Collisional decay and revival of the grating stimulated echo

P. R. Berman
Physics Department, University of Michigan, Randall Laboratories, Ann Arbor, Michigan 48103
(Received 18 October 1993)

A theory is presented that describes collision-induced modifications of the grating stimulated echo (GSE) signal. In the GSE [B. Dubetsky, P. R. Berman, and T. Sleator, Phys. Rev. A **46**, 2213 (1992)], a spatially modulated atomic population is created which undergoes Doppler dephasing and rephasing as a result of the interaction of the atoms with a number of input pulses. At low pressures, there is a rapid decay of the GSE signal owing to a collisional breaking of the dephasing-rephasing process. At high perturber pressures, however, there can be a revival of the GSE signal owing to a collisional inhibition of the Doppler dephasing. These collisional processes are described in detail, with a special emphasis given to the distinction between "open" and "closed" atom-field systems. The manner in which the GSE can be used to determine collision kernels and diffusion coefficients is also discussed.

PACS number(s): 42.50.Md, 51.10.+y

I. INTRODUCTION

Coherent transient phenomena often provide an effective means for measuring relaxation in vapors, liquids, and solids. Recently, an echo scheme, the grating stimulated echo (GSE), was proposed as a technique which was especially well suited to monitor atomic ground-state relaxation in vapors [1]. In the GSE, two counterpropagating pulses separated in time by T_{21} create a spatial ground-state population grating, varying as $\cos(2\mathbf{k}\cdot\mathbf{R})$, where $\mathbf{k}$ is the propagation vector of the first pulse. Owing to Doppler dephasing of the different atomic velocity groups, the population grating for the ensemble rapidly decays following the second pulse. As in all echo phenomena, however, this decay is not an irreversible process, and the application of a standing-wave pulse at a time T following the second pulse starts a rephasing of the different velocity groups that can contribute to the grating. The rephasing is complete at a time T following the third pulse, at which time the macroscopic ground-state grating created by the first two pulses is restored. The application of a traveling-wave pulse at this time leads to an echo signal at a time T_{21} following the application of this traveling-wave pulse. The pulse sequence is shown in Fig. 1.

The time T_{21} must be chosen smaller than the lifetime of the atomic dipole coherence, but the interval T is limited only by some effective ground-state relaxation time. For example, the effective ground-state lifetime might be determined by the time the atoms spend in the atom-field interaction volume. On the other hand, it could be related to a collision-induced change in the Doppler phase associated with the ground-state grating. As has been pointed out in Ref. [1], the GSE can serve as an extremely sensitive probe of ground-state relaxation.

Preliminary results related to the influence of collisions on the GSE signal were presented in [1] to indicate the sensitivity of the GSE to small velocity changes. Friedberg and Hartmann [2] extended these results by considering the limiting case in which $T_{21}=0$; in this manner, spatial harmonics varying as $\exp(\pm 2in\mathbf{k}\cdot\mathbf{R})$ (n =a positive integer) are created which, for $n>2$, may lead to a sensitivity greater than that of the GSE. The limiting case of $T_{21}=0$ had been discussed earlier in theories of echo phenomena in standing-wave fields [3–5], where the influence of collisions on the signals was also discussed. A renewed interest in this type of echo phenomena was kindled by the atom-interferometric scheme of Kasevich

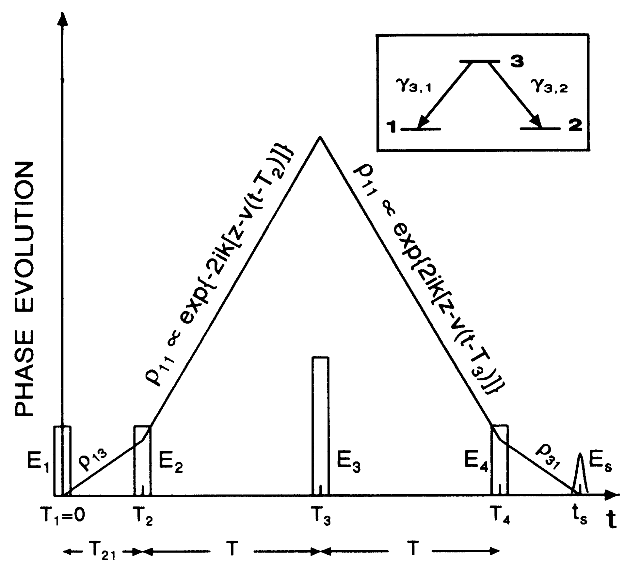

FIG. 1. Doppler phase associated with various density-matrix elements leading to a grating stimulated echo (GSE). The four input pulses E_i ($i=1$–4) give rise to an echo E_s at $t=t_s$. The field propagation vectors for pulses 1, 2, and 4 are $\mathbf{k}_1$, $\mathbf{k}_2$, and $\mathbf{k}_4=\mathbf{k}_2$, respectively, while the third pulse consists of two traveling waves having wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$. The signal propagates in the $\mathbf{k}_1$ direction. The atomic-energy-level diagram is shown in the inset. Each level can also decay to some external reservoir. The incident fields drive only the 1-3 transition. The diagram is drawn for $\mathbf{k}_1=-\mathbf{k}_2=k_1\hat{z}$.

and Chu [6]. They proposed a pulse sequence closely related to that associated with grating echoes [4,5]; however, as applied to atoms at microkelvin temperatures rather than to atoms at 450 K (so that the interval T between pulses could be increased owing to the longer time the atoms spend in the interaction volume), they demonstrated that this pulse sequence has great potential in atomic interferometry owing to its exceptional sensitivity to small velocity changes. It should also be noted that there has been considerable work [7] involving the use of optical transient spectroscopy other than the GSE to probe ground-state relaxation.

In this paper, the role that collisions play in modifying the GSE signal is examined in some detail. Most earlier theories of grating echoes [1–5] incorporated a collision model in which the collision kernel $W(\mathbf{v},\mathbf{v}')$, defined as the probability density per unit time that collisions result in a velocity change from $\mathbf{v}'$ to $\mathbf{v}$, is a function of ($\mathbf{v}$-$\mathbf{v}'$) only. While such a kernel adequately describes weak collisions in many cases of experimental interest, it is known that this kernel cannot give rise to phenomena associated with the collisional narrowing of spectral profiles [8]. It may seem that the grating echoes have very little connection with the collisional narrowing of spectral profiles, but, in fact, there is a very close correspondence.

A necessary [9] condition for the narrowing of spectral profiles is that the mean free path of the atoms is less than the transition wavelength. This condition can be written as

$$k\delta u/\Gamma \ll 1 \ , \tag{1}$$

where δu is $\sqrt{2}$ times the (one-dimensional) rms velocity change per collision, and Γ is a collision rate. Condition (1) implies that, as a result of collisions, the Doppler phase factor always remains much less than unity, resulting in a suppression of the Doppler width. In the case of the grating echoes, we are not dealing with an atomic transition. Rather, we are dealing with a ground-state population density that is modulated as $\cos(2\mathbf{k}\cdot\mathbf{R})$; however, just as in the case of spectral profiles, this modulation gives rise to a Doppler dephasing. The Doppler phase factor for the modulated population varies as $\exp(\pm 2i\mathbf{k}\cdot\mathbf{v}t)$ [whereas the Doppler dephasing associated with spectral profiles varies as $\exp(\pm i\mathbf{k}\cdot\mathbf{v}t)$]. To be specific, the GSE amplitude varies as $\langle \exp[2i\mathbf{k}\cdot(\int_{T_{21}}^{T_{21}+T} - \int_{T_{21}+T}^{T_{21}+2T}\mathbf{v}(t)dt)]\rangle$, where the average is over collision histories. With no collisions, the two integrals cancel and this factor is unity. On the other hand, if $k\delta uT \gg 1$, collisions tend to destroy the echo amplitude by interfering with the dephasing-rephasing process. The collisional destruction occurs unless the mean free path is less than $1/(2k)$; for perturber pressures such that

$$2k\delta u/\Gamma \ll 1 \ , \tag{2}$$

the Doppler phase factor ceases to be important and the GSE signal can be restored. Thus, if one wishes to investigate the collisional suppression of Doppler dephasing in the GSE, one cannot use a difference kernel to describe the collisions. As an alternative model at high perturber densities, a diffusion model can be used. It should be noted that collisional narrowing effects associated with spatially modulated ground-state populations has already been observed experimentally in the frequency domain in four-wave mixing experiments [10,11]. In contrast to frequency domain experiments in which the fields are on at all times and can lead to power broadening of the two-photon resonance, the populations evolve in field-free regions in the GSE and power broadening does not influence the signal.

