Slow collisions between identical atoms in a laser field: The spectrum of redistributed light

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The resonance fluorescence from a system of interacting, identical, and cold (trapped) atoms driven by a laser field is derived. The effects of dressing by the laser field and radiative decay are all included. We examine in detail the applicability of the binary-collision approximation for this problem, which is a special case of resonance broadening. The final result allows a clear physical interpretation and shows interesting features due to the resonant exchange of coherences and excitations. Correspondence with previous work is also discussed.

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I. INTRODUCTION

The resonance fluorescence from a system of interacting, identical, and cold atoms driven by a laser field is interesting for a number of reasons. It can be an important probe of the collisions between two cold identical atoms in a driving laser field. Because the atoms are moving so slowly (about 1.0 m/s in a typical trap), spontaneous decay during a collision becomes an important process. Another consequence of this small speed is the interaction range becomes of the order of wavelengths, where the usual $1/r_{ab}^3$ form of dipole-dipole interaction is no longer valid [1]. Emission during a collision can also result in radiative escape [2], which is one of the mechanisms for trap loss. For typical densities of trapped atoms, we will show that we can make the binary-collision approximation (BCA). This is because two-atom coherences formed during a collision, which decay on the time scale of radiative decay, do not survive until a subsequent collision. This is in contrast to "normal" resonance broadening, where multiatom coherences can occur [3]. In spite of the BCA, there are many interesting features in the spectrum that are due to the resonant nature of the exchange of coherences and excitations. Finally, this problem is closely related to the understanding of the effects of collisions on the cooling of the atoms [4]. We believe that the formal techniques established here will expedite the analysis of this issue.

Section II presents the master equation for a system of N identical atoms in a laser field [1]. In this section we identify the single-atom operators and the interactions. We show that it is possible, in spite of relative motion, to go to a rotating frame in which all the single-atom operators are completely independent of time. In Sec. III we define the proper projection operator for the ensemble average to be done most efficiently. In Sec. IV, we discuss the characteristics of cold-atom collisions and make the binary-collision approximation. We also find the steady-state density matrix for an atom in the laser field and under the influence of collisions. The spectrum is derived in two different ways in Sec. V: first, using the more traditional method based on the dipole correlation function, and then using the approach of Mollow [5]. Finally, in Sec. VI, the spectrum valid to first order in the collisional width is presented. We discuss the physical interpretation of all the terms and the connection to previous approaches.

II. RADIATION DAMPING

In paper I [1] we examined the effects of the electromagnetic interaction between two identical atoms. We started with the density matrix for the full system, which contains both atoms and all modes of the quantized electromagnetic field. As we were interested in the atomic system only, we reduced the problem to the atomic degrees of freedom for pairs of interacting atoms by applying a standard projection-operator technique. We then showed that under certain conditions (which are well satisfied for a system of identical slow atoms undergoing mutual collisions), we could introduce the Born and Markov approximations for radiation relaxation in the usual manner. After implementing these approximations, the equation of motion for the reduced density matrix became a first-order ordinary differential equation [1]. The effect of the contraction of the spontaneous radiation modes was to introduce a modification of the spontaneous decay rate (depending on the interatomic distance) and an effective pairwise-interaction potential between the atoms in our system.

The reduced-density-matrix equation for a system of N identical two-level atoms with $J_g=0$ and $J_e=1$ in a laser field, averaged over spontaneous field modes, is given by [1]

$$\partial_t\rho(t)=-i\frac{\omega_0}{2}\sum_a[S_a^z,\rho(t)]+i\sum_a\frac{\Omega(\mathbf{r}_a(t))}{2}[(S_a^{0+}e^{-i[\omega_L t-\mathbf{k}_L\cdot\mathbf{r}_a(t)]}+\text{H.c.}),\rho(t)]$$
$$-\frac{1}{2}\sum_{a,b}\sum_{m_a,m_b}\Gamma_{am_a}^{bm_b}(\mathbf{r}_{ab}(t))[S_a^{m_a+}S_b^{m_b-}\rho(t)+\rho(t)S_a^{m_a+}S_b^{m_b-}-2S_b^{m_b-}\rho(t)S_a^{m_a+}]$$
$$-i\frac{1}{2}\sum_{\substack{a,b\\a\neq b}}\sum_{m_a,m_b}\Omega_{am_a}^{bm_b}(\mathbf{r}_{ab}(t))[S_a^{m_a+}S_b^{m_b-},\rho(t)]\,, \tag{1}$$

where we have taken the laser to be linearly polarized along the z axis. The sums $\sum_a$ and $\sum_b$ are taken over all atoms in our system, $\sum_{m_a}$ includes degeneracy of the excited state within each atom. ω_0 is already a renormalized atom frequency (with Lamb shift included). $\Omega(\mathbf{r}_a(t))$ is the Rabi frequency which measures the interaction of the atoms with external laser field. S_a^z and $S_a^{m_a\pm}$ are atomic operators defined by

$$S_a^z=\left[\sum_{m_a=-1}^{1}|m_a\rangle\langle m_a|\right]-|g\rangle\langle g|\,, \tag{2a}$$
$$S_a^{m_a+}=|m_a\rangle\langle g|=(S_a^{m_a-})^\dagger\,. \tag{2b}$$

$\Omega_{am_a}^{bm_b}(\mathbf{r}_{ab}(t))$ (for $a\neq b$) describes the interaction potential between atom a and atom b; and $\Gamma_{am_a}^{am_a}$ $[\Gamma_{am_a}^{bm_b}(\mathbf{r}_{ab}(t))]$ corresponds to the spontaneous emission rates as modified by the mutual interactions between the atoms,

$$\Gamma_{am_a}^{am_a}=\gamma=\frac{4}{3}\frac{k_0^3|d|^2}{h}(ck_0=\omega_0)\,, \tag{3a}$$
$$\Gamma_{am_a}^{bm_b}(\mathbf{r}_{ab}(t))=\gamma\left[j_0(x)\delta_{m_am_b}+\sqrt{6\pi}(-1)^{m_b}\begin{bmatrix}1&1&2\\-m_b&m_a&q\end{bmatrix}j_2(x)Y_{2,q}(\theta,\varphi)\right]\,, \tag{3b}$$
$$\Omega_{am_a}^{bm_b}(\mathbf{r}_{ab}(t))=\Gamma_{am_a}^{bm_b}(j_k(x)\leftrightarrow y_k(x))\,. \tag{3c}$$

Here we have set $x=k_0r_{ab}(t)$ and $q=m_b-m_a$; $j_k(x)$ and $y_k(x)$ are spherical Bessel functions of the first and second kind, respectively [6], $Y_{k,q}(\theta,\varphi)$ is a spherical harmonic, θ and φ are the spherical angles of the interatomic vectors $\mathbf{r}_{ab}(t)$ in the laboratory frame defined by the choice of the quantization axis (the direction of laser polarization in this case), and

$$\begin{bmatrix}a&b&c\\a_1&b_1&c_1\end{bmatrix}$$

are Wigner 3-j symbols. $\Omega_{am_a}^{bm_b}$ is obtained from $\Gamma_{am_a}^{bm_b}$ by simply substituting $j_k(x)$ with $y_k(x)$ as indicated in Eq. (3c).

The properties of this interaction potential and collective decay rate are discussed in detail elsewhere [1]. It is sufficient to note that the $\Omega_{am_a}^{bm_b}$ and $\Gamma_{am_a}^{bm_b}$ operators for $a\neq b$ are explicit functions of the interatomic separation $\mathbf{r}_{ab}(t)$, and that the usual $1/r_{ab}^3$-type dipole-dipole interaction is correct only at small distances, i.e., where $x\ll 1$. We have specified $J_g=0$, $J_e=1$, and the laser to be linearly polarized, but the generalization to other cases is straightforward.

In the reduced density matrix Eq. (1) we will now distinguish between two types of terms. Specifically, we will separate terms referring to one atom only from those containing two-atom operators, so that Eq. (1) may be written as

$$\partial_t\rho(t)=\sum_a[L_{0a}+L_{La}(t)-\Gamma_a/2]\rho(t)$$
$$-\frac{1}{2}\sum_{\substack{a,b\\a\neq b}}[\Gamma_{ab}(t)+i\Omega_{ab}(t)]\rho(t) \tag{4}$$

with two-atom operators defined by

$$\Omega_{ab}=\sum_{m_a,m_b}\Omega_{am_a}^{bm_b}[S_a^{m_a+}S_b^{m_b-},\cdot]\,, \tag{5a}$$
$$\Gamma_{ab}=\sum_{m_a,m_b}\Gamma_{am_a}^{bm_b}(S_a^{m_a+}S_b^{m_b-}\cdot+\cdot S_a^{m_a+}S_b^{m_b-}-2S_b^{m_b-}\cdot S_a^{m_a+})\,, \tag{5b}$$

and one-atom operators L_{0a} and Γ_a defined by

$$L_{0a}=-i\frac{\omega_0}{2}[S_a^z,\cdot]\,, \tag{5c}$$
$$\Gamma_a=\sum_{m_a}\gamma(S_a^{m_a+}S_a^{m_a-}\cdot+\cdot S_a^{m_a+}S_a^{m_a-}-2S_a^{m_a-}\cdot S_a^{m_a+})\,. \tag{5d}$$

$L_{La}(t)$ describes interaction of an atom with the external laser field mode. The laser light is assumed to be monochromatic, and its polarization is collinear with the quantization axis. In this work we treat only a traveling laser beam described by

$$\mathbf{E}_L(r,t)=\tfrac{1}{2}E_0\epsilon_L e^{i(\mathbf{k}_L\cdot\mathbf{r}-\omega_L t)}+\text{c.c.} \tag{6}$$

The tetradic operator characterizing the atom-laser interaction is already in the rotating-wave approximation (RWA) and also has the form of a commutator:

$$L_{La}(t)=i\frac{\Omega}{2}[(S_a^{0+}e^{i[\mathbf{k}_L\cdot\mathbf{r}_a(t)-\omega_L t]}+\text{H.c.}),\cdot] \tag{7}$$

with Ω defined by $\Omega=E_0(\epsilon_L\cdot\mathbf{d}_a^{0+})/\hbar$.

