Free precession decay in selective reflection

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A theory of free precession in selective reflection is presented. In contrast to standard theories of free precession in which transmission signals decay in a time on the order of the inverse of the inhomogeneous width, the selective reflection signal decays in a time of order of the inverse of the *homogeneous* width. Both short-pulse excitation and continuous-wave (cw) excitation are considered. In the case of cw excitation, it is possible to observe beating between the contributions to the signal that are first and third order in the amplitude of the excitation field.

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I. INTRODUCTION

Selective reflection at a dielectric-vapor interface [1–3] has proven to be an interesting technique for probing the effects of collisions between atoms in the vapor and the dielectric surface, as well as collisional effects in optically dense media [4,5]. There are basically two features of the atom-dielectric interaction that are important, the van der Waals interaction between the atoms and the dielectric [6] which modifies the atomic response and collisions with the surface that quench any atomic excitation and change the velocity of the atoms. There have been several studies of these phenomena including both linear [2,3] and nonlinear interactions [7–9] between the atoms and the incident fields. One somewhat surprising feature in the linear response is a sub-Doppler structure that manifests itself in selective reflection [1–4]. This feature can be attributed to wall collisions that quench the atomic excitation and change the atomic velocity. Owing to the quenching and the velocity change, there is a fundamental difference in the response of atoms moving towards or away from the interface. Despite this asymmetry, the contribution to the reflected field from atoms moving towards or away from the surface is the same in linear response for a low-density vapor [2,3,7,10].

Most of the studies of selective reflection have involved continuous excitation by an incident field. In this paper, we consider selective reflection involving *transient* fields. In particular, we calculate the free-precession (FP) signal for atoms in the vapor excited by pulsed or cw excitation. In the case of cw excitation, the transient response is achieved by a sudden turning off of the field—only the response to third order in the excitation field is considered for the cw case. If the effects of wall collisions are neglected, the FP response for short-pulse excitation or following cw excitation (in linear response) lasts only for a time equal to the inverse Doppler width $(ku)^{-1}$ associated with the vapor ($k=\omega/c$, where ω is the atomic transition frequency and u is the most probable atomic speed). In the Bloch vector picture, this rapid decrease in the FP signal is attributed to different precession rates for Bloch vectors corresponding to different velocity subgroups of atoms. What may not be appreciated fully, however, is that the rapid decay depends critically on the fact that there is a symmetric precession corresponding to atoms moving with both positive and negative velocities. If this symmetry is broken, it is possible for the linear response to last for a time equal to the inverse of the *homogeneous* decay rate rather than the inhomogeneous one (Doppler width). Since selective reflection provides exactly this type of symmetry breaking, one expects to see a qualitatively different FP response in selective reflection than in traditional FP. This slow decay is the transient analog of the sub-Doppler structure in selective reflection in the frequency domain. The response for short-pulse excitation and the linear response for cw excitation turns out to be the same for atoms approaching and leaving the interface, as in the frequency domain [2–4]. The third-order response for cw fields differs for atoms approaching and leaving the interface, and it is possible to observe beating between the first- and third-order responses.

II. FP TRANSMISSION SIGNAL NEGLECTING WALL COLLISIONS

It is perhaps useful to recall the standard FP transmission results for both short pulse and cw field excitation [11]. In both cases, the atoms are modeled as having two levels, 1 and 2, separated in frequency by ω. The decay rate of the upper level is denoted by γ_2, the coherence ρ_{12} decays at rate γ, and the population $(\rho_{11}+\rho_{22})$ is conserved.

Short pulse excitation. An atomic vapor is irradiated with a field having electric vector

$$\mathbf{E}(Z,t)=\tfrac{1}{2}\hat{\imath}Ef(t)\,e^{i(k_0Z-\Omega t)}+\text{c.c.},$$

where $k_0=\Omega/c\approx k$ is the field propagation constant, Ω is the field frequency, E is the field amplitude (assumed real), $f(t)$ is the pulse envelope function centered at $t=0$ having temporal width τ normalized such that $\int_{-\infty}^{\infty}dt\,f(t)=\tau$, and c.c. stands for complex conjugate. It is assumed that $\gamma\tau\ll ku\tau\ll1$ and that $|\Delta|\tau\ll1$, where $\Delta=\Omega-\omega$ is the atom-field detuning. This field gives rise to a polarization in the vapor that can be written as

