

## Comparison of recoil-induced resonances and the collective atomic recoil laser

P. R. Berman

*Physics Department, University of Michigan, Ann Arbor, Michigan 48109-1120*

(Received 23 July 1998)

The theories of recoil-induced resonances (RIR) [J. Guo, P. R. Berman, B. Dubetsky, and G. Grynberg, *Phys. Rev. A* **46**, 1426 (1992)] and the collective atomic recoil laser (CARL) [R. Bonifacio and L. De Salvo, *Nucl. Instrum. Methods Phys. Res. A* **341**, 360 (1994)] are compared. Both theories can be used to derive expressions for the gain experienced by a probe field interacting with an ensemble of two-level atoms that are simultaneously driven by a pump field. It is shown that the underlying formalisms of the RIR and CARL are equivalent. Differences between the RIR and CARL arise because the theories are typically applied for different ranges of the parameters appearing in the theory. The RIR limit is one in which the time derivative of the probe field amplitude,  $dE_2/dt$ , depends locally on  $E_2(t)$  and the gain depends linearly on the atomic density, while the CARL limit is one in which  $dE_2/dt = \int_0^t f(t,t')E_2(t')dt'$ , where  $f$  is a kernel, and the gain has a nonlinear dependence on the atomic density. Validity conditions for the RIR or CARL limits are established in terms of the various parameters characterizing the atom-field interaction. The probe gain for a probe-pump detuning equal to zero is analyzed in some detail, in order to understand how gain arises in a system which, at first glance, appears to have a symmetry that would preclude the possibility for gain. Moreover, it is shown that these calculations, carried out in perturbation theory, have a range of applicability beyond the recoil problem. Experimental possibilities for observing CARL are discussed. [S1050-2947(99)05601-2]

PACS number(s): 42.50.Vk, 32.80.Lg, 32.80.Pj, 42.50.Md

### I. INTRODUCTION

In recent years, there has been increased interest in spectral features that can be attributed directly to the recoil atoms undergo on the absorption, emission, or scattering of radiation. Among topics that have been discussed that fall into this category are recoil-induced resonances (RIR) [1–9] and the collective atomic recoil laser (CARL) [10–13]. These processes appear to have much in common, although they are described quite differently. It is the purpose of this article to compare the RIR and CARL and to demonstrate that the underlying *formalisms* of the RIR and CARL are equivalent. The reason that this equivalence is not readily apparent is twofold. First, the RIR have been discussed using a density matrix approach in the Schrödinger picture, whereas CARL has been discussed using an operator approach in the Heisenberg picture. Secondly, the RIR and CARL have generally been examined for different ranges of the various parameters characterizing the atom-field interactions. In order to facilitate the discussion, it is useful to review briefly the RIR and CARL.

Since both the RIR and CARL relate to probe field absorption or gain in the presence of a pump field, it is perhaps best to recall the features of the probe absorption or gain spectrum, *neglecting all effects associated with recoil*. Consider an ensemble of two-level atoms interacting with both a pump and probe field. The probe absorption or gain is monitored as a function of the probe-pump detuning  $\delta$  for various pump field strengths, characterized by the pump field Rabi frequency  $\chi_1$ . It is assumed that all collisional effects can be neglected and that the two-level atomic system is *closed* in the sense that the sum of ground- and excited-state populations is conserved for *each* velocity subclass of atoms. Moreover, it is assumed that the pump field detuning  $\Delta$  from

atomic resonance is much larger than any decay rates, Rabi frequencies, or Doppler shifts in the problem. In this limit, the probe absorption spectrum consists of three features, centered near  $\delta = 0, \pm \Delta$  [14].

Of primary concern here is the structure centered near  $\delta = 0$ . In the absence of recoil, this line feature has a dispersionlike shape, is proportional to  $\chi_1^4$ , and has a width of order of the excited-state decay rate [15]. Its origin can be traced to an interference effect involving processes in which *two* photons are emitted into previously unoccupied vacuum mode states [16]. As such, it is linked to spontaneous emission, rather than to a conservative exchange of energy between the pump and probe fields. The fact that the two-level system is “closed” plays a critical role here. Had the system been “open,” the amplitude of this component would vary as  $\chi_1^2$  rather than  $\chi_1^4$  and its width could be determined by some effective ground-state decay rate (e.g., inverse transit time) rather than the excited-state decay rate [17].

The RIR refer to a class of spectroscopic features in nonlinear spectroscopy that can be attributed to a recoil-induced “opening” of an otherwise closed, two-level system [1]. In the presence of recoil, the atomic velocity is changed on the absorption or emission of radiation. As a consequence the sum of ground- and excited-state populations for specific velocity classes is no longer conserved. In the limit of large detuning  $\Delta$ , the RIR can be interpreted as arising from Raman transitions between different center-of-mass states [1]. If the pump and probe fields have propagation vectors  $\mathbf{k}_1$  and  $\mathbf{k}_2$ , respectively, then probe absorption occurs on a Raman transition between center-of-mass momentum states  $|\mathbf{P}\rangle$  and  $|\mathbf{P} - \hbar\mathbf{q}\rangle$  and gain between states  $|\mathbf{P}\rangle$  and  $|\mathbf{P} + \hbar\mathbf{q}\rangle$ , where  $\mathbf{q} = \mathbf{k}_1 - \mathbf{k}_2$ . Depending on the detuning  $\delta$  between the probe and pump fields, one of these processes is favored. For  $\delta$

$<0$ , there is probe gain, for  $\delta > 0$ , there is probe absorption, and for  $\delta = 0$ , the nonlinear probe absorption and gain vanish. In general, the RIR have been formulated as a *stationary* process in which the field achieves a steady-state value at each position  $\mathbf{R}$ —gain occurs as the probe field propagates through the medium. Steady state is achieved by assuming that there is some effective ground-state decay rate or, alternatively, by assuming that the Doppler width associated with the Raman transitions,  $qP_0/M$ , where  $M$  is an atomic mass and  $P_0$  is the width of the momentum distribution, is larger than the recoil frequency,  $\omega_q = \hbar q^2/2M$  [6,8,18]. The gain is normally expressed as  $dE_2/dZ = \alpha E_2$ , where  $E_2$  is the probe field's amplitude and  $\alpha$  is a constant proportional to the atomic density. The point to note here is that the derivative of  $E_2$  depends *locally* on  $E_2$ . Transient theories of the RIR have also been given [6,8,18]; the field dependence in these theories is also taken to be local.

CARL has been formulated as a *transient* problem. It is assumed that the pump and probe fields are modes of an optical cavity in which the atoms are located. As a result of the atom-field interaction, the probe field builds up in the cavity as a function of time. In the limit of large  $\Delta$ , one obtains an equation of the form  $dE_2/dt = \beta \int_0^t f(t,t') E_2(t') dt'$ , where  $\beta$  is a constant proportional to the atomic density and  $f$  is a kernel. In general, the time derivative of  $E_2$  can depend on the past history of the field, rather than locally on the field. This leads to very different dynamics than those encountered in the RIR. One finds threshold conditions for probe gain that depend nonlinearly on the atomic density, as does the gain itself. CARL is a cooperative effect in the sense that gain occurs only for some minimum atomic density for most values of the detuning  $\delta$ . It is possible to have gain for  $\delta \geq 0$ , which is not possible for the RIR.

In certain limits, the CARL equation for  $dE_2/dt$  becomes local and the gain characteristics of CARL become identical to those of the RIR. Thus the CARL equations contain the RIR results as a limiting case. Bonifacio and Verkerk [11] and Bonifacio *et al.* [12] have shown that the CARL equations reproduce the RIR results in the limit of large Doppler width,  $qP_0/M \gg \omega_q$ ; however, the approach they follow differs considerably from the one to be presented herein. On the other hand, as normally formulated, the RIR theories can never reproduce the nonlocal field dependence encountered in CARL. If a somewhat more general derivation of the RIR equations is undertaken, however, as is done in Sec. III, one can obtain equations that are totally equivalent to the CARL equations. Although the RIR and CARL formalisms are equivalent, it is convenient to distinguish the RIR and CARL *limits* of these equations. The RIR limit is defined as one in which the time derivative of the probe field amplitude,  $dE_2/dt$ , depends locally on  $E_2(t)$  and the gain depends linearly on the atomic density, while the CARL limit is defined as one in which  $dE_2/dt$  depends nonlocally on  $E_2(t)$  and the gain has a nonlinear dependence on the atomic density.

In Sec. II the basic equations are derived. The RIR and CARL limits of these equations are obtained in Sec. III. The case of equal pump and probe frequencies is analyzed in Sec. IV using an effective five-level atom. When expressed in this form the calculation has a range of applicability beyond the recoil problem. The results are discussed in Sec. V. Special

emphasis is placed on the distinction between ‘‘atom bunching’’ and ‘‘matter gratings.’’ Experimental implications of the results are also explored. Although the general theory is developed for arbitrary field strengths, the specific calculations are carried out in a perturbation theory limit.

## II. BASIC EQUATIONS

The problem under consideration consists of a pump field and a probe field interacting with an ensemble of two-level atoms. In some applications in CARL, it may be of interest to use a quantized description of the fields to follow the buildup of the probe field from noise, but, in the present discussion, the fields are taken to be classical. The pump field,

$$\mathbf{E}_1(\mathbf{R}, t) = \frac{1}{2} \boldsymbol{\epsilon}_1 [E_1(\mathbf{R}, t) e^{i(\mathbf{k}_1 \cdot \mathbf{R} - \Omega_1 t)} + E_1^*(\mathbf{R}, t) e^{-i(\mathbf{k}_1 \cdot \mathbf{R} - \Omega_1 t)}], \quad (1)$$

has polarization  $\boldsymbol{\epsilon}_1$ , slowly varying electric field amplitude  $E_1(\mathbf{R}, t)$ , propagation vector  $\mathbf{k}_1$ , and frequency  $\Omega_1 = k_1 c$ , while the probe field,

$$\mathbf{E}_2(\mathbf{R}, t) = \frac{1}{2} \boldsymbol{\epsilon}_2 [E_2(\mathbf{R}, t) e^{i(\mathbf{k}_2 \cdot \mathbf{R} - \Omega_2 t)} + E_2^*(\mathbf{R}, t) e^{-i(\mathbf{k}_2 \cdot \mathbf{R} - \Omega_2 t)}], \quad (2)$$

has polarization  $\boldsymbol{\epsilon}_2$ , slowly varying electric field amplitude  $E_2(\mathbf{R}, t)$ , propagation vector  $\mathbf{k}_2$ , and frequency  $\Omega_2 = k_2 c$ . As a result of the nonlinear interaction with the fields, the probe field can be amplified. If the pump field is initially much more intense than the probe field, as is assumed, pump depletion during the early stages of probe amplification can be neglected. Since the calculation in this paper is limited to the early stages of probe amplification, I take the pump field amplitude to be constant,  $E_1(\mathbf{R}, t) \sim E_1$ .

