Transit-time effects in optically pumped coupled three-level systems

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A density-matrix calculation of the line shape for an optically pumped coupled three-level system subjected to two traveling wave-laser fields with Gaussian intensity profiles is presented to third-order in perturbation theory. A closed-form expression for the line shape in the extreme transit-time limit is obtained. The line shape of the narrow resonance (Raman-type processes) is found to be an exponential function of the detuning, with half-width at half maximum $\Delta^{(\mathrm{Hz})}_{\mathrm{HWHM}} \simeq 0.98(u/2\pi D)$ (D is the intensity $1/e$ diameter and u is the thermal speed). The saturation intensity is also modified by the transit rate $\gamma_t = \sqrt{2}u/D$ (rad/sec).

I. INTRODUCTION

Doppler-free laser spectroscopy[1-5] of optically pumped atomic and molecular systems[6-8] exhibits a number of unique features.[6-9] Specifically, for coupled three-level systems [see Fig. 1(a)] in atomic barium and sodium, three novel effects have been observed (i) saturation signals of anomalous sign and amplitude, (ii) greatly reduced saturation thresholds, and (iii) linewidths far narrower than the natural radiative limit.[6,9] For an experiment utilizing weakly saturating copropagating pump and probe beams [see Fig. 1(b)], the line shape consists of a narrow (Raman-type) resonance superimposed on a broader (rate-equation effect) background, which in turn is superimposed on a much broader Doppler background [Fig. 1(c)]. Each of the three features mentioned above are readily explained in terms of the optical pumping and the very long lower-level lifetimes for these systems.[6,9]

Since the experiments are typically performed at very low pressures, one expects that the transit time of an atom across the laser beam will limit the effective lower-level lifetime. Hence, the width of the narrow Raman resonance and the saturation threshold will be determined by the beam transit time. However, the width of the rate-equation background will be essentially unaffected, since it is determined by the (short) upper-level radiative lifetime.

In the present paper, the shape of the Raman resonance is found to be an exponential for the important limiting case in which levels (1) [see Fig. 1(a)] and (2) are nearly degenerate and where the upper-level (0) lifetime is short compared to the beam transit time, while the ground-state lifetimes are long. The half-width at half maximum (HWHM) is given by $\Delta^{(\mathrm{Hz})}_{\mathrm{HWHM}} = (\sqrt{2}\,\ln 2)u/2\pi D \simeq 0.98\,u/2\pi D$, where u is the thermal speed $(\sqrt{2kT/m})$ and D is the intensity $1/e$ diameter. As expected, the effective saturation parameter depends on the transit rate $\gamma_t (=\sqrt{2}\,u/D)$. The rate-equation background is Lorentzian in shape, with the width being determined by the upper-level lifetime.

The above results were obtained by calculating, to third-order in perturbation theory, the line shape of a coupled three-level system [Fig. 1(a)] subjected to two traveling wave-laser fields with Gaussian intensity profiles. As depicted in Fig. 1, the 0-2 transition is pumped by a strong (though weakly saturating) field $\vec{E}_2$, while the 0-1 transition is simultaneously probed by the weaker field $\vec{E}_1$. By assumption, $\mu_{12}=0$ and $\vec{E}_1$ and $\vec{E}_2$ are such that they only interact with their respective transitions. Transit-time effects arise from the Gaussian radial dependence of the fields. Optical pumping is included by introducing spontaneous decay rates into the density-matrix equations (see the Appendix). The results of the general calculation are contained in Sec. II, where a closed-form expression is presented. The details of the calculation can be found in the Appendix, where the method employed is analogous to that of Refs. 10 and 11 (below).

Previously, Rautian and Shalagin[12] have calculated the transit-time contribution to the linewidth of the Lamb dip using third-order perturbation theory. They found that in the extreme transit-time regime the line shape is non-Lorentzian. In addition, the linewidth does not broaden as rapidly as might be expected by varying the effective beam transit rate. As the beam is made smaller and smaller those atoms with low-transverse velocities interact longest with the beam. Hence, the contribution from these atoms increases and the beam acts as a low-transverse-velocity selector. However, this effect is limited due to the fact that the number of atoms with radial velocity v_r is proportional to v_r. This indicates that the contribution from slowly moving atoms is weighted by number and interaction time.