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$$\mathbf{P}(Z,t)=\tfrac{1}{2}\hat{i}P(t)\,e^{i(kZ-\Omega t)}+\text{c.c.}$$
$$=\hat{i}\,\mu\mathcal{N}[\tilde{\rho}_{21}(t)\,e^{i(kZ-\Omega t)}+\text{c.c.}],$$

where μ is a dipole matrix element (taken to be real), $\mathcal{N}$ is the atomic density, and $\tilde{\rho}_{21}(t)=\langle\tilde{\rho}_{21}(\mathbf{v},t)\rangle$ is a density-matrix element, written in an interaction representation, averaged over the velocity distribution

$$W(\mathbf{v})=\frac{1}{(\pi u^2)^{3/2}}e^{-v^2/u^2}. \tag{1}$$

In turn, the polarization field gives rise to a signal field

$$\mathbf{E}_s(Z,t)=\tfrac{1}{2}\hat{i}E_s(t)\,e^{i(kZ-\Omega t)}+\text{c.c.}$$

Using Maxwell's equations, one can relate the signal field to the polarization as

$$E_s(t)=\frac{ik}{2\epsilon_0}\int_0^L dZ\,P(Z,t)$$
$$=\frac{i\mathcal{N}\mu}{\epsilon_0}I_0(t),$$

where

$$I_0(t)=k\int_0^L dZ\,\tilde{\rho}_{21}(Z,t) \tag{2}$$

and L is the sample length. The radiated signal is proportional to $|I_0|^2$. In order to neglect superradiant collective effects, we assume that $(\mathcal{N}\lambda^2L)(\gamma_2/ku)<1$ [12].

The evolution equations for density-matrix elements are

$$d\tilde{\rho}_{21}/dt=i\chi[2\rho_{22}-W(\mathbf{v})]-\eta(v)\tilde{\rho}_{21}, \tag{3a}$$

$$d\rho_{22}/dt=i\chi[\tilde{\rho}_{21}-\tilde{\rho}_{12}]W(\mathbf{v})-\gamma_2\tilde{\rho}_{22}, \tag{3b}$$

where $\tilde{\rho}_{12}=\tilde{\rho}_{21}^{*}$, $\chi=-(\mu E/2\hbar)$ is a Rabi frequency, and

$$\eta(v_z)=\gamma-i\Delta+ikv_z. \tag{4}$$

It is easy to show that, following excitation by the pulse,

$$\tilde{\rho}_{21}(t)=-(i/2)\sin(2\chi\tau)e^{-(\gamma-i\Delta)t}\int d\mathbf{v}\,e^{-ikv_zt}W(\mathbf{v})$$
$$=-(i/2)\sin(2\chi\tau)\,e^{-(\gamma-i\Delta)t}(1/\sqrt{\pi})\int_{-\infty}^{\infty}dx\,e^{-iyx}e^{-x^2}$$
$$=-(i/2)\sin(2\chi\tau)\,e^{-(\gamma-i\Delta)t}\,e^{-y^2/4}, \tag{5}$$

where

$$y=kut. \tag{6}$$

From Eq. (2), it follows that $I_0=kL\tilde{\rho}_{21}(t)$; as a consequence, the FP signal decays in a time of order $(ku)^{-1}$. The factor $e^{i\Delta t}$ in Eq. (5) is indicative of the fact that the atoms radiate at the atomic frequency.

cw excitation. In this case a cw field is applied up until time $t=0$, at which time it is abruptly turned off. At $t=0$, to third order in the incident field, the density-matrix element $\tilde{\rho}_{21}(\mathbf{v},t)$ has achieved its steady-state value

$$\tilde{\rho}_{21}(\mathbf{v})=\tilde{\rho}_{21}^{(1)}(\mathbf{v})+\tilde{\rho}_{21}^{(3)}(\mathbf{v}), \tag{7}$$

where

$$\tilde{\rho}_{21}^{(1)}(\mathbf{v})=\frac{-i\chi}{(\gamma-i\Delta)+ikv_z}W(\mathbf{v}) \tag{8}$$

and

$$\tilde{\rho}_{21}^{(3)}(\mathbf{v})=\frac{4i\chi^3\gamma}{\gamma_2[(\gamma-i\Delta)+ikv_z][\gamma^2+(\Delta-kv_z)^2]}W(\mathbf{v}). \tag{9}$$

For $t>0$, one finds

$$\tilde{\rho}_{21}^{(1)}(t)=-i\chi e^{-(\gamma-i\Delta)t}\int d\mathbf{v}\frac{e^{-ikv_zt}W(\mathbf{v})}{(\gamma-i\Delta)+ikv_z}$$
$$=-i\frac{\chi}{ku}\sqrt{\pi}\exp\left[\left(\frac{\gamma-i\Delta}{ku}\right)^2\right]\left[1-\Phi\left(\frac{y}{2}+\frac{\gamma-i\Delta}{ku}\right)\right], \tag{10}$$

where Φ is the error function. The linear response in this case also decays in a time of order $(ku)^{-1}$, since, for $y\gg1$,

$$\sqrt{\pi}\,\exp\left[\left(\frac{\gamma-i\Delta}{ku}\right)^2\right]\left[1-\Phi\left(\frac{y}{2}+\frac{\gamma-i\Delta}{ku}\right)\right]$$
$$\sim\frac{e^{-(\gamma-i\Delta)t}e^{-y^2/4}}{\frac{y}{2}+\frac{\gamma-i\Delta}{ku}}.$$

