# Effect of scattered radiation on sub-Doppler cooling

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In this paper we discuss the effect of scattered radiation generated by a gas of cold atoms on the temperature of the gas. We show that by treating the reradiated field of the atoms as a fluctuating background field we can derive a master equation for a single atom where the effect of the surrounding medium is included in an effective relaxation operator. This relaxation operator is of second order in the interaction with the medium and in the binary-collision approximation can be written as the sum of two-body interactions with a correlation time equal to the natural lifetime of the atomic transition. The effect of the medium on the two most important sub-Doppler cooling mechanisms, Sisyphus and motion-induced orientation cooling, is investigated analytically in a one-dimensional model. In this we restrict ourselves to the limit of low saturation, weak background field and take the rate-equation limit for the collision operator. We find that both mechanisms are very sensitive to such a background field and that the temperature increases approximately as the number of atoms to the one-third power.

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## I. INTRODUCTION

At the high densities that are now routinely produced in magneto-optical traps, interactions between ultracold atoms become important. These ultracold collisions can be divided into two categories depending on whether or not the presence of laser light during the interaction is critical. In optical traps the type of collisions in which atoms are simultaneously driven by the laser is dominant. (Collisions where both atoms are in the excited state or the ground state are usually neglected in optical traps.) We shall assume that the collisions are sufficiently long range that the resonant dipoledipole interaction is dominant rather than the higher-order (in  $r^{-1}$ ) van der Waals interaction. The excited-state-groundstate collisions that we are considering here can crudely be divided into two regimes. The first one is a dynamic regime, where the presence of the perturbing atom significantly changes the dynamic evolution of the radiating atom during the collision and therefore correlations develop between a colliding pair of atoms. The second is a radiative regime, where the atom-atom interaction is due to the exchange of photons scattered from the laser beam (radiation trapping) and the effect of the interatomic potential can be treated by perturbation theory, i.e., correlations between the colliding atoms can be neglected. In the dynamic regime, which is restricted to interatomic distances  $r \leq \chi_0$ , the potential is described by the highest-order term in 1/r, i.e.,  $V \propto 1/r^3$ . For a laser detuned to the red of the atomic resonance transition, the detuning is of the same order of magnitude as the shift induced by the dipole-dipole interaction at some distance rand there is a significant increase of the excitation probability of an excited-state-ground-state pair. Once such a pair is produced its movement is strongly affected by the large gradient of the excited-state-ground-state potential surface leading to a strong correlation between the colliding atoms and it is therefore necessary to look in detail at the evolution of the pair during the collision. Collision-induced heating and trap loss in this regime occur through radiative escape and through fine- and hyperfine-structure changing processes. Theoretical investigations of these mechanisms have been carried out by Gallagher and Pritchard [1] and Julienne and Vigué [2], and recently alternative approaches have been developed, see, for example, Refs. [3-6] for some recent work on this subject. Little work has been done in the intermediate regime where the far-field approximation breaks down but the separation of the atoms is not small enough to concentrate fully on the  $1/r^3$  term of the dipole-dipole potential. Smith and Burnett [7-9] have carried out simulations on a pair of colliding atoms using the full dipole-dipole interaction, but in their final result they average over a distribution of nearest neighbors rather than taking into account the cumulative effect of the interactions with all atoms in a gas in a proper way by averaging over the whole gas volume. In the radiative regime, we assume that the mean separation of the atoms r is much larger than the atomic transition wavelength  $r \gg \chi_0$  and the interaction between a pair of atoms is proportional to the lowest-order term of the dipole-dipole potential between two driven atoms  $V \propto 1/r$  and this is the regime we focus on in this paper. Using a two-level model we showed in a previous paper [10] that the photon-exchange interaction in this regime not only produces radiation trapping forces, which have been discussed by Sesko et al. [11] and more recently in a generalized treatment by Ellinger et al. [12], but also gives rise to an extra heating term. In the regime of a constant density this extra heating scales as the number of atoms to the one-third power, when the average over all pairs in the cloud of cold atoms is taken. However, because of the restriction of that model to a two-state atom, we could not investigate the effect of radiation trapping on the sub-Doppler cooling mechanisms that rely on the multilevel structure of the atoms. Recent measurements on trapped cesium atoms by Drewsen et al. [13] and Cooper et al. [14] show that the temperature depends on the number of atoms to the one-third power. In this paper, we show that even small intensities of scattered background radiation strongly affect the efficiency of the Sisyphus and motion-induced orientation cooling mechanism [15] and that the minimum temperature increases nearly linearly as the number of atoms in

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the cloud to the one-third power. The physical picture we develop is that of a medium of well localized atoms that generates a background of scattered photons as part of their response to the driving field. This field is then treated as a fluctuating background field. (We note that we do not symmetrize the atomic wave functions as we assume that they are spatially well separated.)