The dynamics of probe field amplification depends on the specific atom-field geometry. One can envision situations in which the probe field amplitude is constant in time but varies in space, or is constant in space but varies in time. The first case is the one considered generally in the RIR, in which cw pump and probe fields interact with atoms in a cell or trap. The probe field amplitude increases in the direction of  $\mathbf{k}_2$  as it propagates through the medium, but is assumed to have evolved to a stationary state. The second case is the one considered generally in CARL, in which the fields correspond to field modes of an optical cavity and the atoms are located in the cavity. For a ring cavity, the probe field intensity is spatially isotropic, but increases in time [12]. In certain limits (to be noted below), the spatial gain coefficient of the RIR multiplied by the speed of light coincides with the temporal gain coefficient of CARL. In other limits, the dynamics of CARL amplification differs from the spatial buildup of the field in the RIR. To compare the RIR and CARL, it is convenient to adopt the cavity model and assume that the probe field amplitude is a function of time only,  $E_2(\mathbf{R}, t) \sim E_2(t)$ . All cavity losses are neglected, as is any ground-state decay.

Although CARL is referred to as a collective effect since conditions for CARL gain depend on the atomic density, each atom in the sample, on average, interacts with the fields in an identical manner. Thus it is sufficient to write the Hamiltonian for a single atom interacting with the fields. The dependence of the field gain on atomic density is included automatically in the coupled Maxwell-Bloch equations. In the dipole and rotating wave approximations, the Hamiltonian for our system is

$$H = \frac{P^2}{2M} + \frac{\hbar\omega}{2}\sigma_z + \hbar \sum_{\mu=1}^2 [\chi_{\mu}(t)e^{i(\mathbf{k}_{\mu}\cdot\mathbf{R}-\Omega_{\mu}t)}\sigma_{+} + (\text{adjoint})], \quad (3)$$

where  $\mathbf{P}$  is the atomic center-of-mass momentum,  $\omega$  is the transition frequency between the ground state  $|g\rangle$  and excited state  $|e\rangle$  of the atom,  $\sigma_z = (|e\rangle\langle e| - |g\rangle\langle g|)$ ,  $\sigma_{+} = |e\rangle\langle g|$ ,

$$\chi_{\mu}(t) = -\frac{\boldsymbol{\mu}\cdot\boldsymbol{\epsilon}_{\mu}E_{\mu}(t)}{2\hbar} \quad (4)$$

is the Rabi frequency of field  $\mu$ , and  $\boldsymbol{\mu} \equiv \langle e|\hat{\boldsymbol{r}}|g\rangle$  is a dipole moment matrix element. Terms related to spontaneous emission are not included in the Hamiltonian (3), for reasons to be discussed below. The Hamiltonian determines the time evolution of the atom, but the atomic evolution must be coupled to the field evolution via Maxwell's equations to arrive at a closed set of equations.

The time evolution of the (complex) probe field amplitude is linked to the polarization of the medium which, in turn, is determined by the atom-field interaction. The medium's polarization can be expressed in terms of atomic density matrix elements as

$$\mathbf{P}(\mathbf{R},t) = N[\boldsymbol{\mu}\rho_{ge}(\mathbf{R},t) + \boldsymbol{\mu}^*\rho_{eg}(\mathbf{R},t)], \quad (5)$$

where  $N$  is the total number of atoms. As a result of the nonlinear atom-field interaction, the density matrix element  $\rho_{eg}(\mathbf{R},t)$  can be written quite generally (see below) as

$$\begin{aligned} \rho_{eg}(\mathbf{R},t) &= \tilde{\rho}_{eg}(t;1)e^{i(\mathbf{k}_1\cdot\mathbf{R}-\Omega_1t)} + \tilde{\rho}_{eg}(t;2)e^{i(\mathbf{k}_2\cdot\mathbf{R}-\Omega_2t)} \\ &+ \sum_{n \neq 0,-1} \tilde{\rho}_{eg}(t;1,n)e^{i(\mathbf{k}_1\cdot\mathbf{R}-\Omega_1t) + in(\mathbf{q}\cdot\mathbf{R} + \delta t)}, \end{aligned} \quad (6)$$

where

$$\mathbf{q} = \mathbf{k}_1 - \mathbf{k}_2, \quad \delta = \Omega_2 - \Omega_1. \quad (7)$$

In the slowly varying amplitude and phase approximation, it follows from Maxwell's equations and Eqs. (5) and (6) that the evolution of the probe field is given by

$$\frac{dE_2(t)}{dt} = \frac{iN\Omega_2}{\epsilon_0}\boldsymbol{\epsilon}_2\cdot\mathbf{P}_2(\mathbf{R},t)e^{-i(\mathbf{k}_2\cdot\mathbf{R}-\Omega_2t)} \quad (8a)$$

$$= \frac{iN\Omega_2}{\epsilon_0}\boldsymbol{\epsilon}_2\cdot\boldsymbol{\mu}^*\tilde{\rho}_{eg}(t;2), \quad (8b)$$

where  $\mathbf{P}_2(\mathbf{R},t)$  is the component of the polarization  $\mathbf{P}(\mathbf{R},t)$  varying as  $e^{i(\mathbf{k}_2\cdot\mathbf{R}-\Omega_2t)}$ . Combining Eq. (8b) with Eq. (4) yields

$$\frac{d\chi_2(t)}{dt} = -\frac{iNd^2\Omega_2}{2\hbar\epsilon_0}\tilde{\rho}_{eg}(t;2), \quad (9)$$

where

$$d \equiv |\boldsymbol{\mu}\cdot\boldsymbol{\epsilon}_2|. \quad (10)$$

An equation for  $\tilde{\rho}_{eg}(t;2)$  is obtained by solving the Schrödinger equation for the Hamiltonian (3).

It is convenient to expand the wave function of the system as

$$\psi(\mathbf{R},\mathbf{r},t) = \sum_{i=g,e} A_i(\mathbf{R},t)\psi_i(\mathbf{r}), \quad (11)$$

where

$$A_j(\mathbf{R},t) = \frac{1}{(2\pi\hbar)^{3/2}} \int d\mathbf{P} A_j(\mathbf{P},t) e^{-i\omega_j t} e^{i\mathbf{P}\cdot\mathbf{R}/\hbar} e^{-iE_P t/\hbar}, \quad (12)$$

$\omega_g = -\omega/2$ ,  $\omega_e = \omega/2$ ,  $E_P = P^2/2M$ , and  $\psi_i(\mathbf{r})$  is the atomic-state eigenfunction associated with state  $i$ . Note that the momentum-state amplitudes  $A_i(\mathbf{P},t)$  are defined in an interaction representation. The state amplitudes  $a_j(\mathbf{P},t)$  in the "normal" representation are related to those in the interaction representation by

$$a_j(\mathbf{P},t) = A_j(\mathbf{P},t) e^{-i\omega_j t} e^{-iE_P t/\hbar}, \quad (13)$$

and density matrix elements in momentum space are given by

$$\begin{aligned} \rho_{jj'}(\mathbf{P},\mathbf{P}',t) &= \overline{a_j(\mathbf{P},t)a_{j'}^*(\mathbf{P}',t)} \\ &= \overline{A_j(\mathbf{P},t)A_{j'}^*(\mathbf{P}',t)} e^{-iE_{PP'}t/\hbar} e^{-i\omega_{jj'}t}, \end{aligned} \quad (14)$$

where  $\omega_{jj'} = \omega_j - \omega_{j'}$ ,  $E_{PP'} = E_P - E_{P'}$ , and the bar indicates an ensemble average. When the wave function (11), (12) is substituted into Schrödinger's equation with the Hamiltonian (3), one finds that the state amplitudes  $A_j(\mathbf{P},t)$  evolve as

$$\begin{aligned} \dot{A}_g(\mathbf{P},t) &= -i \sum_{\mu=1,2} \chi_{\mu}^*(t) \exp[i\Delta_{\mu}t - i\omega_{k_{\mu}}t] \\ &\quad - i\mathbf{k}_{\mu}\cdot\mathbf{P}t/M] A_e(\mathbf{P} + \hbar\mathbf{k}_{\mu},t), \end{aligned} \quad (15a)$$

$$\begin{aligned} \dot{A}_e(\mathbf{P},t) &= -i \sum_{\mu=1,2} \chi_{\mu}(t) \exp[-i\Delta_{\mu}t - i\omega_{k_{\mu}}t] \\ &\quad + i\mathbf{k}_{\mu}\cdot\mathbf{P}t/M] A_g(\mathbf{P} - \hbar\mathbf{k}_{\mu},t), \end{aligned} \quad (15b)$$

where

$$\Delta_{\mu} = \Omega_{\mu} - \omega \quad (16)$$

is an atom-field detuning and

$$\omega_{k_\mu} = \hbar k_\mu^2 / 2M \quad (17)$$

is a frequency associated with atomic recoil. In terms of these state amplitudes, the density matrix element  $\rho_{eg}(\mathbf{R}, t) = A_e(\mathbf{R}, t)A_g^*(\mathbf{R}, t)$  can be written as

$$\rho_{eg}(\mathbf{R}, t) = \frac{1}{(2\pi\hbar)^3} \int d\mathbf{P} d\mathbf{P}' \overline{A_e(\mathbf{P}, t)A_g^*(\mathbf{P}', t)} \times e^{-i\omega t} e^{i(\mathbf{P}-\mathbf{P}') \cdot \mathbf{R}/\hbar} e^{-iE_{PP'}/\hbar}. \quad (18)$$

Together, Eqs. (9), (6), (18), and (15) form a closed set of equations which can be solved numerically to obtain the time evolution of the probe field.

To simplify the analysis of both the RIR and CARL, it is assumed that

$$|\Delta_2| \approx |\Delta_1| \equiv |\Delta| \gg k_\mu u, |\chi_\mu|, \gamma_e, \quad |\delta| \ll |\Delta|, \quad (19)$$

where  $\gamma_e$  is the excited-state decay rate and  $u$  is the most probable atomic speed. If inequalities (19) hold, both the RIR and CARL can be interpreted in terms of *stimulated* processes involving the pump and probe fields—spontaneous emission plays a negligible role. Both the pump and probe fields are turned on and brought adiabatically to their “initial” values at  $t=0$  in a time long compared with  $|\Delta|^{-1}$ , but small compared with all other evolution times in the problem. Thus, at  $t=0$ , the values for the Rabi frequencies are

$$\chi_1(0) = \chi_1, \quad \chi_2(0) = \chi_2(0), \quad (20)$$

while the density matrix for the atoms is taken as

$$\varrho_{ij}(\mathbf{P}, \mathbf{P}'; 0) = \overline{a_i(\mathbf{P}, 0)a_j^*(\mathbf{P}', 0)} = \overline{A_i(\mathbf{P}, 0)A_j^*(\mathbf{P}', 0)} = (2\pi\hbar)^3 V^{-1} W(\mathbf{P}) \delta(\mathbf{P}-\mathbf{P}') \delta_{ij} \delta_{ig}, \quad (21)$$

where  $V$  is the sample volume and  $W(\mathbf{P})$  is the initial momentum distribution [19]. This initial density matrix corresponds to a Wigner function  $W(\mathbf{R}, \mathbf{P}; 0) = W(\mathbf{P})/V$ , which is the product of the momentum distribution times a uniform spatial density (recall that this is a single particle density). The calculation can still be carried out using an amplitude picture. The initial condition (21) is invoked once the density matrix element  $\varrho_{ij}(\mathbf{R}, t)$  has been evaluated.