For $\gamma/ku\ll1$ and $|\Delta|/ku\lesssim1$, the third-order response is given by

$$\tilde{\rho}_{21}^{(3)}(t)=e^{-(\gamma-i\Delta)t}$$
$$\times\int d\mathbf{v}\frac{4i\chi^3\gamma W(\mathbf{v})e^{-ikv_zt}}{\gamma_2[(\gamma-i\Delta)+ikv_z][\gamma^2+(\Delta-kv_z)^2]}$$
$$\approx i\left(\frac{\chi^3}{\gamma^2ku}\right)\left(\frac{2\gamma}{\gamma_2}\right)\sqrt{\pi}e^{-\Delta^2/k^2u^2}e^{-2\gamma t}. \tag{11}$$

The third-order response decays in a time of order $1/2\gamma$. The velocity subgroup of atoms having $kv_z=\Delta\pm\gamma$ provides the major contribution to the third-order response. These selected atoms act as a *homogeneous* sample, having characteristic frequency width γ. Note the absence of a factor $e^{i\Delta t}$; in the laboratory frame, the selected atoms radiate at the laser frequency.

For both short-pulse and cw excitation (in linear response), there is an important cancellation in the contributions from atoms having $v_z>0$ and $v_z<0$ for times $y>1$. As is seen below, in selective reflection, this cancellation no longer occurs and the linear response, FP signal persists for times $y>1$.

III. SELECTIVE REFLECTION

We choose a geometry in which the dielectric-vapor interface coincides with the $Z=0$ plane, with the vapor confined between the planes $Z=0$ and $Z=L$. At $Z=L$, there is a second vapor-dielectric interface. For simplicity, it is assumed that the incident field is totally absorbed at this second interface [13]. The field entering the vapor from the dielectric is given by

$$\mathbf{E}(Z,t)=\tfrac{1}{2}\hat{\imath}E(t)e^{i(kZ-\Omega t)}+\text{c.c.},$$

where $E(t)=[2n/(n+1)]E_d(t)$, n is the index of refraction of the dielectric, and $E_d(t)$ is the incident field amplitude in the dielectric [$E_d(t)=0$ for $t>0$]. It is assumed that the vapor is optically thin over a distance $\lambda=2\pi/k$. The coherent response in the backward direction constitutes the reflected signal. This response is limited to a diffraction cone having an angle of order λ/a, where a is the diameter of the incident beam and $\lambda=2\pi/k_0$. The reflected field amplitude in the backward direction, obtained from the Maxwell-Bloch equations, is [3,7]

$$\begin{aligned}E_r&=\frac{1}{n+1}\frac{ik}{\epsilon_0}\int_0^L dZ\, e^{2ikZ}P(Z,t)\\&=\frac{2}{n+1}\frac{ik\mathcal{N}\mu}{\epsilon_0}\int_0^L dZ\, e^{2ikZ}\tilde{\rho}_{21}(Z,t),\end{aligned}\tag{12}$$

where $P(Z,t)$ and $\tilde{\rho}_{21}(Z,t)$ now depend on Z as well as t. The reflected field can be detected by heterodyning it with a reference field. Here we calculate simply the reflected FP field intensity, which is proportional to

$$S=|E_r|^2=\left(\frac{2}{n+1}\frac{\mathcal{N}\mu}{\epsilon_0}\right)^2|I|^2,$$

where

$$I(t)=k\int_0^L dZ\, e^{2ikZ}\tilde{\rho}_{21}(Z,t).\tag{13}$$

It is assumed that after a collision at an interface, atoms leave in their ground state. Consequently, in calculating the atomic response, one must distinguish atoms moving to the right from those moving to the left. At any given (Z,t), the contribution to the polarization from atoms moving to the right arises only from those atoms that collided with the $Z=0$ interface at time $t-Z/v_z$ while the contribution from atoms moving to the left arises only from those atoms that collided with the $Z=L$ interface at time $t+(L-Z)/v_z$(v_z is negative for atoms moving to the left). For both short pulse and cw excitation, there is no contribution to the polarization if $Z<v_zt$ for atoms moving to the right, since an atom arriving at position Z at time t must have collided with the interface at $Z=0$ at a time $(t-Z/v_z)>0$; as there is no external field for $t>0$ and since any atomic state coherence is destroyed at the interface, an atom arriving at (Z,t) for $(Z-v_zt)<0$ necessarily is in its ground state. Similarly, there is no contribution to the polarization if $(L-Z)<-v_zt$ for atoms moving to the left.