It is not always possible to observe long-lived ground-state transients (i.e., transients whose lifetime is determined by some effective ground-state lifetime). For example, if the atoms interacting with the fields can be modeled as "two-level" atoms, and if the sum of excited and ground-state populations is conserved, then the lifetime of the ground-state transients is determined by the *excited* rather than the ground-state lifetime for such a closed system [12–14]. In order to monitor long-time ground-state relaxation, the atom-field system must be "open" in some respect. This can be accomplished in a number of ways. For example, if the ground state consists of two hyperfine levels, and the incident radiation fields drive transitions between only one of these states and an excited state, then the atom-field system is open via decay to the other hyperfine level. Alternatively, the ground and excited states can each consist of a number of magnetic sublevels, and the atom-field interaction can probe properties such as magnetic-state orientation and alignment which are not conserved [13]. Finally, even for a two-level atom, the sum of excited and ground-state populations for each velocity subclass of atoms is not conserved if effects such as collisions with perturber atoms [14] or recoil on the absorption and emission of radiation [15] are taken into account. In this paper, both homogeneous and inhomogeneous opening of the atom-field system are considered; homogeneous processes (such as nonconservation of magnetic alignment in the absence of collisions) are those which are the same for all atoms in the ensemble, while inhomogeneous processes (such as velocity-changing collisions) differ for atoms having different velocities.

The paper is organized as follows: In Sec. II, the physical system is described and several approximations are introduced. The calculation of the GSE signal is presented in Sec. III for an arbitrary collision kernel. In Sec. IV, a discussion of the low and high perturber pressure limits is given. A difference collision kernel is used for the low-pressure calculations and a diffusion model for the high-pressure calculations. Special emphasis is placed on the roles played by effects related to collisional suppression of Doppler dephasing and the dependence of the GSE amplitude on the homogeneous or inhomogeneous opening of the atom-field system.

II. PHYSICAL SYSTEM AND APPROXIMATIONS

An ensemble of "active" atoms in a vapor cell which undergo binary collisions with a reservoir of foreign gas atoms is subjected to a number of laser pulses. The level scheme for the active atoms is shown in Fig. 1. The pulses, having temporal width τ_p, drive transitions between states 1 and 3. Each of the pulses is assumed to be sufficiently long to guarantee that its spatial extent is larger than the sample size. For a cell of length L, this condition can be stated as

$$\tau_p > L/c \ . \tag{3}$$

For a 1.0-cm sample, this condition implies pulse durations greater than 30 ps. Although condition (3) is not critical, it does simplify the calculations somewhat. The pulse sequence is shown in Fig. 1. Pulses 1, 2, and 4 are traveling-wave pulses having propagation vectors $\mathbf{k}_1$, $\mathbf{k}_2$, and $\mathbf{k}_4=\mathbf{k}_2$, respectively, while pulse 3 consists of two traveling-wave pulses having propagation vectors $\mathbf{k}_1$ and $\mathbf{k}_2$. Pulse i is centered at $t=T_i$, and the pulse intervals

$$T_{ij}=T_i-T_j \tag{4}$$

are chosen such that

$$T_{43}=T_{32}=T \ . \tag{5}$$

The signal is monitored for times $t>T_4$.

For atom-field detunings,

$$\Delta=\Omega-\omega \ , \tag{6}$$

and collision rates Γ, the following inequalities are assumed to hold:

$$\Gamma T_{21}<1 \ , \tag{7a}$$

$$\gamma_3 T>1 \ , \tag{7b}$$

$$\kappa u T \gg 1 \ , \tag{7c}$$

$$\gamma_3 T_{21} \ll 1 \ , \tag{7d}$$

$$\mathbf{k}_1 u\tau_p \ll 1 \ , \quad \gamma_3\tau_p \ll 1 \ , \quad \Gamma\tau_p \ll 1 \ ,$$
$$|\Delta|\tau_p \ll 1 \ , \quad \tau_p \ll T_{ij} \ , \tag{7e}$$

where

$$\boldsymbol{\kappa}=(\mathbf{k}_1-\mathbf{k}_2)/2 \ . \tag{8}$$

Inequalities (7e) allow one to neglect all relaxation and dephasing of atomic density matrix elements (in an interaction representation) during the applied pulses. Inequalities (7a) and (7d) ensure that the signal does not decay owing to homogeneous decay of the atomic dipole coherence in the time interval between the first two pulses, while inequality (7c) guarantees that different velocity subgroups of atoms lead to an inhomogeneous decay of the spatially modulated part of the atomic state populations in the time interval between the second and third pulses. Inequality (7b) implies that the entire signal is associated with the ground-state gratings that are formed and persist in this excitation scheme; any excited-state gratings quickly decay away on a time scale $\gamma_3^{-1}<T$.

Levels 1 and 3 form a "two-level" system which interacts with the external fields. In order to observe ground-state transients on a time scale $T>\gamma_3^{-1}$, it is necessary for the system to be "open" in some respect. The system can be opened in two ways. In the absence of velocity-changing collisions, the system is open if the branching ratio to level 2 is nonzero, i.e., if $\gamma_{3,2}\neq 0$. In the present calculation, level 2 is an all purpose level. It can represent another hyperfine level to which level 3 can decay, or it can simulate other magnetic-state sublevels degenerate with level 1. In the latter case, the magnetic-state alignment or orientation for the system need not be conserved, and the system is "open" for these properties [13].

When $\gamma_{3,2}\neq 0$, spontaneous emission results in a homogeneous opening of the system for all velocity classes of atoms. Velocity-changing collisions can result in an *inhomogeneous* opening of the system, even when $\gamma_{3,2}=0$. As long as the ground- and excited-state collision kernels differ, the sum $\rho_{11}(\mathbf{v})+\rho_{33}(\mathbf{v})$ is not conserved for each velocity subclass of atoms. As a result, the system is open, and transients characterized by ground-state decay rates can be observed [14]. In this paper, both homogeneous and inhomogeneous openings of the system are examined.

The ground-state decay occurs at a rate γ_1 which may be due to transit time effects (the atom spends a finite time in the atom-field interaction volume) or collisions which transfer population from states 1 to 2. In this calculation, it is assumed that γ_1^{-1} represents the longest time scale in the problem. In particular, it is assumed that

$$\gamma_1 \ll \gamma_3 \ . \tag{9}$$

III. CALCULATION OF THE SIGNAL

The pulse sequence and the atomic level scheme is given in Fig. 1. The field amplitude is given by

$$\mathbf{E}(\mathbf{R},t)=(\tfrac{1}{2})\epsilon\sum_j \mathcal{E}_j(\mathbf{R})e^{-i\Omega t}\psi(t-T_j)+\text{c.c.} \ , \tag{10a}$$

where

$$\mathcal{E}_j(\mathbf{R})=\begin{cases}\mathcal{E}_j\exp(i\mathbf{k}_j\cdot\mathbf{R}) \ , & j=1,2,4 \\ \mathcal{E}_3[\exp(i\mathbf{k}_1\cdot\mathbf{R})+\exp(i\mathbf{k}_2\cdot\mathbf{R})] \ , & j=3 \ .\end{cases} \tag{10b}$$

$\psi(t)$ is a pulse envelope function centered at $t=0$ having a temporal width of order τ_p, ϵ is a polarization vector in a direction perpendicular to the plane of $\mathbf{k}_1$ and $\mathbf{k}_2$, $\mathbf{k}_4=\mathbf{k}_2$, and c.c. stands for complex conjugate. The echo signal is proportional to a quantity $I(t)$ defined by

$$I(t)=\left|\int \tilde{\rho}_{31}(\mathbf{v},t)d\mathbf{v}\right|^2 \ , \tag{11}$$

where $\tilde{\rho}_{31}$ is a density-matrix element, written in an interaction representation in which the state amplitudes are defined by $a_1=\tilde{a}_1\exp(i\Omega t/2)$, $a_3=\tilde{a}_3\exp(-i\Omega t/2)$, and density-matrix elements by

$$\tilde{\rho}_{13}=\tilde{a}_1\tilde{a}_3^*=\rho_{13}\exp(-i\Omega t)=\tilde{\rho}_{31}^* \ . \tag{12}$$

In order to calculate the GSE, one follows the density-matrix chain shown in Fig. 1. That is, only those components of the various density-matrix elements having the spatial dependence shown in Fig. 1 contribute to the GSE signal.

The calculation is carried out conveniently by breaking up the problem into time regions where the external fields act and intervals that are field free. During the field interaction, one can neglect the motion of the atoms and any relaxation processes owing to conditions (7e). As a consequence one can obtain the atomic response to the jth pulse by using the following equations for the state amplitudes:

$$\dot{\tilde{a}}_1 = i[\chi_j(\mathbf{R})]^* \psi(t-T_j)\tilde{a}_3 , \tag{13a}$$

$$\dot{\tilde{a}}_3 = i\chi_j(\mathbf{R})\psi(t-T_j)\tilde{a}_1 , \tag{13b}$$

where

$$\chi_j(\mathbf{R}) = \mu \mathcal{E}_j(\mathbf{R})/2\hbar , \tag{14}$$

μ is the 1−3 matrix element (assumed real) of the component of the atomic dipole moment in the direction ϵ. In writing Eqs. (13), an atom-field interaction of the form $-\boldsymbol{\mu}\cdot\mathbf{E}(\mathbf{R},t)$ has been assumed, ($\boldsymbol{\mu}$ is the atomic dipole moment operator) and the rotating-wave approximation has been made.