When inequalities (19) hold, it is possible to solve Eq. (15b) adiabatically for the upper state amplitude in terms of the lower state amplitude. Explicitly, one finds

$$A_e(\mathbf{P}, t) = \sum_{\mu=1,2} [\chi_\mu(t)/\Delta] \exp[-i\Delta_\mu t - i\omega_k t + i\mathbf{k}_\mu \cdot \mathbf{P}t/M] A_g(\mathbf{P} - \hbar\mathbf{k}_\mu, t), \quad (22)$$

where  $\omega_k \equiv \omega_{k_1}$  and terms of order  $|(\Delta_1 - \Delta_2)/\Delta_1|$  and  $|(\omega_{k_1} - \omega_{k_2})/\omega_{k_1}|$  have been ignored. Substituting this expression back into Eq. (15a), one obtains

$$\dot{A}_g(\mathbf{P}, t) = -i \sum_{\mu, \nu=1,2} \frac{\chi_\mu(t)\chi_\nu^*(t)}{\Delta} \exp[i\Delta_{\nu\mu} t - i\omega_{k_\nu} t - i\mathbf{k}_{\nu\mu} \cdot \mathbf{P}t/M] A_g(\mathbf{P} + \hbar\mathbf{k}_{\nu\mu}, t), \quad (23)$$

where

$$\Delta_{\nu\mu} = \Delta_\nu - \Delta_\mu, \quad \mathbf{k}_{\nu\mu} = \mathbf{k}_\nu - \mathbf{k}_\mu. \quad (24)$$

Since  $\mathbf{k}_{\nu\mu}$  can take on the values  $[0, \pm\mathbf{q} = \pm(\mathbf{k}_1 - \mathbf{k}_2)]$  only, it is clear that a solution to this equation can be written as

$$A_g(\mathbf{P}, t) = \sum_{n=-\infty}^{\infty} S_n(\mathbf{P}, t) A_g(\mathbf{P} - n\hbar\mathbf{q}, 0). \quad (25)$$

Combining Eqs. (18), (22), (25), and (21), one finds that the density matrix element  $\rho_{eg}(\mathbf{R}, t)$  is given by

$$\rho_{eg}(\mathbf{R}, t) = \sum_{\mu=1,2} \sum_{n, n'=-\infty}^{\infty} \frac{\chi_\mu(t)}{V\Delta} e^{i(\mathbf{k}_\mu \cdot \mathbf{R} - \Omega_\mu t)} e^{i(n-n')\mathbf{q} \cdot \mathbf{R}} \times e^{-i(n^2 - n'^2)\omega_q t} \int d\mathbf{P} W(\mathbf{P}) e^{-i(n-n')\mathbf{P} \cdot \mathbf{q}t/M} \times S_n(\mathbf{P} + n\hbar\mathbf{q}, t) S_{n'}^*(\mathbf{P} + n'\hbar\mathbf{q}, t). \quad (26)$$

Equation (26) proves that the general form for  $\rho_{eg}(\mathbf{R}, t)$  is correctly given by Eq. (6). Extracting the coefficient of  $e^{i(\mathbf{k}_2 \cdot \mathbf{R} - \Omega_2 t)}$  provides us with the value of  $\tilde{\rho}_{eg}(t; 2)$  needed in Eq. (9). Two types of terms in the summation vary as  $e^{i(\mathbf{k}_2 \cdot \mathbf{R} - \Omega_2 t)}$ , those with  $\mu=2$  and  $n=n'$  and those with  $\mu=1$  and  $n'=n+1$ . Denoting the  $n=n'$  term as  $\tilde{\rho}_{eg}(t; a)$ , the  $n'=n+1$  as  $\tilde{\rho}_{eg}(t; b)$ , and using the normalization condition [19]

$$1 = \sum_{i=1,2} \int d\mathbf{R} |A_i(\mathbf{R}, t)|^2 \approx \int d\mathbf{R} |A_g(\mathbf{R}, t)|^2 = \sum_{n=-\infty}^{\infty} \int d\mathbf{P} W(\mathbf{P}) |S_n(\mathbf{P} + n\hbar\mathbf{q}, t)|^2 \quad (27)$$

one finds

$$\tilde{\rho}_{eg}(t; 2) = \tilde{\rho}_{eg}(t; a) + \tilde{\rho}_{eg}(t; b), \quad (28)$$

with

$$\tilde{\rho}_{eg}(t; a) = \frac{\chi_2(t)}{V\Delta} \sum_{n=-\infty}^{\infty} \int d\mathbf{P} W(\mathbf{P}) |S_n(\mathbf{P} + n\hbar\mathbf{q}, t)|^2 \approx \frac{\chi_2(t)}{V\Delta}, \quad (29a)$$

$$\tilde{\rho}_{eg}(t; b) = \frac{\chi_1 e^{i\delta t}}{V\Delta} \sum_{n=-\infty}^{\infty} e^{i(2n+1)\omega_q t} \int d\mathbf{P} W(\mathbf{P}) e^{i\mathbf{P} \cdot \mathbf{q}t/M} \times S_n(\mathbf{P} + n\hbar\mathbf{q}, t) S_{n+1}^*[\mathbf{P} + (n+1)\hbar\mathbf{q}, t]. \quad (29b)$$

The first term represents linear dispersion of the medium. The second term, which is at the heart of the RIR and CARL, represents a scattering of the pump field off the atomic density distribution created by both the pump and probe fields. We now have all the ingredients necessary to derive the RIR and CARL equations.

### III. CARL AND RIR

In the large detuning limit, both the RIR and CARL equations are most easily derived using an effective Hamiltonian for ground-state atoms. An effective Hamiltonian of the form

$$H_{\text{eff}} = \frac{P^2}{2M} + \frac{\hbar}{\Delta} [|\chi_1|^2 + |\chi_2(t)|^2] + \frac{\hbar}{\Delta} [\chi_1 \chi_2^*(t) e^{i(\mathbf{q} \cdot \mathbf{R} + \delta t)} + \chi_1^* \chi_2(t) e^{-i(\mathbf{q} \cdot \mathbf{R} + \delta t)}] \quad (30)$$

leads to the evolution equation (23) for the ground-state amplitude  $A_g(\mathbf{P}, t)$ . The second term in the Hamiltonian is the spatially homogeneous light shift potential and is not of interest here. The third term is the optical potential formed by the pump and probe fields which gives rise to RIR and CARL. The atoms' center-of-mass motion in this optical potential can be treated classically for times  $\omega_q t \ll 1$ . This is the limit considered by Bonifacio and co-workers [10] and will be referred to as CARLSC, with the "SC" denoting the semiclassical limit. For  $\omega_q t \gtrsim 1$ , a quantized description of the motion is needed. A theory of CARL in which the atomic motion is fully quantized has been given recently by Moore and Meystre [13]. It will be referred to as CARLQ, when it is

necessary to distinguish between CARLSC and CARLQ. The theory of the RIR has been developed using a quantized description of the center-of-mass motion; consequently, the RIR and CARLQ theories should produce identical results, and the RIR and CARLSC theories should produce the same results when  $\omega_q t \ll 1$ . It should be noted that, although the atomic motion in the optical potential is treated classically in CARLSC, the gain in CARLSC results from recoil-induced processes. This point is discussed in more detail in Sec. V.

In calculating expectation values of operators that are diagonal in the internal states, one can neglect contributions from the excited state since the excited-state population is assumed to be negligibly small. The CARL equations are derived using the Heisenberg equations of motion while the RIR are derived using a density matrix approach. I now show that these methods yield equivalent results, as they must.

#### A. CARL

To make connection with CARL, one defines a Heisenberg operator, "bunching parameter,"  $B(t) = e^{i\mathbf{q} \cdot \mathbf{R}(t)}$ . Using Eqs. (14), (25), (21), and (29b), one can write the average value of the bunching parameter as [19]

$$\langle B(t) \rangle = \langle e^{i\mathbf{q} \cdot \mathbf{R}(t)} \rangle = \int d\mathbf{P} d\mathbf{P}' \rho_{gg}(\mathbf{P}, \mathbf{P}'; t) \langle \mathbf{P}' | e^{i\mathbf{q} \cdot \mathbf{R}} | \mathbf{P} \rangle \quad (31a)$$

$$= \frac{1}{(2\pi\hbar)^3} \int d\mathbf{R} d\mathbf{P} d\mathbf{P}' \overline{A_g(\mathbf{P}, t) A_g^*(\mathbf{P}', t)} e^{i\mathbf{q} \cdot \mathbf{R}} e^{i(\mathbf{P} - \mathbf{P}') \cdot \mathbf{R}/\hbar} e^{-iE_{PP'} t/\hbar} \quad (31b)$$

$$= \int d\mathbf{P} A_g(\mathbf{P}, t) A_g^*(\mathbf{P} + \hbar\mathbf{q}, t) e^{-iE_{P, |\mathbf{P} + \hbar\mathbf{q}|} t/\hbar} \quad (31c)$$

$$= \sum_{n, n' = -\infty}^{\infty} \int d\mathbf{P} S_n(\mathbf{P}, t) S_{n'}^*(\mathbf{P} + \hbar\mathbf{q}, t) \rho_{gg}[\mathbf{P} - n\hbar\mathbf{q}, \mathbf{P} - (n' - 1)\hbar\mathbf{q}; 0] e^{-iE_{P, |\mathbf{P} + \hbar\mathbf{q}|} t/\hbar} \quad (31d)$$

$$= \sum_{n = -\infty}^{\infty} e^{i(2n+1)\omega_q t} \int d\mathbf{P} W(\mathbf{P}) e^{i\mathbf{P} \cdot \mathbf{q} t/M} S_n(\mathbf{P} + n\hbar\mathbf{q}, t) S_{n+1}^*[\mathbf{P} + (n+1)\hbar\mathbf{q}, t] \quad (31e)$$

$$= \frac{\Delta V e^{-i\delta t}}{\chi_1} \tilde{\rho}_{eg}(t; b), \quad (31f)$$

