory is calculated including the effects of spatial harmonics in the intermediate populations. In Sec. III, by specializing to a collision dominated regime, a closed-form expression for the lowest order transit-time effects on the power-broadened linewidth is obtained. Due to the approximations which are necessary to calculate the linewidth in the collision dominated limit, the contribution of the transit-time effects must be limited to a few percent of the linewidth in this regime. Thus the primary reason for the calculation is to determine the manner in which various transit-time effects are manifested in the linewidth. The results demonstrate that phenomenological transit-time rates cannot be simply added to the decay rates appearing in the density-matrix equations.

In Sec. IV, an analysis of the fifth-order contribution to the linewidth for a general mean-free path to beam radius ratio is presented using computer evaluation of the multidimensional integrals which emerge from the theory. The resulting plots of linewidth at fixed pressure and intensity versus transit-time frequency (defined below) indicate that the power-broadening first decreases and then increases as the transit-time frequency is increased. The line shapes in fifth order do not vary appreciably from the third-order line shapes which are given in Ref. (7) and are not shown.

II. CALCULATION OF THE LINE SHAPE

In this section the third- and fifth-order perturbation contributions to the line shape are calculated. The resulting expressions are valid to all orders in the transit-time effects which are characterized by the small parameter $\alpha u^2/\gamma_{12}^2$ (defined below). The derivation closely follows the method used by Lamb in his theory of the laser.[12]

The density-matrix equations for a two-level system traveling with velocity $\vec{v}$ and interacting with an external field are given by

$$\dot{\rho}_{11} = -(i/\hbar)\, V_{12}(\rho_{21}-\rho_{12}) - \gamma_1\rho_{11}\,, \tag{1}$$

$$\dot{\rho}_{22} = (i/\hbar)\, V_{12}(\rho_{21}-\rho_{12}) - \gamma_2\rho_{22}\,, \tag{2}$$

$$\dot{\rho}_{12} = -(i/\hbar)\, V_{12}(\rho_{22} - \rho_{11}) + i\omega_0 \rho_{12} - \gamma_{12}\rho_{12}\,, \tag{3}$$

$$\rho_{21} = \rho_{12}^{*}\,, \tag{4}$$

where γ_1 and γ_2 are energy decay rates of levels 1 and 2, respectively, and γ_{12} is the polarization decay rate. ρ_{11} is the probability that the two-level system is in the lower level and ρ_{22} is the corresponding probability for the upper level. ρ_{12} is the off-diagonal density matrix element and is related to the induced polarization. ω_0 is the resonance frequency and $\hbar$ is Planck's constant divided by 2π. $V_{12}(t)$ denotes the interaction with the external field and is given by

$$V_{12}(t) = -\vec{\mu} \cdot \vec{\mathrm{E}} = -\mu E\,, \tag{5a}$$

$$E = E_0 \cos\omega t \sin k\,[z_0 + v_z(t - t_0)] \times U(x_0 + v_x(t - t_0))\, U(y_0 + v_y(t - t_0))\,, \tag{5b}$$

where μ is the transition electric dipole moment, ω is the laser frequency, and $k = \omega/c$ is the wave vector. $V_{12}(t)$ represents the potential as seen by an atom which was created at position $\vec{x}_0$ at time t_0 with velocity $\vec{v}$. By a suitable choice of phase for the wave functions, one may take $V_{12} = V_{21}$. The electric field amplitude, E_0, is assumed to be a slowly varying function of z, i.e., $(dE_0/dz)\, l \ll E_0$, where l is the order of a mean-free path. Thus E_0 is taken to be locally constant and its z dependence may be neglected in the calculation of the absorbed power per unit length as discussed below [see Eq. (29)]. The Gaussian intensity profile of the beam is contained in the U factors in Eq. (5b), where

$$U(x) = e^{-\alpha x^2}\,, \quad \alpha = 1/R^2\,, \tag{6}$$

and R is the electric field $1/e$ radius. E_0 is normalized so that the traveling wave power, P_{TW}, is given by

$$P_{\mathrm{TW}} = \int_0^\infty \frac{C}{8\pi} \left(\frac{E_0}{2}\right)^2 \exp(-2\alpha r^2)\, 2\pi r\, dr\,. \tag{7}$$

For an atom created in state 1 at position $\vec{x}_0$ and time t_0 with velocity $\vec{v}$, one can obtain the following expressions for ρ_{12} to third and fifth order in the interaction V_{12}:

$$\begin{aligned}\rho_{12}^{(3)}(\vec{v}, t; \vec{x}_0, t_0; 1) = (i/\hbar)^3 \int_{t_0}^{t} dt_3 \int_{t_0}^{t_3} dt_2 \int_{t_0}^{t_2} dt_1 \exp[-(\gamma_{12} - i\omega_0)(t - t_3)]\, V_{12}(t_3) \\ \times\{\exp[-\gamma_2(t_3 - t_2)] + \exp[-\gamma_1(t_3 - t_2)]\} V_{12}(t_2) \\ \times\{\exp[-(\gamma_{12} - i\omega_0)(t_2 - t_1)] + \text{c.c.}\} V_{12}(t_1) \exp[-\gamma_1(t_1 - t_0)]\,.\end{aligned} \tag{8}$$

$$\begin{aligned}\rho_{12}^{(5)}(\vec{v}, t; \vec{x}_0, t_0; 1) = (i/\hbar)^5 \int_{t_0}^{t} dt_5 \int_{t_0}^{t_5} dt_4 \int_{t_0}^{t_4} dt_3 \int_{t_0}^{t_3} dt_2 \int_{t_0}^{t_2} dt_1 \exp[-(\gamma_{12} - i\omega_0)(t - t_5)]\, V_{12}(t_5) \\ \times\{\exp[-\gamma_2(t_5 - t_4)] + \exp[\gamma_1(t_5 - t_4)]\} V_{12}(t_4) \{\exp[-(\gamma_{12} - i\omega_0)(t_4 - t_3)] + \text{c.c.}\} \\ \times V_{12}(t_3) \{\exp[-\gamma_2(t_3 - t_2)] + \exp[-\gamma_1(t_3 - t_2)]\} V_{12}(t_2) \\ \times\{\exp[-(\gamma_{12} - i\omega_0)(t_2 - t_1)] + \text{c.c.}\} V_{12}(t_1) \exp[-\gamma_1(t_1 - t_0)]\,.\end{aligned} \tag{9}$$

Since integral perturbation techniques are well-known only the final results above are presented. The contribution from atoms originating in state 2 is neglected in determining the absorbed power, since the ratio of the number of atoms initially in state 2 to the number of atoms initially in state 1 is determined by the Boltzmann factor which is small for an infrared or optical transition.