It is an easy matter to solve Eqs. (13). As a result of pulse j, the change in density-matrix elements $\rho_{ii} = \tilde{a}_i \tilde{a}_i^*$ (unless noted otherwise, indices take on the values 1 and 3) and $\tilde{\rho}_{13}$ is given by

$$\begin{aligned}\rho_{11}(T_j^+) = (\tfrac{1}{2})\{1+\cos[\alpha_j(\mathbf{R})\theta_j]\}\rho_{11}(T_j^-) + (\tfrac{1}{2})\{1-\cos[\alpha_j(\mathbf{R})\theta_j]\}\rho_{33}(T_j^-) \\ -(i/2)\sin[\alpha_j(\mathbf{R})\theta_j]\tilde{\rho}_{13}(T_j^-)e^{i\mathbf{k}_j\cdot\mathbf{R}} + (i/2)\sin[\alpha_j(\mathbf{R})\theta_j]\tilde{\rho}_{31}(T_j^-)e^{-i\mathbf{k}_j\cdot\mathbf{R}}\end{aligned} \tag{15a}$$

$$\begin{aligned}\tilde{\rho}_{13}(T_j^+) = (\tfrac{1}{2})\{1+\cos[\alpha_j(\mathbf{R})\theta_j]\}\tilde{\rho}_{13}(T_j^-) + (\tfrac{1}{2})\{1-\cos[\alpha_j(\mathbf{R})\theta_j]\}\tilde{\rho}_{31}(T_j^-)e^{-2i\mathbf{k}_j\cdot\mathbf{R}} \\ -(i/2)\sin[\alpha_j(\mathbf{R})\theta_j]e^{-i\mathbf{k}_j\cdot\mathbf{R}}\rho_{11}(T_j^-) + (i/2)\sin[\alpha_j(\mathbf{R})\theta_j]\rho_{33}(T_j^-)e^{-i\mathbf{k}_j\cdot\mathbf{R}} ,\end{aligned} \tag{15b}$$

where $T_j^{\pm}(j=1-4)$ are times immediately after and before the application of pulse j,

$$\theta_j = 2\chi_j \int_{T_j^-}^{T_j^+} \psi_j(t-T_j)dt , \quad j=1-4 , \tag{16}$$

$$\alpha_3(\mathbf{R}) = \cos(\kappa\cdot\mathbf{R}) , \quad \alpha_1=\alpha_2=\alpha_4=1 , \tag{17}$$

$$\chi_j = \mu\mathcal{E}_j/2\hbar , \quad j=1-4 , \tag{18}$$

$$\mathbf{k}_3 = (\mathbf{k}_1+\mathbf{k}_2)/2 , \tag{19}$$

$$\kappa = (\mathbf{k}_1-\mathbf{k}_2)/2 ,$$

and equations for $\tilde{\rho}_{31}$ and ρ_{33} can be obtained by interchanging the subscripts 1 and 3 and making the replacement $\mathbf{k}_j \leftrightarrow -\mathbf{k}_j$ in Eqs. (15a) and (15b).

In the field-free regions between pulses 2 and 3 and between pulses 3 and 4, the atomic-state populations evolve as

$$\begin{aligned}\dot{\rho}_{11}(\mathbf{v},t) = -[\gamma_1+\Gamma_1(\mathbf{v})]\rho_{11}(\mathbf{v},t) + \gamma_{3,1}\rho_{33}(\mathbf{v},t) \\ + \int W_1(\mathbf{v},\mathbf{v}')\rho_{11}(\mathbf{v}',t)d\mathbf{v}' ,\end{aligned} \tag{20a}$$

$$\begin{aligned}\dot{\rho}_{33}(\mathbf{v},t) = -[\gamma_3+\Gamma_3(\mathbf{v})]\rho_{33}(\mathbf{v},t) \\ + \int W_3(\mathbf{v},\mathbf{v}')\rho_{33}(\mathbf{v}',t)d\mathbf{v}' ,\end{aligned} \tag{20b}$$

where the overdot signifies $\partial/\partial t + \mathbf{v}\cdot\nabla$, $W_j(\mathbf{v},\mathbf{v}')$ is the probability density per unit time that collisions with perturber atoms change the velocity of an atom in state j from $\mathbf{v}'$ to $\mathbf{v}$, and

$$\Gamma_j(\mathbf{v}) = \int W_j(\mathbf{v},\mathbf{v}')d\mathbf{v}' \tag{21}$$

is the rate of such collisions.

In the field-free regions between pulses 1 and 2 and between pulse 4 and the echo, the atomic-state coherences evolve as

$$\begin{aligned}\dot{\tilde{\rho}}_{13}(\mathbf{v},t) &= [\partial/\partial t + \mathbf{v}\cdot\nabla]\tilde{\rho}_{13}(\mathbf{v},t) \\ &= [\dot{\tilde{\rho}}_{31}(\mathbf{v},t)]^* = 0 ,\end{aligned} \tag{22}$$

since all relaxation can be neglected, owing to conditions (7a) and 7(d).

It is now possible to piece together the solution for the density-matrix chain depicted in Fig. 1. Using Eq. (15b) for the action of the first pulse, (22) for the interval between the first and second pulses, and (15a) for the action of the second pulse, one finds that, immediately following the second pulse,

$$\begin{aligned}\rho_{11}(\mathbf{R},\mathbf{v},T_2^+;-) &= -\rho_{33}(\mathbf{v},T_2^+;-) \\ &= -A_1A_2n_1W_0(\mathbf{v})\exp(i\mathbf{k}_1\cdot\mathbf{v}T_{21}) \\ &\quad\times\exp(-2i\kappa\cdot\mathbf{R}) ,\end{aligned} \tag{23}$$

where

$$A_j \equiv (\tfrac{1}{2})\sin(\theta_j) , \quad j=1,2,4 , \tag{24}$$

$$W_0(\mathbf{v}) = (\pi u^2)^{-3/2}\exp[-(v/u)^2] \tag{25}$$

is the initial velocity distribution in state 1, n_1 is the initial population of level 1, and the minus sign labels the component of ρ_{ii} varying as $\exp(-2i\kappa\cdot\mathbf{R})$ needed for the time interval between the second and third pulses (see Fig. 1).

In the interval between pulses 2 and 3, one uses Eq. (23) to write the formal solution to Eqs. (20) as

$$\rho_{11}(\mathbf{R},\mathbf{v},t;-)=\int G_1(\mathbf{v},\mathbf{v}';t-T_2^+;-)\times\rho_{11}(\mathbf{R},\mathbf{v}',T_2^+;-)d\mathbf{v}' \; , \tag{26a}$$

$$\rho_{33}(\mathbf{R},\mathbf{v},t;-)=-\int G_3(\mathbf{v},\mathbf{v}';t-T_2^+;-)\times\rho_{11}(\mathbf{R},\mathbf{v}',T_2^+;-)d\mathbf{v}' \; , \tag{26b}$$

where the propagators $G_j(\mathbf{v},\mathbf{v}';t;-)$ satisfy the equations

$$\begin{aligned}\partial G_1(\mathbf{v},\mathbf{v}';t;-)/\partial t-2i\boldsymbol{\kappa}\cdot\mathbf{v}G_1(\mathbf{v},\mathbf{v}';t;-)&=-[\gamma_1+\Gamma_1(\mathbf{v})]G_1(\mathbf{v},\mathbf{v}';t;-)\\&\quad-\gamma_{3,1}G_3(\mathbf{v},\mathbf{v}';t;-)\\&\quad+\int W_1(\mathbf{v},\mathbf{v}'')G_1(\mathbf{v}'',\mathbf{v}';t;-)d\mathbf{v}'' \; ,\end{aligned} \tag{27a}$$

$$\begin{aligned}\partial G_3(\mathbf{v},\mathbf{v}';t;-)/\partial t-2i\boldsymbol{\kappa}\cdot\mathbf{v}G_3(\mathbf{v},\mathbf{v}';t;-)&=-[\gamma_3+\Gamma_3(\mathbf{v})]G_3(\mathbf{v},\mathbf{v}';t;-)\\&\quad+\int W_3(\mathbf{v},\mathbf{v}'')G_3(\mathbf{v}'',\mathbf{v}';t;-)d\mathbf{v}'' \; ,\end{aligned} \tag{27b}$$

subject to the initial conditions

$$G_j(\mathbf{v},\mathbf{v}';0;-)=\delta(\mathbf{v}-\mathbf{v}') \; . \tag{27c}$$