where the brackets denote a quantum-mechanical average. Therefore, by combining Eqs. (9), (28), (29), and (31f), one finds

$$\frac{d\chi_2(t)}{dt} = -i \frac{\mathcal{N} d^2 \Omega_2}{2\hbar \epsilon_0} \left\{ \frac{\chi_2(t)}{\Delta} + \frac{\chi_1 e^{i\delta t}}{\Delta} \langle B(t) \rangle \right\}, \quad (32)$$

where  $\mathcal{N} = N/V$  is the atomic density. This is one of the basic CARL equations in the limit of large  $\Delta$  [10]. The remaining CARL equations, obtained from the Heisenberg equations of motion with the Hamiltonian (30), are

$$\frac{dB}{dt} = i \frac{\mathbf{q}}{M} \cdot [\mathbf{P}(t) B(t) + B(t) \mathbf{P}(t)] = i \left( \frac{\mathbf{q} \cdot \mathbf{P}(t)}{M} - \omega_q \right) B(t), \quad (33a)$$

$$\frac{d\mathbf{P}}{dt} = -i \frac{\hbar \mathbf{q}}{\Delta} [\chi_1 \chi_2^*(t) e^{i\delta t} B(t) - \chi_2(t) \chi_1^* e^{-i\delta t} B^\dagger(t)], \quad (33b)$$

where the last equality in Eq. (33a) follows from the commutation properties of  $\mathbf{P}(t)$  and  $B(t) = e^{i\mathbf{q} \cdot \mathbf{R}(t)}$ . These equations do not form a closed set since the equation for  $\langle dB(t)/dt \rangle$  involves  $\langle \mathbf{P}(t) B(t) \rangle$ . One must generate a series

of equations for these higher moments or use some approximation techniques (such as assuming classical motion in the effective potential) to obtain a solution to the equations. The linear dispersion parameter  $\alpha = \mathcal{N}d^2\Omega_2/2\hbar\epsilon_0\Delta$  results in a shift of the probe field frequency. By setting  $\chi_2(t) = \tilde{\chi}_2(t)e^{i\delta t}$  in Eqs. (32) and (33), it is easy to see that the detuning  $\delta$  enters only in the combination  $\delta + \alpha$ . Thus one can redefine the detuning  $\delta$  to include the dispersion shift and replace Eq. (32) by

$$\frac{d\chi_2(t)}{dt} = -i \frac{\mathcal{N}d^2\Omega_2\chi_1 e^{i\delta t}}{2\hbar\epsilon_0\Delta} \langle B(t) \rangle. \quad (34)$$

To examine the small signal gain regime, one can solve Eqs. (33) in perturbation theory. To zeroth order in the fields,

$$\mathbf{P}(t) = \mathbf{P}(0) \equiv \mathbf{P}, \quad (35a)$$

$$\begin{aligned} B^{(0)}(t) &= \exp\left\{i\left(\frac{\mathbf{q}\cdot\mathbf{P}}{M} - \omega_q\right)t\right\} B(0) \\ &= B(0) \exp\left\{i\left(\frac{\mathbf{q}\cdot\mathbf{P}}{M} + \omega_q\right)t\right\}, \end{aligned} \quad (35b)$$

$$\begin{aligned} [B^{(0)}(t)]^\dagger &= \exp\left\{-i\left(\frac{\mathbf{q}\cdot\mathbf{P}}{M} + \omega_q\right)t\right\} B^\dagger(0) \\ &= B^\dagger(0) \exp\left\{-i\left(\frac{\mathbf{q}\cdot\mathbf{P}}{M} - \omega_q\right)t\right\}, \end{aligned} \quad (35c)$$

where the last equalities in Eqs. (35b) and (35c) follow from the commutation properties of  $\mathbf{P}$  and  $B(0) = e^{i\mathbf{q}\cdot\mathbf{R}(0)} \equiv e^{i\mathbf{q}\cdot\mathbf{R}}$ . Note that  $\mathbf{P} = \mathbf{P}(0)$  and  $\mathbf{R} = \mathbf{R}(0)$  are normal Schrödinger operators. Substituting Eqs. (35b) and (35c) into Eq. (33b), one finds, to first order in  $|\chi_1\chi_2|$ ,

$$\begin{aligned} \mathbf{P}^{(1)}(t) &= -i \frac{\hbar\mathbf{q}}{\Delta} \int_0^t dt' \\ &\times \begin{pmatrix} \chi_1\chi_2^*(t') e^{i[(\mathbf{q}\cdot\mathbf{P})/M + \delta - \omega_q]t'} B(0) \\ -\chi_2(t')\chi_1^* e^{-i[(\mathbf{q}\cdot\mathbf{P})/M + \delta + \omega_q]t'} B^\dagger(0) \end{pmatrix}, \end{aligned} \quad (36)$$

and, when this result is substituted in Eq. (33a), one obtains

$$\begin{aligned} B^{(1)}(t) &= \frac{2\omega_q e^{-i\delta t}}{\Delta} \int_0^t dt' \exp\left[i\left(\frac{\mathbf{q}\cdot\mathbf{P}}{M} + \delta - \omega_q\right)(t-t')\right] \int_0^{t'} dt'' \\ &\times \begin{pmatrix} \chi_1\chi_2^*(t'') e^{i[(\mathbf{q}\cdot\mathbf{P})/M + \delta - \omega_q]t''} B(0) e^{i[(\mathbf{q}\cdot\mathbf{P})/M + \delta - \omega_q]t'} B(0) \\ -\chi_2(t'')\chi_1^* e^{-i[(\mathbf{q}\cdot\mathbf{P})/M + \delta + \omega_q]t''} B^\dagger(0) e^{i[(\mathbf{q}\cdot\mathbf{P})/M + \delta - \omega_q]t'} B(0) \end{pmatrix} \end{aligned} \quad (37a)$$

$$= \frac{2\omega_q}{\Delta} \int_0^t dt' e^{i[(\mathbf{q}\cdot\mathbf{P})/M - \omega_q](t-t')} \int_0^{t'} dt'' \begin{pmatrix} \chi_1\chi_2^*(t'') e^{i[(\mathbf{q}\cdot\mathbf{P})/M + \delta](t'+t'')} e^{i\omega_q(t'-t'')} [B(0)]^2 \\ -\chi_2(t'')\chi_1^* e^{i[(\mathbf{q}\cdot\mathbf{P})/M + \delta + \omega_q](t'-t'')} \end{pmatrix}, \quad (37b)$$

where the commutation properties of  $B(0)$  [or  $B^\dagger(0)$ ] and  $\mathbf{P}$  and the relationship  $B^\dagger(0)B(0) = 1$  have been used to go from Eq. (37a) to Eq. (37b). The bunching parameter  $B(t) \sim B^{(0)}(t) + B^{(1)}(t)$  can now be averaged with the initial density matrix (21). The average of Eq. (35b) for  $B^{(0)}(t)$  vanishes as does the first term in Eq. (37b) for  $B^{(1)}(t)$ . On averaging the remaining term in Eq. (37b) for  $B^{(1)}(t)$  with the initial density matrix (21), interchanging the order of integration, and carrying out the integration over  $t'$ , one obtains

$$\begin{aligned} \langle B(t) \rangle &\sim -\frac{2\chi_1^* e^{-i\delta t}}{\Delta} \int d\mathbf{P} W(\mathbf{P}) \\ &\times \int_0^t dt' \chi_2(t') e^{i[(\mathbf{q}\cdot\mathbf{P})/M + \delta](t-t')} \sin[\omega_q(t-t')]. \end{aligned} \quad (38)$$

I will return to this equation after showing that an identical equation is reached using a density matrix approach.

## B. RIR

The RIR are usually calculated in the context of a density matrix approach. From Eq. (31a), it follows that

$$\langle B(t) \rangle = \int \rho_{gg}(\mathbf{P}, \mathbf{P} + \hbar\mathbf{q}; t) d\mathbf{P}, \quad (39)$$

which gives the alternative form for Eq. (34),

$$\frac{d\chi_2(t)}{dt} = -i \frac{\mathcal{N}d^2\Omega_2\chi_1 e^{i\delta t}}{2\hbar\epsilon_0\Delta} \int \rho_{gg}(\mathbf{P}, \mathbf{P} + \hbar\mathbf{q}; t) d\mathbf{P}. \quad (40)$$

To complete the RIR equations, one uses the Hamiltonian (30), along with Eqs. (11)–(14), to obtain density matrix equations

$$\begin{aligned}
\partial \rho_{gg}(\mathbf{P}, \mathbf{P}'; t) / \partial t = & -i \omega_{PP'} \rho_{gg}(\mathbf{P}, \mathbf{P}'; t) \\
& - i \frac{\chi_1^* \chi_2(t) e^{-i\delta t}}{\Delta} [\rho_{gg}(\mathbf{P} + \hbar \mathbf{q}, \mathbf{P}'; t) \\
& - \rho_{gg}(\mathbf{P}, \mathbf{P}' - \hbar \mathbf{q}; t)] \\
& - i \frac{\chi_1 \chi_2^*(t) e^{i\delta t}}{\Delta} [\rho_{gg}(\mathbf{P} - \hbar \mathbf{q}, \mathbf{P}'; t) \\
& - \rho_{gg}(\mathbf{P}, \mathbf{P}' + \hbar \mathbf{q}; t)], \quad (41)
\end{aligned}$$

where  $\omega_{PP'} = E_{PP'}/\hbar$ . It is easily verified that equations for the quantities  $d\langle B(t) \rangle / dt = \int \dot{\rho}_{gg}(\mathbf{P}, \mathbf{P} + \hbar \mathbf{q}; t) d\mathbf{P}$  and  $d\langle \mathbf{P}(t) \rangle / dt = \int \mathbf{P} \dot{\rho}_{gg}(\mathbf{P}, \mathbf{P}; t) d\mathbf{P}$ , obtained using Eq. (41), are identical to Eqs. (33). Thus the generalized RIR density matrix equations are *totally equivalent* to the corresponding operator CARL equations.

To evaluate  $\langle B(t) \rangle$  in the perturbative limit using Eq. (39), one sets  $\mathbf{P}' = \mathbf{P} + \hbar \mathbf{q}$  in Eq. (41) and replaces the density matrix elements in the right hand side of that equation by their zeroth order values,

$$\rho_{gg}^{(0)}(\mathbf{P}, \mathbf{P}'; 0) = (2\pi\hbar)^3 V^{-1} W(\mathbf{P}) \delta(\mathbf{P} - \mathbf{P}'), \quad (42)$$

to obtain [19]

$$\begin{aligned}
\partial \rho_{gg}^{(1)}(\mathbf{P}, \mathbf{P} + \hbar \mathbf{q}; t) / \partial t = & -i \omega_{P|\mathbf{P} + \hbar \mathbf{q}} \rho_{gg}^{(1)}(\mathbf{P}, \mathbf{P} + \hbar \mathbf{q}; t) \\
& - i \frac{\chi_1^* \chi_2(t) e^{-i\delta t}}{\Delta} \\
& \times [W(\mathbf{P} + \hbar \mathbf{q}) - W(\mathbf{P})]. \quad (43)
\end{aligned}$$

This equation is consistent with the RIR picture of Raman transitions between center-of-mass momentum states differing by  $\hbar \mathbf{q}$ . Integrating Eq. (43) over  $\mathbf{P}$  and  $t$ , and using Eq. (39), one reproduces Eq. (38) for  $\langle B(t) \rangle$ . Since  $\langle B(t) \rangle \sim 0$  as  $\omega_q \sim 0$ , the probe gain in the RIR and CARL is a recoil-induced effect.