Only atoms which arrive at position $\vec{x}$ at time t influence a measurement of the polarization at $(\vec{x}, t)$. Thus one must replace $\vec{x}_0$ by $\vec{x} - \vec{v}(t - t_0)$ in the potentials. Therefore, let $V_{12}(t_n) \to V_{12}'(\vec{x}, t; \vec{v}, t_n)$ in Eqs. (8) and (9), where

$$V_{12}'(\vec{x}, t; \vec{v}, t_n) = -\mu E_0 \cos\omega t_n \sin k[z - v_z(t - t_n)] \times U(x - v_x(t - t_n))\, U(y - v_y(t - t_n))\,. \tag{10}$$

This result merely expresses the time dependence of the potential as seen by a contributing atom with velocity $\vec{v}$ in terms of its final coordinates $(\vec{x}, t)$ instead of its initial coordinates $(\vec{x}_0, t_0)$.

For a gas in thermal equilibrium, the number of atoms created in state 1 with velocity $\vec{v}$ at time t_0 is given by $N_1^{(0)}(\vec{v})\gamma_1 dt_0$, where

$$N_1^{(0)}(\vec{v}) = N_1^{(0)} W(\vec{v})\,, \tag{11}$$

and $W(\vec{v})$ is a normalized Maxwellian velocity distribution. Therefore, the total contribution to $\rho_{12}(\vec{x}, \vec{v}, t; 1)$ due to atoms created at all initial times, t_0, is given by

$$\rho_{12}(\vec{x}, \vec{v}, t; 1) = \int_{-\infty}^{t} N_1^{(0)}(\vec{v})\gamma_1 dt_0\, \rho_{12}(\vec{x}, \vec{v}, t; t_0; 1)\,, \tag{12}$$

where the cell diameter is large compared to the mean free path so that the lower limit may be approximated by $-\infty$. The t_0 integration can be performed first by interchanging the order of integration in the standard way, i.e.,

$$\int_{-\infty}^{t} dt_0 \int_{t_0}^{t} dt_5 \ldots \to \int_{-\infty}^{t} dt_5 \int_{-\infty}^{t_5} dt_0 \ldots . \tag{13}$$

Since V'_{12} is independent of t_0, according to Eq. (10), the only t_0 dependence appears in the factor $\exp[-\gamma_1(t-t_0)]$. Using Eq. (14),

$$\int_{-\infty}^{t_1} \gamma_1 dt_0 \exp[-\gamma_1(t_1-t_0)] = 1 . \tag{14}$$

The t_0 integration is rendered trivial. Making the following change of variables for $\rho_{12}^{(5)}$ [in Eq. (9)]:

$$\tau_5 = t - t_5; \quad t \text{ const}, \tag{15a}$$

$$\tau_4 = t_5 - t_4; \quad t_5 \text{ const}, \tag{15b}$$

$$\tau_3 = t_4 - t_3; \quad t_4 \text{ const}, \tag{15c}$$

$$\tau_2 = t_3 - t_2; \quad t_3 \text{ const}, \tag{15d}$$

$$\tau_1 = t_2 - t_1; \quad t_2 \text{ const}, \tag{15e}$$

and making a similar change of variables for $\rho_{12}^{(3)}$ [Eq. (8)], one utilizes Eqs. (13) and (14) and recasts Eqs. (8) and (9) into the following forms:

$$\begin{aligned}\rho_{12}^{(3)}(\vec{x}, \vec{v}, t; 1) = &(-i\mu E_0/\hbar)^3 N_1^{(0)}(\vec{v}) \int_0^\infty d\tau_3 \int_0^\infty d\tau_2 \int_0^\infty d\tau_1 \exp[-(\gamma_{12}-i\omega_0)\tau_3] \\ &\times \cos\omega(t-\tau_3) F(\vec{x}-\vec{v}\tau_3)[\exp(-\gamma_2\tau_2)+\exp(-\gamma_1\tau_2)]\cos\omega(t-\tau_3-\tau_2) F(\vec{x}-\vec{v}(\tau_3+\tau_2)) \\ &\times \{\exp[-(\gamma_{12}-i\omega_0)\tau_1]+\text{c.c.}\}\cos\omega(t-\tau_3-\tau_2-\tau_1) F(\vec{x}-\vec{v}(\tau_3+\tau_2+\tau_1)),\end{aligned} \tag{16}$$

$$\begin{aligned}\rho_{12}^{(5)}(\vec{x}, \vec{v}, t; 1) = &(-i\mu E_0/\hbar)^5 N_1^{(0)}(\vec{v}) \int_0^\infty d\tau_5 \int_0^\infty d\tau_4 \int_0^\infty d\tau_3 \int_0^\infty d\tau_2 \int_0^\infty d\tau_1 \exp[-(\gamma_{12}-i\omega_0)\tau_5] \\ &\times \cos\omega(t-\tau_5) F(\vec{x}-\vec{v}\tau_5)[\exp(-\gamma_2\tau_4)+\exp(-\gamma_1\tau_4)]\cos\omega(t-\tau_5-\tau_4) F(\vec{x}-\vec{v}(\tau_5+\tau_4)) \\ &\times \{\exp[-(\gamma_{12}-i\omega_0)\tau_3]+\text{c.c.}\}\cos\omega(t-\tau_5-\tau_4-\tau_3) F(\vec{x}-\vec{v}(\tau_5+\tau_4+\tau_3)) \\ &\times [\exp(-\gamma_2\tau_2)+\exp(-\gamma_1\tau_2)]\cos\omega(t-\tau_5-\tau_4-\tau_3-\tau_2) F(\vec{x}-\vec{v}(\tau_5+\tau_4+\tau_3+\tau_2)) \\ &\times \{\exp[-(\gamma_{12}-i\omega_0)\tau_1]+\text{c.c.}\}\cos\omega(t-\tau_5-\tau_4-\tau_3-\tau_2-\tau_1) F(\vec{x}-\vec{v}(\tau_5+\tau_4+\tau_3+\tau_2+\tau_1)),\end{aligned} \tag{17}$$

where

$$F(\vec{x}-\vec{v}\tau) = \sin k(z-v_z\tau)\exp[-\alpha(x-v_x\tau)^2] \times \exp[-\alpha(y-v_y\tau)^2]. \tag{18}$$

