Recoil-induced resonances in nonlinear spectroscopy

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It is shown that, as a result of the atomic recoil associated with the absorption and emission of radiation, resonances can appear in nonlinear spectroscopy. The theory is illustrated by calculating the line shapes associated with nearly degenerate four-wave mixing and pump-probe spectroscopy when laser fields interact with an ensemble of two-level atoms. The recoil-induced resonances should be observable for atoms cooled below the Doppler limit of laser cooling, and the line shapes may provide a means for measuring the velocity distribution of these cooled atoms.

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I. INTRODUCTION

When an atom absorbs or emits radiation, there is a corresponding change in the atom's center-of-mass momentum. This "atomic recoil" or "photon recoil" is known to have interesting spectroscopic implications. In 1968, Kol'chenko, Rautian, and Sokolovskii [1] predicted that the recoil effect could lead to a splitting in saturated absorption line shapes. Experimental evidence for this effect was obtained by Hall, Bordé, and Uehara in 1976 [2]. In 1975, Hänsch and Schawlow suggested that the recoil effect could lead to cooling of an atomic vapor when atoms are irradiated by laser radiation tuned to the red side of an atomic resonance [3]. This method, referred to as Doppler cooling, was used to longitudinally cool an atomic beam in 1982 [4] and to cool an atomic vapor in three dimensions in 1985 [5]. More recent interest in the photon-recoil effect has been directed towards the limitations it puts on the minimum temperature achievable by laser cooling [6] and the role it plays in atomic interferometry [7].

In this paper, it is shown that the recoil effect can lead to new resonant structures in four-wave mixing and pump-probe spectroscopic line shapes. These recoil-induced resonances are distinct from the recoil splitting resonances observed in saturation spectroscopy. They are more closely related to the pressure-induced extra resonances first predicted by Bloembergen and co-workers [8]. To understand the origin of the recoil-induced resonances, it may prove helpful to review some features of the pressure-induced resonances.

As a specific example, consider four-wave mixing in an atomic vapor. Three laser beams having wave vectors $\mathbf{k}_1$, $\mathbf{k}_2$, and $\mathbf{k}_3$ are incident on an ensemble of two-level atoms. Beams 1 and 2 are counterpropagating and have frequency Ω, while beam 3 makes a small angle with beam 1 and has frequency $(\Omega+\delta)$ (see Fig. 1). A signal beam is generated in the direction $-\mathbf{k}_3$ having frequency $(\Omega-\delta)$. The intensity of the signal as a function of δ is monitored. Let the atom-field detuning $\Delta=\Omega-\omega$ (ω is the atomic transition frequency) be such that $|\Delta|$ is much greater than the Doppler width ku ($k=k_1$, u is the most probable atomic velocity) associated with the atomic transition. Moreover, let δ be sufficiently small to satisfy $|\delta|\le ku \ll |\Delta|$. In these limits, one can ask whether or not there is a resonant structure in the signal centered at $\delta=0$ and, if so, what is the width that characterizes the resonance.

When the system is "closed" in the sense that population of the two-level atoms is conserved, there is no resonance centered at $\delta=0$ in the absence of collisions, to lowest order in the applied fields. In the presence of dephasing collisions, a resonance having a width equal to the excited-state decay rate Γ appears [8]. The physical origin of this pressure-induced extra resonance has been discussed by several authors [8,9]. Even in the presence of collisions, no resonant structure having a width equal to some effective ground-state width is present.

In order to have a resonance characterized by the ground-state width, the system must be "open" in some manner. For example, if state 1 were not the true ground state but decayed to another level at rate $\gamma_t \ll \Gamma$ (and if there was some incoherent pump to repopulate level 1 as it decays), a resonance centered at $\delta=0$ having width γ_t would appear in the signal. Alternatively, if level 1 actually consisted of a number of degenerate magnetic sublevels, narrow (widths of order of some effective ground-

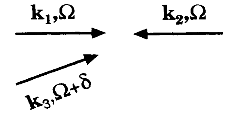

FIG. 1. The incident fields for four-wave mixing. Fields 1, 2, and 3 are the forward pump field, the backward pump field, and the probe field, respectively.

state width determined by optical pumping rates or an inverse transit time) resonances could also be produced for certain polarizations of the applied fields [10,11]. The appearance of these resonances is connected with the fact that magnetic-state orientation or alignment for the multilevel ground state need not be conserved [11]. The relation of these narrow resonances to laser cooling below the Doppler limit has also been noted [12].

One might also think that velocity-changing collisions can lead to a narrow resonance. In the presence of velocity-changing collisions, the population for each velocity subclass of atoms, $\rho_{11}(\mathbf{v})+\rho_{22}(\mathbf{v})$, is not conserved ($\mathbf{v}$ is the atomic velocity) if the collision rates or strengths differ for the ground and excited states. This condition does, in fact, lead to a narrow resonance for detunings $|\Delta| \leq ku$ when specific velocity subclasses are selected by the field [13]; however, for the case considered in which $|\Delta| \gg ku$, the signal is proportional to the velocity-integrated population, which *is* conserved. As a result, no narrow resonance appears.

We are ready to return to recoil-induced resonances, and pose the question, "For the same conditions outlined above, does a narrow resonance (characterized by some effective ground-state width) exist in the absence of collisions for an ensemble of 'closed' two-level atoms?" In considering the recoil effect, one quantizes the atomic center-of-mass motion. Density matrix elements are then written as $\rho_{ij}(\mathbf{p},\mathbf{p}')$, where $\mathbf{p}$ (or $\mathbf{p}'$) labels the atomic momentum. In the presence of recoil, one can show that the total population $\rho_{11}(\mathbf{p},\mathbf{p})+\rho_2(\mathbf{p},\mathbf{p})$ is not conserved; however, since, for $|\Delta| \gg ku$, the signal is proportional to the velocity-integrated population (which is conserved), one might conclude that no narrow resonance exists. This reasoning is not correct, owing to the fact that the signal under consideration depends on *off-diagonal* (in momentum space) elements such as $\rho_{11}(\mathbf{p}+\hbar\mathbf{k}_1,\mathbf{p}+\hbar\mathbf{k}_3)+\rho_{22}(\mathbf{p}+\hbar\mathbf{k}_1,\mathbf{p}+\hbar\mathbf{k}_3)$, rather than the (diagonal) atomic-state population. The velocity-integrated sum, $\rho_{11}(\mathbf{p}+\hbar\mathbf{k}_1,\mathbf{p}+\hbar\mathbf{k}_3)+\rho_{22}(\mathbf{p}+\hbar\mathbf{k}_1,\mathbf{p}+\hbar\mathbf{k}_3)$, is *not* conserved, leading to a narrow resonance. The amplitude of the narrow resonance turns out to be larger than that of a "background" signal for atomic ensembles that have been cooled below the Doppler limit of laser cooling [14]. The line shapes can actually serve as a probe of the atomic velocity distribution in these cases.

This paper is arranged as follows: In Sec. II the generalized master equations for two-level atoms interacting with classical fields is given as well as its third-order perturbation solution.. In Sec. III we analyze in detail the results as applied to four-wave mixing and pump-probe spectroscopy. Finally, a discussion of the results is given in Sec. IV.

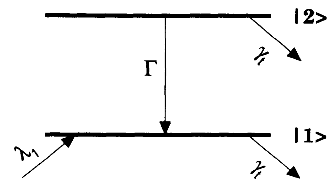

FIG. 2. Atomic level diagram. The ground state $|1\rangle$ is incoherently pumped at rate λ_1. Both the ground state $|1\rangle$ and the excited state $|2\rangle$ decay to the reservoir at rate γ_t, and state $|2\rangle$ decays to state $|1\rangle$ at rate Γ.