Having established that the RIR and CARL formalisms lead to an equivalent set of equations, I now proceed to obtain the RIR and CARL limits of these equations, in the small signal gain (perturbation theory) limit. Recall that the RIR limit, as defined in this paper, is one in which the  $d\chi_2/dt$  depends locally on  $\chi_2(t)$ . The small signal gain regime is limited to those times for which  $\chi_2(t)/\chi_2(0) \lesssim e^{-1}$ . It is convenient to combine Eqs. (34) and (38) into the single equation

$$\begin{aligned}
d\chi_2/dt = & iQ\omega_q^2 \int d\mathbf{P} W(\mathbf{P}) \\
& \times \int_0^t dt' \chi_2(t') e^{i[(\mathbf{q}\cdot\mathbf{P})/M + \delta](t-t')} \sin[\omega_q(t-t')], \quad (44)
\end{aligned}$$

where  $Q$  is the dimensionless CARL parameter [10] defined as

$$Q = \frac{\mathcal{N} d^2 \Omega_2 |\chi_1|^2}{\hbar \Delta^2 \epsilon_0 \omega_q^2}. \quad (45)$$

### C. RIR limit

The RIR limit can be obtained by assuming that  $\chi_2(t')$  is slowly varying compared with the other terms in the integrand of Eq. (44), solving the resulting equation for  $\chi_2(t)$ , and then checking for self-consistency. If  $\chi_2(t')$  is slowly varying compared with the other terms in the integrand of Eq. (44), one can evaluate  $\chi_2(t')$  at  $t' = t$  to arrive at

$$d\chi_2/dt = g(t)\chi_2(t), \quad (46)$$

where the (complex) gain parameter is

$$g(t) = iQ\omega_q^2 \int d\mathbf{P} W(\mathbf{P}) \int_0^t d\tau e^{i[(\mathbf{q}\cdot\mathbf{P})/M + \delta]\tau} \sin(\omega_q \tau). \quad (47)$$

It is assumed that the momentum distribution  $W(\mathbf{P})$  can be written as the product of a one-dimensional, symmetric distribution  $W_q(P_q)$  and a two-dimensional distribution  $W_{\perp}(P_{\perp})$  for momenta  $\mathbf{P}_{\perp}$  transverse to the  $\hat{\mathbf{q}}$  direction. For the sake of definiteness, I take

$$W_q(P_q) = \frac{1}{\sqrt{\pi}P_0} e^{-(P_q/P_0)^2}, \quad (48)$$

although the calculations can be carried through for arbitrary distribution functions. Introducing dimensionless integration variables  $x = P_q/P_0$  and  $y = qu\tau$  in Eq. (47), where  $u = P_0/M$  is the most probable atomic speed, one can rewrite the equation as

$$g(t) = i \frac{Q\omega_q}{\sqrt{\pi}} \left( \frac{\omega_q}{qu} \right) \int_{-\infty}^{\infty} dx e^{-x^2} \int_0^{qu} dy e^{i(xy + \delta y/qu)} \sin\left( \frac{\omega_q}{qu} y \right). \quad (49)$$

The value of the integral depends critically on the ratio

$$r = \frac{qP_0}{M\omega_q} = \frac{2Mu}{\hbar\omega_q} = \frac{qu}{\omega_q}. \quad (50)$$

#### 1. $r \gg 1$

If  $r \gg 1$ , Eq. (49) reduces to

$$g(t) \sim i \frac{Q\omega_q}{\sqrt{\pi}} \left( \frac{\omega_q}{qu} \right) \int_{-\infty}^{\infty} dx e^{-x^2} \int_0^{qu} dy \left( \frac{\omega_q}{qu} y \right) e^{i(xy + \delta y/qu)} \quad (51a)$$

$$\begin{aligned}
& = \frac{Q\omega_q^2}{\sqrt{\pi}} \left( \frac{\omega_q}{qu} \right) d \left\{ \int_{-\infty}^{\infty} dx e^{-x^2} \int_0^{qu} dy \right. \\
& \quad \left. \times e^{i(xy + \delta y/qu)} \right\} / d\delta \quad (51b)
\end{aligned}$$

$$\begin{aligned}
& = Q\omega_q^2 \left( \frac{\omega_q}{qu} \right) d \left\{ \int_0^{qu} dy e^{-y^2/4} e^{i\delta y/qu} \right\} / d\delta. \quad (51c)
\end{aligned}$$

One sees that, in general, the exponential buildup of the field is not linear with time. However, if  $qut \gg 1$ , the expression for  $g(t)$  reduces to

$$g = \pi Q \omega_q^3 \left( \frac{M}{q} \right) d \left\{ W_q(P_q = \delta M/q) + \frac{i}{\pi} \int_{-\infty}^{\infty} dx W_q(P_0 x) P[1/(x + \delta/qu)] \right\} / d\delta, \quad (52)$$

where  $P$  indicates a principal value, giving a small signal gain [1]

$$\text{Re}(g) = \pi Q \omega_q^3 \left( \frac{M}{q} \right) \frac{dW_q(\delta M/q)}{d\delta}. \quad (53)$$

The gain depends linearly on the atomic density and the pump field intensity. The gain is positive for  $\delta < 0$ , negative (absorption) for  $\delta > 0$ , and vanishes at  $\delta = 0$ . This is a "single particle" gain in that each atom contributes separately to the gain and there is no threshold condition for gain that depends on atomic density.

The maximum gain occurs for  $\delta \approx -qu$ , for which  $|g| \approx Q \omega_q (\omega_q/qu)^2$ . For arbitrary values of  $qut$ , it follows from Eq. (51) that the maximum gain cannot exceed this value. Thus, for arbitrary times  $t$ , the RIR gain in the limit that  $r \gg 1$  is of order  $Q \omega_q (\omega_q/qu)^2$ . For self-consistency of the approach, it is necessary that  $|g| \ll qu$ , since it has been assumed that  $\chi_2(t')$  is slowly varying with respect to  $\exp(iqut')$  in Eq. (44). As a consequence, the RIR limit when  $r \gg 1$  is

$$r = \frac{qu}{\omega_q} \gg Q^{1/3}, \quad r \gg 1. \quad (54)$$

## 2. $r \ll 1$

The situation changes when  $r \ll 1$ . In that limit one can replace  $W(\mathbf{P})$  in Eq. (47) by  $\delta(\mathbf{P})$  to obtain

$$g(t) = iQ \omega_q^2 \int_0^t dt' e^{i\delta\tau} \sin(\omega_q \tau). \quad (55)$$

For both  $|\delta t| \ll 1$  and  $|\omega_q t| \ll 1$ ,  $|g(t)| = Q \omega_q^3 t^2/2$ , and  $\chi_2(t) = \chi_2(0) \exp(iQ \omega_q^3 t^3/6)$ , while, for  $\delta/\omega_q = -1$ , the gain is  $g(t) = Q \omega_q t/2 + iQ(1 - e^{-2i\omega_q t})/4$  and  $\chi_2(t) = \chi_2(0) \exp[Q \omega_q^2 t^2/4 + iQ \omega_q t/4 - Q(1 - e^{-2i\omega_q t})/8]$ . If  $|\delta \pm \omega_q| t > 1$ , the gain coefficient is of order  $Q \omega_q t$ . For self-consistency, the requirement that  $\chi_2(t')$  be slowly varying with respect to the integrand in Eq. (55) translates into the requirement that  $\int_0^t |g(t')| dt'$  be small compared with unity. If  $|\delta/\omega_q| \lesssim 1$ , one arrives at the RIR limits

$$Q \omega_q t \ll 1, \quad |\delta \pm \omega_q| t > 1, \quad r = \frac{qu}{\omega_q} \ll 1, \quad (56a)$$

$$Q^{1/2} \omega_q t \ll 1, \quad \delta/\omega_q = -1, \quad \omega_q t > 1, \quad r = \frac{qu}{\omega_q} \ll 1, \quad (56b)$$

$$Q^{1/3} \omega_q t \ll 1, \quad \omega_q t, |\delta t| \ll 1; \quad r = \frac{qu}{\omega_q} \ll 1. \quad (56c)$$

There is always a time range over which the RIR limit is valid, irrespective of the value of  $Q$ . Since  $Q$  is proportional to the atomic density, the RIR limit is valid for arbitrarily long times as the density is reduced. In the RIR limit, the gain is always proportional to the density.

## D. CARL limit

The CARL limit is obtained by assuming that one can set  $\exp[i\mathbf{q} \cdot \mathbf{P}(t-t')/M] \approx 1$  in Eq. (44) for times in which the small signal gain theory is applicable. This assumption is valid for all cases in which the RIR limit is *not* valid. In other words, the CARL limit holds when  $|\delta/qu| \lesssim 1$  and

$$Q^{1/3} \gtrsim r = \frac{qu}{\omega_q} \gg 1 \quad (57)$$

or when  $|\delta/\omega_q| \lesssim 1$  and

$$Q \omega_q t \gtrsim 1, \quad |\delta \pm \omega_q| t > 1, \quad Q < 1, \quad r = \frac{qu}{\omega_q} \ll 1, \quad (58a)$$

$$Q^{1/2} \omega_q t \gtrsim 1, \quad \delta/\omega_q = -1, \quad Q < 1, \quad r = \frac{qu}{\omega_q} \ll 1, \quad (58b)$$

$$Q^{1/3} \omega_q t \gtrsim 1, \quad Q \gtrsim 1, \quad r = \frac{qu}{\omega_q} \ll 1. \quad (58c)$$