The polarization induced in the medium is given by

$$\vec{P} = \vec{\mu}_{21}\rho_{12} + \vec{\mu}_{12}\rho_{21} \quad (\vec{\mu}_{21} = \vec{\mu}_{12} \equiv \vec{\mu}). \tag{19}$$

In terms of the polarization, the absorbed power per unit volume is $\vec{P}\cdot\vec{E}$ where $\vec{E}$ is the applied external field. When $\vec{P}\cdot E$ is spatially and temporally averaged only the terms in $\vec{P}$ proportional to $\exp(\pm ikz)\exp(\pm i\omega t)$ survive, assuming that the electric field amplitude E_0 varies slowly over distances comparable to a wavelength and that the mean free path is large compared to a wavelength. Thus only first harmonic terms need be considered in Eqs. (16) and (17).

Consider first the $\exp(\pm i\omega t)$ terms which are completely contained in the cosine functions appearing in Eqs. (16) and (17). Factoring out $(1/2)^5$ from the product of five cosines in Eq. (17), one obtains a contribution proportional to $\exp(i\omega t)$ by multiplying together three positive-frequency terms and two negative-frequency terms. This results in $10\,(=5!/3!2!)$ components proportional to $\exp(i\omega t)$. Since the cosine product is real, the complex conjugate of these ten components yields the result proportional to $\exp(-i\omega t)$. Inspection of the factor $\exp[-(\gamma_{12}-i\omega_0)\tau_5]$ involved in the τ_5 integral shows that terms containing $\exp(-i\omega\tau_5)$ are slowly varying in τ_5 as ω tends to ω_0, while terms containing $\exp(i\omega\tau_5)$ rapidly vary and may be neglected by comparison (rotating-wave approximation). Each of the first ten components described above is proportional to $\exp[i\omega(t-\tau_5)]$ [$t-\tau_5$ appears in the argument of each cosine function in Eq. (17)], and is slowly varying in τ_5. Thus the ten complex conjugate terms, which are proportional to $\exp[-i\omega(t-\tau_5)]$, are rapidly varying and may be dropped. In addition, of the ten contributions which have been retained from the cosine product, those containing τ_2 or τ_4 in their argument are rapidly oscillating since ω_0 is not present in the factors $\exp(-\gamma_2\tau_4)+\exp(-\gamma_1\tau_4)$ and $\exp(-\gamma_2\tau_2)+\exp(-\gamma_1\tau_2)$. Therefore, these terms may be eliminated.

The product of the five sine functions (from the F functions) in Eq. (17) may be treated analogous to the cosine product except that the complex conjugate expressions which are proportional to $\exp(-ikz)$ are subtracted. All first harmonic terms are retained until the v_z integration is performed. The sign of the $\exp(ikz)$ contributions is positive since each results from the product of three positive and two negative terms.

The $\rho_{12}^{(3)}$ expression may be treated in a similar manner, except that the $\exp(ikz)$ contributions are negative, resulting from a product of two positive terms and one negative term.

A further reduction in the number of terms arising from the sine product can be accomplished by performing the v_z integration next. The total contribution to ρ_{12} from all velocity groups is given by

$$\rho_{12}(\vec{x}, t; 1) = \int_{-\infty}^{\infty} dv_x \int_{-\infty}^{\infty} dv_y \int_{-\infty}^{\infty} dv_z \rho_{12}(\vec{x}, \vec{v}, t; 1) . \tag{20}$$

The Maxwellian velocity distribution defined by

$$W(\vec{v}) = (1/u\sqrt{\pi})^3 \exp[-(v_x^2 + v_y^2 + v_z^2)/u^2] ; \tag{21}$$

$u = (m/2kT)^{-1/2}$ is used in what follows. Consider the v_z integration. A typical expression from the sine product looks like $\exp[ikv_z(\tau - \tau')]$, where $\tau - \tau'$ is some combination of τ_1 to τ_5. Integrating this exponential with the v_z part of Eq. (21) yields

$$I(\tau - \tau') \equiv \frac{1}{u\sqrt{\pi}} \int_{-\infty}^{\infty} dv_z \exp[ikv_z(\tau - \tau')] \exp(-v_z^2/u^2)$$
$$= \exp[-(ku/2)^2 (\tau - \tau')^2] . \tag{22}$$

Because the rapidly oscillating terms in the τ integrals appearing in Eqs. (16) and (17) have been removed, the remaining terms vary slowly over times of order $1/ku$ provided that the Doppler width, ku, is much larger than all other frequencies in the integrands.[13] Thus $I(\tau - \tau')$ [Eq. (22)] is a sharply peaked function in the time domain, and it can be approximated by a delta function, $M\delta(\tau - \tau')$, where M is a normalization constant. To determine M consider the integral

$$\int_0^{\infty} \exp[-(ku/2)^2 (\tau - \tau')^2] d\tau$$
$$= \int_{-ku\tau'/2}^{\infty} dq \exp(-q^2)(2/ku)$$
$$\cong 2\sqrt{\pi}/ku , \tag{23}$$

where $ku\tau'/2$ may be approximated by ∞ since the range of integration over τ' is much greater than $1/ku$. This gives

$$I(\tau - \tau') \cong (2\sqrt{\pi}/ku)\, \delta(\tau - \tau') \tag{24}$$

for integrals having the form of Eq. (22). Expressions of the form $\exp[ikv_z(\tau + \tau')]$ give sharply peaked functions proportional to $\delta(\tau + \tau')$ after v_z integration. Therefore these expressions contribute only for $\tau = \tau' = 0$, since τ and τ' are $\geqslant 0$. Hence the contribution from these expressions is negligible compared to expressions of form of Eq. (24) which contribute for all $\tau = \tau'$. Note the result, Eq. (24), does not depend on the sign of v_z, so that the complex conjugate terms proportional to $\exp(-ikz)$ contribute with equal amplitude. Thus the v_z integration of the first harmonic components yields