Atomic evolution in the absence of collisions is characterized by the one-atom operators in Eq. (4), in which only the $L_{La}(t)$ defined by Eq. (7) has a time dependence. It is convenient to transform to a rotating frame where one has removed the time dependence from the free-evolution operator. We note that due to the motion of atoms $\mathbf{r}_a(t)$ is also time dependent. The standard approach to remove the time-dependent phase from $L_{La}(t)$ consists in the unitary transformation to the rotating frame [7],

$$\rho^R(t)=R(t)\rho(t)\ , \tag{8}$$

where $R(t)$ is defined for a many-atom systems by

$$R(t)=\prod_a R_a(t)$$
$$=\prod_a \exp\{-i[\mathbf{k}_L\cdot\mathbf{r}_a(t)-\omega_L t][S_a^z,\cdot]/2\}\ . \tag{9}$$

The transformation R_a is thus made in each of the atomic subspaces. To make this transformation tractable, we assume straight-line trajectories so that

$$\mathbf{r}_a(t)=\mathbf{r}_{a0}+\mathbf{v}_a t\ . \tag{10}$$

By applying this transformation to our density matrix Eq. (4), we obtain the equation of motion for the density matrix in the rotating frame:

$$\partial_t\rho^R(t)=\sum_a L_a^R\rho^R(t)-\frac{1}{2}\sum_{\substack{a,b\\a\neq b}}[\Gamma_{ab}^R(t)+i\Omega_{ab}^R(t)]\rho^R(t)\ . \tag{11}$$

Here

$$L_a^R=(L_{0a}^R+L_{La}^R-\Gamma_a/2)\ , \tag{12}$$

and the new operators with superscript R are defined by (Γ_a is not changed by the rotating frame transformation)

$$L_{0a}^R(t)=L_{0a}(\omega_0\leftrightarrow\Delta)\ , \tag{13a}$$

where $\Delta=\omega_0-(\omega_L-\mathbf{k}_L\cdot\mathbf{v}_a)$,

$$L_{La}^R=i\frac{1}{2}[S_a^{0+}+S_a^{0-},\cdot]\ , \tag{13b}$$

$$\begin{Bmatrix}\Gamma_{ab}^R(t)\\ \Omega_{ab}^R(t)\end{Bmatrix}=\begin{Bmatrix}\Gamma_{ab}\\ \Omega_{ab}\end{Bmatrix}\times e^{i\mathbf{k}_L\cdot[\mathbf{r}_b(t)-\mathbf{r}_a(t)]}\ . \tag{13c}$$

Note that the one-atom operator L_a^R is now completely independent of time, as a result of making the straight-line-trajectory approximation. Note that Doppler effects show up not only in the definition of Δ, but also in the definitions of $\Gamma_{ab}^R,\Omega_{ab}^R$ via the $e^{i\mathbf{k}L\cdot\mathbf{r}(t)}$ factors.

The last two terms on the right-hand side of Eq. (11) are two-atom operators. For the rest of the paper, we will refer to them as "collisional operators." Setting together the real and the imaginary parts into

$$V_{ab}^R(t)=-\tfrac{1}{2}\{\Gamma_{ab}^R(t)+\Gamma_{ba}^R(t)+i[\Omega_{ab}^R(t)+\Omega_{ba}^R(t)]\}\ , \tag{14}$$

Eq. (11) now becomes

$$\partial_t\rho^R(t)=\sum_a L_a^R\rho^R(t)+\sum_{\substack{a,b\\a<b}}V_{ab}^R(t)\rho^R(t)\ . \tag{15}$$

The phrase collisional operators is well justified in this case because both the collective decay rates $\Gamma_{am_a}^{bm_b}(\mathbf{r}_{ab}(t))$ and the interaction potentials $\Omega_{am_a}^{bm_b}(\mathbf{r}_{ab}(t))$ are only effectively nonzero when the atoms are close together, i.e., during the duration of a collision. A collision in the sense understood here can occur only when one atom is excited and the second is in the ground state. Otherwise, they only interact via weak van der Waals forces, which have a $1/r_{ab}^6$ dependence [8]. During a collision, the radiative decay process is disturbed by the presence of the other atom. As we will see later, for atoms in a typical trap, the average spontaneous emission time is of the same order or even less than the duration of a collision. As a result emission during a collision, and also reexcitation (in the strong laser field), are all possible.

III. COLLISIONAL AVERAGE

By averaging over the spontaneous electromagnetic-field modes we have included the stochastic properties of the radiation field. Our aim is to derive a theory to describe the behavior of an ensemble of atoms, consequently we have also to account for the stochastic aspects of the collisions. We use the same projection-operator techniques that were employed to describe radiation damping.

In our system all atoms are alike. Consequently, the collisions between atoms, which lead for example, to the redistribution of radiation, may be described as self- or resonance broadening. The issue of indistinguishability is worth discussing. In the present approach we assume (for convenience) that the atoms are moving along straight-line classical trajectories. With these trajectories are associated Doppler shifts, and in this picture, we can in principle follow the motion of each individual atom. During the collisions of interest for redistribution the wave functions representing the nuclear motion of the participating atoms do not overlap significantly; i.e., we are above the recoil limit. It follows from this observation that atoms are no longer indistinguishable in the quantum-mechanical sense. Therefore, as long as this classical approximation to the external degrees of freedom is used we do not have to worry specifically about symmetrizing the formulas used in the present calculations.

After we realize that the identical nature of the atoms

is not essential to our approach, we find that our problem can be treated by equivalent methods to those used in foreign gas broadening, although, as in resonance broadening, exchange of excitation can and will occur. To account for the stochastic aspects of collisions in our ensemble we will apply the following procedure: Initially, we will artificially single out one particle that we will call the radiator (as a subscript referring to this special atom we will use r, rather than a or b as we have done so far). The evaluation of all the atoms in our ensemble is described by the same equation of motion, so what we are doing is equivalent to selecting one atom to represent the characteristic behavior in our sample. The rest of the atoms we will call perturbers (using subscript p for them) and we will treat them in a similar manner to perturbers in foreign gas broadening. It is important to notice, however, that the perturbers have internal structure and they interact with the laser light in exactly the same way as the radiator. Perturbers form an environment for the radiator and by collisions disturb the process of radiation. Since we will be interested in the light coming from the ensemble of identical atoms, we will be able to average our master equation over all perturber and collisional variables. By doing so we obtain equations for the populating density and dipole moment in the radiator subspace only. We can then solve these equations formally, substitute the results into the definition of the spectrum of scattered light, and finally average over radiator degrees of freedom. We focus here on the redistribution of radiation, but any observable quantity for the system can be found in this manner.

We have first to define the proper projection operator. We are interested in the equation of motion for the radiator only, hence we must contract the full Hilbert space to the proper subspace. Since we are dealing here with pairwise interactions which depend on interatomic distances, we not only have to average the reduced matrix equation over perturber internal degrees of freedom, but also the perturber translational degrees of freedom. The evaluation of any observable requires such averages.

To summarize the above comments, we expect that in order to construct the proper projection operator we will need to put together the following elements.

(a) A proper initial state of all the perturbers at some initial time $t=-T$, with no correlations present.

(b) The trace over all perturbers internal degrees of freedom. In other words, the final density-matrix equation after collisional average should be expressed in the radiator subspace only.

(c) The projection operator should contain the average over translational degrees of freedom of the perturbers described by $\mathbf{r}_{p0}$ and $\mathbf{v}_p$ [cf. Eq. (10)], or equivalently, the average over impact parameters, times of closest approach, and velocities [9].

According to the above list, we have to start the construction of the projection operator from the initial density matrix for perturbers. We assume that at time $t=-T$ all atoms were uncorrelated, and allow correlations to build up with time [10]. We notice also that what we call perturbers are only artificially distinguished from radiators; they all interact with external laser light, which is present all the time.

At time $t=-T$, all the atoms are assumed to be uncorrelated. Therefore the initial density matrix for the system of perturbers is a product of density matrices for all the uncorrelated perturbers, each of them being a stationary solution $\rho_p^{\mathrm{st},R}$ of the equation of motion of an atom in the external laser field (in the absence of collisions):

$$L_p^R\rho_p^{\mathrm{st},R}\equiv(L_{0p}^R+L_{Lp}^R-\Gamma_p/2)\rho_p^{\mathrm{st},R}=0\ . \tag{16}$$

Since we have ignored the coupling between the translational and the internal degrees of freedom of the atoms [4], and have made the classical straight-line-trajectory approximation, the translational part of freedom is simply described by some prescribed distribution functions which are independent of time. Even though we could have separated out explicitly the translational part, it is more convenient to have it included in all density matrices. Hence, $\rho_p^{\mathrm{st},R}$ includes a translational distribution function given by $w(\mathbf{r}_{p0},\mathbf{v}_p)$; $\rho(t)$ [the same with $\rho^R(t)$ and $\rho^{IR}(t)$] includes a translational part given by $\prod_a w(\mathbf{r}_{a0},\mathbf{v}_a)$. An example of $w(\mathbf{r}_{a0},\mathbf{v}_a)$ would be $f_M(\mathbf{v}_a)/V$, meaning that the atoms are uniformly distributed over a volume V, and that they obey the Maxwell velocity distribution.

In Eq. (16) all operators are taken in the rotating frame, and hence none of them carry any time dependence. Note also that L_{Lp}^R does not depend on the phase of the field. In this frame then $\rho_p^{\mathrm{st},R}$, is time independent. It depends on the velocity $\mathbf{v}_p$, however. This velocity dependence comes from the definition of the L_{0p}^R operator. Equation (16) is a simple algebraic equation, complex, with the real part described by the damping Γ_p. We also note that tetradic states which diagonalize L_a^R are precisely the nonorthogonal complex dressed states (see, for example, Armstrong and Baker [11]).

We now define our projection operator as

$$P_c=\prod_{p(\text{all perturbers})}P_{cp}\ , \tag{17}$$

where

$$P_{cp}(\ )=\rho_p^{\mathrm{st},R}\,\mathrm{Tr}_p(\ )\ , \tag{18}$$

in which Tr_p include not only the trace over the internal states, but also the integration over the translation variables. More specifically,

$$\mathrm{Tr}_p(\ )=\int d\mathbf{r}_{p0}\,d\mathbf{v}_p\,\mathrm{Tr}_{p(\text{internal})}(\ )\ .$$

Since $\mathrm{Tr}_a(\rho_a^{\mathrm{st},R})=1$, we have $P_{cp}^2=1$ and also $P_c^2=1$.