The third pulse converts $\rho_{11}(\mathbf{R},\mathbf{v},T_3^-;-)$ and $\rho_{33}(\mathbf{R},\mathbf{v},T_3^-;-)$ into $\rho_{ii}(\mathbf{R},\mathbf{v},T_3^+;+)$, where "+" labels the component of the density-matrix element varying as $\exp(2i\boldsymbol{\kappa}\cdot\mathbf{R})$. Owing to condition (7b) ($\gamma_3T\gg1$), one can set $\rho_{33}(\mathbf{R},\mathbf{v},T_3^-;-)=0$. Explicitly, from Eqs. (15), one then finds

$$\rho_{11}(\mathbf{R},\mathbf{v},T_3^+;+)=-\rho_{33}(\mathbf{R},\mathbf{v},T_3^+;+)=B\rho_{11}(\mathbf{R},\mathbf{v},T_3^-;-) \; , \tag{28}$$

where

$$B=(\tfrac{1}{2})J_4(\theta_3) \; , \tag{29}$$

and J_4 is a fourth-order Bessel function. Taking into account the evolution from $t=T_3^+$ to times $t\geq T_4$ using Eqs. (5), (26), (28), (15b), and (23), one finds that, for $t\geq T_4$, the component of $\tilde{\rho}_{31}$ contributing to the GSE signal propagating in the $\mathbf{k}_1$ direction is

$$\tilde{\rho}_{31}(\mathbf{R},\mathbf{v},t)=iA_1A_2A_3Bn_1e^{i\mathbf{k}_1\cdot\mathbf{R}}e^{-i\mathbf{k}_1\cdot\mathbf{v}(t-T_4)}\times\int d\mathbf{v}''d\mathbf{v}'G_1(\mathbf{v},\mathbf{v}';T;+)G_1(\mathbf{v}',\mathbf{v}'';T;-)e^{i\mathbf{k}_1\cdot\mathbf{v}''T_{21}}W_0(\mathbf{v}'') \; , \tag{30}$$

where

$$G_1(\mathbf{v},\mathbf{v}';T;+)=[G_1(\mathbf{v},\mathbf{v}';T;-)]^* \; . \tag{31}$$

A. Collisions

The influence of collisions is contained in the propagators $G_{ii}(\mathbf{v},\mathbf{v}';T;\pm)$. From Eqs. (11) and (30), it follows that the GSE signal intensity is proportional to $|S(t)|^2$, where

$$S(t)=\int d\mathbf{v}\,d\mathbf{v}'\,d\mathbf{v}''e^{-i\mathbf{k}_1\cdot\mathbf{v}(t-T_4)}\times G_1(\mathbf{v},\mathbf{v}';T;+)G_1(\mathbf{v}',\mathbf{v}'';T;-)\times e^{i\mathbf{k}_1\cdot\mathbf{v}''T_{21}}W_0(\mathbf{v}'') \; . \tag{32}$$

In general it is impossible to obtain analytic forms for $G_i(\mathbf{v},\mathbf{v}';T;\pm)$. For the purposes of this discussion, it will be sufficient to consider only low- and high-pressure regimes (to be defined below) for which analytical expressions can be obtained, provided that we adopt a collision model in which the characteristic velocity change per collision δu is much less than the most probable active atom speed u; that is,

$$\delta u_i/u\ll1 \; , \tag{33}$$

where the subscript i allows for a state-dependent characteristic velocity change. Since the specific form of the collision kernel is not critical to the present discussion, a kernel is assumed for which condition (33) is satisfied (weak collision model). Before examining the low- and high-pressure regimes, it is useful to point out the importance of open versus closed systems in determining the signal.

B. Open versus closed systems

A "closed" system is defined here as one in which

$$\gamma_3=\gamma_{3,1}+\gamma_1 \; , \tag{34}$$

which implies that the total population density $[\rho_{11}(\mathbf{v},t)+\rho_{33}(\mathbf{v},t)]$ decays to the reservoir at rate γ_1 in the absence of collisions. In the limit that $\gamma_1\sim0$, (recall that γ_1^{-1} represents the longest time scale in the problem) the population density $[\rho_{11}(\mathbf{v},t)+\rho_{33}(\mathbf{v},t)]$ is a conserved quantity.

The significance of a closed system is connected to the fact that any field-induced modifications of the population densities relax at the excited-state decay rate and *not* the ground-state decay rate. This result can be seen directly in Eqs. (27) for the propagators which, in the absence of collisions, can be rewritten as

$$\partial[G_1(\mathbf{v},\mathbf{v}';t;\pm)-G_3(\mathbf{v},\mathbf{v}';t;\pm)]/\partial t\pm 2i\boldsymbol{\kappa}\cdot\mathbf{v}[G_1(\mathbf{v},\mathbf{v}';t;\pm)-G_3(\mathbf{v},\mathbf{v}';t;\pm)]=-\gamma_1[G_1(\mathbf{v},\mathbf{v}';t;\pm)-G_3(\mathbf{v},\mathbf{v}';t;\pm)]\ , \quad (35)$$

provided that condition (34) holds. Since the initial condition is $[G_1(\mathbf{v},\mathbf{v}';0;\pm)-G_3(\mathbf{v},\mathbf{v}';0;\pm)]=0$, one sees that

$$G_1(\mathbf{v},\mathbf{v}';t;\pm)=G_3(\mathbf{v},\mathbf{v}';t;\pm) \quad (36)$$

for closed systems. Consequently $G_1(\mathbf{v},\mathbf{v}';t;\pm)$ decays at the same rate as $G_3(\mathbf{v},\mathbf{v}';t;\pm)$, namely γ_3. From the assumption that $\gamma_3 T\gg 1$, it follows that $G_3(\mathbf{v},\mathbf{v}';T;\pm)\sim 0$, and there is a negligible GSE signal for a closed system in the absence of collisions.

How do collisions modify this result? For systems that are already open as a result of spontaneous emission ($\gamma_{3,2}\neq 0$), collisions alter the GSE signal, but do not play a critical role in opening or closing the system. On the other hand, for closed systems, collision *do* play a critical role in opening the system. If the collision kernels and/or rates for levels 1 and 3 differ, the total population density $[\rho_{11}(\mathbf{v},t)+\rho_{33}(\mathbf{v},t)]$ is no longer conserved (since collisions affect the velocity distributions of states 1 and 3 differently) and the system is "opened" by collisions, leading to a collision-induced GSE signal. [If the collision kernels for levels 1 and 3 were identical, Eq. (35), modified to include collisional effects, remains an homogeneous equation with the same solution (36).] At very high pressures, collisions redistribute the velocities over the entire Maxwellian distribution, and the system can "reclose." These features are seen in the examples given below.

C. Low pressure

The low-pressure limit is defined as one in which collisions do not lead to rethermalization on the time scale ($2T$) of the experiment. For weak collisions defined by (33), the effective rate at which collisions rethermalize the sample is given by [16]

$$\Gamma_i'=\Gamma_i(\delta u_i/u)^2/2\ , \quad (37)$$

and the low-pressure limit by

$$\Gamma_i'T=\Gamma_i(\delta u_i/u)^2T\ll 1\ . \quad (38)$$

From condition (7c) it follows that $\Gamma_i(\delta u_i/u)^2/(\kappa u)\ll 1$ when condition (38) is satisfied.