Baklanov *et al.*[10] have given the linewidth as a

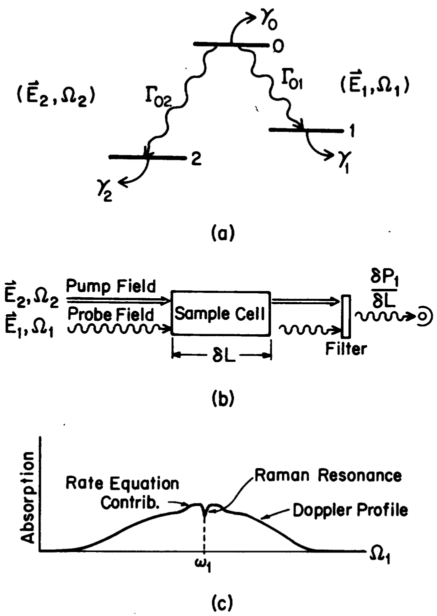

FIG. 1. (a) The optically pumped coupled three-level system being studied. Γ_{01}, Γ_{02} are the spontaneous decay (optical pumping) rates for the 0-1 and 0-2 transitions, respectively, whereas γ_0, γ_1, and γ_2 are the nonradiative rates for levels 0, 1, and 2. (b) Schematic of experimental setup for observing Raman resonance. (c) Typical line shape including optical pumping.

function of the ratio of collision frequency to transit-time frequency. They also present closed-form expressions for the linewidth in the extreme collision dominated and transit-time limits. The effects of a Gaussian beam in a gas-laser cavity have been studied by Maeda and Shimoda.[13] An asymmetry of the shape of the Lamb dip and a shift in its center frequency was found. Bordé *et al.*[14] considered the transit-time problem for a Gaussian beam including the curvature of the phase front and found a shift in the line center. This result was verified experimentally by Hall and Bordé.[14] More recently, Thomas *et al.*[11] calculated to fifth order the line shape for a Doppler-broadened two-level system interacting with a plane-standing-wave-laser field with a Gaussian intensity profile. It was found that the power-broadening contribution to the line shape can be distorted due to the interaction with a beam of finite width.

II. DERIVATION OF LINE SHAPE

For simplicity, only a brief outline of the important features and results will be given in this section. As mentioned above, the details of the derivation have been deferred to the Appendix.

The probe (0-1 transition) and the pump (0-2 transition) fields are, respectively, given by

$$\vec{E}_1 = \vec{E}_1^{(0)} \exp(-2r^2/D^2) \cos(k_1 Z - \Omega_1 t) \tag{1}$$

and

$$\vec{E}_2 = \vec{E}_2^{(0)} \exp(-2r^2/D^2) \cos(k_2 Z - \Omega_2 t), \tag{2}$$

where D is the intensity $1/e$ diameter. The interaction (in the dipole approximation) is of the form

$$V = -\vec{\mu} \cdot (\vec{E}_1 + \vec{E}_2) \tag{3}$$

with $\vec{\mu}$ the dipole moment.

The field amplitudes $E_1^{(0)}$ and $E_2^{(0)}$ are determined from the traveling wave power in each beam, which is given by

$$\begin{aligned} P_i^{(0)} &= \frac{c}{8\pi} (E_i^{(0)})^2 \int_0^\infty 2\pi r\, dr \exp(-4r^2/D^2) \\ &= \pi \left(\frac{D}{2}\right)^2 \frac{c}{8\pi} (E_i^{(0)})^2 \\ &\equiv \pi (D/2)^2 I_i^{(0)}, \quad i = 1, 2. \end{aligned} \tag{4}$$

The density-matrix equations (with optical pumping) given by Eqs. (A1) are integrated using time-dependent perturbation theory (third order in V), subject to the initial condition that the contributing atoms originate in level 2. Atoms originating in level 1 give only Doppler-broadened contributions to the line shape in third order and will not be considered. Proceeding as in the Appendix, the result for the average absorbed probe power per unit length is given by (specializing to nearly degenerate levels 1 and 2 and copropagating waves)

$$\frac{\delta P_1}{\delta L} = \Lambda \operatorname{Re} \int_0^\infty dx_1 \int_0^\infty dx_2 \left\{ \frac{e^{(i\Delta' - \gamma'_{01} - \gamma'_{02})x_1}}{1 + 2x_1^2 + 2x_1 x_2 + x_2^2} \left[\frac{-\Gamma_{01}}{\Gamma_0 - \gamma_1} e^{-\gamma'_1 x_2} + \left(1 + \frac{\Gamma_{01}}{\Gamma_0 - \gamma_1}\right) e^{-\Gamma'_0 x_2} + e^{(i\Delta' - \gamma'_{12})x_2} \right] \right\}, \tag{5}$$