Short-pulse excitation, $\gamma\tau\ll ku\tau\ll1;|\Delta|\tau\ll1$. In this case, $E(t)=Ef(t)$. In analogy with Eq. (5), the density-matrix element $\tilde{\rho}_{21}(Z,t)$ is given by

$$\begin{aligned}\tilde{\rho}_{21}(Z,t)=&-(i/2)\sin(2\chi\tau)\,e^{-\xi t}\int d\mathbf{v}e^{-ikv_zt}W(\mathbf{v})\\&\times[\Theta(-v_z)\Theta(L-Z+v_zt)+\Theta(v_z)\Theta(Z-v_zt)],\end{aligned}\tag{14}$$

where

$$\xi=\gamma-i\Delta,\tag{15}$$

and $\Theta(x)=1$ for $x\geqslant0$ and is 0 otherwise. The integral (13) can be evaluated as

$$\begin{aligned}I=&\tfrac{1}{4}\sin(2\chi\tau)e^{-\xi t}\int d\mathbf{v}[(e^{-ikv_zt}-e^{ikv_zt}e^{2ikL})\Theta(-v_z)\\&+(e^{ikv_zt}-e^{-ikv_zt}e^{2ikL})\Theta(v_z)]W(\mathbf{v})\\=&\tfrac{1}{2}\sin(2\chi\tau)e^{-\xi t}\int d\mathbf{v}(e^{ikv_zt}-e^{-ikv_zt}e^{2ikL})\Theta(v_z)W(\mathbf{v})\\=&\tfrac{1}{4}\sin(2\chi\tau)e^{-\xi t}e^{-y^2/4}\{[1-\Phi(-iy/2)]\\&-e^{2ikL}[1-\Phi(iy/2)]\},\end{aligned}\tag{16}$$

assuming $u/\gamma<L$. It is interesting to note that the contributions from atoms moving towards and away from the interfaces are equal, as in [2,3]. The time scale for the decay is now γ^{-1}, as can be seen from the asymptotic limit, valid for $y=kut\gg1$, given by

$$I\sim\frac{(i/2)(1+e^{2ikL})\sin(2\chi\tau)\,e^{-(\gamma-i\Delta)t}}{\sqrt{\pi}kut}.\tag{17}$$

The factor $e^{i\Delta t}$ is a signature of emission at the atomic resonance frequency.

The reflected field given by Eqs. (12) and (13) originates from the slab of vapor that was excited by the pulse, subject to the boundary condition that there be minimal reflection at $Z=L$ [13]. In most theories of selective reflection, k is assumed to have a small imaginary part $ik\epsilon$, and the assumption $\epsilon kL>1$ is used to eliminate the contribution from the e^{2ikL} term. This approach is justified if the vapor is optically dense over a distance equal to L and if $\epsilon\ll1$. In this limit, the contributions to the reflected field average to zero for atoms having positions $Z\gtrsim(k)^{-1}$. For simplicity, we shall assume that terms involving e^{2ikL} can be neglected [14]. The solid

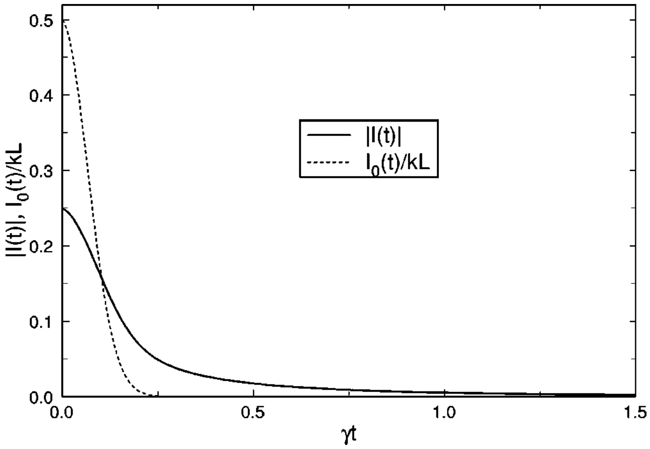

FIG. 1. Graph of the selective reflection, free-precession signal $|I(t)|$ as a function of γt for pulsed excitation (solid line) and the conventional free-precession transmission signal $|I_0(t)|/kL$ for pulsed excitation (dashed line). The signal is independent of the value of Δ provided $|\Delta|\tau \ll 1$, where τ is the pulse duration.

curve in Fig. 1 is a plot of $|I(t)|$ vs $\gamma t=(\gamma/ku)y$ for $2\chi\tau=\pi/2$ and $\gamma/ku=0.05$. For comparison, a graph of a conventional FP signal, $|I_0(t)|$ (divided by kL), is shown as a dashed curve in Fig. 1.

cw excitation. In this case, $E(t)=E[1-\Theta(t)]$. The density-matrix element $\tilde{\rho}_{21}(Z,t)$ is given by

$$\begin{aligned}\tilde{\rho}_{21}(Z,t)=&\int d\mathbf{v}[\Theta(-v_z)\Theta(L-Z+v_zt)\\&+\Theta(v_z)\Theta(Z-v_zt)]\tilde{\rho}_{21}(Z,\mathbf{v},t)\\=&e^{-\xi t}\int d\mathbf{v}e^{-ikv_zt}[\Theta(-v_z)\Theta(L-Z+v_zt)\\&+\Theta(v_z)\Theta(Z-v_zt)]\tilde{\rho}_{21}(Z-v_zt,\mathbf{v},0),\end{aligned}\tag{18}$$