This paper is organized as follows. In Sec. III we use the projection operator methods developed by Zwanzig [16] to derive the master equation for a single atom, subsequently referred to as the radiator, which is driven by a laser field and interacts with a surrounding cloud of similar atoms. Such projection operator methods have also been employed by several authors to study collisional redistribution of light by interactions with atoms of a different species that do not interact with the driving field (foreign gas broadening problem). We shall for convenience refer to those by Burnett et al. [17,18]. We derive a collision operator in which both the radiator and the perturbers interact with the driving field to all orders. We discuss the collision operator in the binarycollision approximation (BCA) and decompose it into a sum over two-body scattering events, each of which has the structure of a time integral over the product of two-time correlation functions of the dipole of the radiator and a perturber. In Sec. IV we discuss the rate-equation limit of the collision operator and in Sec. V give an estimate for the coupling strength of the background field to the radiator. The rateequation limit enables us to obtain analytical results for the influence of the scattered radiation on the two sub-Doppler cooling mechanisms that are most important in magnetooptical traps: Sisyphus cooling and motion-induced orientation cooling. This will give us a qualitative insight into the physical mechanisms leading to the reduction of cooling. These calculations are done in Secs. VI and VII. Finally, in Sec. VIII we summarize the results obtained in this paper and discuss some of the assumptions made in deriving them in conjunction with experimental data. Before we commence with the derivation, we introduce our mathematical notation in Sec. II.

#### **II. NOTATIONAL MATTERS**

In this section we briefly introduce our notational conventions. The equation of motion for the density matrix of the complete system, the radiator, the perturbers, and the vacuum field modes, is given by

$$\frac{d}{dt}\hat{\rho} = -\frac{i}{\hbar}\tilde{L}(t)\hat{\rho},\qquad(1)$$

where  $\tilde{L}(t)$  is the time-dependent Liouville operator

$$\tilde{L}(t)\hat{\rho} = [\hat{H}(t), \hat{\rho}].$$
<sup>(2)</sup>

The time-dependent Hamiltonian for the full system is given by

$$\hat{H}(t) = \hat{H}_{s} + \hat{H}_{R} + \hat{V}_{s-L}(t) + \hat{V}_{s-R}, \qquad (3)$$

where  $\hat{H}_s$  is the free Hamiltonian for a system of N atoms

$$\hat{H}_{s} = \sum_{i=1}^{N} \left( \frac{\hat{\mathbf{P}}^{2}}{2M} + \hbar \,\omega_{0} \hat{P}_{i,e} \right), \tag{4}$$

 $\hat{H}_R$  is the free Hamiltonian of the vacuum field

$$\hat{H}_{R} = \sum_{\mathbf{k},\lambda} \hbar \omega_{k} \hat{a}^{\dagger}_{\lambda,\mathbf{k}} \hat{a}_{\lambda,\mathbf{k}}, \qquad (5)$$

and  $P_{i,e}$  is the projection operator onto the excited state of the *i*th atom.  $\hat{a}^{\dagger}_{\lambda,\mathbf{k}}$  and  $\hat{a}_{\lambda,\mathbf{k}}$  are the photon creation and annihilation operators for the vacuum field mode  $\{\lambda, \mathbf{k}\}$ . They satisfy the usual commutation relation

$$[\hat{a}_{\lambda,\mathbf{k}},\hat{a}^{\dagger}_{\lambda',\mathbf{k}'}] = \delta_{\mathbf{k},\mathbf{k}'}\delta_{\lambda,\lambda'}.$$
(6)

 $\hat{V}_{s-L}(t)$  is the interaction of the atomic system with the laser, i.e.,

$$\hat{V}_{s-L}(t) = \sum_{i=1}^{N} \hat{V}_{i,L}, \qquad (7)$$

with

$$\hat{V}_{i,L} = -[\hat{\mathbf{D}}_i \cdot \mathbf{E}_L(r_i, t)].$$
(8)