where dimensionless variables (x) have been introduced for convenience. Dimensionless frequencies (see Table I below for decay-rate definitions) are denoted by a prime and are defined relative to the beam transit rate $\gamma_t = \sqrt{2}\, u/D$. The detuning is $\Delta' \equiv \Delta/\gamma_t = (\Omega_2 - \Omega_1 - \omega_{12})/\gamma_t$ and $\gamma' \equiv \gamma/\gamma_t$ for each rate γ. Λ is given as

$$\Lambda = \pi \left(\frac{D}{2}\right)^2 N_2^{(0)} \frac{\hbar \Omega_1}{ku/\sqrt{\pi}} \left| \frac{\vec{\mu}_{01} \cdot \vec{E}_1}{2\hbar} \right|^2 \left| \frac{\vec{\mu}_{02} \cdot \vec{E}_2}{2\hbar} \right|^2 \frac{1}{\gamma_t^2}. \tag{6}$$

The integrals in Eq. (5) can be done numerically (e.g., see Ref. 14). However, this is not required when considering the extreme transit-time regime.

TABLE I. Decay-rate definitions.

$\gamma_0, \gamma_1, \gamma_2$	Nonradiative level decay rates.
Γ_{01}, Γ_{02}	Spontaneous decay (optical pumping) rates for the 0-1 and 0-2 transitions, respectively.
$\Gamma_0 = \gamma_0 + \Gamma_{01} + \Gamma_{02}$	Total decay rate for level 0.
$\gamma_{01} = \frac{1}{2}(\gamma_0 + \gamma_1 + \Gamma_{01} + \Gamma_{02})$	Total coherence decay rate, 0-1 transition.
$\gamma_{02} = \frac{1}{2}(\gamma_0 + \gamma_2 + \Gamma_{01} + \Gamma_{02})$	Total coherence decay rate, 0-2 transition.
$\gamma_{12} = \frac{1}{2}(\gamma_1 + \gamma_2)$	Total coherence decay rate, 1-2 transition.

In the extreme transit-time limit $\gamma_1', \gamma_{12}' \ll 1$, but $\gamma_{01}', \gamma_{02}', \Gamma_0' \gg 1$ (i.e., long-lived lower levels, short-lived upper levels, relative to the beam transit time), one can set $x_1' \simeq 0$ in the denominator of Eq. (5) and take $\gamma_1' \simeq 0$, $\gamma_{12}' \simeq 0$. The integrals are then easily done and yield (to lowest order in small quantities) the resonant contribution to the probe absorption in third order

$$\frac{\delta P_1}{\delta L} = \tfrac{1}{2}\alpha_0 S P_1^{(0)} \mathcal{L}(\Delta), \tag{7}$$

where

$$\mathcal{L}(\Delta) = -\frac{\Gamma_{01}}{\Gamma_0}\frac{(\gamma_{01}+\gamma_{02})^2}{\Delta^2 + (\gamma_{01}+\gamma_{02})^2} + \exp\left(-\left|\frac{\Delta}{\gamma_t}\right|\right). \tag{8}$$

The saturation parameter S, is defined as

$$S \equiv I_2^{(0)}/I_{\text{sat}}, \tag{9}$$

and I_{sat} the saturation intensity is

$$I_{\text{sat}} = \frac{c\hbar^2}{2\pi|\vec{\mu}_{02}|^2}\left(\frac{2}{\pi}\gamma_t\right)(\gamma_{01}+\gamma_{02}) \simeq \frac{c\hbar^2}{2\pi|\vec{\mu}_{02}|^2}\left(0.89\frac{u}{D}\right)(\gamma_{01}+\gamma_{02}), \tag{10}$$

where u is the thermal speed and D is the $1/e$ intensity diameter. $P_1^{(0)}$ is the incident traveling wave power of the probe. α_0 is the small signal absorption coefficient for the 0-1 transition and is given by

$$\alpha_0 = 4\pi k\frac{(\mu_{01})^2}{\hbar}\frac{N_2^{(0)}}{ku/\sqrt{\pi}}. \tag{11}$$

Note that the Doppler factor must be included in Eq. (11) for detunings far from line center.

Equation (7) is a closed-form expression for the line shape in the extreme transit-time regime, with fast upper-level decay rates. The exponential form of the Raman term in Eq. (8) is valid for $\Delta' \gg \gamma_{12}'$ (i.e., $\Delta \gg \gamma_{12}$). This is not a serious restriction since the $1/e$ width is γ_t which is very large compared to γ_{12}, at low pressures. The exact second derivative of this term at the origin ($\Delta' = 0$) is

$$-\frac{1}{\delta P_1^{(\text{RAMAN})}}\frac{d^2\delta P_1^{(\text{RAMAN})}}{d\Delta^2} = \frac{1}{\gamma_t}\frac{1}{\gamma_{12}}, \tag{12}$$

which is finite but large ($\gamma_{12} \to 0$).