where the result has been expressed in terms of $\tilde{\rho}_{21}(Z-v_zt,\mathbf{v},0)$, which is a function of $\chi=-(\mu E/2\hbar)$. The density-matrix element $\tilde{\rho}_{21}(Z-v_zt,\mathbf{v},0)$ differs for atoms having positive and negative v_z. Combining Eqs. (13) and (18), one finds

$$I=I_-+I_+,\tag{19}$$

where

$$\begin{aligned}I_-=&k\,e^{-\xi t}\int_0^L dZ\,e^{2ikZ}\int d\mathbf{v}e^{-ikv_zt}\Theta(-v_z)\\&\times\Theta(L-Z+v_zt)\tilde{\rho}_{21}(Z-v_zt,\mathbf{v},0)\\=&k\,e^{-\xi t}\int d\mathbf{v}\,e^{ikv_zt}\Theta(-v_z)\int_{-v_zt}^{L}dZ\,e^{2ikZ}\tilde{\rho}_{21}(Z,\mathbf{v},0)\end{aligned}\tag{20}$$

and

$$\begin{aligned}I_+=&k\,e^{-\xi t}\int_0^L dZ\,e^{2ikZ}\int d\mathbf{v}e^{-ikv_zt}\Theta(v_z)\\&\times\Theta(Z-v_zt)\tilde{\rho}_{21}(Z-v_zt,\mathbf{v},0)\\=&k\,e^{-\xi t}\int d\mathbf{v}\,e^{ikv_zt}\Theta(v_z)\int_0^{L-v_zt}dZ\,e^{2ikZ}\tilde{\rho}_{21}(Z,\mathbf{v},0)\end{aligned}\tag{21}$$

represent the contributions from atoms having $v_z<0$ and $v_z>0$, respectively.

For atoms having positive v_z, an atom reaching $(Z,t=0)$ underwent a collision at the $Z=0$ interface and was projected into its ground state at time $t=-Z/v_z$. For this velocity subclass of atoms, the solution of Eqs. (3) to third order in χ is

$$\tilde{\rho}_{21}(Z,\mathbf{v},0;v_z>0)=\tilde{\rho}_{21}^{(1)}(Z,\mathbf{v},0;v_z>0)+\tilde{\rho}_{21}^{(3)}(Z,\mathbf{v},0;v_z>0),\tag{22}$$

where

$$\begin{aligned}\tilde{\rho}_{21}^{(1)}(Z,\mathbf{v},0;v_z>0)=&-i\chi W(\mathbf{v})\int_{-Z/v_z}^{0}e^{\eta(v_z)t'}dt'\\=&-i\chi W(\mathbf{v})v_z^{-1}\int_0^Z e^{-\eta(v_z)(Z-Z')/v_z}dZ'\end{aligned}\tag{23}$$

and

$$\begin{aligned}&\tilde{\rho}_{21}^{(3)}(Z,\mathbf{v},0;v_z>0)\\&=2i\chi^3W(\mathbf{v})\int_{-Z/v_z}^{0}dt'\,e^{\eta(v_z)t'}\int_{-Z/v_z}^{t'}dt''e^{-\gamma_2(t'-t'')}\\&\quad\times\int_{-Z/v_z}^{t''}dt'''[e^{-\eta(v_z)(t''-t''')}+e^{-\eta^*(v_z)(t''-t''')}]\\&=2i\chi^3W(\mathbf{v})v_z^{-3}\int_0^Z dZ'\,e^{-\eta(v_z)(Z-Z')/v_z}\\&\quad\times\int_0^{Z'}dZ''e^{-\gamma_2(Z'-Z'')/v_z}\\&\quad\times\int_0^{Z''}dZ'''[e^{-\eta(v_z)(Z''-Z''')/v_z}+e^{-\eta^*(v_z)(Z''-Z''')/v_z}].\end{aligned}\tag{24}$$

Combining Eqs. (21)–(24), assuming $\gamma L/v_z\gg 1$, and going over to dimensionless variables, one finds after considerable algebra,

$$I_+=I_+^{(1)}+I_+^{(3)},\tag{25}$$

where

$$I_+^{(1)}=\frac{\tilde{\chi}e^{-\xi t}}{2\sqrt{\pi}}\left[\int_0^\infty\frac{e^{-x^2}e^{ixy}}{\tilde{\xi}-ix}dx-e^{2ikL}\int_0^\infty\frac{e^{-x^2}e^{-ixy}}{\tilde{\xi}+ix}dx\right],\tag{26a}$$

$$I_+^{(3)} = -\frac{4\tilde{\chi}^3 e^{-\xi t}}{2\sqrt{\pi}} \times \left[\begin{array}{c} \int_0^\infty dx \frac{e^{-x^2} e^{ixy}(\tilde{\gamma}-2ix)}{(\tilde{\xi}-ix)^2(\tilde{\xi}^*-3ix)(\tilde{\gamma}_2-2ix)} \\ -e^{2ikL}\frac{\gamma}{\gamma_2}\int_0^\infty dx \frac{e^{-x^2}e^{-ixy}}{\tilde{\xi}+ix}\frac{1}{\tilde{\gamma}^2+(\tilde{\Delta}-x)^2} \end{array} \right], \tag{26b}$$

$y = kut$ and $\tilde{\beta} = \beta/ku$ for any variable β.