Setting  $\exp[i\mathbf{q} \cdot \mathbf{P}(t-t')/M] \approx 1$  and  $\chi_2(t) = \tilde{\chi}_2(t) e^{i\delta t}$  in Eq. (44), one obtains the CARL equation

$$d\tilde{\chi}_2/dt = -i\delta\tilde{\chi}_2 + iQ\omega_q^2 \int_0^t dt' \tilde{\chi}_2(t') \sin[\omega_q(t-t')], \quad (59)$$

which is equivalent to the differential equation

$$\frac{d^3\tilde{\chi}_2}{dt^3} = -i\delta \frac{d^2\tilde{\chi}_2}{dt^2} - \omega_q^2 \frac{d\tilde{\chi}_2}{dt} + i\omega_q^2(\omega_q Q - \delta)\tilde{\chi}_2, \quad (60)$$

subject to the initial conditions  $\tilde{\chi}_2(0) = \tilde{\chi}_2(0)$ , and  $d\tilde{\chi}_2(0)/dt = -i\delta\tilde{\chi}_2(0)$ ,  $d^2\tilde{\chi}_2(0)/dt^2 = -\delta^2\tilde{\chi}_2(0)$  [corresponding to  $\langle B(0) \rangle = d\langle B(0) \rangle/dt = 0$ ]. The small signal behavior is determined from this equation, which has already been analyzed by Bonifacio and co-workers for CARLSC with neglect of the  $\omega_q^2$  term [10] and by Moore and Meystre [13] including this term. Gain occurs if one of the roots of the cubic indicial equation,

$$s^3 + i\delta s^2 + \omega_q^2 s - i\omega_q^2(\omega_q Q - \delta) = 0, \quad (61)$$

has a positive real value. The condition for gain is [13]

$$9(\delta/\omega_q) - (\delta/\omega_q)^3 + [3 + (\delta/\omega_q)^2]^{3/2} < 27Q/2. \quad (62)$$

If  $Q \ll 1$ , gain occurs in the range  $|\delta/\omega_q + 1| < \sqrt{2Q}$  and the gain coefficient is approximately equal to



$\sqrt{2Q - (\delta/\omega_q + 1)^2} \omega_q/2$ . When  $Q \geq 1$ , gain occurs for  $\delta/\omega_q \geq 0$ , which is impossible in the RIR limit. Moreover, in both cases the gain depends nonlinearly on the atomic density and there is a threshold condition for all values of  $\delta/\omega_q \neq -1$ . As such, CARL is a collective effect in the sense that gain does not occur for atomic densities below a certain critical value. For  $\delta/\omega_q = -1$ , which is the resonance condition for Raman transitions between center-of-mass states having momenta  $\mathbf{P} = \mathbf{0}$  and  $\mathbf{P} = \hbar \mathbf{q}$ , there is gain irrespective of the value of  $Q$ . For  $Q \gg 1$  and  $|\delta/\omega_q| \ll Q^{1/3}$ , the gain varies as  $Q^{1/3} \omega_q \cos(\pi/6) = (\sqrt{3}/2) Q^{1/3} \omega_q$ , and the probe field undergoes exponential gain linear in time for  $t \geq (Q^{1/3} \omega_q)^{-1}$ . The CARLSC theory can be used for  $Q \gg 1$ , whereas the CARLQ theory must be used for  $Q \leq 1$ .

#### IV. DEGENERATE PUMP AND PROBE FIELDS, $\delta = 0$

An interesting limiting case is one in which  $W(\mathbf{P}) \sim \delta(\mathbf{P})$ , and the pump and probe field frequencies are degenerate,  $\Omega_1 \approx \Omega_2 \equiv \Omega$ ;  $\delta = \Omega_2 - \Omega_1 = 0$  (recall that the detuning  $\delta$  has been redefined to include the shift arising from linear dispersion). When  $\delta = 0$ , the probe gain vanishes identically in the RIR limit  $r \gg 1$ , but grows exponentially for CARLSC. For CARLQ, it follows from Eq. (62) that the probe gain is exponential provided that

$$Q > \frac{2}{3\sqrt{3}}. \quad (63)$$

This qualitative difference between the RIR and CARL limits is reason enough to consider the  $\delta = 0$  case in some detail, but it is not the only reason. The existence of exponential gain when  $\delta = 0$  is surprising at first glance. It would seem that processes in which a pump photon is absorbed and a probe photon emitted would be exactly canceled by processes in which a probe photon is absorbed and a pump photon emitted, owing to the symmetry of the interaction when  $W(\mathbf{P}) \sim \delta(\mathbf{P})$ . It is the purpose of this section to investigate the origin of exponential gain in CARL when  $\delta = 0$ . Calculations are carried out in a perturbative limit, that is, to lowest order in the pump field intensity. As is shown below, the results are also applicable to a wider range of problems.

In perturbation theory, there are five states that enter the calculation, starting from atoms in their ground state having  $\mathbf{P} = \mathbf{0}$ . The relevant states are  $|g; \mathbf{P} = 0\rangle$ ,  $|e; \mathbf{P} = \hbar \mathbf{k}_1\rangle$ ,  $|e; \mathbf{P} = \hbar \mathbf{k}_2\rangle$ ,  $|g; \mathbf{P} = \pm \hbar \mathbf{q}\rangle$ , having energies  $0, \hbar(\omega + \omega_k), \hbar(\omega + \omega_k)$ , and  $\hbar\omega_q$ , respectively (recall that  $\omega_k \equiv \omega_{k_1} \approx \omega_{k_2}$ ). It is convenient to relabel these states as

$$\begin{aligned} |g; \mathbf{P} = 0\rangle &\equiv |0\rangle, & |e; \mathbf{P} = \hbar \mathbf{k}_1\rangle &\equiv |1\rangle, \\ |e; \mathbf{P} = \hbar \mathbf{k}_2\rangle &\equiv |-1\rangle, & |g; \mathbf{P} = \pm \hbar \mathbf{q}\rangle &\equiv |\pm 2\rangle. \end{aligned} \quad (64)$$

The energy levels associated with these states are shown in Fig. 1. The pump field drives the  $|0\rangle$  to  $|1\rangle$  and  $|-1\rangle$  to  $|-2\rangle$  transitions while the probe field drives the  $|0\rangle$  to  $|-1\rangle$  and  $|1\rangle$  to  $|2\rangle$  transitions. The Hamiltonian for the system can be obtained by expanding the Hamiltonian (3) in a momentum-state basis for the subspace (64). Using the relationship  $\langle \mathbf{P} | e^{i\mathbf{k} \cdot \mathbf{R}} | \mathbf{P}' \rangle = \delta(\mathbf{P} - \mathbf{P}' - \hbar \mathbf{k})$ , one finds

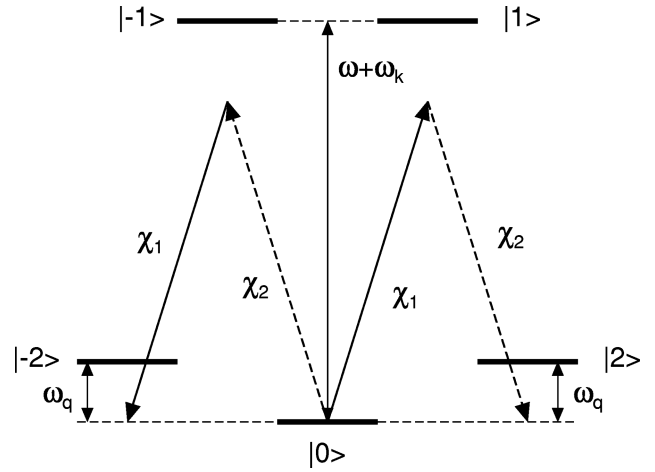


FIG. 1. In perturbation theory, the relevant momentum states can be represented as an equivalent five-level system, interacting with two fields as shown. The initial momentum distribution is taken to be a  $\delta$  function and the detuning is  $\delta = 0$ . State  $|0\rangle$  corresponds to  $|g; \mathbf{P} = 0\rangle$ , state  $|1\rangle$  to  $|e; \mathbf{P} = \hbar \mathbf{k}_1\rangle$ , state  $|-1\rangle$  to  $|e; \mathbf{P} = \hbar \mathbf{k}_2\rangle$ , and states  $|\pm 2\rangle$  to  $|g; \mathbf{P} = \pm \hbar \mathbf{q}\rangle$ . Although derived for the recoil problem, the conclusions reached in the text for this level scheme are applicable to any problem where a similar level scheme is encountered.

$$\begin{aligned} H &= \hbar(\omega + \omega_k)(|1\rangle\langle 1| + |-1\rangle\langle -1|) \\ &+ \hbar\omega_q(|2\rangle\langle 2| + |-2\rangle\langle -2|) \\ &+ \hbar\{\chi_1 e^{-i\Omega t}(|1\rangle\langle 0| + |-1\rangle\langle -2|) + (\text{adjoint})\} \\ &+ \hbar\{\chi_2(t) e^{-i\Omega t}(|1\rangle\langle 2| + |-1\rangle\langle 0|) + (\text{adjoint})\}. \end{aligned} \quad (65)$$

Note that this level scheme could equally well describe an atom in a Stark field, driven by circularly polarized pump and probe fields. The energy  $\hbar\omega_q$  would then correspond to the Stark shifts of the  $m = \pm 2$  ground-state, Zeeman sublevels.

Since decay is neglected, the calculation is most conveniently carried out using state amplitudes rather than density matrix elements. No ensemble average is needed here since we start in an eigenstate of momentum,  $\mathbf{P} = \mathbf{0}$ . It is convenient to work in a field interaction representation in which the state amplitudes  $a_j(\mathbf{P}, t)$  ( $j = -2, 2$ ) are written as

$$a_0(\mathbf{P}, t) = [(2\pi\hbar)^3/V]^{1/2} \tilde{a}_0(t) \delta(\mathbf{P}), \quad (66a)$$

$$a_1(\mathbf{P}, t) = [(2\pi\hbar)^3/V]^{1/2} \tilde{a}_1(t) e^{-i\Omega t} \delta(\mathbf{P} - \hbar \mathbf{k}_1), \quad (66b)$$

$$a_{-1}(\mathbf{P}, t) = [(2\pi\hbar)^3/V]^{1/2} \tilde{a}_{-1}(t) e^{-i\Omega t} \delta(\mathbf{P} - \hbar \mathbf{k}_2), \quad (66c)$$

$$a_{\pm 2}(\mathbf{P}, t) = [(2\pi\hbar)^3/V]^{1/2} \tilde{a}_{\pm 2}(t) \delta(\mathbf{P} \mp \hbar \mathbf{q}), \quad (66d)$$

with density matrix elements given by

$$\tilde{\rho}_{i,j}(t) = \tilde{a}_i(t) \tilde{a}_j^*(t), \quad (67a)$$

$$\rho_{j,j'}(\mathbf{R},t) = \frac{1}{(2\pi\hbar)^3} \int d\mathbf{P} d\mathbf{P}' a_j(\mathbf{P},t) a_{j'}^*(\mathbf{P}',t) e^{i(\mathbf{P}-\mathbf{P}')\cdot\mathbf{R}/\hbar}. \quad (67b)$$

In Eq. (8a) for the probe field evolution, one needs the component of polarization  $\mathbf{P}_2$ , varying as  $e^{i(\mathbf{k}_2\cdot\mathbf{R}-\Omega t)}$ . From Eqs. (66) and (67), it follows that

$$\mathbf{P}_2(\mathbf{R},t) = \mathcal{N}[\rho_{0,-1}\tilde{\rho}_{-1,0}(t) + \rho_{2,1}\tilde{\rho}_{1,2}(t)]e^{i(\mathbf{k}_2\cdot\mathbf{R}-\Omega t)}, \quad (68)$$

where the  $\rho$ 's are dipole matrix elements. For the level scheme corresponding to the states (64),

$$\rho_{0,-1} = \rho_{2,1} = \rho_{ge} \equiv \rho^*. \quad (69)$$

Thus the problem reduces to calculating  $\tilde{\rho}_{-1,0}(t)$  and  $\tilde{\rho}_{1,2}(t)$  to order  $|\chi_1|^2\chi_2$ .