In the low-pressure limit, it is convenient to choose a kernel of the form

$$W_i(\mathbf{v},\mathbf{v}')\equiv W_i(\mathbf{v}-\mathbf{v}')\equiv\Gamma_i F(\mathbf{v}-\mathbf{v}')\ , \quad (39)$$

where $F(\mathbf{v})$ is an even function of $\mathbf{v}$. Although this difference kernel does not satisfy detailed balance, it can still be used provided condition (38) holds. The advantage of the difference kernel is that it allows for an analytical solution of Eqs. (27). Setting

$$G_i(\mathbf{v},\mathbf{v}';t;\pm)=\tilde{G}_i(\mathbf{v},\mathbf{v}';t;\pm)\exp(\mp 2i\boldsymbol{\kappa}\cdot\mathbf{v}t)\ , \quad (40)$$

one finds that the $\tilde{G}_i(\mathbf{v},\mathbf{v}';t;+)$ satisfy

$$\partial\tilde{G}_1(\mathbf{v},\mathbf{v}';t;+)/\partial t=-(\gamma_1+\Gamma_1)\tilde{G}_1(\mathbf{v},\mathbf{v}';t;+)-\gamma_{3,1}\tilde{G}_3(\mathbf{v},\mathbf{v}';t;+) + \int W_1(\mathbf{v}-\mathbf{v}'')\exp[2i\boldsymbol{\kappa}\cdot(\mathbf{v}-\mathbf{v}'')t]\tilde{G}_1(\mathbf{v}'',\mathbf{v}';t;+)d\mathbf{v}''\ , \quad (41a)$$

$$\delta\tilde{G}_3(\mathbf{v},\mathbf{v}';t;+)/\partial t=-(\gamma_3+\Gamma_3)\tilde{G}_3(\mathbf{v},\mathbf{v}';t;+) + \int W_3(\mathbf{v}-\mathbf{v}'')\exp[2i\boldsymbol{\kappa}\cdot(\mathbf{v}-\mathbf{v}'')t]\tilde{G}_3(\mathbf{v}'',\mathbf{v}';t;+)d\mathbf{v}''\ . \quad (41b)$$

It is clear from these equations that $\tilde{G}_i(\mathbf{v},\mathbf{v}';t;+)\equiv\tilde{G}_i(\mathbf{v}-\mathbf{v}';t;+)$ is a function of $(\mathbf{v}-\mathbf{v}')$ only. Consequently, the signal amplitude $S(t)$ given by Eq. (32) may be written as

$$S(t)=(2\pi)^3 n_1\mathcal{G}_1\{[2\boldsymbol{\kappa}T+\mathbf{k}_1(t-T_4)];T;+\}\mathcal{G}_1[\mathbf{k}_1(t-T_4);T;+]\exp[-\mathbf{k}_1^2u^2(t-T_4-T_{21})^2/4]\ , \quad (42)$$

where

$$\mathcal{G}_i(\beta;t;+)=(2\pi)^{-3/2}\int d\bar{\mathbf{v}}\,\tilde{G}_i(\bar{\mathbf{v}};t;+)\exp(-i\boldsymbol{\beta}\cdot\bar{\mathbf{v}}) \quad (43)$$

is the Fourier transform of $\tilde{G}_i(\bar{\mathbf{v}};t;+)$, and the relationship $\mathcal{G}(\beta;t;-)=\mathcal{G}^*(\beta;t;+)=\mathcal{G}(-\beta;t;+)$ has been used, the last equality following from the fact that the kernel is an even function of $\mathbf{v}$.

Equations (41) are readily solved by transform techniques to obtain $\mathcal{G}_i(\beta;t;+)$. For $\gamma_3T\gg 1$, one finds

$$\mathcal{G}_1(\beta;T;+)=(2\pi)^{-3/2}\exp[-\gamma_1(\boldsymbol{\beta},\boldsymbol{\kappa},T)T]\left\{1-\gamma_{3,1}\int_0^T d\tau\exp\{[\gamma_1(\boldsymbol{\beta},\boldsymbol{\kappa},\tau)-\gamma_3(\boldsymbol{\beta},\boldsymbol{\kappa},\tau)]\tau\}\right\} \quad (44)$$

where

$$\gamma_i(\boldsymbol{\beta},\boldsymbol{\kappa},\tau)=\gamma_i+\Gamma_i[1-H_i(\boldsymbol{\beta},\boldsymbol{\kappa},\tau)]\ , \quad (45)$$

$$H_i(\boldsymbol{\beta},\boldsymbol{\kappa},\tau)=(1/\tau)\int_0^{\tau}d\tau'\int d\bar{\mathbf{v}}\,F_i(\bar{\mathbf{v}})\exp(-i\boldsymbol{\beta}\cdot\bar{\mathbf{v}}+2i\boldsymbol{\kappa}\cdot\bar{\mathbf{v}}\tau')$$
$$=(2\pi)^{3/2}(1/\tau)\int_0^{\tau}d\tau'\,\mathcal{F}_1(\boldsymbol{\beta}-2\boldsymbol{\kappa}\tau')\ , \quad (46)$$

and $\Gamma_i\mathcal{F}_i(\boldsymbol{\beta})$ is the Fourier transform of the collision kernel. This result is discussed in detail in Sec. III D. In general terms, however, collisions result in a decrease in the GSE signal if $\kappa\delta u T \gg 1$.

D. High-pressure regime

The high-pressure regime is defined by

$$r\equiv\Gamma_i'/(2\kappa u)=\Gamma_i(\delta u_i/u)^2/(4\kappa u)\gg 1\ . \quad (47)$$

Condition (47) can be interpreted as a requirement that the effective mean free path is much smaller than the wavelength $\lambda_{\text{eff}}=|\mathbf{k}_1-\mathbf{k}_2|/k_1k_2$ associated with two-photon transitions involving fields 1 and 2. In stationary spectroscopy of two-photon transitions, this would be a regime of collisional narrowing of the two-photon spectral profile (absorption or emission). As is shown below, in transient spectroscopy this pressure regime can correspond to a revival of the GSE signal.

Owing to conditions (7c) and (47), it follows that $\Gamma_i'T\gg 1$. If, in addition, it is assumed that

$$\Gamma_i'/\gamma_3\gg 1\ , \quad (48)$$

the thermalization in both the ground and excited states can be considered to be complete in the time interval T_{42}. All memory of the initial velocity is lost on a time scale of order $(\Gamma_i')^{-1}\ll\gamma_3^{-1}$. Therefore, for times $t\gg\gamma_3^{-1}$, the propagator $G_i(\mathbf{v},\mathbf{v}';t;+)$ can be written as

$$G_i(\mathbf{v},\mathbf{v}';t;+)=W_0(\mathbf{v})G_i(t)[1+O(r^{-1})]\ , \quad (49)$$

where G_i is a function that is determined below.

Since the atomic velocity distribution is thermalized on the time scale of the experiment, it is no longer valid to use a difference kernel to describe collisional relaxation. On the other hand, one *can* use a diffusion model to describe collisions in the time interval T_{42} since the collisional mean free path is much smaller than the characteristic distance [of order $(2\kappa u)^{-1}$] over which $\rho_{ii}(\mathbf{R},\mathbf{v},t)$ varies when condition (47) holds. In the diffusion model, one writes the atomic current density $\mathbf{j}_i$ as

$$\mathbf{j}_i(\mathbf{R},t)=-D_i\nabla\rho_{ii}(\mathbf{R},t)\ , \quad (50)$$

where D_i is the diffusion coefficient for scattering of atoms in level i by the perturbers and $\mathbf{j}_i(\mathbf{R},t)$ and $\rho_{ii}(\mathbf{R},t)$ are velocity-averaged quantities. Combining the divergence of Eq. (50) with the equation of continuity, one finds that the collisional time rate of change of $\rho_{ii}(\mathbf{R},t)$ is given by

$$\partial\rho_{ii}(\mathbf{R},t)/\partial t=D_i\nabla^2\rho_{ii}(\mathbf{R},t)\ . \quad (51)$$

In the intervals T_{32} and T_{43}, $\rho_{ii}(\mathbf{R},\mathbf{v},t;\pm)$ varies as $\exp(\pm 2i\boldsymbol{\kappa}\cdot\mathbf{R})$, so that the collisional decay of $\rho_{ii}(\mathbf{R},t;\pm)$ resulting from *spatial* diffusion is given by

$$\partial\rho_{ii}(\mathbf{R},t;\pm)/\partial t=-d_i\rho_{ii}(\mathbf{R},t;\pm)\ , \quad (52)$$

where

$$d_i=4\kappa^2D_i\ . \quad (53)$$

It then follows from Eqs. (26), (27), (30), and (52) that the $G_i(t)$ satisfy

$$dG_1(t)/dt=-(\gamma_1+d_1)G_1(t)-\gamma_{3,1}G_3(t)\ , \quad (54a)$$

$$dG_3(t)/dt=-(\gamma_3+d_3)G_3(t)\ . \quad (54b)$$

Equations (49) and (54) also follow from a rigorous solution of the Fokker-Planck equation in phase space [17], when the limit $\Gamma_i'\gg T^{-1}$, γ_3, is taken. Moreover, they can be obtained directly from Eqs. (27) by using a trial solution of the form

$$G_i(\mathbf{v},\mathbf{v}';t;+)$$
$$=G_i(t)W_0(\mathbf{v})[1+i(\boldsymbol{\kappa}\cdot\mathbf{v}/\kappa u)g_i+O(r^{-2})]\ , \quad (55)$$

where g_i is a constant of order $r^{-1}\ll 1$ [18].