$$\rho_{12}^{(3)}(\vec{x}, t; 1) = -\left(\frac{-\mu E_0}{4\hbar}\right)^3 \frac{2\sqrt{\pi}}{ku} N_1^{(0)}$$
$$\times \int_0^{\infty} d\tau_3 \int_0^{\infty} d\tau_2 \int_0^{\infty} d\tau_1 [\exp(-\gamma_2\tau_2) + \exp(-\gamma_1\tau_2)] G_3'(\vec{x}, t; \tau_1, \tau_2, \tau_3) , \tag{25a}$$

where

$$G_3'(\vec{x}, t; \tau_1, \tau_2, \tau_3) = H_3(x; \tau_1, \tau_2, \tau_3) H_3(y, \tau_1, \tau_2, \tau_3) \exp(i\omega t) [\exp(ikz) - \exp(-ikz)]$$
$$\times [\exp\{-[\gamma_{12} + i(\omega - \omega_0)]\tau_3\} (\exp\{-[\gamma_{12} + i(\omega - \omega_0)]\tau_1\} + \text{c.c.})\, \delta(\tau_1 - \tau_3)] , \tag{25b}$$

with

$$H_3(x; \tau_1, \tau_2, \tau_3) = (1/u\sqrt{\pi}) \int_{-\infty}^{\infty} dv_x \exp(-v_x^2/u^2) \exp\{-\alpha(x - v_x\tau_3)^2 - \alpha[x - v_x(\tau_3 + \tau_2)]^2 - \alpha[x - v_x(\tau_3 + \tau_2 + \tau_1)]^2\} , \tag{25c}$$

and a similar expression for $H_3(y; \tau_1, \tau_2, \tau_3)$.

$$\rho_{12}^{(5)}(\vec{x}, t; 1) = \left(-\frac{\mu E_0}{4\hbar}\right)^5 \frac{2\sqrt{\pi}}{ku} N_1^{(0)} \int_0^{\infty} d\tau_5 \int_0^{\infty} d\tau_4 \int_0^{\infty} d\tau_3 \int_0^{\infty} d\tau_2 \int_0^{\infty} d\tau_1$$
$$\times \{[\exp(-\gamma_2\tau_4) + \exp(-\gamma_1\tau_4)][\exp(-\gamma_2\tau_2) + \exp(-\gamma_1\tau_1)] G_5'(\vec{x}, t; \tau_1 \ldots \tau_5)\} , \tag{26a}$$

where

$$G_5'(\vec{x}, t; \tau_1 \ldots \tau_5)$$
$$= H_5(x; \tau_1 \ldots \tau_5) H_5(y; \tau_1 \ldots \tau_5) \exp(i\omega t) [\exp(ikz) - \exp(-ikz)]$$
$$\times [\exp\{-[\gamma_{12} + i(\omega - \omega_0)]\tau_5\} (\exp\{-[\gamma_{12} - i(\omega - \omega_0)](\tau_1 + \tau_3)\} + \exp\{-[\gamma_{12} + i(\omega - \omega_0)]\tau_1 - [\gamma_{12} - i(\omega - \omega_0)]\tau_3\}$$
$$+ \exp\{-[\gamma_{12} - i(\omega - \omega_0)]\tau_1 - [\gamma_{12} + i(\omega - \omega_0)]\tau_3\} + \exp\{-[\gamma_{12} + i(\omega - \omega_0)](\tau_1 + \tau_3)\})$$
$$\times [\delta(\tau_5 - \tau_1 - \tau_3) + \delta(\tau_3 - \tau_1 - \tau_5) + \delta(\tau_1 - \tau_3 - \tau_5) + \delta(\tau_5 - \tau_1 - 2\tau_2 - \tau_3) + \delta(\tau_1 - \tau_3 - 2\tau_4 - \tau_5)]], \tag{26b}$$

with

$$H_5(x; \tau_1 \ldots \tau_5)$$
$$= \frac{1}{u\sqrt{\pi}} \int_{-\infty}^{\infty} dv_x \exp(-v_x^2/u^2) \exp\{-\alpha(x - v_x\tau_5)^2 - \alpha[x - v_x(\tau_5 + \tau_4)]^2 \cdots - \alpha[x - v_x(\tau_5 + \tau_4 + \tau_3 + \tau_2 + \tau_1)]^2\}, \tag{26c}$$

and a similar expression for $H_5(y; \tau_1 \ldots \tau_5)$.

The integrals in Eqs. (25c) and (26c) are easily evaluated yielding:

$$H_3(x; \tau_1, \tau_2, \tau_3) H_3(y; \tau_1, \tau_2, \tau_3)$$
$$= \frac{\exp\{-[3\alpha - \alpha^2 D^2/(\alpha C + 1/u^2)](x^2 + y^2)\}}{1 + \alpha u^2 C}, \tag{27a}$$

where

$$C = \tau_3^2 + (\tau_3 + \tau_2)^2 + (\tau_3 + \tau_2 + \tau_1)^2, \tag{27b}$$

$$D = 3\tau_3 + 2\tau_2 + \tau_1, \tag{27c}$$

$$H_5(x; \tau_1 \ldots \tau_5) H_5(y; \tau_1 \ldots \tau_5)$$
$$= \frac{\exp\{-[5\alpha - \alpha^2 A^2/(\alpha B + 1/u^2)](x^2 + y^2)\}}{1 + \alpha u^2 B}, \tag{28a}$$

where

$$B = \tau_5^2 + (\tau_5 + \tau_4)^2 + \cdots + (\tau_5 + \tau_4 + \tau_3 + \tau_2 + \tau_1)^2, \tag{28b}$$

$$A = 5\tau_5 + 4\tau_4 + 3\tau_3 + 2\tau_2 + \tau_1. \tag{28c}$$

In order to obtain the line shape, consider the power absorbed per unit length

$$\overline{P}_L = \int_{-\infty}^{\infty} dx \int_{-\infty}^{\infty} dy \langle \dot{\vec{P}} \cdot \vec{E} \rangle, \tag{29}$$