From Eq. (21) it is seen that the integral over Z can be considered to be a function of $L' = L - v_z t$. For atoms having negative v_z, one can change variables $[v_z \to -v_z, Z \to Z - v_z t]$ to express the integrand in Eq. (20) as a function of L' and use the iterative solutions of Eqs. (3) to show that

$$I_-(\tilde{\xi}) = -e^{2ikL}[I_+(\tilde{\xi}^*)]^*. \tag{27}$$

In this manner, one finds

$$I_- = I_-^{(1)} + I_-^{(3)}, \tag{28}$$

where

$$I_-^{(1)} = \frac{\tilde{\chi} e^{-\xi t}}{2\sqrt{\pi}}\left[\int_0^\infty \frac{e^{-x^2}e^{ixy}}{\tilde{\xi}-ix}dx - e^{2ikL}\int_0^\infty \frac{e^{-x^2}e^{-ixy}}{\tilde{\xi}+ix}dx\right] = I_+^{(1)}, \tag{29a}$$

$$I_-^{(3)} = -\frac{4\tilde{\chi}^3 e^{-\xi t}}{2\sqrt{\pi}} \times \left[\begin{array}{c} \frac{\gamma}{\gamma_2}\int_0^\infty dx \frac{e^{-x^2}e^{ixy}}{\tilde{\xi}-ix}\frac{1}{[\tilde{\gamma}^2+(\tilde{\Delta}+x)^2]} \\ -e^{2ikL}\int_0^\infty dx \frac{e^{-x^2}e^{-ixy}(\tilde{\gamma}+2ix)}{(\tilde{\xi}+ix)^2(\tilde{\xi}^*+3ix)(\tilde{\gamma}_2+2ix)} \end{array} \right]. \tag{29b}$$

Let us analyze the linear and third-order contributions separately, again neglecting contributions from the e^{2ikL} terms. The linear response is the same for the atoms moving in both directions. Explicitly, one finds

$$I^{(1)} = I_+^{(1)} + I_-^{(1)} = \frac{\tilde{\chi} e^{-\xi t}}{\sqrt{\pi}}\int_0^\infty \frac{e^{-x^2}e^{ixy}}{\tilde{\xi}-ix}dx. \tag{30}$$

In general, this integral must be evaluated numerically; however, for $y \gg 1$, it is possible to obtain an asymptotic expression. For $y \gg 1$, the major contribution to the integral comes from values of $x \ll 1$, unless a resonance denominator appears in the integrand. The integrand contains an energy denominator of the form $[\tilde{\gamma} - i(x+\tilde{\Delta})]$. For $\tilde{\Delta} > 0$, there is no resonance; on the other hand, for $\tilde{\Delta} < 0$, one can rewrite the integral as

$$\int_0^\infty \frac{e^{-x^2}e^{ixy}}{\tilde{\xi}-ix}dx = \int_{-\infty}^\infty \frac{e^{-x^2}e^{ixy}}{\tilde{\xi}-ix}dx - \int_{-\infty}^0 \frac{e^{-x^2}e^{ixy}}{\tilde{\xi}-ix}dx.$$

The first term has been evaluated as Eq. (10) and does not contribute for $y \gg 1$, while the second term no longer has a resonant denominator. Thus, regardless of the sign of $\tilde{\Delta}$, for $y \gg 1$ the major contribution to the integral comes from values of $x \ll 1$, and one can approximate the linear response as

$$I^{(1)} \sim \frac{\tilde{\chi} e^{-\xi t}}{\sqrt{\pi}}\int_0^\infty \frac{e^{ixy}}{\tilde{\xi}-ix}dx = \frac{i\tilde{\chi}}{\sqrt{\pi}}E_1(\tilde{\xi}y), \tag{31}$$

where E_1 is the exponential integral, $E_1(z) = \int_z^\infty (e^{-x}/x)dx$. For $|\tilde{\xi}| y \gg 1$,

$$I^{(1)} \sim \frac{i\tilde{\chi} e^{-\xi t}}{\sqrt{\pi}\xi t}. \tag{32}$$

The signal is emitted at the atomic resonance frequency and decays more rapidly than $e^{-\gamma t}$, but less rapidly than $e^{-(kut)^2/4}$.