To zeroth order in r^{-1}, the final expression for $G_1(\mathbf{v},\mathbf{v}';t;+)$ obtained from Eqs. (49), (54), and (53) is

$$G_1(\mathbf{v},\mathbf{v}';T;+)=\frac{\gamma_{3,2}+4\kappa^2(D_3-D_1)}{\gamma_3+4\kappa^2(D_3-D_1)}$$
$$\times W_0(\mathbf{v})\exp[-(\gamma_1+4\kappa^2D_1)T]\ , \quad (56)$$

where Eq. (5) has been used. The diffusion coefficient is related to the effective collision rate Γ' by [19]

$$D_i=u^2/(2\Gamma_i')\ . \quad (57)$$

At very high pressures where $D_i\sim 0$, $G_1(\mathbf{v},\mathbf{v}';T;+)$ is restored to its collision-free value.

IV. DISCUSSION

A. Low pressure—decay

The low-pressure limit is defined by condition (38). To avoid any collisional decay associated with the Doppler phase acquired by the optical dipoles in the interval between the first and second pulses, and following the final pulse, it is assumed that

$$\Gamma_iT(k\delta uT_{\text{ph}})^2\ll 1\ , \quad (58)$$

where

$$T_{\text{ph}}=\text{Max}(T_{21},t-T_4)\ . \quad (59)$$

The signal is proportional to $|S(t)|^2$, where $S(t)$, as given by Eq. (42), depends on the product $\mathcal{G}_1\{[2\kappa T+\mathbf{k}_1(t-T_4)];T;+\}\mathcal{G}_1[\mathbf{k}_1(t-T_4);T;+]$, with $\mathcal{G}_1(\beta;T;+)$ given by Eqs. (44)–(46). Each of the $\mathcal{G}_1(\beta;T;+)$'s appearing in Eq. (42) contains an exponential factor. When condition (58) holds, the exponential factor is the same for both terms and is equal to [see Eqs. (44)–(46)]

$$E(\kappa,T)\equiv\exp\left[-\left\{\gamma_1+\Gamma_1\left[1-(T)^{-1}\int_0^T d\tau\int d\bar{\mathbf{v}}\,F_1(\bar{\mathbf{v}})\exp(2i\boldsymbol{\kappa}\cdot\bar{\mathbf{v}}\tau)\right]\right\}T\right] . \tag{60}$$

The exponential decay of the signal depends on the magnitude of $\kappa\delta uT$, where δu is $\sqrt{2}u$ times the (one-dimensional) rms velocity change per collision. This phase factor is related to the collision-induced change in the phase associated with the modulated ground-state population. If $2\kappa\delta uT\ll 1$, Eq. (60) becomes

$$E(\kappa,T)\equiv\exp(-\{[\gamma_1T+(\tfrac{1}{3})\Gamma_1T(\kappa\delta uT)^2]\}) . \tag{61}$$

On the other hand, if $\kappa\delta uT\gg 1$, any collision destroys the echo signal and only those atoms which do not collide in a time T contribute to the signal. As a consequence, the exponential factor reduces to

$$E(\kappa,T)\equiv\exp[-(\gamma_1+\Gamma_1)T] . \tag{62}$$

For arbitrary values of $\kappa\delta uT$, the exponential factor is directly related to the Fourier transform of the collision kernel through Eq. (60). As such, one can probe the collision kernel by varying either κ (by varying the angle between $\mathbf{k}_1$ and $\mathbf{k}_2$) or varying T. The signal intensity varies as $|E(\kappa,t)|^4$.

As was noted previously [1], the GSE becomes an extremely sensitive probe of weak velocity changes when one chooses $\mathbf{k}_1$ and $\mathbf{k}_2$ to be counterpropagating such that $\kappa=2k_1$. In that case, one can measure velocity changes as small as $\gamma_1/2\kappa$, where γ_1 is determined by the rate at which atoms leave the interaction volume.

In addition to the exponential factors, each of the $\mathcal{G}_1(\beta;T;+)$'s contains an amplitude factor given by

$$A(\boldsymbol{\beta},\boldsymbol{\kappa},T)=1-\gamma_{3,1}\int_0^T d\tau\exp\{[\gamma_1(\boldsymbol{\beta},\boldsymbol{\kappa},\tau)-\gamma_3(\boldsymbol{\beta},\boldsymbol{\kappa},\tau)]\tau\} , \tag{63}$$

where $\gamma_i(\boldsymbol{\beta},\boldsymbol{\kappa},\tau)$ is defined by (45) and (46). The amplitude factors reflect the degree to which the system has been opened by spontaneous decay or collisions. The system must open on a time scale of order γ_3^{-1}. This is seen in Eq. (63), where only times $\tau\le\gamma_3^{-1}$ contribute to the integral. After that time, the atom is back in its ground state. The ground-state population must retain some memory of the spatial modulation if the GSE signal is to be nonvanishing.

If $\gamma_{3,2}\neq 0$, spontaneous emission leads to an opening of the system on a time scale γ_3^{-1} of order $\gamma_{3,2}/\gamma_3$. Collisions open the system only if, on a time scale of order γ_3^{-1}, the velocity changes in the intervals T_{32} and T_{43} lead to significant changes in the Doppler phase acquired by the optical dipoles or the spatially modulated populations. This condition is different for the amplitudes corresponding to the time intervals T_{32} and T_{43} for which

$$\boldsymbol{\beta}_1\equiv\mathbf{k}_1(t-T_4)\quad\text{and}\quad\boldsymbol{\beta}_2\equiv[2\boldsymbol{\kappa}T+\mathbf{k}_1(t-T_4)] , \tag{64}$$

respectively. Let us consider these two terms separately:

$$\boldsymbol{\beta}_1=\mathbf{k}_1(t-T_4) .$$

From Eqs. (45) and (46), it follows that

$$\gamma_i(\boldsymbol{\beta}_1,\boldsymbol{\kappa},\tau)=\gamma_i+\Gamma_i\left[1-(\tau)^{-1}\int_0^\tau d\tau'\int d\bar{\mathbf{v}}\,F_i(\bar{\mathbf{v}})\exp(2i\boldsymbol{\kappa}\cdot\bar{\mathbf{v}}\tau'-i\boldsymbol{\beta}_1\cdot\bar{\mathbf{v}})\right] . \tag{65}$$

One sees the contribution from the collision-induced change in the phase of the optical dipoles [$\boldsymbol{\beta}_1\cdot\mathbf{v}=\mathbf{k}_1\cdot\mathbf{v}(t-T_4)$ term] and that from the phase acquired owing to the spatial modulation of the populations ($2\boldsymbol{\kappa}\cdot\mathbf{v}\tau'$ term). As an example, consider the limiting case in which

$$\kappa\delta u/\gamma_3\ll 1 ,\quad \mathbf{k}\delta uT_{\text{ph}}\ll 1 ,\quad \Gamma_i(k_1\delta u)^2/\gamma_3^3\ll 1 . \tag{66}$$

In this limit, it follows from Eq. (65) and (63) that

$$\gamma_i(\boldsymbol{\beta}_1,\boldsymbol{\kappa},\tau)=\gamma_i+[\Gamma_i(\delta u)^2/4\tau]\times\int_0^\tau d\tau'|2\boldsymbol{\kappa}\tau'-\boldsymbol{\beta}_1|^2 . \tag{67}$$

and

$$A(\boldsymbol{\beta}_1,\boldsymbol{\kappa},T)=(\gamma_{3,2}/\gamma_3) +(\gamma_{3,1}/\gamma_3^2)[\Gamma_3(\delta u_3)^2-\Gamma_1(\delta u_1)^2] \times[2(\kappa/\gamma_3)^2-(\boldsymbol{\beta}_1\cdot\boldsymbol{\kappa}/\gamma_3)+(\beta_1)^2/4] . \quad (68)$$

If κ is of order k_1, the terms involving $\boldsymbol{\beta}_1=\mathbf{k}_1(t-T_4)$ can be dropped owing to condition (7d) [20]. If, instead of the limit (66), one takes

$$k_1\delta u/\gamma_3 \gg 1 , \quad (69)$$

then it follows from Eqs. (63) and (65) that

$$A(\boldsymbol{\beta}_1,\boldsymbol{\kappa},T)=[\gamma_{3,2}+\Gamma_3-\Gamma_1]/[\gamma_3+\Gamma_3-\Gamma_1] . \quad (70)$$

If $\gamma_{3,2}=0$ (closed system) it may be possible to see the collisional opening of the system; however, the collisional terms are always small in regions where the exponential factor $E(\kappa,T)$ is non-negligible—the same collisional mechanism that is responsible for opening the system also leads to decay of the GSE signal. To observe such small collisional terms, the time T must be chosen sufficiently large to ensure that they are larger than terms of order $\exp(-\gamma_3 T)$ which contribute to the signal but have been neglected throughout this discussion.