Atomic sodium is one example of a system in which the limiting case of Eq. (8) is applicable. Reasonable numbers for Na are typically of the order $\Gamma_{01} \simeq \Gamma_{02} \simeq 10$ MHZ and $\gamma_t \simeq 0.1$ MHZ. Figure

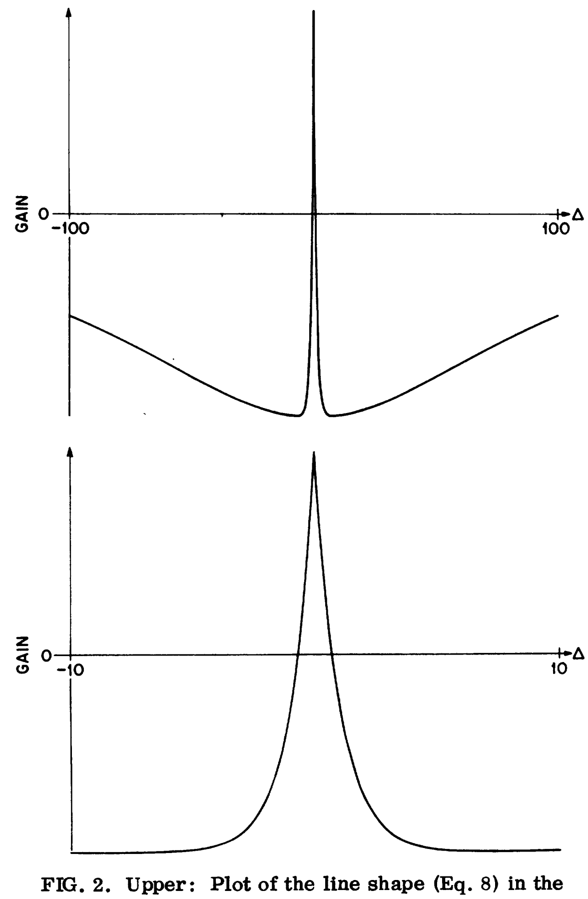

FIG. 2. Upper: Plot of the line shape (Eq. 8) in the extreme transit-time limit for $(\gamma_{01}+\gamma_{02}) = 100\gamma_t$ and $\Gamma_{01} = 0.5\Gamma_{03}$ where $\Delta = (\Omega_2 - \Omega_1 - \omega_{12})/\gamma_t$. Lower: Detailed plot of the narrow resonance.

2(a) is a plot of $\mathcal{L}(\Delta)$ for $(\gamma_{01}+\gamma_{02})=100\gamma_t$ and $\Gamma_{01}=0.5\,\Gamma_0$. As expected, it shows a narrow resonance (Raman term) superimposed on a broader Lorentzian (rate-equation term) of opposite sign. At zero detuning there is gain due to the Raman transition 2-1. With increasing detuning, however, optical pumping (fast radiative decay) becomes important and absorption begins to dominate.

The most interesting feature of Fig. 2 is the exponential form of the narrow resonance. Figure 2(b) shows this resonance in greater detail. The half-width at half maximum $\Delta_{\rm HWHM}$ is given by

$$\Delta_{\rm HWHM}^{\rm (Hz)}=\gamma_t \ln 2 \simeq 0.98\,u/2\pi D\,, \tag{13}$$

where u is the thermal speed and D the $1/e$ intensity diameter.

It is important to note that, in the extreme transit-time limit, only the *shape* of the narrow resonance is appreciably affected by the finite time of flight of the atom through the beam, whereas the *shape* of the population-saturation resonance remains essentially unaffected. Thus, for the case considered here, Raman-type or coherent double-quantum processes appear quite sensitive to transit-time effects. However, population-saturation processes are relatively insensitive.

Equation (10) for the saturation intensity and Eq. (13) for the half-width, together with the above discussion, suggest that in this limit a plane-wave theory can be used to calculate the approximate line shape. In this case, the plane-wave theory yields a narrow *Lorentzian* Raman-type resonance superimposed on a broader Lorentzian rate-equation background. Although the precise *shape* of the Raman resonance is incorrectly predicted (i.e., Lorentzian instead of exponential shape), the intensities and widths of the broad and narrow resonances are given to approximately a few percent accuracy if the effective lower-level decay rates γ_1, γ_2 are taken to be $\sim u/D$ (rad/sec).