where the polarization, $\vec{P}$, is given by Eq. (19) and the electric field, $\vec{E}$, is given by Eq. (5b). The angular brackets in Eq. (29) denote a time average as well as a spatial average of $\sin^2 kz = \frac{1}{2}$. In the spatial average it is assumed that E_0 is a slowly varying function of z and that $l/\lambda \gg 1$, where l is the mean free path and λ is the wavelength of the incident radiation field. The limits of integration in Eq. (29) are approximated by $\pm\infty$ assuming that the cell diameter and detector field of view are much larger than the electric field $1/e$ radius. Performing the x and y integrations gives for the absorbed power per unit length[14]:

$$\overline{P}_L = \overline{P}_L^{(4)} + \overline{P}_L^{(6)}, \tag{30}$$

with

$$\overline{P}_L^{(4)} = -\frac{\pi^{3/2}}{\alpha} \left(\frac{\mu E_0}{4\hbar}\right)^4 \frac{\hbar\omega}{ku} N_1^{(0)} \int_0^{\infty} d\tau_2 \int_0^{\infty} d\tau_3 \left(\frac{(e^{-\gamma_2\tau_2} + e^{-\gamma_1\tau_2})\{\exp[-2(\gamma_{12} + i(\omega - \omega_0))]\tau_3 + \exp(-2\gamma_{12}\tau_3)\}}{1 + \alpha u^2(\tau_2^2 + 2\tau_2\tau_3 + 2\tau_3^2)} \right) + \text{c.c.}, \tag{31a}$$

where the integration over τ_1 has been performed using the delta function appearing in Eq. (25b); the superscript (4) on $\overline{P}_L$ refers to fourth order in the field amplitude E_0.

$$\bar{P}_L^{(6)} = \frac{2\pi^{3/2}}{3\alpha}\left(\frac{\mu E_0}{4\hbar}\right)^6 \frac{\hbar\omega}{ku} N_1^{(0)} \int_0^\infty d\tau_1 \int_0^\infty d\tau_2 \int_0^\infty d\tau_3 \int_0^\infty d\tau_4 \,(e^{-\gamma_2\tau_4}+e^{-\gamma_1\tau_4})$$
$$\times\big(\exp\{-2[\gamma_{12}+i(\omega-\omega_0)](\tau_1+\tau_3)\}+\exp\{-2\gamma_{12}\tau_1-2[\gamma_{12}+i(\omega-\omega_0)]\tau_3\}$$
$$+\exp\{-2\gamma_{12}\tau_3-2[\gamma_{12}+i(\omega-\omega_0)]\tau_1\}+\exp[-2\gamma_{12}(\tau_1+\tau_3)]\big)$$
$$\times\left(\frac{2(e^{-\gamma_2\tau_2}+e^{-\gamma_1\tau_2})}{1+\frac{2}{3}\alpha u^2(3\tau_1^2+2\tau_2^2+5\tau_3^2+2\tau_4^2+3\tau_1\tau_2+6\tau_1\tau_3+3\tau_1\tau_4+4\tau_2\tau_3+2\tau_2\tau_4+5\tau_3\tau_4)}\right.$$
$$+\frac{e^{-\gamma_2\tau_2}+e^{-\gamma_1\tau_2}}{1+\frac{2}{3}\alpha u^2(5\tau_1^2+2\tau_2^2+5\tau_3^2+2\tau_4^2+5\tau_1\tau_2+8\tau_1\tau_3+4\tau_1\tau_4+4\tau_2\tau_3+2\tau_2\tau_4+5\tau_3\tau_4)}$$
$$\left.+\frac{2\big(\exp\{-2[\gamma_{12}+\gamma_2/2+i(\omega-\omega_0)]\tau_2\}+\exp\{-2[\gamma_{12}+\gamma_1/2+i(\omega-\omega_0)]\tau_2\}\big)}{1+\frac{2}{3}\alpha u^2(3\tau_1^2+9\tau_2^2+5\tau_3^2+2\tau_4^2+9\tau_1\tau_2+6\tau_1\tau_3+3\tau_1\tau_4+12\tau_2\tau_3+6\tau_2\tau_4+5\tau_3\tau_4)}\right)+\text{c.c.}, \tag{31b}$$

where use has been made of the symmetry of the integrand under the interchange $\tau_1\leftrightarrow\tau_5$, $\tau_2\leftrightarrow\tau_4$, and $\tau_3\leftrightarrow\tau_3$, after the x and y integrals are calculated and where the resulting integrations have been performed using the delta functions appearing in Eq. (26b). The superscript (6) on $\bar{P}_L$ in Eq. (31) refers to sixth order in the field amplitude E_0. Equations (31a) and (31b) give (to sixth order in $\mu E_0\hbar$) the exact contribution to the absorbed power per unit length, including spatial harmonic effects in the population, valid to all orders in the transit-time effects. The entire transit-time dependence is contained in the denominators of Eqs. (31a) and (31b) in the terms proportional to αu^2.

III. TRANSIT-TIME EFFECTS IN THE COLLISION DOMINATED REGIME

The integrals in Eqs. (31a) and (31b) are easily evaluated in the limit $\alpha u^2/\gamma_c^2 \ll 1$ (where $\gamma_c=\gamma_1$, γ_2, or γ_{12}), using a first-order Taylor expansion of the denominators. Note that the last two terms of Eq. (31b) which contain the factor $\exp[-2i(\omega-\omega_0)\tau_2]$ arise from spatial harmonic effects in the populations. In order to facilitate the calculation of transit-time contributions to the linewidth in the collision dominated regime, it is convenient to specialize to a rate-equation approximation (REA).[15] Thus the last two terms of Eq. (31b) are dropped.[16] By performing the integrations one obtains $\bar{P}^{(4)}+\bar{P}^{(6)}$ correct to lowest order in the parameter $\alpha u^2/\gamma_c^2$.