Before undertaking this calculation, it is useful to obtain an expression for the time evolution of the probe field's energy density  $\mathcal{W}_2 = \epsilon_0|E_2|^2/2$ . Using Eqs. (8a), (4), (68), and (69), one finds

$$d\mathcal{W}_2/dt = i\mathcal{N}\Omega\rho^* \cdot \boldsymbol{\epsilon}_2 E_2^*(t)(\tilde{\rho}_{-1,0} + \tilde{\rho}_{1,2})/2 + \text{c.c.} \quad (70a)$$

$$= -i\mathcal{N}\hbar\Omega\chi_2^*(t)(\tilde{\rho}_{-1,0} + \tilde{\rho}_{1,2}) + \text{c.c.} \quad (70b)$$

$$= \mathcal{N}\hbar\Omega[\dot{\rho}_{2,2} - (\dot{\rho}_{-1,-1} + \dot{\rho}_{-2,-2})]. \quad (70c)$$

The last line follows from the density matrix equations of motion for the Hamiltonian (65) and can be given an obvious physical interpretation. Population of state  $|2\rangle$  implies gain on the probe field, while population in states  $|-1\rangle$  or  $|-2\rangle$  implies loss for the probe field. The time rate of change in energy density is simply the difference  $[\dot{\rho}_{2,2} - (\dot{\rho}_{-1,-1} + \dot{\rho}_{-2,-2})]$  multiplied by the product of energy,  $\hbar\Omega$ , gained or lost by the probe field in each elementary process and the atomic density. In the adiabatic limit considered in this paper, the excited-state population is negligible and one has

$$d\mathcal{W}_2/dt \sim \mathcal{N}\hbar\Omega(\dot{\rho}_{2,2} - \dot{\rho}_{-2,-2}). \quad (71)$$

One might expect that  $\dot{\rho}_{2,2} = \dot{\rho}_{-2,-2}$ , owing to the symmetry of the level scheme, but we will see that this is not the case.

Assuming that the detuning of the fields  $\Delta$  from the ground- to excited-state atomic resonance is sufficiently large to adiabatically eliminate the excited states, one uses the Hamiltonian (65) to show that the excited-state amplitudes are given by

$$\tilde{a}_1(t) \sim \frac{\chi_1(t)}{\Delta}\tilde{a}_0(t) + \frac{\chi_2(t)}{\Delta}\tilde{a}_2(t), \quad (72a)$$

$$\tilde{a}_{-1}(t) \sim \frac{\chi_2(t)}{\Delta}\tilde{a}_0(t) + \frac{\chi_1(t)}{\Delta}\tilde{a}_{-2}(t), \quad (72b)$$

and that the state amplitudes  $\tilde{a}_0(t), \tilde{a}_{\pm 2}(t)$  evolve as

$$d\tilde{a}_0(t)/dt = -i\chi_1^*\tilde{a}_1(t) - i\chi_2^*(t)\tilde{a}_{-1}(t), \quad (73a)$$

$$d\tilde{a}_2(t)/dt = -i\omega_q\tilde{a}_2(t) - i\chi_2^*(t)\tilde{a}_1(t), \quad (73b)$$

$$d\tilde{a}_{-2}(t)/dt = -i\omega_q\tilde{a}_{-2}(t) - i\chi_1^*\tilde{a}_{-1}(t). \quad (73c)$$

It is a straightforward exercise to solve Eqs. (73) in perturbation theory to third order in the fields, starting from  $\tilde{a}_0(0) = 1$ . One finds

$$\tilde{a}_0^{(0)}(t) = 1, \quad (74a)$$

$$\tilde{a}_1^{(1)}(t) = \frac{\chi_1}{\Delta}, \quad (74b)$$

$$\tilde{a}_{-1}^{(1)}(t) = \frac{\chi_2(t)}{\Delta}, \quad (74c)$$

$$\tilde{a}_2^{(2)}(t) = -i \int_{-\infty}^t dt' \frac{\chi_2^*(t')\chi_1}{\Delta} e^{-i\omega_q(t-t')}, \quad (74d)$$

$$\tilde{a}_{-2}^{(2)}(t) = -i \int_{-\infty}^t dt' \frac{\chi_1^*\chi_2(t')}{\Delta} e^{-i\omega_q(t-t')}, \quad (74e)$$

$$\tilde{a}_0^{(2)}(t) = -i \int_{-\infty}^t dt' \frac{|\chi_1|^2 + |\chi_2(t')|^2}{\Delta}, \quad (74f)$$

$$\tilde{a}_1^{(3)}(t) \sim \frac{\chi_1}{\Delta}\tilde{a}_0^{(2)}(t) + \frac{\chi_2(t)}{\Delta}\tilde{a}_2^{(2)}(t), \quad (74g)$$

$$\tilde{a}_{-1}^{(3)}(t) \sim \frac{\chi_2(t)}{\Delta}\tilde{a}_0^{(2)}(t) + \frac{\chi_1}{\Delta}\tilde{a}_{-2}^{(2)}(t), \quad (74h)$$

where the superscripts denote the order of the fields.

Consider, first, Eq. (71) for the probe field intensity, which depends on

$$d(\rho_{2,2} - \rho_{-2,-2})/dt = d[|\tilde{a}_2^{(2)}(t)|^2 - |\tilde{a}_{-2}^{(2)}(t)|^2]/dt. \quad (75)$$

By inspecting Eqs. (74d) and (74e), one can understand the manner in which  $|\tilde{a}_2^{(2)}(t)|^2$  can grow more rapidly than  $|\tilde{a}_{-2}^{(2)}(t)|^2$ . Suppose  $\chi_2(t)$  acquires a positive, time-dependent phase as a result of the atom-field interaction. In this case, the quantity  $\chi_2^*(t')e^{i\omega_q t'}$  appearing in the integrand of Eq. (74d) varies more slowly than the quantity  $\chi_2(t')e^{i\omega_q t'}$  appearing in the integrand of Eq. (74e). As a result, state  $|2\rangle$  population builds up more rapidly than that of state  $|-2\rangle$ , leading to probe gain. In other words, the nonlinear phase modulation of the probe field effectively favors the  $|0\rangle$  to  $|2\rangle$  transition over the  $|0\rangle$  to  $|-2\rangle$  transition if the phase is positive.

It follows from Eq. (59) with  $\delta=0$  that  $\chi_2(t)/\chi_2(0) \sim 1 + i(Q\omega_q t^3)/6 \sim e^{iQ\omega_q t^3/6}$  for  $\omega_q t \ll 1$ . At early times, the phase is positive, favoring probe gain. Whether or not the field continues to grow depends on the value of  $Q$ . If  $Q \ll 1$ , one can solve Eq. (60) to first order in  $Q$  to obtain

$$\chi_2(t) \sim [e^{iQ\omega_q t} - iQe^{-iQ\omega_q t/2} \sin(\omega_q t)] \chi_2(0). \quad (76)$$

For  $Q \ll 1$ , the intensity of the probe field remains approximately constant. If  $Q\omega_q t \ll 1$ , one recovers the RIR limit, in which the time development of the field is approximately local, and there is neither absorption nor gain. On the other hand, for  $Q \geq 1$ , the field buildup occurs sufficiently rapidly to ensure that

$$\begin{aligned} (\dot{\rho}_{2,2} - \dot{\rho}_{-2,-2}) &= -2\text{Re}[\chi_2^*(t)(\tilde{\rho}_{-1,0}^{(3)} + \tilde{\rho}_{1,2}^{(3)})] \\ &= 4\text{Re}\left\{ i \frac{|\chi_1|^2 \chi_2^*(t)}{\Delta^2} \int_0^t dt' \chi_2(t') \right. \\ &\quad \left. \times \sin[\omega_q(t-t')] \right\} \end{aligned} \quad (77)$$

remains positive for all  $t$ . In this case, there is exponential gain for the probe, linear in time and nonlinear in the atomic density, for times  $\omega_q t Q^{1/3} > 1$ . This is the CARL limit.

In summary, the probe gain that occurs for  $\delta=0$  and large  $\Delta$  is clearly *not* a single particle effect. It is more closely related to a ‘‘propagation’’ effect in which the phase modulation of the probe field produced by the nonlinear atom-field interaction drives the probe gain.

## V. DISCUSSION

It has been shown that the density-matrix-RIR and Heisenberg-operator-CARL formalisms lead to equivalent equations. The RIR and CARL limits refer simply to different regions of parameter space of these equations. For a given experimental situation, one must determine whether one is in the RIR limit, the CARL limit, or neither limit (as is most often the case). The experimental implications of the RIR and CARL are discussed below, but first I would like to discuss the distinction between the terms ‘‘matter grating’’ used in discussions of the RIR and ‘‘atomic bunching’’ used in discussions of CARL.

The term ‘‘matter grating’’ refers to spatially modulated atom distributions resulting from a nonlinear atom-field interaction. The term ‘‘atomic bunching’’ refers to a redistribution or focusing of atoms in an optical potential. For the RIR and CARL, *these terms are synonymous*. If recoil effects are neglected, that is, if the center-of-mass motion is treated classically from the outset, the *total atomic density* is conserved for each velocity subclass of atoms (neglecting collisions). A homogeneous atomic density remains homogeneous to all orders in the atom-field interaction. Recoil effects allow for a modification of the total atomic density. Whether one calls this ‘‘atomic bunching’’ or ‘‘matter grating’’ production is a matter of personal preference [5]. The key point is that the modification of the total atomic density results entirely from effects related to recoil on the absorption, emission, or scattering of radiation. To lowest order in the atom-field interaction, the matter grating or atomic bunching consists of a spatial modulation of the atomic density having period  $2\pi/|\mathbf{k}_1 - \mathbf{k}_2|$ . With increasing field strength, higher order spatial harmonics are produced, corresponding to ‘‘higher order matter gratings’’ or ‘‘focusing’’ or ‘‘atomic bunching.’’ Of course it is possible to derive an

effective potential of the form

$$\begin{aligned} H_{\text{eff}} &= \left\{ \frac{\hbar}{\Delta} [|\chi_1|^2 + |\chi_2(t)|^2] + \frac{\hbar}{\Delta} [\chi_1 \chi_2^*(t) e^{i(\mathbf{q} \cdot \mathbf{R} + \delta t)} \right. \\ &\quad \left. + \chi_1^* \chi_2(t) e^{-i(\mathbf{q} \cdot \mathbf{R} + \delta t)}] \right\} |1\rangle \langle 1| \end{aligned} \quad (78)$$

without quantization of the center-of-mass motion and to consider *classical* motion in this potential. However, since the effective potential is proportional to  $\hbar$ , any changes in the atomic density vanish in the classical limit. This is in contrast to bunching in the free electron laser where the effective potential does not vanish in the classical limit.