Before we can apply the projection operator to the density-matrix equation, we first introduce the interaction picture in all one-atom operators; for both the perturbers and the radiator,

$$\rho^{IR}(t)=\exp\left[-\sum_a L_a^R t\right]\rho^R(t)\ . \tag{19}$$

The master equation in the interaction picture is simply

$$\partial_t\rho^{IR}(t)=\sum_{\substack{a,b\\a<b}}V_{ab}^{IR}(t)\rho^{IR}(t)\ , \tag{20}$$

where $V_{ab}^{IR}(t)$ can be defined as

$$V_{ab}^{IR}(t)=e^{-(L_a^R+L_b^R)t}V_{ab}^R(t)e^{(L_a^R+L_b^R)t} . \tag{21}$$

Now the equation of motion for $P_c\rho^{IR}(t)$, averaged over collisions, may be obtained in precisely the same way that was used to contract over the continuum of the spontaneous modes [1],

$$\partial_t P_c\rho^{IR}(t)=P_c V^{IR}(t)P_c\rho^{IR}(t)+P_c V^{IR}(t)Q_c\rho^{IR}(t) , \tag{22a}$$

$$\partial_t Q_c\rho^{IR}(t)=Q_c V^{IR}(t)Q_c\rho^{IR}(t)+Q_c V^{IR}(t)P_c\rho^{IR}(t) , \tag{22b}$$

where $Q_c=1-P_c$, and

$$V^{IR}(t)=\sum_{\substack{a,b\\a<b}} V_{ab}^{IR}(t) . \tag{23}$$

$Q_c\rho^{IR}(t)$ can be solved by a Green function in terms of $P_c\rho^{IR}$. With our definition of projection operator, the statement that the atoms are not correlated at $t=-T$ is simply transformed to $Q_c\rho^{IR}(-T)=0$. For definiteness, we take $T=0$ for the time being (we can do so only if the results we are looking for are evaluated at $t=\infty$, which will be the case until Sec. V, where we will have to take $-T$ to be $-\infty$). With this initial condition we get

$$Q_c\rho^{IR}(t)=\int_0^t G^{IR}(t,t')Q_c V^{IR}(t')P_c\rho^{IR}(t')dt' , \tag{24}$$

where the propagator $G^{IR}(t,t')$ is defined through a time-ordered exponential,

$$G^{IR}(t,t')=T\exp\left[Q_c\int_{t'}^t V^{IR}(\tau)d\tau\right] . \tag{25}$$

We therefore obtain a closed equation for $P_c\rho^{IR}(t)$,

$$\begin{aligned}\partial_t P_c\rho^{IR}(t)=&P_c V^{IR}(t)P_c\rho^{IR}(t)\\&+P_c V^{IR}(t)\int_0^t G^{IR}(t,t')Q_c V^{IR}(t')\\&\times P_c\rho^{IR}(t')dt' .\end{aligned} \tag{26}$$

This equation is still very complicated, since the operator contains all possible collisions. The significant simplification can be obtained when we realize that the strong collisions between the radiator and the perturbers in our system are well separated in time and that an atom in the trap always has enough time to reach its steady state before it encounters the next strong collision. Thus we can use the binary-collision approximation. The distant weak collisions can be treated perturbatively. It should be noted, however, that, due to the long-range $1/r_{ab}$ type of interactions, correlations between distant perturbers can occur, which means that, as in the case of Stark broadening [12], a self-consistent solution will replace these perturbers by shielded quasiparticles.

IV. BINARY-COLLISION APPROXIMATION

In the context of foreign gas broadening, the binary-collision approximation (BCA) is based on the condition that strong collisions do not overlap, which requires that the time between (strong) collisions be much longer than the duration of a (strong) collision. However, it will become evident later that such a condition is not sufficient for BCA in resonance broadening. We have to require in addition that the time between (strong) collisions is much longer than the radiative decay time of the excited state of the atom. In this section, we will show qualitatively that these two conditions are indeed satisfied for atoms in a laser trap; we then show how these conditions are used to make the binary-collision approximation to our equation of motion. Finally, we will solve for the steady-state density matrix for an atom under both the influence of the laser field and the influence of collisions.

For laser traps that exist today [13], a typical density of trapped atoms, n, is of order of 10^{10} cm^{-3}. The average atomic velocity v is roughly 1.0 m/s [13]. For an estimate of the dipole-dipole interaction strength we will take $V(r)=C_3/r^3$ in which C_3 will be taken for the specific case of cesium atoms: $C_3=50$ eV Å^3 [13].

The data presented above were selected for an estimate of the order of magnitude of the Weisskopf radius r_W. The concept of the Weisskopf radius [14] is very useful for a rough estimate of the different time scales characterizing the colliding atoms. It is defined as the radius corresponding to unit phase shift,

$$\int_0^t V(\tau)d\tau\sim V(r_W)\tau_{\rm coll}\sim\hbar , \tag{27}$$

where $\tau_{\rm coll}$ is the duration of the collision. On the other hand,

$$\tau_{\rm coll}\sim r_W/v . \tag{28}$$

From these two equations we can estimate for our case of slowly moving atoms that the collision time $\tau_{\rm coll}$ is roughly 10^{-7} s; and the Weisskopf radius is roughly $\sim 10^3$ Å. A collision will be regarded as strong if two atoms are within the Weisskopf radius of each other. In subsequent discussions, the term collision will always be used to refer to a strong collision in this sense. Consequently, an estimate of collisional cross section is πr_W^2; and the frequency of collisions $\nu_{\rm coll}$ is

$$\nu_{\rm coll}=n\pi r_W^2 v , \tag{29}$$

which leads to an estimate of the time between collisions $T_{\rm coll}\sim 1/\nu_{\rm coll}$ of the order of 10^{-3} s. Finally, the spontaneous emission time $1/\gamma$ for, e.g., $6P_{3/2}(F'=5)$ to $6S_{1/2}(F=4)$ transition in cesium, may be estimated as being of the order 10^{-8} s.

This procedure gives a crude upper estimate of the strong collision radius. By using Eq. (28) we have assumed the collision can be completed, whereas the time scale of importance in Eq. (27) for redistribution is roughly $1/\gamma$ (or more generally the inverse of the detuning) which is less than our estimate for $\tau_{\rm coll}$. In addition, the fact that r_W is of order $\lambda/2\pi$ indicates that $1/r^2$ and $1/r$ terms in $V(r)$ are important. However, our estimate is quite sufficient to establish the overall time scales. More detail estimates again indicate that for distances much greater that $\lambda/2\pi$ the collisions are weak.

We have therefore shown that for the cold atom traps

available today, the time between collisions of importance for redistribution is much greater than both the duration of the collision and the spontaneous decay time of the transition. The first of these two inequalities establishes that strong collisions do not overlap in time. Since it takes only a few $1/\gamma$'s for an atom to reach its steady state in the laser field [15], the second of these inequalities establishes the loss of memory of atoms which have suffered collisions before they come into subsequent interactions with other perturbers.

Since the width associated with collisions is roughly ν_{coll}, we have therefore also established that the collisional widths γ_c are much smaller than the radiative decay width γ. This fact will be used in Sec. VI to treat the effects of collisions on the redistribution spectrum perturbatively.

Before making the BCA we remark on the role of coherences. Atoms are driven by the external field of the laser. As long as they do not interact they are affected by the field independently and one would be dealing with one-atom field-induced coherences only (at least if we ignore coherent coupling of all atoms which occurs only in the forward direction). However, suppose now that an atom A, which is initially excited, collides with another atom B, which is initially in its ground state. During the collision, the atoms exchange information about their excitation and dipole moments, and after the collision a correlation between their final states arises, so that the excitation can be shared between the atoms. This sharing is quantified as a two-atom coherence. Ben-Reuven [3] has pointed out that if this coherence survives to the collision of atom B (or A) with some third atom C, the information about the excitation of A would reach C and consequently a three-atom coherence would emerge. In this way a complicated many-body coherence would be set up. These many-atom coherences, of course, may occur in resonance broadening. The crux of our physical argument for cold collisions will be that the time between collisions is long compared to $1/\gamma$ so that the two-atom coherences which occur after a collision will, since a steady state is established, die out during the time between subsequent collisions.

We are now ready to make the binary-collision approximation to the equation of motion for $P_c\rho^{IR}(t)$. Using the fact that $\mathrm{Tr}_a\,\mathrm{Tr}_b V_{ab}^{IR}\cdots=0$ and that all the perturbers are equivalent, we can rewrite Eq. (26) as

$$\begin{aligned}\partial_t P_c\rho^{IR}(t)&=NP_cV_{rp}^{IR}(t)P_c\rho^{IR}(t)+NP_cV_{rp}^{IR}(t)\int_0^t G^{IR}(t,t')Q_cV^{IR}(t')P_c\rho^{IR}(t')dt'\\&=NP_cV_{rp}^{IR}(t)P_c\rho^{IR}(t)+NP_cV_{rp}^{IR}(t)\int_0^t G^{IR}(t,t')Q_cV_{rp}^{IR}(t')P_c\rho^{IR}(t')dt'\\&\quad+NP_cV_{rp}^{IR}(t)\int_0^t G^{IR}(t,t')Q_c\bar{V}^{IR}(t')P_c\rho^{IR}(t')dt'\,,\end{aligned}\tag{30}$$

where

$$\bar{V}^{\text{IR}}(t)=\sum_{(a,b)\neq(r,p)}V_{ab}^{IR}(t)\,.$$

The subscripts r and p refer to the radiator and a particular perturber, respectively. This equation is still exact.

Using again $\mathrm{Tr}_a\,\mathrm{Tr}_b V_{ab}^{IR}\cdots=0$, we have

$$P_cV_{rp}^{IR}(t)\cdots Q_cV_{rp}^{IR}(t_1)Q_cV_{p'p''}^{IR}(t_2)\cdots P_c\rho^{IR}(t')=0\,,\tag{31}$$

where $p',p''\neq p$. Since we have shown that strong collisions do not overlap in time, the following two equations are valid:

$$P_cV_{rp}^{IR}(t)\cdots Q_cV_{rp}^{IR}(t_1)\cdots Q_cV_{rp'}^{IR}(t_2)\cdots Q_cV_{rp}^{IR}(t_3)\times\cdots P_c\rho^{IR}(t')=0\,,\tag{32}$$

$$P_cV_{rp}^{IR}(t)\cdots Q_cV_{rp}^{IR}(t_1)\cdots Q_cV_{pp'}^{IR}(t_2)\cdots Q_cV_{rp}^{IR}(t_3)\times\cdots P_c\rho^{IR}(t')=0\,,\tag{33}$$

where $p'\neq p$ in both equations. The physical interpretation of these two equations is as follows. If the radiator is interacting strongly with the perturber p during (t',t), neither the radiator nor the perturber p can be interacting strongly with any other perturbers, say p', in the same time interval. Note that the collision represented in, e.g., Eq. (32) should not be understood as the sequence in which p interacts with r at t' and then comes back to interact with r again after r has suffered some other collisions. These types of events occur very rarely and incoherently. They are accounted for within the BCA by the average over $(\mathbf{r}_{0p},\mathbf{v}_p)$, which determines the frequency and directions the perturbers come in to interact with the radiator.

Expanding the Green function G^{IR} in terms of V_{ab}^{IR}, Eqs. (31)–(33) immediately give

$$P_cV_{rp}^{IR}(t)G^{IR}(t,t')Q_cV_{rp}^{IR}(t')P_c\rho^{IR}(t')=P_cV_{rp}^{IR}(t)G_{rp}^{IR}(t,t')Q_cV_{rp}^{IR}(t')P_c\rho^{IR}(t')\,,\tag{34}$$

where

$$G_{rp}^{IR}(t,t')=T\exp\left[Q_c\int_{t'}^t V_{rp}^{IR}(\tau)d\tau\right]\,.\tag{35}$$

Expanding $G^{IR}(t,t')$ according to

$$G^{IR}(t,t')=G_{rp}^{IR}(t,t')+\int_{t'}^t G_{rp}^{IR}(t,t_1)Q_c\bar{V}^{IR}(t_1)\times G^{IR}(t_1,t')dt_1\,,$$

and using again Eqs. (31)–(33), we obtain

$$P_c V_{rp}^{IR}(t)\int_0^t G^{IR}(t,t')Q_c\bar{V}^{IR}(t')P_c\rho^{IR}(t')dt'=P_c V_{rp}^{IR}(t)Q_c\bar{\rho}^{IR}(t)+P_c V_{rp}^{IR}(t)\int_0^t G_{rp}^{IR}(t,t_1)Q_c\bar{V}_{rp}^{IR}(t_1)Q_c\bar{\rho}^{IR}(t_1)dt_1 \ . \tag{36}$$

Here $Q_c\bar{\rho}^{IR}(t)$ is given by Eqs. (24) and (25) with V^{IR} being replaced by $\bar{V}^{IR}$ in both equations, i.e., $Q_c\bar{\rho}^{IR}(t)$ is generated by all collisions prior to the collision (rp). This term is easily shown to be zero for foreign gas broadening. It is generally nonzero, however. We now show that it can be set equal to zero if the time between collisions satisfies $T_{\rm coll}\gg 1/\gamma$.