Setting the resonant part of the total line shape (sum of $\bar{P}^{(4)}+\bar{P}^{(6)}$) equal to half its maximum height ($\omega=\omega_0$) allows the linewidth to be determined. The equation to be solved contains two independent small parameters,

$$\delta_1 \equiv \frac{1}{2\gamma_{12}}\left(\frac{1}{\gamma_1}+\frac{1}{\gamma_2}\right)\left(\frac{\mu E_0}{2\hbar}\right)^2, \tag{32}$$

where δ_1 is the saturation parameter at beam center and

$$\delta_2 \equiv \alpha u^2/\gamma_{12}^2. \tag{33}$$

Assuming a half-width at half-maximum of the following form:

$$\Gamma_{\rm HWHM}^2 = \gamma_{12}^2 + g_1\delta_1 + g_2\delta_2 + g_{12}\delta_1\delta_2, \tag{34}$$

then by Taylor expansion of each Lorentzian to appropriate order and by comparison of the terms proportional to δ_1, δ_2, and $\delta_1\delta_2$, one obtains the coefficients g_1, g_2, and g_{12}. Note that retaining terms proportional to $\delta_1\delta_2$ does not imply that terms of order $(\delta_1)^2$ or $(\delta_2)^2$ should be retained, since the perturbation theory and Taylor expansion of the denominators are valid only to orders linear in δ_1 and δ_2. In addition, if a higher-order perturbation treatment and/or higher-order Taylor expansion is carried out, the additional contributions obviously cannot affect the results which are linear in δ_1 and δ_2. Using the fact that δ_1 and δ_2 are independent, one may calculate the "g" coefficients by putting $\delta_2=0$ to determine g_1 and then putting $\delta_1=0$ to determine g_2. Then, after considerable algebra, when both δ_1 and δ_2 are nonzero, one may obtain g_{12} by using Eq. (34) and the previously calculated g_1 and g_2. The resulting HWHM is given by

$$\Gamma_{\rm HWHM}=\gamma_{12}+\frac{1}{4}\left(\frac{1}{\gamma_1}+\frac{1}{\gamma_2}\right)\left(\frac{\mu E_0}{2\hbar}\right)^2+\frac{\alpha u^2}{2\gamma_{12}}\left[3+2\frac{\gamma_{12}}{\gamma_1+\gamma_2}\left(\frac{\gamma_1}{\gamma_2}+\frac{\gamma_2}{\gamma_1}\right)\right]-\frac{\alpha u^2}{2\gamma_{12}^2}\left(\frac{1}{\gamma_1}+\frac{1}{\gamma_2}\right)\left(\frac{\mu E_0}{2\hbar}\right)^2$$
$$\times\left[\frac{209}{72}+\frac{5}{3}\gamma_{12}^2\frac{\gamma_1\gamma_2}{\gamma_1+\gamma_2}\left(\frac{1}{\gamma_1^3}+\frac{1}{\gamma_2^3}\right)+\frac{47}{12}\frac{\gamma_{12}}{\gamma_1+\gamma_2}\left(\frac{\gamma_1}{\gamma_2}+\frac{\gamma_2}{\gamma_1}\right)+\frac{2}{3}\left(\frac{\gamma_{12}}{\gamma_1+\gamma_2}\right)^2\left(\frac{\gamma_1}{\gamma_2}+\frac{\gamma_2}{\gamma_1}\right)^2\right]. \tag{35}$$

The first term in Eq. (35) gives the pure collision-broadening contribution to the linewidth. The next expression is the power-broadening contribution to the linewidth in the REA for a plane standing wave with a Gaussian profile. More interesting is the increase in the linewidth due to transit-time effects represented by the third term. For the case where $\gamma_1=\gamma_2=\gamma_{12}$, this result is identical to that given by Baklanov *et al.* (Ref. 8). Note that if the decay rates are linearly dependent on pressure then this result varies inversely with pressure. The validity of Eq. (35) is, of course, restricted to the collision dominated regime where it is necessary to have $[\alpha u^2/\gamma_c^2]^{1/2}\lesssim 0.1$ (where $\gamma_c=\gamma_1$, γ_2, or γ_{12}) in order that the Taylor series expansion of the denominators converge. The last term shows that transit-time effects cause a decrease in the power broadening in the collision dominated regime. Note that the terms in the square brackets are pressure independent if the γ's vary linearly with pressure.

In the next section, the linewidth is given as a function of $(\alpha u^2)^{1/2}\gamma_c$ by numerical evaluation.

IV. COMPUTER EVALUATION OF THE POWER-BROADENED TRANSIT-TIME LINEWIDTH

As in Sec. III, it is convenient to specialize to a rate-equation approximation, and drop the last two terms of Eq. (31b). This is not necessary, but is done to considerably reduce the required computer time. In addition, let $\gamma_1=\gamma_2=\gamma_{12}=\gamma$ to avoid introducing an extra parameter into the numerical evaluation. Consider the following function,

$$F(S,\beta,\Delta)=\underline{\tilde{P}}^{(4)}(\Delta)-\tfrac{1}{2}\underline{\tilde{P}}^{(4)}(\Delta=0)+\underline{\tilde{P}}^{(6)}(\Delta)-\tfrac{1}{2}\underline{\tilde{P}}^{(6)}(\Delta=0)\,, \tag{36}$$

where $\underline{\tilde{P}}$ denotes the resonant part of Eqs. (31a) and (31b) with all the γ's $=\gamma$; where $\beta=\gamma/(\alpha u^2)^{1/2}$ and

$$S=\frac{1}{\gamma^2}\left(\frac{\mu E_0}{2\hbar}\right)^2, \tag{37}$$

$$\Delta=2(\omega-\omega_0)/\gamma\,. \tag{38}$$

Since $\Delta=0$ defines the maximum of the resonance, the zeros of $F(\Delta)$ holding S,β fixed represent the full-width at half maximum, Δ_F, in units of the collision frequency. Explicitly, $F(S,\beta,\Delta_F)$ is given by