In studying the transit-time regime as presented here, the Raman term makes it possible to use long lower-level lifetimes as a means of studying transit-time effects. As a result, one is provided with an excellent method of investigating the manner in which beam transit-time enters into three-level line shapes.

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APPENDIX: DETAILS OF LINE SHAPE CALCULATION

The density-matrix equations for the system shown in Fig. 1 are

$$\begin{aligned}
\dot\rho_{00}&=-(i/\hbar)(V_{01}\rho_{10}-\rho_{01}V_{10})-(i/\hbar)(V_{02}\rho_{20}-\rho_{02}V_{20})-\Gamma_0\rho_{00}\,,\\
\dot\rho_{11}&=-(i/\hbar)(V_{10}\rho_{01}-\rho_{10}V_{01})-\gamma_1\rho_{11}+\Gamma_{01}\rho_{00}\,,\\
\dot\rho_{22}&=-(i/\hbar)(V_{20}\rho_{02}-\rho_{20}V_{02})-\gamma_2\rho_{22}+\Gamma_{01}\rho_{00}\,,\\
\dot\rho_{01}&=-(\gamma_{01}+i\omega_{01})\rho_{01}-(i/\hbar)(\rho_{11}-\rho_{00})V_{01}-(i/\hbar)V_{02}\rho_{21}\,,\\
\dot\rho_{02}&=-(\gamma_{02}+i\omega_{02})\rho_{02}-(i/\hbar)(\rho_{22}-\rho_{00})V_{02}-(i/\hbar)V_{01}\rho_{12}\,,\\
\dot\rho_{12}&=-(\gamma_{12}+i\omega_{12})\rho_{12}-(i/\hbar)(V_{20}\rho_{02}-\rho_{10}V_{02})\,,\\
\rho_{ji}&=\rho_{ij}^*\,,
\end{aligned}\tag{A1a}$$

where

$$\begin{aligned}
\Gamma_0&=\gamma_0+\Gamma_{01}+\Gamma_{02}\,,\\
\gamma_{01}&=\tfrac{1}{2}(\gamma_0+\gamma_1+\Gamma_{01}+\Gamma_{02})\,,\\
\gamma_{02}&=\tfrac{1}{2}(\gamma_0+\gamma_2+\Gamma_{01}+\Gamma_{02})\,,\\
\gamma_{12}&=\tfrac{1}{2}(\gamma_1+\gamma_2)\,,
\end{aligned}\tag{A1b}$$

as defined in Table I. The γ_0, γ_1, and γ_2 are the nonradiative decay rates of levels 0, 1, and 2, respectively. Γ_{01} and Γ_{02} are the spontaneous decay rates (optical pumping) for the 0-1 and 0-2 transitions, respectively. $\hbar$ is Planck's constant divided by 2π. The interactions are $V_{01}=-\vec\mu_{01}\cdot\vec{\rm E}_1$ and $V_{02}=-\vec\mu_{02}\cdot\vec{\rm E}_2$, where the slowly varying envelope approximation $(1/E_0)(dE_0/dZ)l\ll 1$ is used with l the order of a mean-free path. By a suitable choice of phase for the wave functions of levels 1 and 2, one may take $\mu_{10}=\mu_{01}$ and $\mu_{20}=\mu_{02}$.

Owing to the Boltzmann factor, for an optical transition, the unperturbed population of level 0 is negligible compared to that of level 2. The population of level 1 produces only nonresonant (Doppler-broadened) contributions to the line shape in third order. Therefore, the initial conditions on the density matrix are taken to be

$$\rho_{ij}^{(0)}(t=t_0;\vec{v},\vec{r}_0,t_0;2)=\delta_{i2}\,\delta_{j2}\,, \tag{A2}$$

which indicates the creation of an atom in state 2 at time t_0 at position $\vec{r}_0$ with velocity $\vec{v}$. It is assumed that the atoms move in straight lines and simply decay out of the appropriate levels. These assumptions lead to the following results in third order:

$$\begin{aligned}\rho_{01}^{(3)}(\vec{x},\vec{v},t;t_0,2)=&\left(\frac{-i}{\hbar}\right)^3\int_{t_0}^{t}dt_3\int_{t_0}^{t_3}dt_2\int_{t_0}^{t_2}dt_1\{[e^{-(\gamma_{01}+i\omega_{01})(t-t_3)}\\&\times V_{01}(t_3)e^{-\Gamma_0(t_3-t_2)}V_{02}(t_2)e^{-(\gamma_{02}-i\omega_{01})(t_2-t_1)}V_{02}(t_1)+\text{c.c.}]\\&+e^{-(\gamma_{01}+i\omega_{01})(t-t_3)}V_{02}(t_3)e^{-(\gamma_{12}-i\omega_{12})(t_3-t_2)}V_{02}(t_2)\\&\times e^{-(\gamma_{02}-i\omega_{02})(t_2-t_1)}V_{02}(t_1)\}e^{-\gamma_2(t_2-t_0)}\\&-\Gamma_{01}\left(\frac{-i}{\hbar}\right)^3\int_{t_0}^{t}dt_3\int_{t_0}^{t_3}dt'\int_{t_0}^{t'}dt_2\int_{t_0}^{t_2}dt_1\,e^{-(\gamma_{01}+i\omega_{01})(t-t_3)}\\&\times[e^{-\gamma_1(t_3-t')}V_{01}(t_3)e^{-\Gamma_0(t'-t_2)}V_{02}(t_2)\\&\times e^{-(\gamma_{02}-i\omega_{02})(t_2-t_1)}V_{02}(t_1)+\text{c.c.}]e^{-\gamma_2(t_1-t_0)}\,,\end{aligned} \tag{A3}$$

where

$$V_{01}(t_n)=-\mu_{01}E_1^{(0)}U(x_0+v_x(t_n-t_0))U(y_0+v_y(t_n-t_0))\cos\{k_1[z_0+v_z(t_n-t_0)]-\Omega_1t_n\}\,, \tag{A4a}$$

$$V_{02}(t_n)=-\mu_{02}E_2^{(0)}U(x_0+v_x(t_n-t_0))U(y_0+v_y(t_n-t_0))\cos\{k_2[z_0+v_z(t_n-t_0)]+\Omega_2t_n\}\,, \tag{A4b}$$

and

$$U(x)=\exp(-2x^2/D^2)\,, \tag{A4c}$$

$$U(y)=\exp(-2y^2/D^2)\,. \tag{A4d}$$

Only atoms which arrive at position $\vec{x}$ at time t influence the measurement of polarization at $(\vec{x},t)$. This is equivalent to replacing $\vec{x}_0$ by $\vec{x}-\vec{v}_x(t-t_0)$ in the potentials, due to the assumed straight-line motion of the atoms. Therefore, in Eq. (A3) let

$$V_{01}(t_n)\to V'_{01}(t_n)=-\mu_{01}E_1^{(0)}U(x-v_x(t-t_n))U(y-v_y(t-t_n))\cos\{k_1[z-v_z(t-t_n)]-\Omega_1t_n\}\,, \tag{A5a}$$

$$V_{02}(t_n)\to V'_{02}(t_n)=-\mu_{02}E_2^{(0)}U(x-v_x(t-t_n))U(y-v_y(t-t_n))\cos\{k_2[z-v_z(t-t_n)]+\Omega_2t_n\}. \tag{A5b}$$

In equilibrium the number of atoms created in state 2 between t_0 and t_0+dt_0 is given by $N_2^{(0)}(\vec{v})d^3\vec{v}\gamma_2dt_0$, where

$$N_2^{(0)}(\vec{v})=N_2^{(0)}1/(u\sqrt{\pi})^3\exp-(v_x^2+v_y^2+v_z^2/u^2)\,. \tag{A6}$$

Then the total contribution to $\rho_{01}^{(3)}(\vec{x},t;t_0,2)$ due to atoms created at all times t_0, with all velocities $\vec{v}$, is

$$\rho_{01}^{(3)}(\vec{x},t;2)=\int d^3\vec{v}\int_{-\infty}^{t}dt_0\gamma_2N_2^{(0)}(\vec{v})\rho_{01}^{(3)}(\vec{x},\vec{v},t,t_0,2)\,, \tag{A7}$$

where the cell diameter is assumed large compared to the mean-free path so that the lower limit may be replaced by $-\infty$.

From Eqs. (A5a) and (A5b) it is evident that the $V(t_n)$ do not depend on t_0 so that the only t_0 dependence appears in $\exp[-\gamma_2(t_1-t_0)]$ and in the integration limits. To perform the t_0 integration first, the order of integration is changed in the usual manner to obtain

$$\int_{-\infty}^{t}dt_0\int_{t_0}^{t}dt_5\int_{t_0}^{t_3}dt_2\int_{t_0}^{t_2}dt_1=\int_{-\infty}^{t}dt_3\int_{-\infty}^{t_3}dt_2\int_{-\infty}^{t_2}dt_1\int_{-\infty}^{t_1}dt_0$$

and

$$\int_{-\infty}^{t}dt_0\int_{t_0}^{t}dt_3\int_{t_0}^{t_3}dt'\int_{t_0}^{t'}dt_2\int_{t_0}^{t_2}dt_1=\int_{-\infty}^{t}dt_3\int_{-\infty}^{t_3}dt'\int_{-\infty}^{t'}dt_2\int_{-\infty}^{t_2}dt_1\int_{-\infty}^{t_1}dt_0\,.$$