The laser field is treated as a *c* number and the dipole operator for the *i*th atom  $\hat{\mathbf{D}}_i$  can be written as

$$\mathbf{\hat{D}}_i \equiv |D| \mathbf{\hat{d}}_i \,. \tag{9}$$

Here |D| is the reduced atomic dipole moment and  $\hat{\mathbf{d}}_i$  is the reduced atomic dipole operator for atom *i*. The coupling of the *i*th atom to the quantized reservoir of the vacuum field modes is given by

$$\hat{V}_{i,R} = -i\hbar \sum_{\mathbf{k},\lambda} \left( |D| \sqrt{\frac{2\pi\omega_k}{\hbar V}} \right) (\hat{\mathbf{d}}_i \cdot \boldsymbol{\epsilon}_\lambda) [e^{i\mathbf{k}\cdot\hat{\mathbf{R}}_i} \hat{a}_{\lambda,\mathbf{k}} - \hat{a}^{\dagger}_{\lambda,\mathbf{k}} e^{-i\mathbf{k}\cdot\hat{\mathbf{R}}_i}],$$
(10)

where V is the normalization volume and we have  $\hat{V}_{s-R} = \sum_{i=1}^{N} \hat{V}_{i,R}$ .

### **III. THE ONE-ATOM MASTER EQUATION**

In this section we show how to derive the master equation for the reduced density matrix of the radiator. The derivation of the equation of motion of the reduced density operator then proceeds in two stages. First the vacuum field is eliminated by introducing the well known spontaneous decay terms as well as giving rise to an atom-atom interaction that is due to the exchange of scattered photons. This elimination using projection operators is discussed in detail by Burnett *et al.* in Ref. [17]. The Liouville space is divided into a subspace in which the density matrix factorizes into the density matrix for the radiation field  $\hat{\rho}_R$  and the reduced density matrix for the radiator and the perturbers  $\hat{\sigma}(t) = \text{Tr}_R[\hat{\rho}(t)]$ and its complement. The final result of this elimination procedure is equal to [cf. Eq. (2.28) in Ref. [17]]

$$\frac{d}{dt}\hat{\sigma}(t) = -\frac{i}{\hbar} [\tilde{L}_s + \tilde{L}_{s-L}(t) + \tilde{V} + \tilde{S}]\hat{\sigma}(t).$$
(11)

Here  $\tilde{L}_s$  and  $\tilde{L}_{s-L}(t)$  describe the free evolution of the atoms and the atom-laser coupling, respectively. The Liouville space operator  $\tilde{S}$  describes the decay of the individual atoms due to the coupling to the vacuum field. It can be written as the sum of the decay terms of the individual atoms  $\tilde{S} = \Sigma_i \tilde{S}_i$ . In the rotating-wave approximation the decay of the individual atoms is described by the well known expression [32]

$$-\frac{i}{\hbar}\tilde{S}_{i}\hat{\sigma}(t) = -\frac{\Gamma}{2}[(\hat{\mathbf{d}}_{i}^{+}\cdot\hat{\mathbf{d}}_{i}^{-})\hat{\sigma}(t) + \hat{\sigma}(t)(\hat{\mathbf{d}}_{i}^{+}\cdot\hat{\mathbf{d}}_{i}^{-})] + \Gamma\sum_{\lambda}\int\frac{d\Omega}{8\pi/3}(\hat{\mathbf{d}}_{i}^{-}\cdot\boldsymbol{\epsilon}_{\lambda}) \times e^{-i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{i}}\hat{\sigma}(t)e^{i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{i}}(\hat{\mathbf{d}}_{i}^{+}\cdot\boldsymbol{\epsilon}_{\lambda}).$$
(12)

The operator  $\tilde{V}$  describes the coupling of the atoms of the gas to each other via the exchange of scattered photons, which is the dominant interaction for large interatomic separations. The interaction  $\tilde{V}$  can be written as a sum over the photon exchange interactions between all possible pairs in the cloud  $\tilde{V} = \sum_i \sum_{j,j>i} \tilde{V}_{ij}$ , where  $\tilde{V}_{ij}$  is the interaction of the *i*th and the *j*th atom in the cloud. The interaction between a pair of atoms comprises two contributions: the first one consists of the dipole of atom *i* scattering photons from the laser field that are absorbed by atom *j*; the second contribution comes from the reverse process, i.e., atom *i* absorbing photons scattered by atom *j*. The detailed expressions for the interatomic potential are given in the Appendix.