$$\begin{aligned}F(S,\beta,\Delta_F)=-\frac{4\pi^{3/2}}{\alpha}\left(\frac{\mu E_0}{4\hbar}\right)^4\frac{\hbar\omega}{ku}\frac{N_1^{(0)}}{\alpha u^2}\Bigg[&\int_0^\infty dx_1\int_0^\infty dx_2\,\frac{e^{-x_2}e^{-2x_1}(\cos\Delta_F x_1-\frac{1}{2})}{\beta^2+(x_2^2+2x_1x_2+2x_1^2)}\\&-\frac{S}{3}\int_0^\infty dx_1\int_0^\infty dx_2\int_0^\infty dx_3\int_0^\infty dx_4\,e^{-(x_2+x_4)}e^{-2(x_1+x_3)}[\cos\Delta_F(x_1+x_3)+\cos\Delta_F x_3+\cos\Delta_F x_1-\tfrac{3}{2}]\\&\times\bigg(\frac{2}{\beta^2+\frac{2}{3}(3x_1^2+2x_2^2+5x_3^2+2x_4^2+3x_1x_2+6x_1x_3+3x_1x_4+4x_2x_3+2x_2x_4+5x_3x_4)}\\&+\frac{1}{\beta^2+\frac{2}{3}(5x_1^2+2x_2^2+5x_3^2+2x_4^2+5x_1x_2+8x_1x_3+4x_1x_4+4x_2x_3+2x_2x_4+5x_3x_4)}\bigg)\Bigg]=0\,,\end{aligned} \tag{39}$$

where dimensionless variables have been introduced.

Equation (39) represents a well defined, if somewhat complicated function of Δ_F. Using Newton's rule, the zeros can in principle be calculated to any desired accuracy. This requires that we determine $\partial F/\partial\Delta$ as well as $F(\Delta)$. To solve Eq. (39) in practice, the integrals are performed using a multidimensional analog of Simpson's rule. Given S,β, and an initial guess of Δ_{F_i}, the integrals for $F(\Delta)$ and $\partial F/\partial\Delta$ may be calculated simultaneously (since differentiation with respect to Δ when taken under the definite integrals merely modifies the cosine functions). A better guess for Δ_F is then

$$\Delta_{F_{i+1}}=\Delta_{F_i}-\frac{F(S,\beta,\Delta_{F_i})}{\partial F(S,\beta,\Delta_{F_i})/\partial\Delta}\,. \tag{40}$$

Iteration then gives the zeros of the approximated $F(S,\beta,\Delta)$ to the required accuracy. Care must, of course, be taken to insure that the approximate function $F(\Delta)$ represents the true integrals with sufficient accuracy. Figures (1) and (2) show the results of the calculation for $S=0.20$. The $S=0.0$ curve of Fig. (1) represents the un-power-broadened (fourth order) linewidth. This is in good agreement with the results of Ref. 8, except that the curve is plotted in a somewhat different manner. The $S=0.20$ curve indicates that the power-broadening contribution first decreases and then

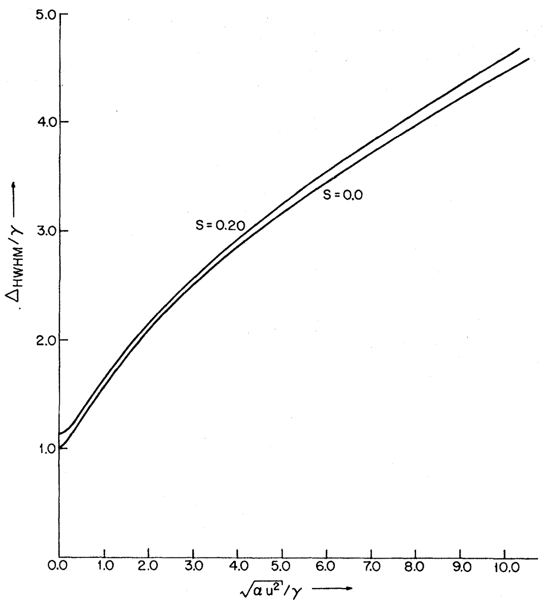

FIG. 1. Linewidth as a function of $(\alpha u^2)^{1/2}/\gamma$ at fixed pressure and intensity. $\gamma_t = \sqrt{\alpha u^2} = u/R$ is the transit-time frequency where $u = (2kT/m)^{1/2}$ and R is the $1/e$ radius of the Gaussian field (not intensity). Δ_{HWHM}/γ is the half-width at half-maximum in units of the collision frequency. The upper and lower curves give the linewidth for $S = 0.20$ and 0.0, respectively, where S is the saturation parameter at beam center defined by $S = (\mu E_0/2\hbar)^2/\gamma^2$. E_0 is defined by Eq. (7).

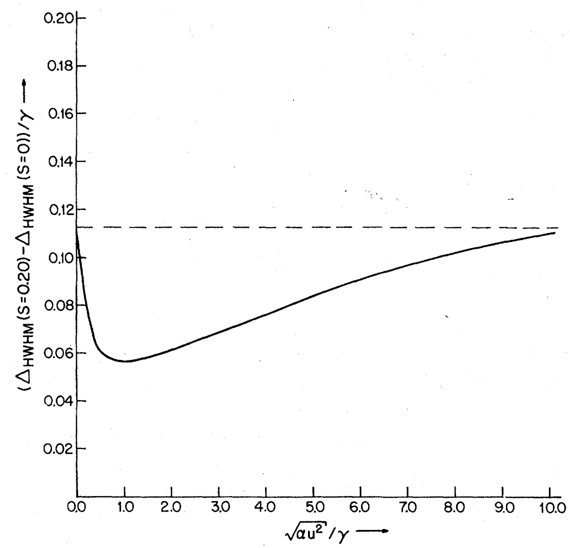

FIG. 2. Power broadening contribution to the linewidth as a function of $(\alpha u^2)^{1/2}/\gamma$ for $S = 0.20$. This curve represents the difference between the upper and lower curves of Fig. 1. The definitions are as in Fig. 1. Note that the power-broadening contribution is reduced by approximately a factor 2 when $(\alpha u^2)^{1/2}/\gamma \sim 1$. The dashed line shows the plane-wave theory prediction for comparison.

increases as the transit-time frequency $\gamma_t = \sqrt{\alpha u^2} = u/R$ is increased at fixed intensity and pressure. To show this clearly, the difference between the two curves is plotted on an expanded scale in Fig. (2). [The zeros of $F(\Delta)$ have been determined with suitable accuracy to make this possible.]