In Eqs. (34)–(36), we can extract $\prod_{p'\neq p}P_{cp}$ from P_c (recall $P_{cp}^2=P_{cp}$) and commute it freely. As a result, all the Q_c's can be replaced by $Q_{cp}=1-P_{cp}$. Equation (36) now becomes

$$P_c V_{rp}^{IR}(t)\int_0^t G^{IR}(t,t')Q_c\bar{V}^{IR}(t')P_c\rho^{IR}(t')dt'=P_c V_{rp}^{IR}(t)Q_{cp}\bar{\rho}_{rp}^{IR}(t)+P_c V_{rp}^{IR}(t)\int_0^t G_{rp}^{IR}(t,t_1)Q_c\bar{V}_{rp}^{IR}(t_1)Q_{cp}\bar{\rho}_{rp}^{IR}(t_1)dt_1 \ , \tag{37}$$

where

$$\bar{\rho}_{rp}^{IR}=\prod_{p'\,(\neq p)}\mathrm{Tr}_{p'}\bar{\rho}^{IR} \ .$$

Since both the radiator r and the perturber p must have been free for a time $\delta t\sim T_{\rm coll}$ before coming into interacting with each other, we have

$$\bar{\rho}_{rp}^{IR}(t)=e^{-(L_r^R+L_p^R)t}[e^{(L_r^R+L_p^R)\delta t}\bar{\rho}_{rp}^{R}(t-\delta t)] \ .$$

Since $\delta t\gg 1/\gamma$, both the radiator and perturber have time to reach a steady state [15], i.e.,

$$\bar{\rho}_{rp}^{IR}(t)=e^{-(L_r^R+L_p^R)t}\rho_r^{\mathrm{st},R}\rho_p^{\mathrm{st},R}=\rho_r^{\mathrm{st},R}\rho_p^{\mathrm{st},R} \ ,$$

which immediately gives

$$Q_{cp}\bar{\rho}_{rp}^{IR}(t)=0 \ . \tag{38}$$

The same argument applies, of course, also to $Q_{cp}\bar{\rho}_{rp}^{IR}(t_1)$. We stress that physically this is equivalent to the statement that correlations due to two-atom coherences, via $Q_{cp}\bar{\rho}_{rp}^{IR}(t)$, die out when steady state can be attained between collisions (i.e., when $T_{\rm coll}\gg 1/\gamma$).

Combining Eqs. (30), (34), (37), and (38), and taking the thermodynamic limit [12] ($Q_{cp}=1-P_{cp}\approx 1$), we have finally

$$\partial_t P_c\rho^{IR}(t)=NP_c V_{rp}^{IR}(t)P_c\rho^{IR}(t)+NP_c V_{rp}^{IR}(t)\int_0^t U_{rp}^{IR}(t,t')V_{rp}^{IR}(t')P_c\rho^{IR}(t')dt' \ , \tag{39}$$

where

$$U_{rp}(t,t')=T\exp\left[\int_{t'}^t V_{rp}^{IR}(\tau)d\tau\right] \ . \tag{40}$$

This is the master equation in the binary-collision approximation. It treats all collisions as if they do not overlap in time. As indicated earlier, weak distant collisions do overlap, but in a perturbative treatment of independent quasiparticles, to lowest order their effects are additive and consequently contribute as if they did not overlap in time. We shall now find its steady-state solution. First, we define

$$\Sigma^{(0)}(t)\equiv e^{L_r^R t}\prod_p \mathrm{Tr}_p\rho^{IR}(t) \ . \tag{41}$$

Using the stationary property of the collisional average [12],

$$P_c V^{IR}(t_1+t)\cdots V^{IR}(t_K+t)P_c =e^{-L_r^R t}P_c V^{IR}(t_1)\cdots V^{IR}(t_K)e^{L_r^R t}P_c \ , \tag{42}$$

we can easily show from Eq. (39) that $\Sigma^{(0)}(t)$ satisfies

$$\begin{aligned}\partial_t\Sigma^{(0)}(t)=&L_r^R\Sigma^{(0)}(t)+N\,\mathrm{Tr}_p V_{rp}^{IR}(0)\rho_p^{\mathrm{st},R}\Sigma^{(0)}(t)\\&+N\,\mathrm{Tr}_p V_{rp}^{IR}(0)\\&\times\int_0^t U_{rp}^{IR}(0,t'-t)V_{rp}^{IR}(t'-t)\\&\times e^{L_r^R(t-t')}\rho_p^{\mathrm{st},R}\Sigma^{(0)}(t')dt' \ .\end{aligned} \tag{43}$$

As $t\to\infty$, $\Sigma^{(0)}(t)$ goes to a steady-state value characterized by $\partial_t\Sigma^{(0)}(t)=0$. We define

$$\Sigma^{(0)}\equiv\lim_{t\to\infty}\Sigma^{(0)}(t) \ ,$$

and use the following property of Laplace transform:

$$\lim_{t\to\infty}X(t)=\lim_{s\to 0}s\tilde{X}(s) \ , \tag{44}$$

where $\tilde{X}(s)$ is the Laplace transform of $X(t)$. This enables us to obtain from Eq. (43) an equation which determines $\Sigma^{(0)}$; i.e.,

$$(L_r^R+L_c)\Sigma^{(0)}=0 \ . \tag{45}$$

Here the "damping operator" L_c can be written in two equivalent forms:

$$L_c = N \lim_{s\to 0} \mathrm{Tr}_p \int_0^\infty V_{rp}^{IR}(0) U_{rp}^{IR}(0,-\tau) \times \rho_p^{\mathrm{st},R} e^{-(s-L_r^R)\tau} d\tau (s-L_r^R) \quad (46a)$$

$$= N \lim_{s\to 0} (s-L_r^R)\mathrm{Tr}_p \int_0^\infty e^{-(s-L_r^R)\tau} U_{rp}^{IR}(\tau,0) \times V_{rp}^{IR}(0)\rho_p^{\mathrm{st},R} d\tau \, . \quad (46b)$$

Using again the stationary property given by Eq. (42) and integrating by parts, we can also write the L_c in forms that do not contain any V_{rp}^{IR}:

$$L_c = N \lim_{s\to 0} (s-L_r^R)\mathrm{Tr}_p \int_0^\infty e^{-(s-L_r^R)\tau} [U_{rp}^{IR}(\tau,0)-1] \times \rho_p^{\mathrm{st},R} d\tau (s-L_r^R) \quad (46c)$$

$$= N \lim_{s\to 0} (s-L_r^R)\mathrm{Tr}_p \int_0^\infty [U_{rp}^{IR}(0,-\tau)-1] e^{-(s-L_r^R)\tau} \times \rho_p^{\mathrm{st},R} d\tau (s-L_r^R) \, . \quad (46d)$$

Again, $\Sigma^{(0)}$ includes, in our notation, a distribution function describing the translation degrees of freedom of the radiator: $w(\mathbf{r}_{r0}, \mathbf{v}_r)$.

V. SPECTRUM OF SCATTERED LIGHT

We now calculate the intensity of light scattered into a given mode of the radiation field. The rate of detection of photons in a particular mode (k,λ) is proportional to the rate at which photons are generated in this mode, i.e., $dN_{k\lambda}(t)/dt$. $N_{k\lambda}(t)$ is the number of photons in mode (k,λ) at time t, and is given formally by

$$N_{k\lambda}(t) = \langle a_{k\lambda}^\dagger(t) a_{k\lambda}(t) \rangle \, . \quad (47)$$

In the Heisenberg picture $a_{k\lambda}^\dagger(t)$ and $a_{k\lambda}(t)$ are related to the atomic dipole operators. If we assume that at initial time $t=-\infty$ there are no photons at the frequency ω present, this relationship can be written in the form

$$a_{k\lambda}(t) = \sum_{a,m_a} \int_{-\infty}^t d\tau [g_{k\lambda}^{am_a} S_a^{m_a +}(\tau) + (g_{k\lambda}^{am_a})^* S_a^{m_a -}(\tau)] \times \exp[-i\mathbf{k}\cdot\mathbf{r}_a(\tau) - i\omega_{k\lambda}(t-\tau)] \, . \quad (48)$$

Here the sum is over all atoms in our system and we have also introduced $g_{k\lambda}^{am_a}$ defined by

$$g_{k\lambda}^{am_a} = \left[\frac{2\pi ck}{\hbar V}\right]^{1/2} (\epsilon_\lambda \cdot \mathbf{d}_a^{m_a +}) \, . \quad (49)$$

The rate of change of photons in the mode (k,λ) is then given by

$$\sigma_{k\lambda}(t) = \frac{dN_{k\lambda}(t)}{dt} = \sum_{a,b} \sum_{m_a,m_b} \int_{-\infty}^t d\tau e^{i\omega_{k,\lambda}(t-\tau)} (g_{k\lambda}^{bm_b})(g_{k\lambda}^{am_a})^* \langle \exp[i\mathbf{k}\cdot\mathbf{r}_b(\tau) - i\mathbf{k}\cdot\mathbf{r}_a(t)] S_b^{m_b +}(\tau) S_a^{m_a -}(t) \rangle + \text{c.c.} \quad (50)$$

We have used $\langle\ \rangle$ to denote the ensemble average, which in our case consists of field, atom (internal degrees of freedom), and collisional degrees of freedom. The collisional part, as discussed in Sec. III, represents the translational degrees of freedom of the atoms, which can be described either by $(\mathbf{r}_{a0}, \mathbf{v}_a)$ or by a set of collisional parameters, namely, time of closest approach, impact parameter, angular configuration, and relative velocity [9]. Notice that to define the translational variables for a given collision it is convenient to work in terms of relative coordinates.