We now turn to the third-order response. In the perturbation theory limit, $(2\gamma/\gamma_2)(\chi^2/\gamma^2) \ll 1$, the third-order response will always be less than the linear response *unless* there is a contribution from a resonant denominator. The presence of a resonance denominator allows the third-order response to be comparable with the linear response within a certain time window. It is easy to see from Eq. (26b) that no such resonance exists for the atoms having $v_z > 0$, if the e^{2ikL} term is neglected; thus, one can set

$$I_+^{(3)} \sim 0. \tag{33}$$

The first term of Eq. (29b) for $I_-^{(3)}$ contains a resonance denominator, but only for $\tilde{\Delta} < 0$, corresponding to atoms that have a velocity $v_z < 0$ that brings them into resonance with the field. Thus the only possibility for a significant contribution from the third-order terms in the perturbation theory

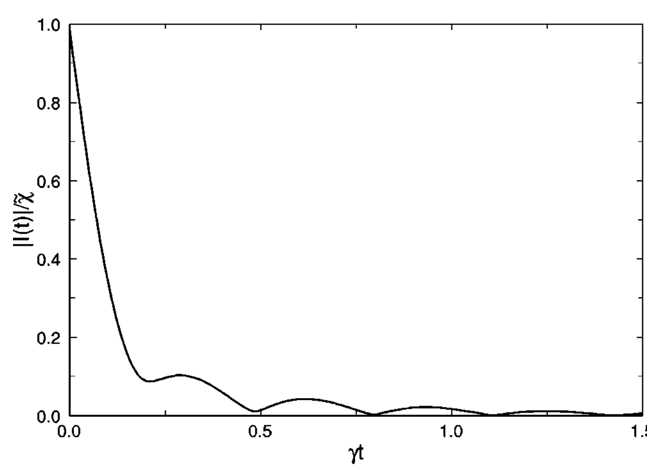

FIG. 2. Graph of the selective reflection, free-precession signal $|I(t)|$ as a function of γt for cw excitation when $\tilde{\Delta} = \Delta/ku = -1$ and $\tilde{\gamma} = \gamma/ku = 0.05$. Note that $y = kut = \gamma t/\tilde{\gamma} = 20\gamma t$.

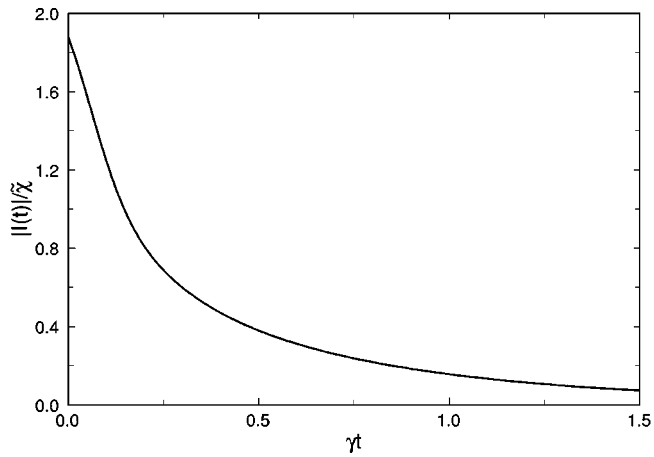

FIG. 3. Graph of the selective reflection, free-precession signal $|I(t)|$ as a function of γt for cw excitation when $\tilde{\Delta}=\Delta/ku=0$ and $\tilde{\gamma}=\gamma/ku=0.05$.

limit occurs for $\tilde{\Delta}<0$ and $y\gg 1$ (for $y\lesssim 1$, the linear response is dominant). In the limit $\tilde{\gamma}\ll 1$, $\tilde{\Delta}\approx -1$, and $y\gg 1$, it follows from Eqs. (29b), (32), and (33), that

$$I\sim \frac{\tilde{\chi}e^{-\gamma t}}{\sqrt{\pi}}\left[-\frac{e^{i\Delta t}}{\Delta t}+\frac{\pi}{2}\left(\frac{\chi}{\gamma}\right)^2\left(\frac{2\gamma}{\gamma_2}\right)e^{-\tilde{\Delta}^2}e^{-\gamma t}\right]. \quad (34)$$

The third-order contribution corresponds to radiation at the laser frequency so there can be beating at frequency Δ between the first- and third-order contributions. This feature is seen in Fig. 2, where $|I(t)|/\tilde{\chi}$, as given by Eqs. (19), (25), (26), (28), and (29), is plotted as a function of γt for $\tilde{\gamma}=0.05$, $(\chi/\gamma)^2(2\gamma/\gamma_2)=0.2$, $(2\gamma/\gamma_2)=1$, and $\tilde{\Delta}=-1$. The first- and third-order contributions are comparable in the range $0.5<\gamma t<1.5$, as is evidenced by the fact that the signal strength falls close to zero as it oscillates.

Similar plots are shown in Figs. 3 and 4 for $\tilde{\Delta}=0$ and $\tilde{\Delta}=1$, respectively. The third-order response is at most 20% of the linear response for these parameters and there is no oscillatory behavior. To a first approximation, $I(t)$ is given by Eq. (31). There is a rapid initial falloff of the signal in a time of order $(ku)^{-1}$ followed by a slower decay that varies as $e^{-\gamma t}/t$ for sufficiently long times. The signal strength at $t=0$ in linear response is given by

$$I(0)=\frac{\tilde{\chi}e^{\tilde{\xi}^2}}{2\sqrt{\pi}}[\pi\{1-\Phi(\tilde{\xi})\}+iE_1(\tilde{\xi}^2)]$$

and is largest for $\Delta=0$.