II. CALCULATION OF SIGNAL

In order to derive a general result which is applicable to both the four-wave mixing and pump-probe spectroscopy problems, we consider an ensemble of two-level atoms, each having the level scheme shown in Fig. 2, which interact with an electric field given by

$$\mathbf{E}=\sum_{\mu=1}^{N} \tfrac{1}{2}\mathbf{E}_\mu[\exp(i\mathbf{k}_\mu\cdot\mathbf{R}-i\Omega_\mu t)+\text{c.c.}] \ . \qquad (1)$$

The electric field consists of N plane-wave fields, having frequencies Ω_μ, propagation directions $\mathbf{k}_\mu$, and amplitudes $\mathbf{E}_\mu$ ($\mu=1,2,\ldots,N$). We expand the atomic density matrix in the coupled basis of internal states $|\alpha\rangle=|1\rangle,|2\rangle$ and external center-of-mass momentum states $|\mathbf{p}\rangle$ as

$$\hat{\rho}=\sum_{\alpha,\beta}\rho_{\alpha\beta}(\mathbf{p},\mathbf{p}')|\alpha,\mathbf{p}\rangle\langle\beta,\mathbf{p}'| \ . \qquad (2)$$

In terms of an interaction representation defined by

$$\rho_{\alpha\alpha}(\mathbf{p},\mathbf{p}')=\tilde{\rho}_{\alpha\alpha}(\mathbf{p},\mathbf{p}')e^{-[(p^2-p'^2)/2m]t} \ , \qquad (3)$$

$$\rho_{12}(\mathbf{p},\mathbf{p}')=\tilde{\rho}_{12}(\mathbf{p},\mathbf{p}')e^{i\omega t}e^{-i[(p^2-p'^2/2m)]t} \ , \qquad (4)$$

etc., we obtain the following generalized equations for $\dot{\tilde{\rho}}_{\alpha\beta}(\mathbf{p},\mathbf{p}')$:

$$\begin{aligned}\dot{\tilde{\rho}}_{12}(\mathbf{p},\mathbf{p}')=&\, i\sum_{\mu=1}^{N}\chi_\mu^*\exp\left[i\Delta_\mu t-i\frac{\mathbf{k}_\mu\cdot\mathbf{p}'}{m}t+i\omega_k t\right]\tilde{\rho}_{11}(\mathbf{p},\mathbf{p}'-\hbar\mathbf{k}_\mu)\\ &-i\sum_{\mu=1}^{N}\chi_\mu^*\exp\left[i\Delta_\mu t-i\frac{\mathbf{k}_\mu\cdot\mathbf{p}}{m}t-i\omega_k t\right]\tilde{\rho}_{22}(\mathbf{p}+\hbar\mathbf{k}_\mu,\mathbf{p}')-\gamma\tilde{\rho}_{12}(\mathbf{p},\mathbf{p}') \ ,\end{aligned} \qquad (5)$$

$$\begin{aligned}\dot{\tilde{\rho}}_{22}(\mathbf{p},\mathbf{p}')=&\, i\sum_{\mu=1}^{N}\chi_\mu^*\exp\left[i\Delta_\mu t-i\frac{\mathbf{k}_\mu\cdot\mathbf{p}'}{m}t+i\omega_k t\right]\tilde{\rho}_{21}(\mathbf{p},\mathbf{p}'-\hbar\mathbf{k}_\mu)\\ &-i\sum_{\mu=1}^{N}\chi_\mu\exp\left[-i\Delta_\mu t+i\frac{\mathbf{k}_\mu\cdot\mathbf{p}}{m}t-i\omega_k t\right]\tilde{\rho}_{12}(\mathbf{p}-\hbar\mathbf{k}_\mu,\mathbf{p}')-\gamma_2\tilde{\rho}_{22}(\mathbf{p},\mathbf{p}') \ ,\end{aligned} \qquad (6)$$

$$\dot{\tilde{\rho}}_{11}(\mathbf{p},\mathbf{p}')=i\sum_{\mu=1}^{N}\chi_\mu \exp\left[-i\Delta_\mu t+i\frac{\mathbf{k}_\mu\cdot\mathbf{p}'}{m}t+i\omega_k t\right]\tilde{\rho}_{12}(\mathbf{p},\mathbf{p}'+\hbar\mathbf{k}_\mu)$$
$$-i\sum_{\mu=1}^{N}\chi_\mu^* \exp\left[i\Delta_\mu t-i\frac{\mathbf{k}_\mu\cdot\mathbf{p}}{m}t-i\omega_k t\right]\tilde{\rho}_{21}(\mathbf{p}+\hbar\mathbf{k}_\mu,\mathbf{p}')$$
$$+\int\Gamma N(\mathbf{q})d\mathbf{q}\tilde{\rho}_{22}(\mathbf{p}+\hbar\mathbf{q},\mathbf{p}'+\hbar\mathbf{q})\exp\left[-i\frac{\mathbf{p}\cdot\mathbf{q}}{m}t+i\frac{\mathbf{p}'\cdot\mathbf{q}}{m}t\right]-\gamma_t\tilde{\rho}_{11}(\mathbf{p},\mathbf{p}')+\gamma_t W(\mathbf{p},\mathbf{p}')\ , \quad (7)$$

$$\tilde{\rho}_{21}(\mathbf{p},\mathbf{p}')=[\tilde{\rho}_{12}(\mathbf{p}',\mathbf{p})]^*\ . \quad (8)$$

The quantities appearing in these equations are defined as follows: The Rabi frequency χ_μ, detuning Δ_μ, and recoil frequency ω_k are given by

$$\chi_\mu=\frac{-\langle 2|\boldsymbol{\mu}|1\rangle\cdot\mathbf{E}_\mu}{2\hbar}\ ,$$
$$\Delta_\mu=\Omega_\mu-\omega\ ,$$

and

$$\omega_k=\frac{\hbar k^2}{2m}\ ,$$

respectively, where $\boldsymbol{\mu}$ is the dipole moment operator. The excited-state decay rate γ_2 and the coherence decay rate γ are given by

$$\gamma_2=\Gamma+\gamma_t\ ,$$
$$\gamma=\frac{\Gamma}{2}+\gamma_t\ , \quad (9)$$

respectively, where Γ is the rate of spontaneous decay from level 2 to level 1 and γ_t is some effective decay time for the atom as a whole, assumed to be much less than Γ. For example, γ_t^{-1} can correspond to a finite interaction time for the atoms in the fields resulting from atoms entering and leaving the interaction region [15]. There is no incoherent pumping of level 2, but level 1 is pumped at rate $\lambda_1=\gamma_t W(\mathbf{p},\mathbf{p}')$, such that the ground-state equilibrium distribution is equal to

$$\tilde{\rho}_{11}^{(0)}(\mathbf{p},\mathbf{p}')=W(\mathbf{p},\mathbf{p}')\ . \quad (10)$$

Finally, the distribution $N(\mathbf{q})$ is the normalized probability density for the emission of a photon having momentum $\hbar\mathbf{q}$. For example, if the atoms have $J_1=0$ and $J_2=1$ ground- and excited-state angular momenta, respectively, then effective "two-level" atoms can be achieved if all the fields have the same linear polarization. In that limit, $N(\mathbf{q})$ is given by

$$N(\mathbf{q})=\frac{3}{8\pi}\sin^2\theta\ , \quad (11)$$

where θ is the angle between $\mathbf{q}$ and the fields' polarization direction $\hat{\mathbf{z}}$. For this distribution $N(\mathbf{q})$, one has

$$\int q_\mu N(\mathbf{q})d\mathbf{q}=0,\quad \mu=x,y,z$$

and

$$\int q_x^2 N(\mathbf{q})d\mathbf{q}=\int q_y^2 N(\mathbf{q})d\mathbf{q}=\tfrac{2}{5}k^2,\quad \int q_z^2 N(\mathbf{q})d\mathbf{q}=\tfrac{1}{5}k^2\ .$$

We are interested in calculating the macroscopic polarization of the medium $P(\mathbf{R},t)$, which is given by

$$\mathbf{P}(\mathbf{R},t)=\langle 2|\boldsymbol{\mu}|1\rangle\rho_{12}(\mathbf{R},t)+\text{c.c.}\ ,$$

where $\rho_{12}(\mathbf{R},t)$ can be written as

$$\rho_{12}(\mathbf{R},t)=\frac{1}{(2\pi\hbar)^3}\int\int d\mathbf{p}\,d\mathbf{p}'\exp\left[i\frac{(\mathbf{p}-\mathbf{p}')\cdot\mathbf{R}}{\hbar}-i\frac{p^2-p'^2}{2m\hbar}t+i\omega t\right]\times\tilde{\rho}_{12}(\mathbf{p},\mathbf{p}')\ . \quad (12)$$

For compactness, we denote $\rho_{12}(\mathbf{R},t)$ simply by ρ_{12} below.