In order to proceed further with the derivation of the oneatom density matrix we now separate the Liouville operator for the atomic system into the operator for the free evolution of the radiator  $\tilde{L}_a$  and the perturbers,  $\tilde{L}_p$ ,

$$\tilde{L}_s = \tilde{L}_a + \tilde{L}_p \,. \tag{13}$$

We will adopt the convention that quantities referring to the radiator have a subscript a and quantities referring to the perturbers as a whole have a subscript p. Operators that refer to individual perturbers are denoted with the letter j, where j runs from 1 to N, N being the number of perturbers. Hence we have

$$\tilde{S} = \tilde{S}_a + \tilde{S}_p \,, \tag{14}$$

$$\tilde{L}_{s-L}(t) = \tilde{L}_{a-L}(t) + \tilde{L}_{p-L}(t).$$
(15)

The two projection operators  $\tilde{P}_c$  and  $\tilde{Q}_c$  are defined by

$$\tilde{P}_c \hat{\sigma}(t) = \hat{\sigma}_p(t) \otimes \operatorname{Tr}_{\text{pert}}[\hat{\sigma}(t)] = \hat{\sigma}_p(t) \otimes \hat{\sigma}_a(t), \quad (16)$$

$$\tilde{Q}_c = 1 - \tilde{P}_c \,, \tag{17}$$

where  $\hat{\sigma}_a(t)$  is the reduced density matrix for the radiator. Note that in contrast to the derivation of the collision operator presented in Ref. [17] the factorized part of the density matrix of the perturbers  $\hat{\sigma}_p(t)$  has the same time argument as the density-matrix of the radiator. This is due to the fact that the perturbers still evolve due to the driving field, the coupling to the vacuum modes, and interactions with each other.

The equation of motion for the factorized part of  $\hat{\sigma}(t)$ ,  $\tilde{P}_c \hat{\sigma}(t)$ , is found by projecting Eq. (11) onto the two subspaces defined by  $\tilde{P}_c$  and  $\tilde{Q}_c$  and substituting the formal solution for  $\tilde{Q}_c \hat{\sigma}(t)$  back into the equation of motion for  $\tilde{P}_c \hat{\sigma}(t)$ . This yields

$$\frac{d}{dt}\tilde{P}_{c}\hat{\sigma}(t) = -\frac{i}{\hbar}\tilde{P}_{c}[\tilde{L}_{a} + \tilde{L}_{a-L}(t) + \tilde{V} + \tilde{S}_{a}]\tilde{P}_{c}\hat{\sigma}(t) + \left(-\frac{i}{\hbar}\right)\tilde{P}_{c}\tilde{V}\tilde{U}_{c}(t,t_{0})\tilde{Q}_{c}\hat{\sigma}(t_{0}) + \left(-\frac{i}{\hbar}\right)^{2}\int_{t_{0}}^{t}dt_{1}\tilde{K}_{c}(t,t_{1})\tilde{P}_{c}\hat{\sigma}(t_{1}).$$
(18)

The collision kernel  $\tilde{K}_c(t,t_1)$  gives the full contribution of the radiator perturber interactions to the evolution of the factorized density matrix  $\tilde{P}_c \hat{\sigma}(t)$ . It is defined as

$$\tilde{K}_{c}(t,t_{1}) \equiv \tilde{P}_{c} \tilde{V} \tilde{Q}_{c} \tilde{U}_{c}(t,t_{1}) \tilde{Q}_{c} \tilde{V} \tilde{P}_{c}, \qquad (19)$$

where

$$\tilde{U}_{c}(t,t_{1}) = T \exp\left[-\frac{i}{\hbar} \int_{t_{1}}^{t} dt' \tilde{Q}_{c} [\tilde{L}_{s} + \tilde{L}_{s-L}(t') + \tilde{V} + \tilde{S}]\right],$$
(20)

where T is the time ordering operator (see Ref. [17]).