$$\boldsymbol{\beta}_2=[2\boldsymbol{\kappa}T+\mathbf{k}_1(t-T_4)] .$$

In this case,

$$\gamma_i(\boldsymbol{\beta}_2,\boldsymbol{\kappa},\tau)=\gamma_i+\Gamma_i\left[1-(\tau)^{-1}\int_0^\tau d\tau'\int d\bar{\mathbf{v}}\,F_i(\bar{\mathbf{v}})\exp\{2i\boldsymbol{\kappa}\cdot\bar{\mathbf{v}}(\tau'-T)-i\boldsymbol{\beta}_1\cdot\bar{\mathbf{v}}\}\right] . \quad (71)$$

This value of γ_i corresponds to the interval between the third and fourth pulses and differs qualitatively from the value given in Eq. (65) corresponding to the interval between the second and third pulses. The origin of the difference can be traced to the fact that the modulated population has acquired a phase $2\boldsymbol{\kappa}\cdot\mathbf{v}T$ at the time of the application of the third pulse. This phase factor can be seen in Eq. (71). With increasing τ', the phase proportional to $\boldsymbol{\kappa}\cdot\mathbf{v}$ decreases; however, since the maximum τ' which contributes is of order γ_3^{-1} and since $\gamma_3 T\gg 1$, the phase factor does not change significantly from its value at $\tau'=0$. In fact, it is possible to use inequalities (7b), (7c), and (58) to show that corrections to the phase factor for times $\tau'\neq 0$ can be ignored, so that

$$\gamma_i(\boldsymbol{\beta}_2,\boldsymbol{\kappa},\tau)=\gamma_i+(2\pi)^{3/2}\Gamma_i\mathcal{F}_i(\beta_2) . \quad (72)$$

One can write the amplitude factor $A(\beta_2,T)$ as

$$A(\beta_2,T)=[\gamma_{3,2}+Y_3(\beta_2)-Y_1(\beta_2)]/[\gamma_3+Y_3(\beta_2)-Y_1(\beta_2)] , \quad (73)$$

where

$$Y_i(\beta)=(2\pi)^{3/2}\Gamma_i\mathcal{F}_i(\beta) . \quad (74)$$

For $\kappa\delta uT\gg 1$, $Y_i(\beta)\simeq\Gamma_i$.

To summarize these low-pressure results, one writes the signal amplitude $S(t)$ as

$$S(t)=A(\boldsymbol{\beta}_1,\boldsymbol{\kappa},T)A(\boldsymbol{\beta}_2,\boldsymbol{\kappa},T)|E(\boldsymbol{\kappa},T)|^2\exp\{-[k_1u(t-T_4-T_{21})]^2/4\} , \quad (75)$$

where $A(\boldsymbol{\beta}_1,\boldsymbol{\kappa},T)$ is given by Eqs. (68) or (70), $A(\boldsymbol{\beta}_2,\boldsymbol{\kappa},T)$ by Eq. (73), and $E(\boldsymbol{\kappa},T)$ by Eq. (60). The echo occurs at $(t-T_4)=T_{21}$, and its decay properties are determined by $E(\boldsymbol{\kappa},T)$ as described above. The collisional contributions to the amplitudes are small whenever $E(\boldsymbol{\kappa},T)$ is not negligibly small (the collisional mechanism which opens the system is also responsible for the decay of the signal). As a consequence, if $(\gamma_{3,2}/\gamma_3)$ is of order unity, the product of amplitudes can be approximated as

$$A(\boldsymbol{\beta}_1,\boldsymbol{\kappa},T)A(\boldsymbol{\beta}_2,\boldsymbol{\kappa},T)\simeq(\gamma_{3,2}/\gamma_3)^2 . \quad (76)$$

On the other hand, for a system that is closed in the absence of collisions ($\gamma_{3,2}=0$), the product of the amplitudes is equal to

$$A(\boldsymbol{\beta}_1,\boldsymbol{\kappa},T)A(\boldsymbol{\beta}_2,\boldsymbol{\kappa},T)=(\gamma_3)^{-1}[\Gamma_3(\delta u_3)^2-\Gamma_1(\delta u_1)^2][2(\kappa/\gamma_3)^2-(\boldsymbol{\beta}_1\cdot\boldsymbol{\kappa}/\gamma_3)+(\beta_1)^2/4] \times[Y_3(\beta_2)-Y_1(\beta_2)]/[\gamma_3+Y_3(\beta_2)-Y_1(\beta_2)] \quad (77)$$

for $\kappa\delta u\ll\gamma_3$, and

$$A(\beta_1,\boldsymbol{\kappa},T)A(\beta_2,\boldsymbol{\kappa},T)=[(\Gamma_3-\Gamma_1)/(\gamma_3+\Gamma_3-\Gamma_1)]^2 \quad (78)$$

for $\kappa\delta u\gg\gamma_3$.

B. High pressures—revival

In the high-pressure regime (47), it follows from Eqs. (32), (53), and (56) that

$$S(t)=[(\gamma_{3,2}+d_3-d_1)/(\gamma_3+d_3-d_1)]^2 \times e^{-2(\gamma_1+d_1)T}e^{-(k_1uT_{21})^2/4} \times e^{-[k_1u(T-T_4)]^2/4} \quad (79)$$

where $d_i=4\kappa^2D_i$, and D_i is a diffusion coefficient inversely proportional to the collision rate. Owing to collisions, all velocity correlation between the initial and final intervals is lost. As a consequence the signal is maximum at $(t-T_4)=0$ and can be referred to as a grating free-induction decay [21] rather than an echo. For the signal to be non-negligible, one must choose T_{21} such that $k_1uT_{21}\ll 1$ and $\Gamma_{13}T_{21}\ll 1$, where Γ_{13} is the collision decay rate associated with the optical dipoles [22].

The role of collisions in the time interval T_{42} is reflected in the amplitude and exponential factors in (79). There is a collisional inhibition of the Doppler dephasing associated with the modulated ground-state populations that leads to a revival of the exponential factor when

$$2d_1T=4\kappa^2D_1T =8\kappa^2u^2T/\Gamma_1'=16\kappa^2u^2T/[\Gamma_1'(\delta u_1/u)^2]\leq 1 \ . \quad (80)$$

The amplitude factor contains contributions from both spontaneous decay and collisions, reflecting the opening of the 1-3 "two-level" system by these processes. In analogy with the low-pressure regime, the collisional process that leads to a revival of the system also "recloses" the collisional contribution to the signal, since the atoms have been redistributed over the entire Maxwellian distribution by collisions [14]. Thus, when condition (80) is satisfied, it is easiest to see the revival of the signal when $\gamma_{3,2}\neq 0$. In that case the amplitude of the signal is proportional to $(\gamma_{3,2}/\gamma_3)^2$ [23].

When condition (80) holds, the signal amplitude varies as $\exp(-2\gamma_1T)$. Typically, γ_1 consists of two parts

$$\gamma_1=\gamma_t+\gamma_s \ , \quad (81)$$

where γ_t is the rate at which atoms leave the interaction volume (inverse transit time), and γ_s is the rate at which collisions transfer population from states 1 to 2 (for an alkali-metal ground state, γ_s would represent the rate for spin-flip collisions). For the moment, we neglect γ_s and determine under what conditions one can reasonably hope to see a revival of the signal.

First of all, we must satisfy condition (47) in the high-pressure region. The revival effect is most easily seen if one takes a small angle θ between $\mathbf{k}_1$ and $\mathbf{k}_2$ such that $\kappa=k_1\theta\ll k_1$. The angle θ must be chosen large enough to spatially separate the two beams. I shall assume a value $\theta^2=1.0\times 10^{-3}$ so that $\theta\simeq 0.03$. With this value of θ, an effective collision cross section $\sigma'=\Gamma'/Nu_r$ on the order of 1.0×10^{-14} cm^2 and $u_r=5.0\times 10^4$ cm/s (u_r is the most probable active atom-perturber relative speed), condition (47) is satisfied provided $N\geq 1.0\times 10^{17}$ atoms/cm^3. Next, to avoid transit time damping, we must require that

$$d_1>\gamma_t \ , \quad (82)$$

when $2d_1T\leq 1$; if these conditions are satisfied, all transit time effects can be ignored. The rate γ_t is given by [24]

$$\gamma_t=5.8D_1/R^2 \ , \quad (83)$$

where R is the radius of the excitation beam. It follows from Eqs. (80) and (83) that condition (82) is valid provided $(\kappa R)^2\gg 1$, which is satisfied for k_1 in the optical region of the spectrum, $\theta>0.01$, and $R=1.0$ cm.

If D_1 is written in terms of a diffusion cross section as [25]

$$\sigma_d\equiv(3\sqrt{\pi}/16)u_r/ND_1 \ , \quad (84)$$

condition (80) for the observation of the revival effect becomes

$$(\tfrac{3}{2})\sqrt{\pi}\kappa^2u_rT/(N\sigma_d)\leq 1 \ . \quad (85)$$

For $\sigma_d=4.0\times 10^{-15}$ cm^2 (typical of alkali-metal-atom–rare-gas collisions [25]) and $N=1.0\times 10^{19}$ atoms/cm^3, condition (85) is satisfied provided $T<0.04$ μs, a value that is often consistent with conditions (7b) and (7c) [26]. Thus collisional revival phenomena should be observable.