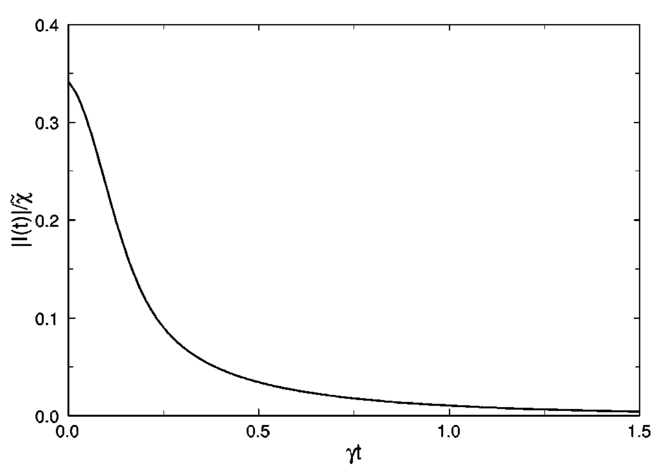

FIG. 4. Graph of the selective reflection, free-precession signal $|I(t)|$ as a function of γt for cw excitation when $\tilde{\Delta}=\Delta/ku=1$ and $\tilde{\gamma}=\gamma/ku=0.05$.

IV. DISCUSSION

It has been shown that the decay time for free precession (FP) in a Doppler-broadened vapor is of order γ^{-1} when the FP signal is viewed in selective reflection and the vapor is excited by either a short pulse or a weak cw field. This result contrasts with the phase-matched, coherent FP emission in the direction of the exciting field, which decays with a time constant of order $(ku)^{-1}\ll\gamma^{-1}$. Moreover, beating between the first- and third-order response is possible if $\tilde{\Delta}$ is of order -1. Of course, the FP signal in the forward direction is much larger in magnitude than that of selective reflection. The relative magnitude of the two fields is L/λ, since the entire vapor contributes to the forward signal but only a layer having length of order λ contributes to the coherent, selective reflection signal [14]. In general, there are contributions to the selective reflective signal resulting from the boundary conditions at both interfaces (although results were plotted neglecting contributions from the interface at $Z=L$). If the loss in the medium is negligible, there is a symmetry between these contributions that can be expressed as $I_-(\Delta)=-e^{2ikL}[I_+(-\Delta)]^*$. In this limit, the selective reflection signal is a symmetric function of Δ, if interference terms between the two contributions are neglected. There is also spontaneous emission emitted in the backwards direction. One can estimate that the ratio of spontaneous emission into the diffraction cone of the selective reflection signal is less than $(\mathcal{N}\lambda^2L)^{-1}(ku/\gamma_2)(L/a)^2$ times that of selective reflection [15]. Since $\mathcal{N}\lambda^2L(\gamma_2/ku)$ can take on a maximum value of order unity if superradiant effects are to be neglected, one must choose $a>L$ to have the selective reflection signal dominate. Alternatively, one could measure the selective reflection signal using heterodyne and phase-sensitive detection to eliminate the spontaneous-emission contribution altogether.

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[13] To have minimum reflection at the second boundary, one can model the dielectric for $Z>L$ as having $n_2\approx 1$. In that case, in the absence of the vapor, the reflected field amplitude is

$$E_r \sim \left[\frac{(n-1)}{(n+1)} - \frac{2n(n_2-1)e^{2ikL}}{(n+1)^2}\right] E_d .$$

For cw field excitation, and when the vapor can be characterized by an index of refraction n_1 (as would be appropriate for a vapor of stationary atoms in linear response), one can show that the polarization field of the vapor gives rise to an electric field that modifies this result such that the ''1'''s are replaced by n_1's. Thus, this approach is consistent with linear-response theory for a field geometry consisting of a dielectric having index of refraction n for $Z<0$, $n_1\approx 1$ for $0\leqslant Z\leqslant L$, and $n_2\approx n_1\approx 1$ for $Z>L$.
[14] The terms involving e^{2ikL} can be included if necessary, but the boundary condition at $Z=L$ should be treated more realistically. Reflections at the $Z=L$ boundary of the forward, *phase-matched* FP signal can swamp the non–phase-matched backward-emitted signal. If $\epsilon kL \lesssim 1$, all atoms in the vapor contribute to the reflected signal. One way to avoid the e^{2ikL} terms is to tilt the interface at $Z=L$ by an angle that results in a washing out of the phase factor at this boundary. For a related discussion, see S. Briaudeau, S. Saltiel, G. Nienhuis, D. Bloch, and M. Ducloy, Phys. Rev. A **57**, R3169 (1998).
[15] When superradiant effects are negligible, the ratio of *total* fluorescence to FP intensity is of order $(\mathcal{N}\lambda^2 L)^{-1}(ku/\gamma_2)>1$; restricting the fluorescence to the diffraction cone adds a factor $(\lambda/a)^2$, and going from FP to selective reflection adds a factor $(L/\lambda)^2$.