The third-order perturbation solution of Eqs. (5)–(8) is

$$\rho_{12}=-iN_0\sum_{\mu,\nu,\sigma=1}^{N}\chi_\mu^*\chi_\nu\chi_\sigma^*\exp(-i\mathbf{k}_s\cdot\mathbf{R}+i\Delta_s t+i\omega t)$$
$$\times\int W(\mathbf{p})d\mathbf{p}\left[\frac{1}{\gamma+i\left[\Delta_\mu-\frac{\mathbf{k}_\mu\cdot\mathbf{p}}{m}-\omega_k\right]}\right]\left[\frac{1}{\xi_s-i\frac{\mathbf{k}_s\cdot\mathbf{p}}{m}+i\omega_d}\frac{1}{\xi_1-i\frac{\mathbf{k}_{\mu\nu}\cdot\mathbf{p}}{m}-i\omega_1}\right.$$
$$\left.-\int\frac{\Gamma N(\mathbf{q})d\mathbf{q}}{\xi_2-i\frac{\mathbf{k}_{\mu\nu}\cdot\mathbf{p}}{m}}\frac{1}{\xi_s-i\frac{\mathbf{k}_s\cdot\mathbf{p}}{m}+i\omega_d'}\frac{1}{\xi_1-i\frac{\mathbf{k}_{\mu\nu}\cdot\mathbf{p}}{m}-i\omega_1'}\right]$$

$$+\frac{1}{\gamma-i\left[\Delta_\nu-\frac{k_\nu\cdot\mathbf{p}}{m}-\omega_k\right]}\left[\frac{1}{\xi_s-i\frac{\mathbf{k}_s\cdot\mathbf{p}}{m}+i\bar{\omega}_d}\frac{1}{\xi_1-i\frac{\mathbf{k}_{\mu\nu}\cdot\mathbf{p}}{m}-i\bar{\omega}_1}-\int\frac{\Gamma N(\mathbf{q})d\mathbf{q}}{\xi_2-i\frac{\mathbf{k}_{\mu\nu}\cdot\mathbf{p}}{m}}\frac{1}{\xi_s-i\frac{\mathbf{k}_s\cdot\mathbf{p}}{m}+i\omega_d'}\frac{1}{\xi_1-i\frac{\mathbf{k}_{\mu\nu}\cdot\mathbf{p}}{m}-i\omega_1'}\right]$$

$$+\frac{1}{\xi_s-i\frac{\mathbf{k}_s\cdot\mathbf{p}}{m}+i\omega_c}\frac{1}{\left[\gamma+i\left[\Delta_\mu-\frac{\mathbf{k}_\mu\cdot\mathbf{p}}{m}-\omega_k\right]\right]\left[\gamma-i\left[\Delta_\nu-\frac{\mathbf{k}_\nu\cdot\mathbf{p}}{m}-\omega_k\right]\right]}\Bigg], \quad (13)$$

where N_0 is the atom number density, and

$$\mathbf{k}_{\mu\nu}=\mathbf{k}_\mu-\mathbf{k}_\nu,\quad \mathbf{k}_s=\mathbf{k}_\mu+\mathbf{k}_\sigma-\mathbf{k}_\nu\ ,$$
$$\Delta_{\mu\nu}=\Delta_\mu-\Delta_\nu,\quad \Delta_s=\Delta_\mu+\Delta_\sigma-\Delta_\nu\ ,$$
$$\xi_s=\gamma+i\Delta_s,\quad \xi_1=\gamma_t+i\Delta_{\mu\nu},\quad \xi_2=\gamma_2+i\Delta_{\mu\nu}\ ,$$
$$\omega_1=2\omega_k-\frac{\hbar}{m}\mathbf{k}_\mu\cdot\mathbf{k}_\nu,\quad \bar{\omega}_1=-\omega_1,\quad \omega_1'=-\frac{\hbar}{m}\mathbf{k}_{\mu\nu}\cdot\mathbf{q}\ , \quad (14)$$
$$\omega_d=\omega'-\frac{\hbar}{m}k_s^2,\quad \bar{\omega}_d=\omega'-\frac{\hbar}{m}\mathbf{k}_s\cdot\mathbf{k}_\sigma\ ,$$
$$\omega_d'=\omega'-\frac{\hbar}{m}\mathbf{k}_s\cdot(\mathbf{k}_\mu+\mathbf{k}_\sigma-\mathbf{q}),\quad \omega'=\omega_k+\frac{\hbar}{m}\mathbf{k}_{\mu\nu}\cdot\mathbf{k}_{\sigma\nu}\ ,$$
$$\omega_c=\omega_k-\frac{\hbar}{m}\mathbf{k}_\sigma\cdot\mathbf{k}_\nu\ .$$

A term of order γ_t/Γ has been neglected in deriving (13). Note that the various ω and ξ in Eq. (14) are implicit functions of $\mathbf{k}_\mu$, $\mathbf{k}_\nu$, $\mathbf{k}_\sigma$, and $\mathbf{q}$, and the ω are all of order of the recoil frequency ω_k, which, for optical frequencies, is of order 100 kHz.

In deriving Eq. (13) we have chosen

$$W(\mathbf{p},\mathbf{p}')=\frac{N}{V}(2\pi\hbar)^3W(\mathbf{p})\delta(\mathbf{p}-\mathbf{p}')\ , \quad (15)$$

where N is the total number of atoms, V is the quantization volume, and $W(\mathbf{p})$ is some distribution function of $\mathbf{p}$. Equation (15) corresponds to a uniform atomic distribution in position space as

$$\rho(\mathbf{R})=\frac{N}{V}\ . \quad (16)$$

This choice of initial state is not restrictive provided that the atoms are not localized to distances less than an optical wavelength [16].

In order to simplify Eq. (13), we proceed as follows. First, terms involving ξ_s are expanded as

$$\frac{1}{\xi_s-i[(\mathbf{k}_s\cdot\mathbf{p})/m]+i\omega_d}=\frac{1}{\xi_s-i[(\mathbf{k}_s\cdot\mathbf{p})/m]}\left[1+\frac{-i\omega_d}{\xi_s-i[(\mathbf{k}_s\cdot\mathbf{p})/m]}\right], \quad (17)$$

assuming that $|\xi_s|\simeq\gamma\gg\omega_d$. Second, the relation $\gamma_2=\Gamma+\gamma_t$ is used to rewrite

$$\frac{\Gamma}{\xi_2-i[(\mathbf{k}_{\mu\nu}\cdot\mathbf{p})/m]}\frac{1}{\xi_1-i[(\mathbf{k}_{\mu\nu}\cdot\mathbf{p})/m]-i\omega_1'}=-\frac{1}{\xi_2-i[(\mathbf{k}_{\mu\nu}\cdot\mathbf{p})/m]}+\frac{1}{\xi_1-i[(\mathbf{k}_{\mu\nu}\cdot\mathbf{p})/m]-i\omega_1'}\left[1-\frac{i\omega_1'}{\xi_2-i[(\mathbf{k}_{\mu\nu}\cdot\mathbf{p})/m]}\right] \quad (18)$$

along with similar relations for other terms involving ξ_2. Third, it is assumed that the atoms are sufficiently cold,

$$ku\equiv\frac{kp_0}{m}\ll\gamma\ , \quad (19)$$

with p_0 being the most probable atomic momentum, to neglect $\mathbf{k}\cdot\mathbf{p}/m$ terms in comparison with γ or γ_2. Finally, we assume that

$$|\Delta|\geq\gamma\gg\omega_k\ , \quad (20)$$

so we can neglect ω_k in comparison with $|\Delta|$. With these modifications, Eq. (13) is reduced to

$$\rho_{12}=-iN_0\sum_{\mu,\nu,\sigma=1}^{N}\chi_\mu^*\chi_\nu\chi_\sigma^*\exp[-i\mathbf{k}_s\cdot\mathbf{R}+i(\omega+\Delta_s)t]\frac{1}{\xi_s}$$
$$\times\left\{\frac{\sqrt{\pi}}{|\mathbf{k}_{\mu\nu}|u}\frac{1}{\gamma+i\Delta_\mu}\left[I\left[\frac{i\xi_1+\omega_1}{|\mathbf{k}_{\mu\nu}|u}\right]-\int N(\mathbf{q})d\mathbf{q}\,I\left[\frac{i\xi_1+\omega_1'}{|\mathbf{k}_{\mu\nu}|u}\right]\right.\right.$$
$$\left.+\frac{-i\omega_d}{\xi_s}I\left[\frac{i\xi_1+\omega_1}{|\mathbf{k}_{\mu\nu}|u}\right]+\int N(\mathbf{q})d\mathbf{q}\left[\frac{i\omega_d'}{\xi_s}+\frac{i\omega_1'}{\xi_2}\right]I\left[\frac{i\xi_1+\omega_1'}{|\mathbf{k}_{\mu\nu}|u}\right]\right]$$
$$+\frac{\sqrt{\pi}}{|\mathbf{k}_{\mu\nu}|u}\frac{1}{\gamma-i\Delta_\nu}\left[I\left[\frac{i\xi_1+\bar{\omega}_1}{|\mathbf{k}_{\mu\nu}|u}\right]-\int N(\mathbf{q})d\mathbf{q}\,I\left[\frac{i\xi_1+\omega_1'}{|\mathbf{k}_{\mu\nu}|u}\right]+\frac{-i\bar{\omega}_d}{\xi_s}I\left[\frac{i\xi_1+\bar{\omega}_1}{|\mathbf{k}_{\mu\nu}|u}\right]\right.$$
$$\left.\left.+\int N(\mathbf{q})d\mathbf{q}\left[\frac{i\omega_d'}{\xi_s}+\frac{i\omega_1'}{\xi_2}\right]I\left[\frac{i\xi_1+\omega_1'}{|\mathbf{k}_{\mu\nu}|u}\right]\right]+\frac{2}{(\gamma+i\Delta_\mu)(\gamma-i\Delta_\nu)}\right\}, \tag{21}$$