Are there situations where matter gratings are produced by fields without any contribution from recoil? The answer to this question is ‘‘yes,’’ provided one considers the matter gratings associated with individual internal atomic states rather than the total atomic density. For example, for an ensemble of *stationary*, closed, two-level atoms there is a spatial modulation in the *population difference* between the excited and ground states produced by the pump and probe fields. Scattering of the pump field from this spatially modulated population difference leads to a dispersion-shaped probe absorption profile centered at a probe-pump detuning  $\delta=0$  in the limit of large atom-field detuning  $\Delta$ . The amplitude of the dispersion profile varies as the square of the pump field intensity and its width is equal to the excited-state decay rate [15]. It is also possible to have gain profiles with a width corresponding to some effective ground-state decay time if one considers open systems in which spontaneous emission also plays a role. For example, if the ground-state consists of two hyperfine states and the fields drive transitions between only one of these hyperfine states and an excited state, then, as a result of spontaneous emission, both hyperfine state sublevel populations can be spatially modulated (although the *total* atomic density—the sum of all ground- and excited-state populations—remains constant in the absence of recoil). It is possible to monitor the atomic gratings in specific ground-state hyperfine levels by using radiation that couples only the targeted ground-state sublevel to an excited state.

Finally, I would like to discuss some experimental implications of the RIR and CARL. To observe the spectral features of the RIR and CARL, it is best to use cold atoms in a collisionless environment. Moreover, to isolate the effects of interest, one must use experiments which involve closed, two-level transitions or signals that depend only on total atomic-state density. The RIR have already been observed in several experiments involving laser-cooled atoms [5–9]. The experiments in which evidence for CARL was claimed [20,21] were all carried out under conditions (collisions, radiation trapping, large Doppler widths) which are outside the CARL limit. The results of these experiments can be explained by conventional theories in which recoil is neglected [22]. For  $Q > 1$ , the CARL limit is reached for  $qu/\omega_q = 2\hbar q/Mu \leq Q^{1/3}$ . For densities of order  $10^{18}$  atoms/m<sup>3</sup>, it is possible to achieve values of  $Q^{1/3}$  of order 100–1000. Thus it may be possible to observe CARL for atoms cooled to or somewhat above the sub-Doppler limit of laser cooling.

The linear gain coefficient in this case is of order  $10^7 \text{ s}^{-1}$ , which must exceed any cavity loss.

Is it possible to observe the RIR in a thermal vapor using pump-probe spectroscopy? The linear absorption rate is  $g_1 = (Nd^2\Omega_2/\hbar\epsilon_0)(\gamma_e/\Delta^2)$ , which implies that the ratio of the recoil-induced gain  $g$  [Eq. (53)] to the linear absorption rate is of order  $(\hbar|\chi_1|^2/\gamma_e)/E_a$ , where  $E_a = mu^2/2$  is the kinetic energy of the atoms. For sub-Doppler-cooled atoms, this ratio can be greater than unity, but it is small at room temperature. Still it might be possible to use modulation techniques to isolate the RIR contribution to the probe absorption. For the RIR signal to be larger than the background, dispersion-like contribution to the probe absorption that varies as  $2\sqrt{\pi}|\chi_1|^4\gamma_e/(\Delta^5qu)$ , one requires that the ratio  $(\Delta^2/|\chi_1|^2)(\hbar q/Mu)(\Delta/\gamma_e)$  be greater than unity. This can be achieved at room temperature for sufficiently large  $\Delta/\chi_1$ .

Perhaps the best way to observe CARL would be to use an atomic beam, transversely cooled below the recoil limit. The beam (or, alternatively, sub-Doppler-cooled atoms from a magneto-optical trap) can be passed through a cavity with some transit time  $\Gamma^{-1}$ . It is not difficult to extend the theory to allow for transit time effects through an effective decay rate  $\Gamma$  for ground-state atoms. For subrecoil-cooled atoms, Eq. (59) is replaced by

$$d\chi'_2/dt = (\Gamma - i\delta)\chi'_2 + iQ\omega_q^2 \int_0^t dt' \chi'_2(t') \sin[\omega_q(t-t')], \quad (79)$$

where  $\chi'_2 = \chi_2 e^{(\Gamma - i\delta)t}$ , Eq. (60) by

$$\frac{d^3\chi'_2}{dt^3} = (\Gamma - i\delta)\frac{d^2\chi'_2}{dt^2} - \omega_q^2\frac{d\chi'_2}{dt} + i\omega_q^2(\omega_q Q - \delta - i\Gamma)\chi'_2, \quad (80)$$

and the indicial equation (61) by

$$s^3 - (\Gamma - i\delta)s^2 + \omega_q^2 s - i\omega_q^2(\omega_q Q - \delta - i\Gamma) = 0. \quad (81)$$

With the inclusion of decay, the CARL limit is still given by Eq. (58) for  $\Gamma t \ll 1$ . For  $\Gamma t \gg 1$ , the CARL limit is  $\Gamma \leq Q^{1/3}\omega_q$  if  $Q \gg 1$  and  $|\delta/\omega_q| \ll Q^{1/3}$ ; and  $\Gamma \leq Q^{1/2}\omega_q$  if  $Q \ll 1$  and  $\delta/\omega_q = -1$ . Decay tends to diminish the gain parameter when  $Q \gg 1$ , but can actually *reduce* the gain threshold when  $Q < 1$ . For example, if  $\delta = 0$ , the threshold condition is reduced from  $Q > 2/(3\sqrt{3})$  to  $Q > 0$  if  $\Gamma \neq 0$ .

#### ACKNOWLEDGMENTS

It is a pleasure to acknowledge helpful discussions with B. Dubetsky and J. L. Cohen, and their suggestions for improving the manuscript. This work was supported by the U.S. Office of Army Research under Grant No. DAAG55-97-0113 and the National Science Foundation under Grant Nos. PHY-9414020 and PHY-9800981.

- 
- [1] J. Guo, P. R. Berman, B. Dubetsky, and P. R. Berman, Phys. Rev. A **46**, 1426 (1992).
- [2] J. Guo and P. R. Berman, Phys. Rev. A **47**, 4128 (1993); J. Guo, *ibid.* **49**, 3934 (1994); **51**, 2338 (1995).
- [3] P. R. Berman, B. Dubetsky, and J. Guo, Phys. Rev. A **51**, 3947 (1995).
- [4] B. Dubetsky and P. R. Berman, Phys. Rev. A **52**, R2519 (1995).
- [5] J. Y. Courtois, G. Grynberg, B. Lounis, and P. Verkerk, Phys. Rev. Lett. **72**, 3017 (1994).
- [6] S. Guibal, C. Triché, L. Guidoni, P. Verkerk, and G. Grynberg, Opt. Commun. **131**, 61 (1996).
- [7] D. R. Meacher, D. Boiron, H. Metcalf, C. Saloman, and G. Grynberg, Phys. Rev. A **50**, R1992 (1994).
- [8] M. Kozuma, Y. Imai, N. Nakagawa, and M. Ohtsu, Phys. Rev. A **52**, R3421 (1995); M. Kozuma, N. Nakagawa, W. Jhe, and M. Ohtsu, Phys. Rev. Lett. **76**, 2428 (1996).
- [9] M. G. Raizen, J. Koga, B. Sundaram, Y. Kishimoto, H. Takuma, and T. Tajima (unpublished).
- [10] R. Bonifacio and L. De Salvo, Nucl. Instrum. Methods Phys. Res. A **341**, 360 (1994); R. Bonifacio, L. De Salvo, L. M. Narducci, and E. J. D'Angelo, Phys. Rev. A **50**, 1716 (1994); R. Bonifacio and L. De Salvo, Opt. Commun. **115**, 505 (1995); L. De Salvo, R. Cannerozzi, R. Bonifacio, E. J. D'Angelo, and L. M. Narducci, Phys. Rev. A **52**, 2342 (1995); R. Bonifacio, L. De Salvo, L. M. Narducci, and E. J. D'Angelo (unpublished).
- [11] R. Bonifacio and P. Verkerk, Opt. Commun. **124**, 469 (1996).
- [12] Propagation effects have been included in the paper of Bonifacio *et al.*, R. Bonifacio, G. R. M. Robb, and B. W. J. McNeil, Phys. Rev. A **56**, 912 (1997).
- [13] M. G. Moore and P. Meystre, Phys. Rev. A **58**, 3248 (1998).
- [14] B. R. Mollow, Phys. Rev. A **5**, 2217 (1972); F. Y. Wu, S. Ezekiel, M. Ducloy, and B. R. Mollow, Phys. Rev. Lett. **38**, 1077 (1977).
- [15] The linewidth actually results from the convolution of a Lorentzian having width equal to the excited-state decay rate  $\gamma_e$  and a Gaussian having width  $qu$ , where  $q$  is the magnitude of the difference of pump and probe field wave vectors and  $u$  is the most probable atomic speed.
- [16] G. Grynberg and C. Cohen-Tannoudji, Opt. Commun. **96**, 150 (1993).
- [17] G. Khitrova, P. R. Berman, and M. Sargent, J. Opt. Soc. Am. B **5**, 160 (1988).
- [18] B. Dubetsky and P. R. Berman, Phys. Rev. A **47**, 1294 (1993).
- [19] The finite quantization volume for the center-of-mass motion implies that one should replace  $\delta(0)$  by  $V/(2\pi\hbar)^3$  whenever it appears. This prescription gives an initial density  $\rho_{gg}(\mathbf{P}, \mathbf{P}; 0) = W(\mathbf{P})$ .
- [20] G. L. Lippi, G. P. Barozzi, S. Barbay, and J. R. Tredicce, Phys. Rev. Lett. **76**, 2452 (1996).
- [21] P. R. Hemmer, N. P. Bigelow, D. P. Katz, M. S. Shahriar, L. De Salvo, and R. Bonifacio, Phys. Rev. Lett. **77**, 1468 (1996).
- [22] W. J. Brown, J. R. Gardner, D. J. Gauthier, and R. Vilaseca, Phys. Rev. A **55**, R1601 (1997).