V. DISCUSSION

The $S = 0.0$ (no power-broadening) curve of Fig. 1 shows that the linewidth increases as the beam radius R is reduced. However, the *rate* at which the linewidth broadens decreases as the beam radius is reduced to a size significantly smaller than the mean free path λ_p. This behavior is discussed in Ref. 7, where it is suggested that for $R \lesssim \lambda_p$, the primary contribution to the nonlinear part of the absorption signal arises from molecules with low transverse velocity which experience the longest interaction time with the laser field.

The power-broadened ($S = 0.20$) curve of Fig. 1 appears similar to that of the $S = 0.0$ curve. In order to study the effects of the finite beam size on the power-broadening contribution to the linewidth, the difference between the two curves of Fig. 1 is plotted on an expanded scale in Fig. 2. The power-broadening contribution to the linewidth, as shown in Fig. 2, exhibits two distinct regimes. First, consider the regime where $(\alpha u^2)^{1/2}/\gamma < 1$, corresponding to a beam radius R which is larger than the mean free path. Define the nonlinear part of the absorption signal as ΔI. In this case, the molecules which make the primary contribution to ΔI interact with the laser field for an average time which is determined by collisions. However, as the beam radius is reduced to a size comparable to the mean free path [i.e., $(\alpha u^2)^{1/2}/\gamma \sim 1$], a large number of molecules from the transverse velocity distribution interact with the laser field for a reduced time (compared to $1/\gamma$) determined by their transverse velocity and the beam size. Since the power-broadening contribution to the signal ΔI arises from a higher-order process (intensity $\propto E^6$) than the basic collision-broadened nonlinear resonance (intensity $\propto E^4$), the reduced interaction time results in a decrease in the relative number of transverse velocity molecules which appreciably contribute to ΔI via the E^6 process. In this way, the power-broadening contribution to the linewidth initially decreases.

Now consider the regime where $(\alpha u^2)^{1/2}/\gamma > 1$, corresponding to a beam radius which is significantly smaller than the mean free path. The interaction time of the molecules which travel across the beam with average thermal speed ($\sim u$) is reduced to such an extent that molecules with small transverse velocities, although reduced in number according to the radial Maxwellian velocity distribution, make the largest contribution to the nonlinear absorption signal ΔI, because of their longer interaction time with the field. Accordingly, the relative number of molecules which make significant E^6 (compared to E^4) contributions is altered by the presence of the low transverse velocity molecules contributing to the power broadening (the E^6 term).

VI. CONCLUSIONS

In summary, this work presents several new features of the transit-time corrections to nonlinear Doppler-free resonances. The most important result is given in Fig. 2, which shows that the power-broadening contribution to the linewidth can be drastically altered by the finite beam diameter. This should be compared to the plane wave result which shows no dependence on beam size at fixed intensity and pressure. It is likely, even at higher intensities, where the perturbation theory is no longer valid, that the effect of the finite beam size in selection of the low transverse velocity molecules will persist. This effect tends, quite generally, to alter the relative numbers of molecules which are able to participate in the various higher-order multiphoton processes contributing to the line shape. Hence, one expects that a complex dependence of the linewidth on pressure, intensity, and beam dimension will continue in the region of strong saturation.

The closed-form expression for the linewidth [Eq. (35)], valid in the collision dominated regime, gives the dependence of the linewidth on pressure, beam size, and intensity for general collision parameters ($\gamma_1, \gamma_2, \gamma_{12}$). The result shows how the transit-time parameter $\alpha u^2 = \gamma_t^2$ enters the linewidth. This should be helpful in introducing phenomenological expressions to fit existing data. Of importance is the fact the square of γ_t enters into the calculation and not γ_t itself. Assuming that the collision parameters vary linearly with pressure, one can see from Eq. (35) that the third-order transit time contribution to the linewidth varies inversely as pressure, whereas the fifth-order transit time correction to the power-broadening varies inversely as the cube of the pressure. The small size of the transit-time corrections in this regime make their effects on the linewidth difficult to observe.

At low pressures, where the mean free path attains centimeter dimensions, the transit-time broadening as well as the transit-time corrections to the power broadening should be observable with beams of reasonable diameter.

In this paper, only a single two-level system has been discussed. More generally, one must consider the contribution from many degenerate two-level systems weighted appropriately by their transition matrix elements μ. For the perturbation regime (weak saturation), the required modifications are simple. Assuming that the collision parameters are identical for all the degenerate two level systems, one replaces μ^4 in Eq. (31a) by a sum over μ^4 for the various contributing degenerate levels. Similarly one modifies μ^6 in Eq. (31b). These modifications will not alter the basic results of Fig. 1 and Fig. 2 in any essential way. However, for strong saturation, where the intensity dependence of the linewidth (even for the plane wave theory) becomes somewhat complex, it is probably best to consider a single two-level system. The $R(0)$ ($J=0$) transitions of CO_2 or N_2O offer single two-level systems which can be utilized experimentally to investigate the effects of the finite beam size both in the weak- and strong-saturation regimes.

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[13]This assumes that $ku \gg \gamma_1, \gamma_2, \gamma_{12}, u/R$, and $(\omega-\omega_0)$ which is valid in the Doppler-broadened regime.

[14]The result given by Eq. (31a) is in agreement with Eq. (7.2) of Ref. 7 and Eq. (2) of Ref. 8.

[15]Treatments which ignore population pulsations in space or in time are called rate-equation approximations. See, for example, W. Culshaw, Phys. Rev. 164, 329 (1967); H. Greenstein, Phys. Rev. 175, 438 (1968); S. G. Rautian, Doctoral dissertation, Works Lebedev Inst., Acad. Sci. (USSR) 43, (1968); B. J. Feldman and M. S. Feld, Phys. Rev. A 1, 1375 (1970); See also Ref. 12, Sec. 18.

[16]The inclusion of terms arising from spatial harmonics in the population would increase the power-broadening contribution to the linewidth by ~25%. Inspection of the last two terms of Eq. (31b) shows that they contain the exponential factors $\exp\{-2[\gamma_{12}+\gamma_a/2+i(\omega-\omega_0)]\tau_2\}$, $(\gamma_a=\gamma_1,\gamma_2)$ which decay much faster than $\exp(-\gamma_a\tau_2)$ when $\gamma_{12} \gtrsim \gamma_a$.