where

$$I(z)=\frac{i}{\sqrt{\pi}}\int_{-\infty}^{\infty}\frac{W_1(p_0t)p_0dt}{z-t}, \tag{22}$$

and $W_1(p_\mu)$ is the atomic-momentum distribution function in one dimension [it is assumed that $W(\mathbf{p})=W_1(p_x)W_1(p_y)W_1(p_z)$]. If $W(\mathbf{p})$ is the Gaussian distribution

$$W(\mathbf{p})=\frac{1}{(\sqrt{\pi}p_0)^3}e^{-\mathbf{p}^2/p_0^2}, \tag{23}$$

then $I(z)$ becomes the well-known complex error function [17]

$$w(z)=\frac{i}{\pi}\int_{-\infty}^{\infty}\frac{e^{-t^2}}{z-t}dt,\quad \mathrm{Im}(z)>0. \tag{24}$$

The perturbation treatment is valid when saturation effects can be neglected. For the second-order corrections to the ground-state population to be small compared with unity, one requires that

$$\frac{\chi^2}{\gamma_t|\Delta|}\frac{\omega_k}{ku}<1, \tag{25}$$

where χ is the Rabi frequency corresponding to the pump field. By adiabatically eliminating $\tilde{\rho}_{12}$, $\tilde{\rho}_{21}$, and $\tilde{\rho}_{22}$ in Eqs. (5)–(8), we obtain an equation for $\tilde{\rho}_{11}(\mathbf{p},\mathbf{p}')$ which leads us to believe that condition (25) also guarantees that contributions of higher order in the applied fields can be neglected.

Finally, in what follows, it is assumed that the Doppler width is much greater than the recoil frequency, i.e.,

$$ku \gg \omega_k. \tag{26}$$

III. LINE-SHAPE ANALYSIS

We wish to analyze both four-wave mixing and pump-probe spectroscopic line shapes using Eq. (13). Note that Eq. (13) will yield the standard results if the photon recoil terms are neglected. In particular, all resonances characterized by width γ_t are canceled (the introduction of an overall decay rate γ_t for the atom does not in itself lead to resonances having width γ_t [10,13]). Inclusion of the recoil terms leads to qualitatively new results as we now demonstrate.

A. Four-wave mixing signal

The geometry for four-wave mixing is shown in Fig. 1. There are three incident fields ($N=3$). Fields 1 and 2 are counterpropagating "pumps" ($\mathbf{k}_1=-\mathbf{k}_2$) having frequency Ω and field 3 is a probe whose wave vector $\mathbf{k}_3$ is at a small angle θ relative to $\mathbf{k}_1$, and whose frequency is $\Omega+\delta$. The signal generated in the direction $-\mathbf{k}_3$ is calculated, corresponding to a density matrix element ρ_{12} which varies as $\exp[i\mathbf{k}_3\cdot\mathbf{R}+i(\Omega-\delta)t]$. Two terms contribute to the signal. The first, called the "backward-grating" term, corresponds to the forward pump scattering off the atomic grating produced by the probe and backward pump fields. In this case, the needed quantities in Eq. (13) are

$$\begin{aligned}&\mathbf{k}_\mu=\mathbf{k}_2=k(-\hat{\mathbf{x}}),\quad \mathbf{k}_\sigma=\mathbf{k}_1=k\hat{\mathbf{x}},\\&\mathbf{k}_\nu=\mathbf{k}_3=k(\hat{\mathbf{x}}+\theta\hat{\mathbf{y}}),\\&\Delta_\mu=\Delta_\sigma=\Delta,\quad \Delta_\nu=\Delta+\delta,\\&\omega_1=4\omega_k,\quad \bar{\omega}_1=-4\omega_k,\quad \omega_1'=\frac{2\hbar k}{m}q_x,\\&\omega_d=-\omega_k,\quad \bar{\omega}_d=3\omega_k,\quad \omega_d'=\omega_k-\frac{\hbar k}{m}(q_x+\theta q_y).\end{aligned} \tag{27}$$

Substituting (27) into (21), one finds that the backward-grating contribution to ρ_{12}, denoted by $\rho_{12}^{(b)}$, is given by

$$\rho_{12}^{(b)} \approx C\frac{\sqrt{\pi}}{2ku}\left\{\frac{1}{\gamma+i\Delta}\left[I\left[\frac{i\gamma_t+\delta+4\omega_k}{2ku}\right]-\int N(\mathbf{q})d\mathbf{q}\,I\left[\frac{i\gamma_t+\delta+(2\hbar k/m)q_x}{2ku}\right]\right]\right.$$
$$\left.+\frac{1}{\gamma-i(\Delta+\delta)}\left[I\left[\frac{i\gamma_t+\delta-4\omega_k}{2ku}\right]-\int N(\mathbf{q})d\mathbf{q}\,I\left[\frac{i\gamma_t+\delta+(2\hbar k/m)q_x}{2ku}\right]\right]\right\}+C\frac{2}{(\gamma+i\Delta)[\gamma-i(\Delta+\delta)]}$$
$$\approx C\sqrt{\pi}\frac{\omega_k}{(ku)^2}\frac{-i2\Delta}{\gamma^2+\Delta^2}I'(z_1)+C\frac{2}{\gamma^2+\Delta^2}\ , \tag{28}$$

where

$$C=-iN_0\chi_1^*\chi_2^*\chi_3\frac{\exp[i\mathbf{k}_3\cdot\mathbf{R}+i(\Omega-\delta)t]}{\gamma+i(\Delta-\delta)}\ ,$$

$$z_1=\frac{i\gamma_t+\delta}{2ku}\ ,$$

and $I'(z_1)$ is the first-order derivative of $I(z_1)$ with respect to z_1. Corrections to Eq. (28) are of order of ku/γ or ω_k/ku.

The second term is the "forward-grating" term, corresponding to the backward pump scattering off the grating formed by the probe and forward pump. In this case, the needed quantities in Eq. (13) are

$$\mathbf{k}_\mu=-\mathbf{k}_\sigma=k\hat{\mathbf{x}},\quad \mathbf{k}_\nu=k(\hat{\mathbf{x}}+\theta\hat{\mathbf{y}})\ ,$$
$$\omega_1=\omega_k\theta^2,\quad \bar{\omega}_1=-\omega_k\theta^2,\quad \omega_1'=\frac{\hbar k}{m}\theta q_y\ , \tag{29}$$
$$\omega_d=\bar{\omega}_d=-\omega_k,\quad \omega_d'=\omega_k-\frac{\hbar k}{m}(q_x+\theta q_y)\ .$$

Substituting (29) into (21), one obtains the forward-grating contribution, denoted by $\rho_{12}^{(f)}$, to be

$$\rho_{12}^{(f)} \approx C4\sqrt{\pi}\frac{\omega_k}{ku\theta}\frac{1}{\gamma^2+\Delta^2}\left[\frac{i\gamma}{\gamma+i\Delta}I(z_2)-\frac{i\Delta\theta}{2ku}I'(z_2)-\frac{2}{5}\frac{\omega_k\gamma}{(ku)^2}I''(z_2)\right]+C\frac{2}{\gamma^2+\Delta^2}\ , \tag{30}$$

where

$$z_2=\frac{i\gamma_t+\delta}{ku\theta}\ ,$$

and $I''(z_2)$ is the second-order derivative of $I(z_2)$ with respect to z_2. Corrections to Eq. (30) are of the order of ω_k/ku.