Defining

$$\hat{D}_a^+(t) = \sum_{m_a} g_{k\lambda}^{am_a} S_a^{m_a +}(t) \, , \quad (51)$$

where $S_a^{m_a +}(t)$ is the Heisenberg picture, we can write the spectrum in the form

$$\sigma_{k\lambda}(t) = \sum_{a,b} \int_0^\infty d\tau e^{i\omega_{k\lambda}\tau} \langle \exp[i\mathbf{k}\cdot\mathbf{r}_b(t-\tau) - i\mathbf{k}\cdot\mathbf{r}_a(t)] \hat{D}_b^+(t-\tau) \hat{D}_a^-(t) \rangle + \text{c.c.} \quad (52)$$

If both $t-\tau$ and τ are times when the system has already reached a stationary state in the presence of the driving field, the ensemble average in Eq. (52) is independent of t. The stationary spectrum is therefore given by

$$\sigma_{k\lambda} = \int_{-\infty}^\infty C_{k\lambda}(\tau) d\tau = \lim_{s\to 0} 2\,\mathrm{Re} \int_0^\infty C_{k\lambda}(\tau) e^{-s\tau} d\tau \, , \quad (53)$$

where

$$C_{k\lambda}(\tau) = \sum_{a,b} e^{i\omega_{k\lambda}\tau} \langle \exp[i\mathbf{k}\cdot\mathbf{r}_{b0} - i\mathbf{k}\cdot\mathbf{r}_a(\tau)] \hat{D}_b^+ \hat{D}_a^-(\tau) \rangle \, , \quad (54)$$

in which $\hat{D}_b^+ = \hat{D}_b^+(0)$ is a dipole operator in the Schrödinger picture and $\mathbf{r}_{b0} \equiv \mathbf{r}(0)$. In Eq. (53), it is implied that the system is in a stationary state from $\tau \sim -1/s$ to $\tau \sim +1/s$, which, upon taking $s \to 0$, goes to $(-\infty, +\infty)$.

Writing out explicitly the ensemble average and the time evolution of $\hat{D}_a^-(\tau)$, we have

$$
\begin{aligned}
C_{k\lambda}(\tau)&=\sum_{a,b} e^{i\omega_{k\lambda}\tau}\mathrm{Tr}_A\{\exp[i\mathbf{k}\cdot\mathbf{r}_{b0}-i\mathbf{k}\cdot\mathbf{r}_a(\tau)]\mathrm{Tr}_F[\rho_{AF}(0)\hat{D}_b^+(0)\hat{U}_{AF}^{-1}(\tau)\hat{D}_a^-\hat{U}_{AF}(\tau)]\}\\
&=\sum_{a,b} e^{-i\omega_{k\lambda}\tau}\mathrm{Tr}_A\{\exp[i\mathbf{k}\cdot\mathbf{r}_{b0}-i\mathbf{k}\cdot\mathbf{r}_a(\tau)]\hat{D}_a^-\,\mathrm{Tr}_F[U_{AF}(\tau)\rho_{AF}(0)\hat{D}_b^+]\}\\
&=\sum_{a,b} e^{i\omega_{k\lambda}\tau}\mathrm{Tr}_A\{\exp[i\mathbf{k}\cdot\mathbf{r}_{b0}-i\mathbf{k}\cdot\mathbf{r}_a(\tau)]\hat{D}_a^-[U(\tau)\rho(0)\hat{D}_b^+]\}\\
&=\sum_{a,b} e^{i\omega_{k\lambda}\tau}\mathrm{Tr}_A\{\exp[i\mathbf{k}\cdot\mathbf{r}_{b0}-i\mathbf{k}\cdot\mathbf{r}_a(\tau)]R(\tau)\hat{D}_a^-R^{-1}(\tau)R(\tau)[U(\tau)\rho(0)\hat{D}_b^+]\}\\
&=\sum_{a,b} e^{i\Delta\omega\tau}\mathrm{Tr}_A\{\exp(i\mathbf{k}\cdot\mathbf{r}_{b0}-i\Delta\mathbf{k}\cdot\mathbf{v}_a\tau-i\Delta\mathbf{k}\cdot\mathbf{r}_{a0})\hat{D}_a^-[U^R(\tau)R(0)\rho(0)\hat{D}_b^+]\}\\
&=\sum_{a,b} e^{i\Delta\omega\tau}\mathrm{Tr}_A\{\exp(i\Delta\mathbf{k}\cdot\mathbf{r}_{b0}-i\Delta\mathbf{k}\cdot\mathbf{v}_a\tau-i\Delta\mathbf{k}\cdot\mathbf{r}_{a0})\hat{D}_a^-e^{L_a^R\tau}[U^{IR}(\tau)\rho^R(0)\hat{D}_b^+]\}\ . \qquad (55)
\end{aligned}
$$

$\mathrm{Tr}_A=\prod_a\mathrm{Tr}_a$ is the trace over all the atoms (their internal as well as translational degrees of freedom). Tr_F stands for trace over all the field modes. $\rho_{AF}(0)$ is an initial density matrix for the complete system of all atoms plus field modes. $\hat{U}_{AF}(\tau)$ is the Hilbert-space evolution operator for this closed system, and $U_{AF}(\tau)$ is the corresponding Liouville-space evolution operator. After tracing over all the field modes, the system of all atoms evolves according to $U(\tau)$: a Liouville-space evolution operator which satisfies Eq. (1) with the initial condition $U(0)=1$. $U^R(\tau)$ and $U^{IR}(\tau)$ are Liouville-space evolution operators defined similarly through Eqs. (15) and (20). After going to the rotating frame by the transformation $R(\tau)$ defined in Eq. (9), the system evolves according to $U^R(\tau)$. In the interaction picture, it evolves according to $U^{IR}(\tau)$.

In Eq. (55), we have defined $\Delta\omega=\omega_{k\lambda}-\omega_L$ and $\Delta\mathbf{k}=\mathbf{k}-\mathbf{k}_L$. It is convenient to also define

$$K_a^R\equiv L_a^R+i(\Delta\omega-\Delta\mathbf{k}\cdot\mathbf{v}_a)\ . \qquad (56)$$

Since all the atoms are equivalent, we can split the double sum and the trace in Eq. (55). Calling one of the atoms the radiator and the rest perturbers, we can write Eq. (55) as

$$
\begin{aligned}
C_{k\lambda}(\tau)&=N\,\mathrm{Tr}_r\left\{\hat{D}_r^-e^{K_r^R\tau}\prod_p\mathrm{Tr}_p[g_1^{IR}(\tau)+g_2^{IR}(\tau)]\right\}\\
&=N\,\mathrm{Tr}_r\left\{\hat{D}_r^-e^{K_r^R\tau}\prod_p\mathrm{Tr}_p[P_cg_1^{IR}(\tau)+P_cg_2^{IR}(\tau)]\right\}\ . \qquad (57)
\end{aligned}
$$

Here g^{IR} are defined by

$$g_1^{IR}(\tau)\equiv U^{IR}(\tau)\rho^R(0)\hat{D}_r^+\ , \qquad (58)$$

and

$$g_2^{IR}(\tau)=U^{IR}(\tau)\rho^R(0)\sum_{p'}\hat{D}_{p'}^+\exp[i\Delta\mathbf{k}\cdot(\mathbf{r}_{p'0}-\mathbf{r}_{r0})]\ . \qquad (59)$$

Substituting Eq. (57) into Eq. (53), we find the stationary spectrum becomes

$$\sigma_{k\lambda}=2N\lim_{s\to0}\mathrm{Re}(\mathrm{Tr}_r\{\hat{D}_r^-[f_1(s)+f_2(s)]\})\ , \qquad (60)$$

where $f(s)$ is defined by

$$f(s)=\int_0^\infty e^{-(s-K_r^R)\tau}\prod_p\mathrm{Tr}_pP_cg^{IR}(\tau)d\tau\ . \qquad (61)$$

The scattered spectrum is therefore given by the Laplace transforms of $\prod_p\mathrm{Tr}_pP_cg_1^{IR}(\tau)$ and $\prod_p\mathrm{Tr}_pP_cg_2^{IR}(\tau)$ at $s'=s-K_r^R$.

From their definitions it is obvious that $P_cg^{IR}(\tau)$ satisfy the same equation [Eq. (26)] as $P_c\rho^{IR}(\tau)$ does. The only difference is that we can only assume now $Q_cg^{IR}(-\infty)=0$ [instead of $Q_cg^{IR}(0)=0$], since at $\tau=0$ the system has already reached a stationary state. With this initial condition, we have, after making a binary-collision approximation the same way as in Sec. IV,

$$\partial_\tau P_cg^{IR}(\tau)=NP_cV_{rp}^{IR}(\tau)P_cg^{IR}(\tau)+NP_cV_{rp}^{IR}(\tau)\int_0^\tau U_{rp}^{IR}(\tau,\tau')V_{rp}^{IR}(\tau')P_cg^{IR}(\tau')d\tau'+NP_cV_{rp}^{IR}(\tau)U_{rp}^{IR}(\tau,0)Q_cg^{IR}(0)\ , \qquad (62)$$

where

$$Q_cg^{IR}(0)=\int_{-\infty}^0G^{IR}(0,\tau')Q_cV^{IR}(\tau')P_cg^{IR}(\tau')d\tau'\ . \qquad (63)$$

We have not made the BCA on $Q_cg^{IR}(0)$ since it is not very useful at this stage.

Using the stationary property of the collisional average given by Eq. (42), and Eqs. (61) and (62), we obtain

$$f(s)=\mathcal{L}^{-1}\left[\prod_p\mathrm{Tr}_pP_cg^{IR}(0)+N\int_0^\infty e^{-(s-K_r^R)\tau}\prod_{\mathrm{Tr}_p}\mathrm{Tr}_pP_cV_{rp}^{IR}(\tau)U_{rp}^{IR}(\tau,0)d\tau Q_cg^{IR}(0)\right]\ . \qquad (64)$$

Here, $\mathcal{L}$ is a line-shape operator given by

$$\mathcal{L}=\mathcal{L}_0-\mathcal{L}_c \ , \tag{65}$$

and $\mathcal{L}_0$ is defined by

$$\mathcal{L}_0\equiv s-K_r^R \ . \tag{66}$$

We have also used

$$\mathcal{L}_c=N\mathcal{L}_0\mathrm{Tr}_p\int_0^\infty e^{-\mathcal{L}_0\tau}[U_{rp}^{IR}(\tau,0)-1]\rho_p^{\mathrm{st},R}d\tau\mathcal{L}_0 \tag{67}$$

$$=N\mathcal{L}_0\mathrm{Tr}_p\int_0^\infty[U_{rp}^{IR}(0,-\tau)-1]e^{-\mathcal{L}_0\tau}\rho_p^{\mathrm{st},R}d\tau\mathcal{L}_0 \ . \tag{68}$$

The task of deriving the spectrum is now reduced to deriving workable expressions for the two source terms (initial conditions) on the right-hand side of Eq. (64). The physical origin of the second source term is in the non-Markoffian nature of the redistribution problem [16]. Away from the line center ω_0, light can be emitted only *during* a collision in which the system is correlated.