Let us now return to γ_s. It is interesting to determine the order of magnitude of the cross sections $\sigma_s=\gamma_s/(Nu_r)$ that one can measure using the GSE. In order to be able to measure γ_s, one must have $2\gamma_sT$ of order unity, i.e.,

$$\sigma_s=\gamma_s/Nu_r\simeq(2Nu_rT)^{-1} \ . \quad (86)$$

Since condition (85) must be satisfied simultaneously, one finds that cross sections

$$\sigma_s\geq(3/4)\sqrt{\pi}\kappa^2/(N^2\sigma_d) \quad (87)$$

can be measured. For the values of κ and σ_d given above, and for $N=1.0\times 10^{19}$ atoms/cm^3, cross sections as small as 2.0×10^{-17} cm^2 can be detected [26].

This might seem like a small cross section, but alkali-metal-atom–rare-gas spin-flip cross sections can be as small as 10^{-26} cm^2 [27]. Actually the GSE is not the best method for an optical detection of these cross sections. If one creates an initial ground-state magnetization with a radiation pulse and probes it at some later time T, the interval T is limited only by the atom's interaction time in the beam, i.e.,

$$\gamma_tT\leq 1 \ . \quad (88)$$

Combining conditions (86) and (88) and using Eq. (83), one finds that cross sections

$$\sigma_s\geq(3/4)5.8\sqrt{\pi}/(N^2\sigma_dR^2) \quad (89)$$

can be measured. For $N=1.0\times 10^{21}$ atoms/cm^3, $R=1.0$ cm, and $\sigma_d=4.0\times 10^{-15}$ cm^2, $\sigma_s\geq 2.0\times 10^{-27}$ cm^2. For these parameters, a time delay of order 10 s is needed if the cross section is $\sigma_s=2.0\times 10^{-27}$ cm^2.

ACKNOWLEDGMENTS

I should like to thank B. Dubetsky for many helpful discussions. This work is supported by the National Science Foundation through Grant No. PHY-9113590 and by the U.S. Army Research Office.

[1] B. Dubetsky, P. R. Berman, and T. Sleator, Phys. Rev. A **46**, 2213 (1992).
[2] R. Friedberg and S. R. Hartmann, Laser Phys. **3**, 526 (1993).
[3] J. L. LeGouët and P. R. Berman, Phys. Rev. A **20**, 1105 (1979).
[4] T. W. Mossberg, R. K. Kachru, E. Whittaker, and S. R. Hartmann, Phys. Rev. Lett. **43**, 851 (1979).
[5] R. Kachru, T. W. Mossberg, E. Whittaker, and S. R. Hartmann, Opt. Commun. **31**, 223 (1979).
[6] M. Kasevich and S. Chu, Phys. Rev. Lett. **67**, 177 (1991).
[7] See, for example, D. Suter and J. Mlynek, in *Advances in Magnetic and Optical Resonance,* edited by W. S. Warren (Academic, New York, 1992), Vol. 16, p. 1, and references therein.
[8] R. H. Dicke, Phys. Rev. **89**, 472 (1953); P. W. Anderson, J. Phys. Soc. Jpn. **9**, 316 (1954); R. Kubo, *ibid.* **9**, 935 (1954).
[9] In order to observe collisional narrowing on a given transition, the collisional interaction cannot differ appreciably for the transition levels. This condition is not normally satisfied for optical transitions, but is automatically satisfied for two-photon transitions if the initial and final states are identical or belong to the same ground-state manifold.
[10] See, for example, J. F. Lam, D. G. Steel, and R. A. McFarlane, Phys. Rev. Lett. **49**, 1628 (1992); **56**, 1679 (1986); L. J. Rothberg and N. Bloembergen, Phys. Rev. A **30**, 820 (1984); J. Liu and D. G. Steel, *ibid.* **38**, 4639 (1988).
[11] Collisional narrowing has also been predicted (V. I. Barantsov, L. T. Bolotskikh, and A. K. Popov, Zh. Eksp. Teor. Fiz. **66**, 866 (1974) [Sov. Phys. JETP **39**, 419 (1974)]; V. I. Barantsov and A. K. Popov, Pis'ma Zh. Eksp. Teor. Fiz. **24**, 555 (1976) [Sov. Phys. JETP Lett. **24**, 512 (1976)]) and observed [S. Saoudi, Ch. Lerminiaux, and M. Dumont, Phys. Rev. Lett. **56**, 2164 (1986)] on Raman transitions, and has been observed in Rayleigh scattering [A.-M. Cazabat and J. Larour, J. Phys. (Paris) **36**, 1209 (1975)].
[12] J. F. Lam, D. G. Steel, and R. A. McFarlane, in *Laser Spectroscopy VII,* edited by T. W. Hansch and Y. R. Shen (Springer, Berlin, 1985), p. 279.
[13] P. R. Berman, D. G. Steel, G. Khitrova, and J. Liu, Phys. Rev. **38**, 252 (1988).
[14] M. Gorlicki, P. R. Berman, and G. Khitrova, Phys. Rev. A **37**, 4340 (1988).
[15] J. Guo, P. R. Berman, B. Dubetsky and P. R. Berman, Phys. Rev. A **46**, 1426 (1992).
[16] See, for example, P. R. Berman, Phys. Rev. **9**, 2170 (1973).
[17] S. Chandrasekhar, Rev. Mod. Phys. **15**, 1 (1943).
[18] The function g_i is strictly constant only if $\int d\mathbf{v}' W_i(\mathbf{v}',\mathbf{v})(\mathbf{v}-\mathbf{v}')$ is a linear function of $\mathbf{v}$ as it is for a Keilson-Storer kernel [J. Keilson and J. E. Storer, Q. Appl. Math. **10**, 243 (1952)] or for a small perturber to active atom mass ratio. Even if the g_is are not constant, Eqs. (53) and (54) retain their validity, but the value of the diffusion coefficient will differ slightly from that calculated using lowest-order Chapman-Enskog theory (see Refs. [19], [24], and [25]).
[19] P. R. Berman, J. E. M. Haverkort, and J. P. Woerdman, Phys. Rev. A **34**, 4647 (1976).
[20] Equation (68) corrects an error in Eq. (8) of Ref. 1. In Eq. (8) of Ref. 1, the terms in the amplitude proportional to κ were omitted. As is noted in the text, the κ term dominates the β_1 term when κ is of order k_1.
[21] B. Dubetsky and P. R. Berman (unpublished).
[22] The restrictions on T_{21} are not critical since one can let T_{21} go to zero (see Refs. 2, 4, 5, and 21).
[23] In the case of magnetic orientation or alignment, collisions destroy all excited-state magnetic coherence in the high-pressure limit. As a consequence, the "in" term no longer contributes to the ground-state magnetic coherence and the amplitude factor is unity. On the other hand, at low pressures, the ratio $\gamma_{3,2}/\gamma_3$ can be positive or negative, when $\gamma_{3,2}$ is used to simulate a decay rate for magnetic coherence (see Ref. 13). As a result, the revived signal may actually be larger than the low-pressure signal in certain cases.
[24] J. E. M. Haverkort, J. P. Woerdman, and P. R. Berman, Phys. Rev. A **36**, 5251 (1987).
[25] W. A. Hamel, J. E. M. Haverkort, H. G. C. Werij, and J. P. Woerdman, J. Phys. B **19**, 4127 (1986).
[26] The density must be chosen consistent with conditions (7e) for a given pulse duration (limitations posed by T_{21} are not fundamental—see Ref. [22]). Condition (3) requiring the pulse to fill the sample is not critical to the calculation. What *is* important is that the two beams of pulse-3 overlap in the sample. For small angles θ between the beams, this does not lead to restrictions on the pulse duration. The pulse duration can then be chosen sufficiently small to guarantee that $\Gamma_{13}\tau_p \leq 1$. For $\tau_p = 1.0$ ps, this condition restricts the density to $N \leq 1.0\times10^{20}$ or 10^{21} atoms/cm^3, which is also the limit of validity of the binary collision approximation. The value $N=1.0\times10^{19}$ atoms/cm^3 used in the examples is consistent with all the conditions stated in the text. If, instead, a value of $N=1.0\times10^{21}$ atoms/cm^3 were used, this would reduce the lower limit on σ_d to 2.0×10^{-21} cm^2 and change the condition on T to $T<4.0$ μs, for which conditions (7b) and (7c) are easily satisfied.
[27] For a review, see W. Happer, Rev. Mod. Phys. **44**, 169 (1972). Additional complications that can arise in determining ground-state depolarization rates are discussed in this paper.