The four-wave mixing signal is proportional to $|\rho_{12}|^2$, with $\rho_{12}=\rho_{12}^{(b)}+\rho_{12}^{(f)}$. For both $\rho_{12}^{(b)}$ and $\rho_{12}^{(f)}$, the recoil-induced terms, proportional to ω_k, exhibit resonant structures at $\delta=0$. In the case of $\rho_{12}^{(b)}$, the width of this resonance is of order $2ku$ when $ku \gg \gamma_t$, and the dimensionless parameter that determines its amplitude relative to that of a background signal [last term in Eq. (28)] is given by [18]

$$\eta_1=\frac{|\Delta|\omega_k}{(ku)^2}\sim\frac{\hbar|\Delta|}{k_BT}\ , \tag{31}$$

where T is the temperature, and k_B is Boltzmann's constant. At room temperature and for $|\Delta|\simeq\Gamma$, $\eta_1 \ll 1$, so recoil effects can be neglected in considering $\rho_{12}^{(b)}$. Only when

$$k_BT \le \hbar|\Delta| \tag{32}$$

is satisfied, does the resonance become visible. For $|\Delta|\simeq\Gamma$, this condition can be recognized as that for sub-Doppler cooling [14]. It is interesting to note that, for $|\Delta|\gg\gamma$, $\rho_{12}^{(b)}\propto\Delta^{-2}$, in contrast to the pressure-induced resonances for which $\rho_{12}^{(b)}\propto\Delta^{-3}$.

In the case of $\rho_{12}^{(f)}$, the resonance, which is peaked at $\delta=0$, has a much narrower width, $\bar{\gamma}_t=\gamma_t * ku\theta$, which arises from the convolution of $[(i\gamma_t+\delta)/k\theta-v]^{-1}$ with the atomic velocity distribution having width of order u. For $|\Delta|\simeq\gamma$, the dimensionless parameter that determines the amplitude of this resonance relative to that of the background is

$$\eta_2=\begin{cases}\omega_k/ku\theta & \text{if } \theta<\dfrac{ku}{|\Delta|}\ ,\\ \omega_k|\Delta|/(ku)^2 & \text{if } \dfrac{ku}{|\Delta|}<\theta\ll 1\ .\end{cases} \tag{33}$$

The amplitude of this recoil-induced structure of $\rho_{12}^{(f)}$ relative to that of $\rho_{12}^{(b)}$ is determined by η_2/η_1.

The quantity $|\rho_{12}|$ is plotted as a function of δ in Fig. 3, with $W(\mathbf{p})$ taken to be a Gaussian [Eq. (23)]. In this case $I(z)$ is replaced by the complex error function $w(z)$, whose first- and second-order derivatives are given by [17]

$$w'(z)=\frac{2i}{\sqrt{\pi}}-2zw(z)\ ,$$
$$w''(z)=-\frac{4iz}{\sqrt{\pi}}-2(1-2z^2)w(z)\ .$$

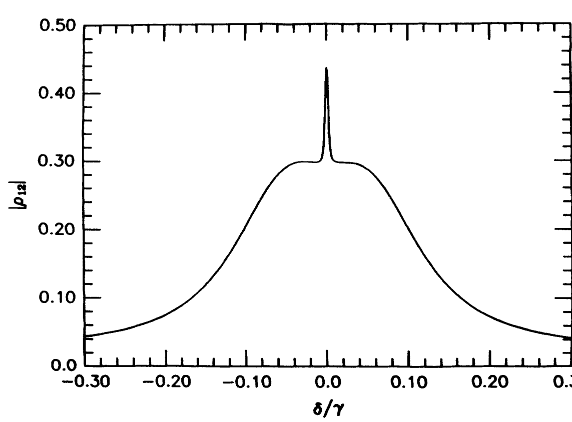

FIG. 3. Density matrix element $|\rho_{12}|$ as a function of dimensionless pump-probe detuning δ/γ for $\Delta/\gamma=-5.0$, $\theta=0.05$, $ku/\gamma=0.05$, $\omega_k/\gamma=0.005$, $\gamma_t/\gamma=1.0\times10^{-4}$. The ratio γ_t/γ is chosen sufficiently small in this and future examples to ensure that the line shape does not depend on γ_t/γ.

As one can see, the line shape consists of two peaks superimposed on each other. The broader one having width of order $2ku$ comes from $\rho_{12}^{(b)}$, while the narrower one whose width is of order $ku\theta$ represents the contribution of $\rho_{12}^{(f)}$.

Equations (28) and (30) are derived from Eq. (21) which in turn is derived assuming that the atoms are cold, $ku \ll \gamma$. In the case of thermal atoms, where $ku \gg \gamma, \chi, |\Delta|$, etc., one must return to the general expression Eq. (13). In this limit, the integral in Eq. (13) can be carried out using contour integration. The backward-grating term is zero after integration because all the poles of the denominators lie in the same half plane. For $ku\theta \ll \gamma$, the forward-grating term becomes

$$\rho_{12} = i2\sqrt{\pi}N_0 \frac{\chi_1^*\chi_2^*\chi_3}{ku} \frac{\exp[i\mathbf{k}_3\cdot\mathbf{R}+i(\Omega-\delta)t]}{2\gamma+i(2\Delta-\delta)} \times \left[\frac{2}{\gamma_2 - i\delta} + \frac{2i\omega_k}{2\gamma+i(2\Delta-\delta)} \times \frac{\sqrt{\pi}}{ku\theta} w\left[\frac{i\gamma_t+\delta}{ku\theta}\right]\right]. \quad (34)$$

Assuming that $ku\theta \gg \gamma_t$, the resonance having width $ku\theta$ is detectable provided that θ is sufficiently small to satisfy

$$\frac{\omega_k}{ku\theta} > 1 , \quad (35)$$

or

$$\theta < \frac{\omega_k}{ku} \sim 10^{-4} .$$

This very small value of θ has probably prevented any fortuitous discovery of this resonance.

B. Pump-probe spectroscopy

From the general solution Eq. (13), one can also obtain the density matrix element needed for calculating the probe absorption coefficient in the presence of a pump field. Again it is assumed that both probe and pump fields are sufficiently weak to ensure that inequality (25) is satisfied and perturbation theory is valid, and that the atoms are sufficiently cold to satisfy condition (19). In this case, only two fields are present ($N=2$), the pump field 1 and the probe field 2, with frequencies $\Omega_1=\Omega$ and $\Omega_2=\Omega+\delta$, and propagation vectors $\mathbf{k}_1$ and $\mathbf{k}_2$, respectively. The probe absorption is related to the imaginary part of the component of ρ_{12} that varies as $\exp[-i\mathbf{k}\cdot\mathbf{R}+i(\Omega+\delta)t]$. To third order in the applied fields this contribution varies as $|\chi|^2\chi'^*$, where χ and χ' are pump and probe Rabi frequencies, respectively. We neglect the linear contribution to the probe absorption, assuming that the nonlinear component can be isolated using modulation techniques.

The appropriate third-order contribution to ρ_{12} can be viewed as arising from the following perturbation channels:

(i) $\rho_{11}^{(0)} \xrightarrow{\chi^*} \rho_{12}^{(1)} \xrightarrow{\chi} \rho_{22}^{(2)} \xrightarrow{\chi'^*} \rho_{12}^{(3)}$,

(ii) $\rho_{11}^{(0)} \xrightarrow{\chi} \rho_{21}^{(1)} \xrightarrow{\chi^*} \rho_{22}^{(2)} \xrightarrow{\chi'^*} \rho_{12}^{(3)}$,

(iii) $\rho_{11}^{(0)} \xrightarrow{\chi'^*} \rho_{12}^{(1)} \xrightarrow{\chi} \rho_{22}^{(2)} \xrightarrow{\chi^*} \rho_{12}^{(3)}$,

(iv) $\rho_{11}^{(0)} \xrightarrow{\chi} \rho_{21}^{(1)} \xrightarrow{\chi'^*} \rho_{22}^{(2)} \xrightarrow{\chi^*} \rho_{12}^{(3)}$.