The source terms for $f_1(s)$ are easy to handle. Keeping in mind that our system is in a steady state from $\tau\sim-1/s$ to $\tau=+1/s$, which eventually goes to $(-\infty,+\infty)$, we obtain

$$\prod_p \mathrm{Tr}_p P_c g_1^{IR}(0)=\left[\prod_p \mathrm{Tr}_p P_c\rho^R(0)\right]\hat{D}_r^+=\Sigma^{(0)}\hat{D}_r^+ \ ,$$

$$X_1\equiv P_c V_{rp}^{IR}(\tau)U_{rp}^{IR}(\tau,0)Q_c g_1^{IR}(0)$$

$$=P_c V_{rp}^{IR}(\tau)U_{rp}^{IR}(\tau,0)[Q_c\rho^R(0)]\hat{D}_r^+ \tag{69}$$

$$=P_c V_{rp}^{IR}(\tau)U_{rp}^{IR}(\tau,0)\left[\left[\int_{-\infty}^0 G^{IR}(0,\tau')Q_c V^{IR}(\tau')P_c\rho^{IR}(\tau')d\tau'\right]\hat{D}_r^+\right] \ .$$

The binary collision approximation on this term can be carried out the same way as in Sec. IV. Noticing that in a steady state $\prod_p \mathrm{Tr}_p\rho^{IR}(\tau)=e^{-L_r^R\tau}\Sigma^{(0)}$, we obtain

$$X_1=P_c V_{rp}^{IR}(\tau)U_{rp}^{IR}(\tau,0)\left[\lim_{s\to 0}\int_{-\infty}^0 U_{rp}^{IR}(0,\tau')V_{rp}^{IR}(\tau')\rho_p^{\mathrm{st},R}e^{(s-L_r^R)\tau'}d\tau'\,\Sigma^{(0)}\right]\hat{D}_r^+ \ . \tag{70}$$

The source terms for $f_2(s)$ are slightly more complicated,

$$\prod_p \mathrm{Tr}_p P_c g_2^{IR}(0)=\prod_p \mathrm{Tr}_p P_c\left[\sum_{p'}\hat{D}_{p'}^+\exp[i\Delta\mathbf{k}\cdot(\mathbf{r}_{p'0}-\mathbf{r}_{r0})]\rho^R(0)\right]$$

$$=\prod_p \mathrm{Tr}_p P_c\left[\sum_{p'}\hat{D}_{p'}^+\exp[i\Delta\mathbf{k}\cdot(\mathbf{r}_{p'0}-\mathbf{r}_{r0})](P_c+Q_c)\rho^R(0)\right] \ .$$

If $w(\mathbf{r}_{p0},\mathbf{v}_{p0})$ does not vary significantly over the range of $1/|\Delta\mathbf{k}|$ (we are not concerned here with the Rayleigh scattering in the forward direction since it cannot be measured experimentally, and $\mathbf{k}=\mathbf{k}_L$ in fact corresponds to radiation in the laser mode which is specifically excluded from our treatment), we have

$$\mathrm{Tr}_p\hat{D}_p^+\exp[i\Delta\mathbf{k}\cdot(\mathbf{r}_{p0}-\mathbf{r}_{r0})]\rho_p^{\mathrm{st},R}=0 \ . \tag{71}$$

Physically, this corresponds to the usual argument that, due to random phase (i.e., the $\exp[i\Delta\mathbf{k}\cdot(\mathbf{r}_{p0}-\mathbf{r}_{r0})]$ factors), the scattering in directions other than the forward direction is proportional to N rather than N^2. Therefore

$$\prod_p \mathrm{Tr}_p P_c g_2^{IR}(0)=\prod_p \mathrm{Tr}_p P_c\left[\sum_{p'}\hat{D}_{p'}^+\exp[i\Delta\mathbf{k}\cdot(\mathbf{r}_{p'0}-\mathbf{r}_{r0})]Q_c\rho^R(0)\right]$$

$$=N\,\mathrm{Tr}_p\left[\hat{D}_p^+\exp[i\Delta\mathbf{k}\cdot(\mathbf{r}_{p0}-\mathbf{r}_{r0})]\lim_{s\to 0}\int_{-\infty}^0 U_{rp}^{IR}(0,\tau')V_{rp}^{IR}(\tau')\rho_p^{\mathrm{st},R}e^{(s-L_r^R)\tau'}d\tau'\,\Sigma^{(0)}\right] \ . \tag{72}$$

If we define

$$X_2=P_c V_{rp}^{IR}(\tau)U_{rp}^{IR}(\tau,0)Q_c g_2^{IR}(0) \ ,$$

we can move all $P_{cp''}$ $(p''\neq p)$ from P_c (remember $P_{cp''}^2=P_{cp''}$) to Q_c to form

$$X_2=P_c V_{rp}^{IR}(\tau)U_{rp}^{IR}(\tau,0)(1-P_{cp})g^{IR}(0)$$
$$=P_c V_{rp}^{IR}(\tau)U_{rp}^{IR}(\tau,0)(1-P_{cp})\{[P_c\rho^R(0)+Q_c\rho^R(0)]\hat{D}_p^+\exp[i\Delta\mathbf{k}\cdot(\mathbf{r}_{p0}-\mathbf{r}_{r0})]\}\ .$$

We therefore obtain in thermodynamic limit [12] $(1-P_{cp}\approx 1)$,

$$X_2=P_c V_{rp}^{IR}(\tau)U_{rp}^{IR}(\tau,0)\left[\left[\rho_p^{\mathrm{st},R}\Sigma^{(0)}+\lim_{s\to 0}\int_{-\infty}^{0}U_{rp}^{IR}(0,\tau')V_{rp}^{IR}(\tau')\rho_p^{\mathrm{st},R}e^{(s-L_r^R)\tau'}d\tau'\,\Sigma^{(0)}\right]\hat{D}_p^+\exp[i\Delta\mathbf{k}\cdot(\mathbf{r}_{p0}-\mathbf{r}_{r0})]\right]\ . \tag{73}$$

In deriving X_2 we have again used Eq. (71).

Combining Eqs. (60), (64), (69), (70), (72), and (73), we can write after some integration by parts our final expression for the spectrum in the form

$$\sigma_{k\lambda}=2N\lim_{s\to 0}\mathrm{Re}\left[\mathrm{Tr}_r\hat{D}_r^-\mathcal{L}^{-1}\left[\Sigma^{(0)}\hat{D}_r^+ +N\,\mathrm{Tr}_p\left[\hat{D}_p^+\exp[i\Delta\mathbf{k}\cdot(\mathbf{r}_{p0}-\mathbf{r}_{r0})]\int_0^\infty[U_{rp}^{IR}(0,-\tau)-1]\right.\right.\right.$$
$$\left.\times e^{-(s-L_r^R)\tau}d\tau(s-L_r^R)\Sigma^{(0)}\rho_p^{\mathrm{st},R}\right]$$
$$+N\mathcal{L}_0\mathrm{Tr}_p\int_0^\infty e^{-\mathcal{L}_0\tau}[U_{rp}^{IR}(\tau,0)-1]d\tau\left[\left[\int_0^\infty[U_{rp}^{IR}(0,-\tau')-1]e^{-(s-L_r^R)\tau'}d\tau'\right.\right.$$
$$\left.\left.\times(s-L_r^R)\Sigma^{(0)}\rho_p^{\mathrm{st},R}\right]\{\hat{D}_r^+ +\hat{D}_p^+\exp[i\Delta\mathbf{k}\cdot(\mathbf{r}_{p0}-\mathbf{r}_{r0}]\}\right]$$
$$\left.\left.\left.+N\mathcal{L}_0\mathrm{Tr}_p\left[\int_0^\infty e^{-\mathcal{L}_0\tau}[U_{rp}^{IR}(\tau,0)-1]d\tau\{\Sigma^{(0)}\rho_p^{\mathrm{st},R}\hat{D}_p^+\exp[i\Delta\mathbf{k}\cdot(\mathbf{r}_{p0}-\mathbf{r}_{r0}]\}\right]\right]\right]\ . \tag{74}$$

This is the more traditional way of deriving the spectrum. We have found it worthwhile to also derive the spectrum following the procedure of Mollow [5,17]. The initial conditions are included more readily in such a procedure, which makes the derivation easier.

The rate of emitting one photon into mode (k,λ) at time t is given by

$$\sigma_{k\lambda}(t)=\partial_t\langle\langle\{1\}_{k\lambda},\{1\}_{k\lambda}|\mathrm{Tr}_A\mathrm{Tr}'_F\rho_{AF}(t)=\partial_t\langle\langle\{1\}_{k\lambda},\{1\}_{k\lambda}|\mathrm{Tr}_A\rho'(t)\ . \tag{75}$$

Tr'_F means tracing over all the field modes except (k,λ). $\rho_{AF}(t)$ is the full density matrix for the complete system of all atoms plus field modes. $\rho'(t)$ satisfies basically the same equation [Eq. (1)] as $\rho(t)$ except we still have the interaction with the mode (k,λ) left over by not tracing over it. Going to the rotating frame and interaction picture as we did earlier, we can write

$$\sigma_{k\lambda}(t)=\langle\langle\{1\}_{k\lambda},\{1\}_{k\lambda}|\mathrm{Tr}_A\partial_t\rho'^{IR}(t)\ , \tag{76}$$

where $\rho'^{IR}(t)$ satisfies [cf. Eq. (20)]

$$\partial_t\rho'^{IR}(t)=\sum_{\substack{a,b\\a<b}}V_{ab}^{IR}(t)\rho'^{IR}(t)+\sum_a L_{ak\lambda}^{IR}(t)\rho'^{IR}(t)\ , \tag{77}$$

where $L_{ak\lambda}^{IR}(t)$ is the interaction of the atom a with the radiation mode (k,λ); it is given in the rotating-wave approximation (RWA) by

$$L_{ak\lambda}^{IR}(t)=e^{-L_a^R t}[\hat{D}_a^-\exp[-i\Delta\mathbf{k}\cdot\mathbf{r}_a(t)+i\Delta\omega t]a_{k\lambda}^\dagger-\mathrm{H.c.},\cdot]e^{L_a^R t}\ . \tag{78}$$

Substituting Eqs. (77) and (78) into Eq. (76), we obtain

$$\sigma_{k\lambda}(t)=\langle\langle\{1\}_{k\lambda},\{1\}_{k\lambda}|\mathrm{Tr}_A\sum_a L_{ak\lambda}^{IR}(t)\rho'^{IR}(t)$$
$$=N\langle\langle\{1\}_{k\lambda},\{1\}_{k\lambda}|\mathrm{Tr}_r L_{rk\lambda}^{IR}(t)\prod_p\mathrm{Tr}_p P_c\rho'^{IR}(t)=2N\,\mathrm{Re}[\mathrm{Tr}_r\hat{D}_r^-F(t)]\ , \tag{79}$$

where we have defined

$$F(t)=\langle\langle\{0\}_{k\lambda},\{1\}_{k\lambda}|\exp(K_r^R t-i\Delta\mathbf{k}\cdot\mathbf{r}_{r0})\prod_p\mathrm{Tr}_p P_c\rho'^{IR}(t)\ . \tag{80}$$

Since eventually we will obtain the stationary spectrum from Eq. (79) by taking t to infinity, i.e.,

$$\sigma_{k\lambda}=2N\lim_{t\to\infty}\mathrm{Re}[\mathrm{Tr}_r\hat{D}_r^- F(t)]=2N\lim_{s\to 0}\mathrm{Re}[\mathrm{Tr}_r\hat{D}_r^- s\tilde{F}(s)]\ , \tag{81}$$

we can assume there is no correlations at $t=0$:

$$Q_c\rho'^{IR}(0)=0\ , \tag{82}$$

meaning no atom-atom correlation at $t=0$, and

$$\langle\langle\{0\}_{k\lambda},\{1\}_{k\lambda}|\rho'^{IR}(t)\equiv\langle\{0\}_{k\lambda}|\rho'^{IR}(t)|\{1\}_{k\lambda}\rangle=0\ , \tag{83}$$

meaning no atom-field correlation at $t=0$. The initial condition for $F(t)$ is therefore simply $F(0)=0$.