Using

$$\int_{-\infty}^{t}dt_0\gamma_2e^{-\gamma_2(t_1-t_0)}=1$$

and by making the following change of variables:

$$\tau_3 = t - t_3,\quad t - \text{const},$$
$$\tau_2 = t_3 - t_2,\quad t_3 - \text{const},$$
$$\tau_1 = t_2 - t_1,\quad t_2 - \text{const},$$
$$\tau' = t_3 - t',\quad t_3 - \text{const},$$
$$\tau_2' = t' - t_2,\quad t' - \text{const},$$

one obtains an expression for $\rho_{01}^{(3)}(\vec{x}, t; 2)$. Further simplification is obtained by defining

$$\tau_+ \equiv \tau_2' + \tau',$$
$$\tau_- \equiv \tau_2' - \tau',$$

or

$$\tau_2' = \tfrac{1}{2}(\tau_+ + \tau_-) \geq 0,$$
$$\tau' = \tfrac{1}{2}(\tau_+ - \tau_-) \geq 0,$$

with a Jacobian $= \frac{1}{2}$.

Then, Eq. (A7) yields

$$\rho_{01}^{(3)}(\vec{x}, t; 2) = (-i/\hbar)^3 \int_0^\infty d\tau_3 \int_0^\infty d\tau_2 \int_0^\infty d\tau_1\, e^{-(\gamma_{01}+i\omega_{02})\tau_3} \times \Big\{ V_{01}'(t-\tau_3)\Big[-\frac{\Gamma_{01}}{\Gamma_0-\gamma_1} e^{-\gamma_1\tau_2} + \Big(1+\frac{\Gamma_{01}}{\Gamma_0-\gamma_1}\Big)e^{-\Gamma_0\tau_2}\Big]$$
$$\times [V_{02}'(t-\tau_2-\tau_3)\, e^{-(\gamma_{02}-i\omega_{02})\tau_1} V_{02}'(t-\tau_1-\tau_2-\tau_3) + \text{c.c.}]$$
$$+ V_{01}'(t-\tau_3) e^{-(\gamma_{12}-i\omega_{12})\tau_2}$$
$$\times V_{02}'(t-\tau_2-\tau_3) e^{-(\gamma_{02}-i\omega_{02})\tau_1} V_{02}'(t-\tau_1-\tau_2-\tau_3)\Big\}, \tag{A8}$$

where $\tau_+ \to \tau_2$, since τ_+ is a dummy variable.

Substituting the explicit potentials into Eq. (A8) results in a somewhat complicated equation for $\rho_{01}^{(3)}$. Simplification is achieved by retaining only the first-harmonic components in $(\Omega_1 t)$, which contribute to the time averaged absorbed power [Eq. (A12) below.] One finds that the combination $\Omega_1(t-\tau_3)$ appears in each term (due to V_{01}') and these in turn are multiplied by the factor $\exp(i\omega_{01}\tau_3)$. Therefore, the resonant (slowly varying) contributions to the τ_3 integration must contain $\exp[i\Omega_1(t-\tau_3)]$. The complex conjugate first-harmonic components are antiresonant and may be neglected.

The number of terms in $\rho_{01}^{(3)}$ can be further reduced by performing the v_z integration in Eq. (A7). A typical term is of the form

$$I(\tau) = \frac{1}{u\sqrt{\pi}} \int_{-\infty}^{\infty} dv_z \exp(ikv_z\tau)\exp[-(v_z/u)^2]$$
$$= \exp(-\tfrac{1}{4}k^2u^2\tau^2). \tag{A9}$$

The terms retained from Eq. (A8) have $\tau = (\tau_3 \pm \tau_1)$, etc., and are slowly varying in τ. Hence, $I(\tau)$ is sharply peaked in τ, since $ku \gg \gamma$ (γ is a homogeneous width). This enables one to take $I(\tau)$ proportional to $\delta(\tau)$. Integrating $I(\tau)$ determines the proportionality constant $2\sqrt{\pi}/ku$. Thus, one has

$$I(\tau) \simeq \frac{2\sqrt{\pi}}{ku}\,\delta(\tau). \tag{A10}$$

Now, a term with $\tau = \tau_3 - \tau_1$ samples whenever $\tau_3 = \tau_1$ while a term containing $(\tau_1 + \tau_3)$ samples only for $\tau_1 = \tau_3 = 0$ since $\tau_i \geq 0$. Hence, contributions of the latter type may be dropped by comparison to the former. Completing the v_x and v_y integrations in Eq. (A7), one finally obtains for copropagating waves