The first two channels are related to population modification by the pump field. These channels correspond to setting $\chi_\mu=\chi_\nu=\chi$ and $\chi_\sigma=\chi'$ in Eq. (13). Channels (i) and (ii) do not lead to any resonance at $\delta=0$, but they can modify the amplitude of the "background" signal. Channels (iii) and (iv) are related to population gratings created by the pump and probe. These channels correspond to setting $\chi_\mu=\chi'$ and $\chi_\nu=\chi_\sigma=\chi$ in Eq. (13). If we let $\rho_{12}=\tilde{\rho}_{12}\exp[-i\mathbf{k}\cdot\mathbf{R}+i(\Omega+\delta)t]$, then the general expression of $\tilde{\rho}_{12}$ for arbitrary directions $\mathbf{k}_1$ and $\mathbf{k}_2$ of the pump and probe fields is given by

$$\begin{aligned}\tilde{\rho}_{12} = -\frac{i\chi'^*N_0}{\gamma+i(\Delta+\delta)} \Bigg\{ & \frac{\sqrt{\pi}}{|\mathbf{k}_{12}|u}\frac{|\chi|^2}{\gamma+i(\Delta+\delta)} \Bigg[I\left[\frac{-\delta+i\gamma_t+\omega_1}{|\mathbf{k}_{12}|u}\right] - \int N(\mathbf{q})d\mathbf{q}\, I\left[\frac{-\delta+i\gamma_t+\omega_1'}{|\mathbf{k}_{12}|u}\right] \\ & + \frac{-i\omega_d}{\gamma+i(\Delta+\delta)} I\left[\frac{-\delta+i\gamma_t+\omega_1)}{|\mathbf{k}_{12}|u}\right] \\ & + \int N(\mathbf{q})d\mathbf{q}\frac{i\omega_d'}{\gamma+i(\Delta+\delta)} I\left[\frac{-\delta+i\gamma_t+\omega_1'}{|\mathbf{k}_{12}|u}\right]\Bigg] \\ & + \frac{\sqrt{\pi}}{|\mathbf{k}_{12}|u}\frac{|\chi|^2}{\gamma-i\Delta}\Bigg[I\left[\frac{-\delta+i\gamma_t+\bar{\omega}_1}{|\mathbf{k}_{12}|u}\right] - \int N(\mathbf{q})d\mathbf{q}\, I\left[\frac{-\delta+i\gamma_t+\omega_1'}{|\mathbf{k}_{12}|u}\right] \\ & + \frac{-i\bar{\omega}_d}{\gamma+i(\Delta+\delta)} I\left[\frac{-\delta+i\gamma_t+\bar{\omega}_1}{|\mathbf{k}_{12}|u}\right] \\ & + \int N(\mathbf{q})d\mathbf{q}\frac{i\omega_d'}{\gamma+i(\Delta+\delta)} I\left[\frac{-\delta+i\gamma_t+\omega_1'}{|\mathbf{k}_{12}|u}\right]\Bigg] \\ & + \frac{2\gamma|\chi|^2}{\gamma^2+\Delta^2}\left[\frac{2}{\gamma_2} - \frac{1}{\gamma+i(\Delta+\delta)}\frac{i[(\hbar\mathbf{k}_1\cdot\mathbf{k}_2)/m]}{\gamma_t}\right] + \frac{2|\chi|^2}{\gamma^2+\Delta^2+\delta\Delta+i\delta\gamma}\Bigg\} , \end{aligned} \quad (36)$$

where

$$\omega_1 = 2\omega_k - \frac{\hbar}{m}\mathbf{k}_1\cdot\mathbf{k}_2, \quad \bar{\omega}_1 = -\omega_1, \quad \omega_1' = \frac{\hbar}{m}\mathbf{k}_{12}\cdot\mathbf{q} ,$$
$$\omega_d = -\omega_k, \quad \bar{\omega}_d = \omega_k - \frac{\hbar}{m}\mathbf{k}_1\cdot\mathbf{k}_2 , \tag{37}$$
$$\omega_d' = \omega_k - \frac{\hbar}{m}\mathbf{k}_2\cdot(\mathbf{k}_1+\mathbf{k}_2-\mathbf{q}) .$$

When pump and probe fields are counterpropagating,

$$\mathbf{k}_1 = -\mathbf{k}_2 = k\hat{\mathbf{x}} ,$$

we have the simplified result

$$\tilde{\rho}_{12} = \frac{i\chi'^* N_0}{\gamma + i(\Delta+\delta)}\left[\frac{i\sqrt{\pi}\omega_k}{(ku)^2}\frac{|\chi|^2(2\Delta+\delta)}{\gamma^2+\Delta^2+\delta\Delta+i\delta\gamma}I'(z) - \frac{4\gamma|\chi|^2}{\gamma^2+\Delta^2}\left[\frac{1}{\gamma_2}+\frac{i}{\gamma+i(\Delta+\delta)}\frac{\omega_k}{\gamma_t}\right] - \frac{2|\chi|^2}{\gamma^2+\Delta^2+\delta\Delta+i\delta\gamma}\right] , \tag{38}$$

where

$$z = \frac{-\delta + i\gamma_t}{2ku} .$$

In the limit

$$|\Delta| \gg \gamma , \quad ku \gg \gamma_t , \quad \eta_1 \gg 1 , \quad \delta \approx 0 , \tag{39}$$

where η_1 is given by Eq. (31), the imaginary part of $\tilde{\rho}_{12}$ reduces to

$$\mathrm{Im}(\tilde{\rho}_{12}) = 2\pi N_0 \frac{|\chi|^2\chi'^*}{\Delta^2}\frac{\omega_k}{(ku)^2}p_0^2 W_1'\left[-\frac{p_0\delta}{2ku}\right] - 8N_0\frac{\gamma^2|\chi|^2\chi'^*}{\Delta^5}\frac{\omega_k}{\gamma_t} , \tag{40}$$

where $W_1(p)$ is the one-dimensional momentum distribution function specified above, and $W_1'(p)$ is its first-order derivative with respect to p. In Fig. 4, we display the absorption coefficient $\mathrm{Im}(\tilde{\rho}_{12})$ as a function of δ for the Gaussian distribution (23) of $W(\mathbf{p})$. It has a dispersion-like line shape centered around $\delta=0$, with the separation between the two peaks of order ku. The line shape is a direct measure of the derivative of the velocity distribution of the atoms. In contrast to the pressure-induced probe absorption signal whose dispersive part varies as Δ^{-3}, the dispersive part of the recoil-induced probe absorption is a symmetric function of Δ, varying as Δ^{-2}.

Equation (36) can also be used to calculate the probe absorption in the presence of a standing-wave pump field. As shown in Fig. 5, the standing-wave pump field can be regarded as two counterpropagating waves having wave vectors $\mathbf{k}_f = k\hat{\mathbf{x}}$ and $\mathbf{k}_b = -\mathbf{k}_f$, the probe-field propagation direction $\mathbf{k}_p$ makes an angle θ with $\mathbf{k}_f$. To second order in pump-field strength, the component of $\tilde{\rho}_{12}$ needed to calculate the probe absorption can be obtained by letting $\mathbf{k}_2 = \mathbf{k}_p$ in Eq. (36) and summing the results with $\mathbf{k}_1$ equal to $\mathbf{k}_f$ and $\mathbf{k}_b$, respectively. For $|\Delta| \gg \delta$, $\eta_1 \gg 1$, one obtains the following expression for $\tilde{\rho}_{12}$:

$$\tilde{\rho}_{12} = -2\sqrt{\pi}N_0\frac{\chi'^*|\chi|^2}{(\gamma+i\Delta)(\gamma^2+\Delta^2)}\frac{\omega_k}{2\sin(\theta/2)ku} \times\left[\frac{2\gamma}{\gamma+i\Delta}I(z_f) + \frac{2\sin(\theta/2)\Delta}{ku}[I'(z_f)+I'(z_b)] - \frac{4}{5}\frac{i\omega_k\gamma}{(ku)^2}I''(z_f)\right] , \tag{41}$$

where

$$z_f = \frac{-\delta+i\gamma_t}{2\sin(\theta/2)ku} , \quad z_b = \frac{-\delta+i\gamma_t}{2\cos(\theta/2)ku} . \tag{42}$$

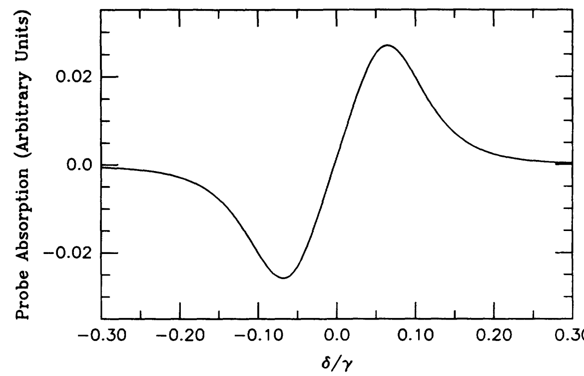

FIG. 4. Probe absorption line shape as a function of dimensionless pump-probe detuning δ/γ for the case of a counterpropagating pump field and $\Delta/\gamma = -20.0$, $\chi/\gamma = 1.0$, $ku/\gamma = 0.05$, $\omega_k/\gamma = 0.05$, $\gamma_t/\gamma = 0.005$.