From Eq. (77), we can derive an equation of motion for $P_c\rho'^{IR}(t)$ in the same way as for $P_c\rho^{IR}(t)$. We obtain [cf. Eqs. (26) and (30)]

$$\begin{aligned}\partial_t P_c\rho'^{IR}(t)=&[L^{IR}_{rk\lambda}(t)+NP_cV^{IR}_{rp}(t)]P_c\rho'^{IR}(t)\\&+NP_c[L^{IR}_{pk\lambda}(t)+V^{IR}_{rp}(t)]\int_0^t G'^{IR}(t,t')Q_c\left[V^{IR}(t')+\sum_{p'}L^{IR}_{p'k\lambda}(t')\right]P_c\rho'^{IR}(t')dt'\ ,\end{aligned} \tag{84}$$

where we have used the initial condition given by Eq. (82). G'^{IR} is defined by

$$G'^{IR}(t,t')=T\exp\left[\int_{t'}^t Q_c\left[V^{IR}(t_1)+\sum_a L^{IR}_{ak\lambda}(t_1)\right]dt_1\right]\ . \tag{85}$$

Expanding G'^{IR} according to

$$G'^{IR}(t,t')=G^{IR}(t,t')+\int_{t'}^t G^{IR}(t,t_1)Q_c\sum_a L^{IR}_{ak\lambda}(t_1)G'^{IR}(t,t')dt_1\ , \tag{86}$$

and keeping only the terms up to first order in $L^{IR}_{ak\lambda}$ [we can use such a perturbative approach as long as there is no accumulation of photons in mode (k,λ)], we have

$$\begin{aligned}\partial_t P_c\rho'^{IR}(t)=&NP_cV^{IR}_{rp}(t)P_c\rho'^{IR}(t)+NP_cV^{IR}_{rp}(t)\int_0^t G^{IR}(t,t')Q_cV^{IR}(t')P_c\rho'^{IR}(t')dt'\\&+L^{IR}_{rk\lambda}(t)P_c\rho'^{IR}(t)+NP_cL^{IR}_{pk\lambda}(t)\int_0^t G^{IR}(t,t')Q_cV^{IR}(t')P_c\rho'^{IR}(t')dt'\\&+NP_cV^{IR}_{rp}(t)\int_0^t G^{IR}(t,t')Q_c\sum_{p'}L^{IR}_{p'k\lambda}(t')P_c\rho'^{IR}(t')dt'\\&+NP_cV^{IR}_{rp}(t)\int_0^t\int_{t'}^t G^{IR}(t,t_1)Q_c\sum_a L^{IR}_{ak\lambda}(t_1)G^{IR}(t_1,t')Q_cV^{IR}(t')P_c\rho'^{IR}(t')dt_1dt'\ .\end{aligned} \tag{87}$$

The binary-collision approximation can be carried out in the same way as in Sec. IV. With the help of Eq. (71), we get

$$\begin{aligned}\partial_t P_c\rho'^{IR}(t)=&NP_cV^{IR}_{rp}(t)P_c\rho'^{IR}(t)+NP_cV^{IR}_{rp}(t)\int_0^t U^{IR}_{rp}(t,t')V^{IR}_{rp}(t')P_c\rho'^{IR}(t')dt'\\&+L^{IR}_{rk\lambda}(t)P_c\rho'^{IR}(t)+NP_cL^{IR}_{pk\lambda}(t)\int_0^t U^{IR}_{rp}(t,t')V^{IR}_{rp}(t')P_c\rho'^{IR}(t')dt'\\&+NP_cL^{IR}_{pk\lambda}(t)P_c\rho'^{IR}(t)+NP_cV^{IR}_{rp}(t)\int_0^t U^{IR}_{rp}(t,t')L^{IR}_{pk\lambda}(t')P_c\rho'^{IR}(t')dt'\\&+NP_cV^{IR}_{rp}(t)\int_0^t\int_{t'}^t U^{IR}_{rp}(t,t_1)[L^{IR}_{rk\lambda}(t_1)+L^{IR}_{pk\lambda}(t_1)]U^{IR}_{rp}(t_1,t')V^{IR}_{rp}(t')P_c\rho'^{IR}(t')dt_1dt'\ .\end{aligned} \tag{88}$$

It is now straightforward to derive an equation of motion for $F(t)$. Using again Eq. (71) and the stationary property given by Eq. (42), we have

$$\begin{aligned}\partial_t F(t)=&K_r^R F(t)+NTr_pV^{IR}_{rp}(0)\rho_p^{st,R}F(t)+N\,\mathrm{Tr}_pV^{IR}_{rp}(0)\int_0^t U^{IR}_{rp}(0,t'-t)V^{IR}_{rp}(t'-t)\rho_p^{st,R}e^{K_r^R(t-t')}F(t')dt'+\Sigma^{(0)}(t)\hat{D}_r^+\\&+N\,\mathrm{Tr}_p\hat{D}_p^+\exp[i\Delta\mathbf{k}\cdot(\mathbf{r}_{p0}-\mathbf{r}_{r0})]\int_0^t U^{IR}_{rp}(0,t'-t)V^{IR}_{rp}(t'-t)\rho_p^{st,R}e^{L_r^R(t-t')}\Sigma^{(0)}(t')dt'\\&+N\,\mathrm{Tr}_p\int_0^t e^{K_r^R(t-t')}V^{IR}_{rp}(t-t')U^{IR}_{rp}(t-t',0)\{\rho_p^{st,R}\Sigma^{(0)}(t')\hat{D}_p^+\exp[i\Delta\mathbf{k}\cdot(\mathbf{r}_{p0}-\mathbf{r}_{r0})]\}dt'\\&+N\,\mathrm{Tr}_p\int_0^t e^{K_r^R(t-t_1)}V^{IR}_{rp}(t-t_1)U^{IR}_{rp}(t-t_1,0)\\&\times\left[\int_0^{t_1}U^{IR}_{rp}(0,t'-t_1)V^{IR}_{rp}(t'-t_1)e^{L_r^R(t_1-t')}\Sigma^{(0)}(t')\rho_p^{st,R}\right]\{\hat{D}_r^+\hat{D}_p^+\exp[i\Delta\mathbf{k}\cdot(\mathbf{r}_{p0}-\mathbf{r}_{r0})]\}]dt'dt_1\ .\end{aligned} \tag{89}$$

In this equation $\Sigma^{(0)}(t)$ is defined by

$$\Sigma^{(0)}(t)=\langle\langle\{0\}_{k\lambda},\{0\}_{k\lambda}|e^{L_r^R t}\prod_p \mathrm{Tr}_p P_c\rho'^{IR}(t)\ .$$

From Eq. (88), it is easy to show that it satisfies the same equation as the $\Sigma^{(0)}$ defined earlier by Eq. (41), and is therefore the same quantity.

Taking the Laplace transform of Eq. (89) [keeping in mind that we have the initial condition $F(0)=0$], we obtain after some integration by parts the same result given by Eq. (74).

The spectrum given by Eq. (74) is the main result of this paper. It forms the basis for calculating, for example, the emission spectrum from a system of cold atoms in a trap. It is also directly related to the probability of one or more photons being emitted during a single collision, which is important for understanding the radiative escape [2] and other fascinating behavior of cold atoms in a laser field [4,18].

VI. DISCUSSION AND CONCLUSIONS

As indicated in Sec. IV, the collisional width γ_c is orders of magnitude smaller than the radiative decay width γ. We can therefore expand our line-shape operator given by Eqs. (65)–(68) to the first order in collisional width according to

$$\mathcal{L}^{-1}=\frac{1}{\mathcal{L}_0}+\frac{1}{\mathcal{L}_0}\mathcal{L}_c\frac{1}{\mathcal{L}}\approx\frac{1}{\mathcal{L}_0}+\frac{1}{\mathcal{L}_0}\mathcal{L}_c\frac{1}{\mathcal{L}_0}\ . \tag{90}$$

The validity of this expansion has to be considered carefully. To evaluate $1/\mathcal{L}_0$ we use eigen-tetradic states (complex dressed states) of the Liouville L_r^R. For a two-state atom these eigenvalues (given by $\det|s-L_r^R|=0$) correspond to $s=0$ and the roots of the Mollow cubic [15]. The damping part of the roots of the cubic is of order γ, which is large compared to elements of L_c (of order at most γ_c with $1/\gamma_c$ the time between collisions T_{coll}). The $s=0$ root corresponds to the steady state, for which $L_r^R\rho_r^{\mathrm{st},R}=0$. The steady state, when collisions are included, however, corresponds to Eq. (45), i.e., to the eigen-tetradic of $L_r^R+L_c$ with eigenvalue $s=0$. Consequently, perturbation theory, to lowest order in L_c, shows that the diagonal tetradic matrix element of L_c in the eigen-tetradic of L_r^R for eigenvalue $s=0$ is zero. This means that no divergent terms proportional to $1/s^2$ (with $s=0$) occur in the second term of Eq. (90). Thus the second term in Eq. (90) is of order γ_c/γ compared to the first.

It should be noted that the $1/s$ pole corresponds to Rayleigh scattering, so that when the first term in Eq. (90) corresponds to this pole, the second term indicates a modification to the Rayleigh scattering of order γ_c/γ.

The stationary density matrix in the presence of collisions defined by Eqs. (45) and (46) can also be expanded to the first order in collisional width,

$$\Sigma^{(0)}=\rho_r^{\mathrm{st},R}+\lim_{s\to0}\left[\frac{1}{s-L_r^R}L_c\Sigma^{(0)}\right]$$
$$\approx\rho_r^{\mathrm{st},R}+\lim_{s\to0}\left[\frac{1}{s-L_r^R}L_c\rho_r^{\mathrm{st},R}\right]\ . \tag{91}$$

After some straightforward algebra, we obtain the stationary spectrum to first order in collisional width,

$$\sigma_{k\lambda}=2N\lim_{s\to0}\mathrm{Re}\Big[\mathrm{Tr}_r\hat{D}_r^-\Big\{\mathcal{L}_0^{-1}(\rho_r^{\mathrm{st},R}\hat{D}_r^+)+N\mathcal{L}_0^{-1}(\mathrm{Tr}_p[(s-L_r^R)^{-1}V_{rp}^{IR}(0)U_{rp}^{IR}(0,-\infty)\rho_r^{\mathrm{st},R}\rho_p^{\mathrm{st},R}]\hat{D}_r^+$$
$$+\mathrm{Tr}_p\{\hat{D}_p^+\exp[i\Delta\mathbf{k}\cdot(\mathbf{r}_{p0}-\mathbf{r}_{r0})][s-L_r^R-L_p^R-i\Delta\mathbf{k}\cdot(\mathbf{v}_p-\mathbf{v}_r)]^{-1}$$
$$\times V_{rp}^{\mathrm{IR}}(0)U_{rp}^{\mathrm{IR}}(0,-\infty)\rho_r^{\mathrm{st},R}\rho^{\mathrm{st},R}\})$$
$$+N\,\mathrm{Tr}_p\int_0^\infty e^{-\mathcal{L}_0\tau}[U_{rp}^{IR}(\tau,0)-1]d\tau$$
$$\times([U_{rp}^{IR}(0,-\infty)\rho_r^{\mathrm{st},R}\rho_p^{\mathrm{st},R}][\hat{D}_r^++\hat{D}_p^+\exp[i\Delta\mathbf{k}\cdot(\mathbf{r}_{p0}-\mathbf{r}_{r0})]])\Big\}\Big]\ . \tag{92}$$