$$\rho_{01}^{(3)}(\vec{x}, t; 2) = N_2^{(0)}\Big(\frac{i}{2\hbar}\Big)^3 \mu_{01}\mu_{02}^2 \frac{2\sqrt{\pi}}{k_1 u} e^{-i(k_1 z - \Omega_1 t)}$$
$$\times \int_0^\infty d\tau_3 \int_0^\infty d\tau_2 \int_0^\infty d\tau_1\, e^{-[\gamma_{01}+i(\omega_{01}-\Omega_1)]\tau_3} e^{-[\gamma_{02}-i(\omega_{02}-\Omega_2)]\tau_1} \times \Big\{\Big[\Big(-\frac{\Gamma_{01}}{\Gamma_0-\gamma_1}\Big)e^{-\gamma_1\tau_2} + \Big(1+\frac{\Gamma_{01}}{\Gamma_0-\gamma_1}\Big)e^{-\Gamma_0\tau_3}\Big] \begin{bmatrix} \delta\Big(\tau_1 - \frac{k_1}{k_2}\tau_3\Big), & k_1 \geq k_2 \\ \frac{k_2}{k_1}\delta\Big(\tau_3 - \frac{k_2}{k_1}\tau_1\Big) & k_2 \geq k_1 \end{bmatrix}$$

$$+e^{-[\gamma_{12}-i(\omega_{12}+\Omega_1-\Omega_2)]\tau_2}\begin{bmatrix}\delta\left(\tau_1-\frac{k_1}{k_2}\tau_3+\epsilon\,\tau_2\right), & k_1\geq k_2\\ \frac{k_2}{k_1}\,\delta\left(\tau_3-\frac{k_2}{k_1}\tau_1+\epsilon\,\tau_2\right), & k_2\geq k_1\end{bmatrix}\Bigg\}\,J(x,\tau_1,\tau_2,\tau_3)J(y,\tau_1,\tau_2,\tau_3)\,, \quad \text{(A11a)}$$

where

$$\epsilon=\begin{cases}1-\frac{k_1}{k_2}\,, & k_1\geq k_2\\ 1-\frac{k_2}{k_1}\,, & k_2\geq k_1\end{cases} \quad \text{(A11b)}$$

and

$$J(x,\tau_1,\tau_2,\tau_3)=\int_{-\infty}^{\infty}\frac{dv_x}{u\sqrt{\pi}}\,e^{-(v_x/u)^2}\,U_x(t_1)U_x(t_2)U_x(t_3)$$

$$=\frac{\exp[-x^2(3\alpha-\alpha^2B^2/1/u^2+\alpha A)]}{(1+\alpha u^2A)^{1/2}}\,. \quad \text{(A11c)}$$

$J(y,\tau_1,\tau_3,\tau_2)$ is the same as above with x and y interchanged; (A11d)

$$\alpha=2/D^2\,, \quad \text{(A11e)}$$

$$A=(\tau_1+\tau_2+\tau_3)^2+(\tau_2+\tau_3)^2+\tau_3^2\,,$$
$$B=3\tau_3+2\tau_2+\tau_1\,. \quad \text{(A11f)}$$

Equation (A11) is valid for any two coupled Doppler-broadened transitions (inverted V configuration).

The time and spatially averaged absorbed power per unit volume is

$$P=\frac{1}{L}\int_0^L dz\langle(\dot{\vec{P}}_{01}\cdot\vec{E}_1)\rangle\,, \quad \text{(A12)}$$

where L is the active length of the cell, $\langle\ \rangle$ denotes time averaging, and $\vec{P}_{01}$ is the polarization on the 0-1 transition, which is

$$\vec{P}_{01}=\vec{\mu}_{01}\rho_{10}+\vec{\mu}_{10}\rho_{01}$$
$$=2\,\mathrm{Re}\,\vec{\mu}_{10}\rho_{01}$$

and

$$\vec{E}_1=\mathrm{Re}\,\vec{E}_1^{(0)}\exp(-2r^2/D^2)\exp[-i(k_1z-\Omega_1t)]\,.$$

The power absorbed per unit length is given by

$$P^{(0,1)}/L=\int_0^\infty 2\pi r\,dr\,P(r)\,. \quad \text{(A13)}$$

For nearly degenerate or degenerate case ($k_1\simeq k_2\equiv k$), Eqs. (A11), (A12), and (A13) readily yield Eq. (6) of the text.

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