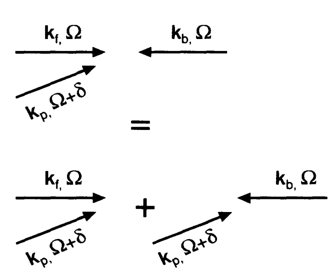

FIG. 5. Geometry of pump-probe absorption in the presence of a standing-wave pump field. To second order in the pump field, interference effects arising from the two traveling-wave components of the pump field do not contribute to the probe absorption.

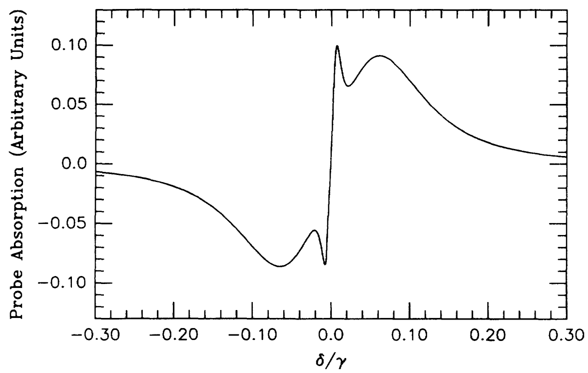

FIG. 6. Probe absorption line shape as a function of dimensionless pump-probe detuning δ/γ for the case of a standing-wave pump field and $\Delta/\gamma=-20.0$, $\chi/\gamma=0.25$, $\theta=0.2$, $ku/\gamma=0.05$, $\omega_k/\gamma=0.005$, $\gamma_t/\gamma=0.0005$.

It is interesting to note that the population modification term in the presence of a single pump field [i.e., the term varying as ω_k/γ_t in Eq. (38)] vanishes in the case of a standing-wave pump field. The line shape of the probe absorption coefficient $\mathrm{Im}(\tilde{\rho}_{12})$ as a function of δ is shown in Fig. 6 for a Gaussian distribution (23) of $W(\mathbf{p})$. It is composed of two dispersionlike curves centered around $\delta=0$, resulting from the existence of the two counterpropagating traveling waves comprising the standing-wave pump field.

The above calculation also suggests that it is possible to obtain gain for the probe field under certain conditions. For example, in the case of counterpropagating pump and probe fields, the recoil-induced probe gain [see Eq. (40)] is of order

$$N_0\frac{\chi'\gamma}{\Delta^2}\left[\frac{\chi^2}{k^2u^2}\frac{\omega_k}{\gamma}\right],$$

when $|\Delta|\gg\gamma$, while the linear loss is of order

$$N_0\frac{\chi'\gamma}{\Delta^2}$$

when $|\Delta|\gg\gamma$. Therefore the condition for probe gain is

$$\frac{\chi^2}{k^2u^2}\frac{\omega_k}{\gamma}>1,$$

or

$$\chi^2>\gamma\frac{k_BT}{\hbar}. \tag{43}$$

For sub-Doppler laser cooling one finds $k_BT\simeq\hbar\chi^2/|\Delta|$, provided $\chi^2/|\Delta|\geq\omega_k$. For this temperature range, Eq. (43) is satisfied when $|\Delta|\gg\gamma$ [16].

IV. DISCUSSION

We have seen that, in certain limits, the photon recoil effect gives rise to an additional type of resonance in nonlinear spectroscopy. Its existence does not depend on whether the system is open ($\gamma_2-\Gamma\neq\gamma_t$) or closed ($\gamma_2-\Gamma=\gamma_t\ll\Gamma$), a condition that is critical in other cases for the existence of resonance characterized by a ground-state decay rate.

As indicated in the Introduction, the appearance of additional resonances with the inclusion of recoil terms is due to the fact that the signal under consideration depends on the off-diagonal elements of $\rho_{\alpha\alpha}(\mathbf{p},\mathbf{p}')$ in momentum space, and the velocity-integrated sum of $\tilde{\rho}_{11}(\mathbf{p},\mathbf{p}+\hbar\mathbf{k}_{\mu\nu})+\tilde{\rho}_{22}(\mathbf{p},\mathbf{p}+\hbar\mathbf{k}_{\mu\nu})$, for $\mathbf{k}_{\mu\nu}\neq0$, is not conserved. This point is illustrated when one writes down the equation of rate of change for this sum as follows:

$$\begin{aligned}\frac{d}{dt}\int[\tilde{\rho}_{11}(\mathbf{p},\mathbf{p}+\hbar\mathbf{k}_{\mu\nu})+\tilde{\rho}_{22}(\mathbf{p},\mathbf{p}+\hbar\mathbf{k}_{\mu\nu})]d\mathbf{p}=\int&\left[\frac{\chi_\mu^*\chi_\nu}{\gamma+i\Delta_\mu}(1-e^{-i\omega_1t})+\frac{\chi_\mu^*\chi_\nu}{\gamma-i\Delta_\nu}(1-e^{-i\bar{\omega}_1t})\right.\\&\left.-\frac{\Gamma\chi_\mu^*\chi_\nu}{(\gamma+i\Delta_\mu)(\gamma-i\Delta_\nu)}\left[1-\int N(\mathbf{q})e^{-i\omega_1't}d\mathbf{q}\right]\right]\\&\times\exp\left[i\Delta_{\mu\nu}t-i\frac{\mathbf{k}_{\mu\nu}\cdot\mathbf{p}}{m}t\right]\tilde{\rho}_{11}^{(0)}(\mathbf{p},\mathbf{p})d\mathbf{p}.\end{aligned} \tag{44}$$

Clearly, when one neglects all the photon recoil terms involving ω_1, $\bar{\omega}_1$, and ω_1', the rate of change is zero and the velocity-integrated sum of these off-diagonal elements is conserved. The correction terms to the above equation come from expansions of the exponential functions containing ω's. This results in the various terms in the final signal that contain ω_1, $\bar{\omega}_1$, and ω_1', which is partially responsible for the additional recoil-related structures.

The differences between ω_1, $\bar{\omega}_1$, and ω_1' can be explained by an argument based on energy conservation conditions for certain stages of various processes that contribute to the signal. The resonance conditions are most easily seen in an amplitude picture [19]. For example, amplitudes leading to the absorption of two pump photons μ and σ, and emission of a ν photon and a signal photon s, are shown in Figs. 7(a) and 7(b). The signal field has frequency Ω_s given by

$$\Omega_s=\Omega_\mu+\Omega_\sigma-\Omega_\nu. \tag{45}$$

We are interested only in the coherent, phase-matched signal, having propagation vector

$$\mathbf{k}_s = \mathbf{k}_\mu + \mathbf{k}_\sigma - \mathbf{k}_\nu \ . \tag{46}$$

The atomic-momentum changes associated with each step of the amplitude diagrams are also shown in Fig. 7. The overall momentum changes of the atom are zero for both processes (a) and (b), as is evident from (46). However, at the intermediate steps [absorption of a photon from field μ and emission of a photon into field ν in (a), and absorption of photon from field σ and emission of a photon into field s in (b)], the atomic-momentum changes differ. The resonance conditions for these intermediate steps can be written as

$$\text{(a)}\quad \frac{\mathbf{p}^2}{2m} + \hbar\Omega_\mu = \frac{(\mathbf{p}+\hbar\mathbf{k}_\mu - \hbar\mathbf{k}_\nu)^2}{2m} + \hbar\Omega_\nu \ ,$$

or

$$\Delta_{\mu\nu} - \frac{\mathbf{k}_{\mu\nu}\cdot\mathbf{p}}{m} - \omega_1 = 0 \ , \tag{47}$$

and

$$\text{(b)}\quad \frac{\mathbf{p}^2}{2m} + \hbar\Omega_\sigma = \frac{(\mathbf{p}+\hbar\mathbf{k}_\sigma - \hbar\mathbf{k}_s)^2}{2m} + \hbar\Omega_s \ ,$$

or

$$\Delta_{\mu\nu} - \frac{\mathbf{k}_{\mu\nu}\cdot\mathbf{p}}{m} - \overline{\omega}_1 = 0 \ , \tag{48}$$

where Eqs. (45) and (46) have been used.