The first term is the spectrum in the absence of collisions. It is the familiar Mollow spectrum [15] for a two-state atom. The $1/s$ pole corresponds to Rayleigh scattering and the roots of the cubic give rise to the Mollow triplet. The second term represents the effect of the collisions on the steady-state density matrix. This modification, which physically described the effect of collisions on the population (in contrast to coherences) of complex dressed states [11], affects both the "triplet" and Rayleigh components of the spectrum. The third term is the "collisional broadening" term corresponding to the destruction of optical coherence. It describes the process of emission during a collision. The $U_{rp}^{IR}(0,-\infty)$ part of this term gives the evolution during a collision of the states of the radiator and the perturber from free states at $\tau=-\infty$ until at $\tau=0$ a dipole coherence is created by $\hat{D}_r^+$ (or $\hat{D}_p^+$). The $U_{rp}^{IR}(\tau,0)-1$ integral then describes the decay of this coherence. Note, in particular, that the evolution from $-\infty$ is specific for a given collision since both $U_{rp}^{IR}(0,-\infty)$ and $U_{rp}^{IR}(\tau,0)$ are calculated for the same single-collision parameters. Due to the $s=0$ steady-state pole of the $\mathcal{L}_0$ operator this third term also contains corrections to the Rayleigh component.

Due to the $e^{i\mathbf{k}\cdot\mathbf{r}}$ factors in V_{rp}^{IR} [see Eq. (13c)] and that multiplying $\hat{D}_p^+$, the above expression specifically includes all Doppler effects. In addition, we emphasize that the modification of the radiative decay is fully included as well as fully retarded interactions.

Another interesting feature of this spectrum is that it includes, in both the second and third term in Eq. (92), the effects of resonant exchange of coherence and excitation. This exchange affects the spectrum through the terms of the type $\hat{D}_r^-[U_{rp}^{IR}(\tau,0)-1]\hat{D}_p^+$. In the context of resonance broadening Berman and Lamb [19] (see also Ben-Reuven [3]) have obtained expressions for the line shape by keeping track of the excitation which, following a collision, may either remain on the original radiator or be transferred to its collision partner. Within appropriate limits, which correspond to making (i) $1/\Delta\omega$ expansion of the resonant broadening line shape [a single-collision expansion equivalent to our expansion of Eq. (90)] and (ii) making an impact limit on our expressions with radiative decay during a collision being ignored and driving field being weak, it can be shown that our results for both direct and exchange of excitation terms agree with those of Berman and Lamb and Ben-Reuven. Of course our results are not restricted to the impact limit, or to the neglect of radiative decay, or to weak field. Further, not only can exchange of excitation occur, but a coherent superposition (atom-atom coherence) can also exist after a collision and is again included in our formalism, i.e., we include two-atom coherences as well as one-atom coherences (excitation).

For $|\omega'-\omega_0| \gg \gamma,\Omega$, emission is possible only during a collision (except for the Rayleigh scattering, of course). In this large-detuning limit, the integration in the third term of the spectrum can be carried out by a stationary phase method [20]. This integration corresponds to emission at the stationary phase point (equivalent to a Frank-Condon transition). In the weak-field limit, when the initial propagation in the collision due to $U_{rp}^{IR}(0,-\infty)$ corresponds to a single absorption before the emission takes place, we can recover the results for absorption and emission during a collision, as given by Gallagher and Pritchard [2]. We note that Gallagher and Pritchard use a trajectory that is modified by the potential, whereas for convenience we used straight lines. In the general theory of redistribution due to foreign gases [16] in the weak-field limit various types of absorption and emission processes occur. Although we choose to work with complex dressed states to describe the free evolution we stress that all such effects are included here. In fact, we emphasize that the use of these complex dressed states includes multiple emission and absorption to all orders. Before the radiative event for the mode of interest (k,λ), characterized by the occurrences of $\hat{D}_r^+$ (or $\hat{D}_p^+$), the $U_{rp}^{IR}(0,-\infty)$ can describe multiple absorption and spontaneous emission into any mode other than (k,λ). Similar physics is also implied by $U_{rp}^{IR}(\tau,0)$ in the third terms of Eq. (92). The treatment of Gallagher and Pritchard [2] does not include these reexcitation events. It is interesting that the reexcitation is related in phase to the driving laser field, thus the different evolution periods in the collision are correlated. This leads, among other interesting features, to the contribution of the third term to the coherent Rayleigh scattering.

Finally, we mention again that we have, for convenience, assumed a straight-line classical trajectory. This appears to omit two potentially important effects. The first is the deviation from a straight line due to close strong interactions, which tend to dominate the spectrum in the far wings (the quasistatic region). These close, small r, collisions are amenable to a quantum treatment (see, for example, Julienne and Vigue [21]) provided suitable connection is made to the large-r behavior where straight lines are appropriate. Alternatively, to a first approximation the particle could follow trajectories determined by forces obtained via Ehrenfest's theorem. In principle, it is straightforward to deal with deviations from a straight line.

The second effect is more subtle, but of crucial importance to the theory of laser cooling; namely, the forces and diffusive motion due to the recoil as a result of spontaneous emission. In principle, one could treat the center-of-mass motion quantum mechanically and our Liouville equations of motion would be replaced by equations for Wigner distributions [22].

Although these are interesting effects, in general, they are not very important for determining the collisional part of the spectrum which is the quantity of interest in this study. For a frequency separation of $\Delta\omega$, the "time of interesting" is of the order of $1/|\Delta\omega|$, which for the collisional part of the spectrum ($\Delta\omega > \gamma$) is at most $1/\gamma$ where γ is the radiative decay rate (and much less in the wings). The narrow features observed by Westbrook *et al.* [23] are due to constraining the free motion within a volume of $\sim\lambda^3$ (i.e., Dicke narrowing). The time scale for these features corresponds to the time to move a distance of the order of a wavelength, which is longer than $1/\gamma$. This "free motion" therefore does not affect regions of the spectrum where $\Delta\omega > \gamma$ (in fact, in this approach we are not concerned with such localization of atomic motion since we are only considering a running wave). The quantity which determines the collisional part of the spectrum is qualitatively the phase shift η due to the interaction, i.e., $\eta = \int_0^t V\,dt'/\hbar$ where V is the appropriate interaction. Simple Weisskopf theory shows that in the impact region of the spectrum the most important regions are those for which $\eta \sim 1$. For times of interest $\sim 1/\gamma$ this gives important impact parameters of the order of $\lambda/2\pi$ for cold collisions (and less for $\Delta\omega$ in the line wings). The relevant question now is: "Will the effect of recoil change the trajectory by a sufficient amount that the phase shift due to a collision as calculated by straight lines is in significant error?" The answer is "no," since the deviation in the trajectory due to recoil in a time of $1/\gamma$ is, for a particle of mass M, approximately $2\pi\hbar/M\gamma\lambda$ which is typically negligible ($\sim 10^{-3}$) compared to $\lambda/2\pi$. Thus during the time of interest deflections have negligible effect on the collisional phase shift (and hence the spectrum). The deflections due to recoil momentum *are,* of course, very significant over the longer periods of time needed for cooling or Dicke narrow. A similar effect occurs in the usual line-broadening theory where often straight lines are fine for calculating broadening cross sections (especially in cases such as that considered here where the potential is very long range) but hopeless for calculating differential cross sections (i.e., deflections).

In conclusion, we have derived the redistribution spec-

trum for a system of cold identical atoms in a driving laser field. Obviously spontaneous decay and long-range interactions play a significant role in cold identical atom collisions; however, more detailed understanding awaits future numerical calculations.

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[1] M. Trippenbach, B. Gao, J. Cooper, and K. Burnett, preceding paper, Phys. Rev. A **45**, 6539 (1992).
[2] A. Gallagher and D. E. Pritchard, Phys. Rev. Lett. **63**, 957 (1989).
[3] A. Ben-Reuven, Phys. Rev. A **22**, 2572 (1980); **22**, 2585 (1980).
[4] A. M. Smith and K. Burnett, J. Opt. Soc. Am. B **8**, 1592 (1991), and references therein.
[5] B. R. Mollow, Phys. Rev. A **12**, 1919 (1975).
[6] M. Abramowitz and I. A. Stegun, *Handbook of Mathematical Functions* (NBS, Washington, DC, 1964), p. 437.
[7] See, for example, K. Burnett, J. Cooper, P. Kleiber, and A. Ben Reuven, Phys. Rev. A **25**, 1345 (1982).
[8] See, for example, H. Margenau and N. R. Kestner, *Theory of Intermolecular Forces* (Pergamon, Oxford, 1969).
[9] E. W. Smith, C. R. Vidal, and J. Cooper, J. Res. Natl. Bur. Stand. **73A**, 405 (1969).
[10] K. Burnett, J. Cooper, R. J. Ballagh, and E. Smith, Phys. Rev. A **22**, 2005 (1980).
[11] L. Armstrong, Jr. and H. C. Baker, J. Phys. B **13**, 4727 (1980).
[12] E. W. Smith, J. Cooper, and R. Vidal, Phys. Rev. **185**, 140 (1969).
[13] D. Sesko, T. Walker, C. Monroe, A. Gallagher, and C. Wieman, Phys. Rev. Lett. **63**, 961 (1989); T. Walker, D. Sesko, and C. Wieman, *ibid.* **64**, 408 (1990).
[14] V. Weisskopf, Z. Phys. **75**, 287 (1932). See also D. Mihalas, *Stellar Atmospheres* (Freeman, New York, 1978), p. 283.
[15] B. R. Mollow, Phys. Rev. **188**, 1969 (1969).
[16] K. Burnett and J. Cooper, Phys. Rev. A **22**, 2044 (1980).
[17] G. Alber and J. Cooper, Phys. Rev. A **33**, 3084 (1986).
[18] P. S. Julienne and F. H. Mies, J. Opt. Soc. Am. B **6**, 2257 (1989).
[19] P. R. Berman and W. E. Lamb, Phys. Rev. **187**, 221 (1969).
[20] See, for example, G. Alber and J. Cooper, Phys. Rev. A **31**, 3644 (1985).
[21] P. S. Julienne and J. Vigué, Phys. Rev. A **44**, 4464 (1991).
[22] J. Dalibard and C. Cohen-Tannoudji, J. Phys. B **18**, 1661 (1985).
[23] C. I. Westbrook, R. N. Watts, C. E. Tanner, S. L. Rolston, W. D. Phillips, P. D. Lett, and P. L. Gould, Phys. Rev. Lett. **65**, 33 (1990).