Additional amplitude diagrams can involve the emission of a $\mathbf{k}_s$ photon along with the emission of one or more spontaneous photons. A detailed analysis of these

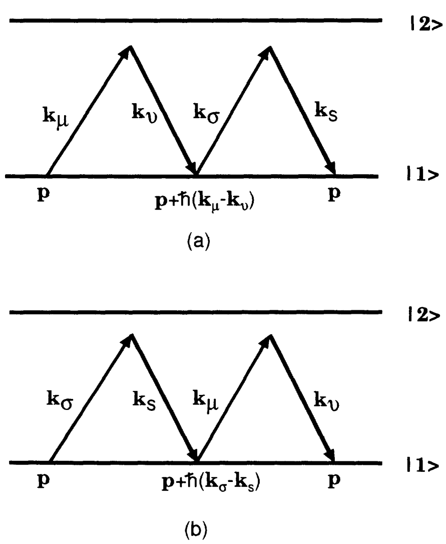

FIG. 7. Diagrams in the amplitude picture showing two distinct four-photon processes that contribute to the signal. Propagation vectors $\mathbf{k}_\mu$, $\mathbf{k}_\nu$, and $\mathbf{k}_\sigma$ are those of the incident fields while $\mathbf{k}_s$ is that of the generated signal. The overall momentum changes of the atom are zero for both cases; however, at the intermediate stage following the initial Raleigh-type processes in (a) and (b), the atomic momenta differ.

diagrams is deferred to a future planned publication. What is relevant to the present analysis is that the signal comes from the interference of processes involving spontaneous decays. For example, there can be an interference between two Rayleigh processes which are the first steps of more elaborate diagrams: (i) absorption of a photon from field μ and emission of a spontaneous photon of momentum $\hbar\mathbf{q}$, and (ii) absorption of a photon from field ν and emission of the same spontaneous photon of momentum $\hbar\mathbf{q}$. The resonance conditions for these processes are

$$\text{(i)}\quad \frac{\mathbf{p}^2}{2m} + \hbar\Omega_\mu = \frac{(\mathbf{p}+\hbar\mathbf{k}_\mu - \hbar\mathbf{q})^2}{2m} + \hbar\omega \ ,$$

$$\text{(ii)}\quad \frac{\mathbf{p}^2}{2m} + \hbar\Omega_\nu = \frac{(\mathbf{p}+\hbar\mathbf{k}_\nu - \hbar\mathbf{q})^2}{2m} + \hbar\omega \ .$$

Since the contribution to the signal comes from the interference of these two processes, both conditions must be satisfied simultaneously, leading to a resonance condition

$$\Delta_{\mu\nu} - \frac{\mathbf{k}_{\mu\nu}\cdot\mathbf{p}}{m} - \omega_1' = 0 \ . \tag{49}$$

Equations (47)–(49) coincide with the resonance conditions for the denominators involving ω_1, $\overline{\omega}_1$, and ω_1' in Eq. (13) [20].

The terms involving ω_d also emerge from an amplitude approach, but cannot be explained in terms of simple Raleigh processes. They arise as a result of different weighting factors for the various Raleigh processes. In other words, the signal actually depends on a weighted sum of the $\tilde{\rho}_{11}(\mathbf{p},\mathbf{p}+\hbar\mathbf{k}_{\mu\nu})$ and $\tilde{\rho}_{22}(\mathbf{p},\mathbf{p}+\hbar\mathbf{k}_{\mu\nu})$. The differences between the weighting factors are of order ω_d/γ compared with the magnitude of the factors themselves. As long as $|\mathbf{k}_{\mu\nu}|$ is of order k, the fact that the velocity-integrated sum of the off-diagonal terms $\tilde{\rho}_{11}(\mathbf{p},\mathbf{p}+\hbar\mathbf{k}_{\mu\nu})+\tilde{\rho}_{22}(\mathbf{p},\mathbf{p}+\hbar\mathbf{k}_{\mu\nu})$ is not conserved determines most of the contribution to the recoil-related effects, and the differences in various weighting factors can be neglected. But as $|\mathbf{k}_{\mu\nu}|\to 0$, the contribution to the signal arising from off diagonality of $\tilde{\rho}_{11}+\tilde{\rho}_{22}$ becomes comparable to that arising from the differences in the various weighting factors. As a result these differences must be maintained, leading to the ω_d terms.

The Δ^{-2} dependence of the recoil-induced resonance in pump-probe spectroscopy [see Eq. (40)] is found also in Raman spectroscopy for stationary atoms without the inclusion of recoil effects [21]. We wish to demonstrate here that it is possible to establish some link between these two cases. A typical probe gain for a Raman process between levels a and b can be written as [22]

$$g = \frac{N_0(P_a - P_b)}{\Delta^2} \frac{\gamma_t |\chi|^2 \chi'^*}{\gamma_t^2 + D_{ab}^2} \ , \tag{50}$$

for $|\Delta| \gg \gamma, \chi, \chi', |D_{ab}|$, etc., where Δ is the pump (or probe) detuning from some intermediate level, P_a and P_b are the normalized populations in levels a and b ($P_a = N_a/N_0$, $P_b = N_b/N_0$), γ_t is the common decay rate for levels a and b, and D_{ab} is the detuning from resonance for the Raman process. If we return to the case of two-level atoms with quantized center-of-mass momentum interacting with counterpropagating pump and probe fields, a corresponds to the atom in the ground state with momentum p, and b to the atom in the ground state with momentum $p+2\hbar k$, while D_{ab} is given by

$$D_{ab}=\delta+\frac{2kp}{m}+4\omega_k \; .$$

The overall Raman gain can be obtained by integrating over p as follows:

$$G=\frac{N_0}{\Delta^2}|\chi|^2\chi'^*\gamma_t\int dp\frac{W_1(p)-W_1(p+2\hbar k)}{\gamma_t^2+(\delta+2kp/m+4\omega_k)^2}$$

$$=-\frac{N_0}{\Delta^2}|\chi|^2\chi'^*2\hbar k\int dp\frac{\gamma_t W_1'(p)}{\gamma_t^2+(\delta+2kp/m)^2} \; . \tag{51}$$

In the limit of Eq. (39), this gives

$$G=-2\pi N_0\frac{|\chi|^2\chi'^*}{\Delta^2}\frac{\omega_k}{(ku)^2}p_0^2W_1'\left[-\frac{p_0\delta}{2ku}\right] ,$$

which is identical to the nonconstant contribution in Eq. (40) except the sign (simply arising from the difference between the definitions of "gain" and "absorption"). This shows that the Δ^{-2} dependence of the recoil-induced resonance in the pump-probe spectroscopy can be regarded as partially arising from Raman resonances between atomic states labeled by center-of-mass momenta p and $p+2\hbar k$, respectively.

It is possible to deduce the shape and width of G as a function of δ without explicitly evaluating it. Assuming that $W_1(p)$ is centered at $p=0$, then from the first line of Eq. (51), if $\delta>0$, the resonance condition requires that $p<-\hbar k$. For this region of p, $W_1(p)<W_1(p+2\hbar k)$, as a result $G<0$. By the same argument one finds that for $\delta<0$, $G>0$. So when δ is tuned across 0, one gets a dispersive curve for G whose width is determined by that of W_1, which is of order ku. The above argument is analogous to that for normal Raman processes between internal atomic states [21].

An interesting result of this theory is that it may provide a way to determine the momentum distribution of cold atoms by laser spectroscopy other than some non-spectroscopic ways such as time-of-flight method. For example, in the pump-probe case with pump and probe counter propagating, the scan of probe absorption as a function of pump-probe detuning has a dispersionlike line shape that directly reflects the atomic-momentum distribution.

Our calculation has been limited to "two-level" atoms. It is important to generalize the calculation to include magnetically degenerate ground states. Such a multilevel structure is necessary to achieve frictional laser cooling of atoms below the Doppler limit. Optical pumping plays an important role in the cooling processes. As the atom moves through the cooling fields, it sees a polarization gradient of the fields. The magnetic orientation or alignment of the ground state need not be conserved in these optical pumping processes. This feature leads to narrow structures in nonlinear spectroscopy even without the inclusion of recoil effects [11]. It remains to be seen whether the recoil-induced structures persist for magnetically degenerate ground states and, if so, how the new level scheme and the finite optical pumping rates alter the structures found in this paper for simple two-level atoms.

It may be possible to observe directly the features predicted in this work. If cooled atoms are optically pumped to a particular level without increasing the temperature too much, a two-level atom can be approximated. The pump-probe spectrum for these atoms can then be obtained. Owing to the narrow nature of these structures, one has to be careful about the presence of saturation effects and keep the field strength sufficiently weak.

In conclusion, we have demonstrated that the inclusion of recoil effects leads to additional resonant structures in nonlinear spectroscopy. Further study is required to see if these resonances are related to recent experimental results on pump-probe absorption of cold atoms [21]. It may also be worthwhile to investigate the implications of this theory to laser cooling of "two-level" atoms.

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