The term proportional to  $\tilde{Q}_c \hat{\sigma}(t_0)$  in Eq. (18) gives the contribution of initial correlations before the start of the collision at time  $t_0$  to the evolution of  $\hat{\sigma}_a(t)$ . These initial correlations evolve from some distant time in the past  $t = -\infty$  according to

$$\tilde{\mathcal{Q}}_{c}\hat{\sigma}(t_{0}) = \tilde{U}_{c}(t_{0}, t = -\infty)\tilde{\mathcal{Q}}_{c}\hat{\sigma}(t = -\infty)$$
$$-\frac{i}{\hbar} \int_{-\infty}^{t_{0}} dt_{1}\tilde{U}_{c}(t_{0}, t_{1})\tilde{\mathcal{Q}}_{c}\tilde{V}\tilde{P}_{c}\hat{\sigma}(t_{1}). \quad (21)$$

We can now set the initial correlations at  $t = -\infty$  equal to zero. This can be justified as follows. Whether correlations between the perturbers and the radiator in the distant past influence the evolution of the combined system of the atoms and the perturbers at time  $t_0$  depends on the memory time of the collision kernel  $\tilde{K}_c(t,t_1)$ . We shall show below that this time is of the order of the natural lifetime of the excited state of the atoms. This means that we can safely neglect initial correlations at  $t = -\infty$  in the distant past.

The term proportional to  $\tilde{P}_c \tilde{V} \tilde{P}_c$  in Eq. (18) gives the contribution of the mean potential between the perturbers and the radiator to the evolution of  $\tilde{P}_c \hat{\sigma}(t)$ . The discussion in the Appendix shows that this interaction has the nature of an electromagnetic field that is a superposition of all the fields generated by the individual perturbers coupled to the dipole moment of the radiator. We will now argue that this term can be neglected under certain circumstances. When the average over the perturbers is taken, all terms in  $\tilde{V}$  that refer to the interaction between two perturbers do not contribute to

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the average. The contribution of the mean potential to the evolution of the density matrix is given by

$$\tilde{P}_{c}\tilde{V}\tilde{P}_{c}\hat{\sigma}(t) = \hat{\sigma}_{p}(t) \otimes [\hat{\sigma}_{a}(t), \hat{\mathbf{d}}_{a}^{+} \cdot \hat{\mathbf{E}}_{p}^{+}(t, \hat{\mathbf{R}}_{a}) + \hat{\mathbf{d}}_{a}^{-} \cdot \hat{\mathbf{E}}_{p}^{-}(t, \hat{\mathbf{R}}_{a})].$$
(22)

 $\hat{\mathbf{E}}_{p}^{+}(t, \hat{\mathbf{R}}_{a})$  and  $\hat{E}_{p}^{-}(t, \hat{\mathbf{R}}_{a})$  are the positive and negative frequency parts of the *mean* electromagnetic field generated by the perturbers as a response to the driving field. Formally they are given by

$$\hat{\mathbf{E}}_{p}^{\pm}(t,\hat{\mathbf{R}}_{a}) = \operatorname{Tr}_{\text{pert}}\left[\sum_{j,j\neq a} \hat{\mathbf{E}}^{\pm}(\hat{\mathbf{R}}_{a},\hat{\mathbf{R}}_{j})\hat{\sigma}_{p}(t)\right], \quad (23)$$

where  $\hat{\mathbf{E}}^{\pm}(\hat{\mathbf{R}}_a, \hat{\mathbf{R}}_j)$  are the positive and negative frequency parts of the field generated by perturber *j*. The detailed expression is given in Eq. (A2) in the Appendix. It should be noted that  $\hat{\mathbf{E}}_p^{\pm}(t, \hat{\mathbf{R}}_a)$  is still an operator in the subspace of the radiator due to its dependence on the position operator  $\hat{\mathbf{R}}_a$ .

The BCA, which is assumed to be valid in almost all calculations involving collisions, has been discussed in detail, for example by Burnett *et al.* [17]. In the BCA the density matrix for the perturbers can be written in a factorized form

$$\hat{\sigma}_p(t) = \prod_{j=1}^{N} \hat{\sigma}_j(t)$$
(24)

and consequently the projection operator  $P_c$  can be written as a product of projection operators of the form

$$\tilde{P}_{c}^{j}\hat{O} = \hat{\sigma}_{j}(t) \otimes \operatorname{Tr}_{j \operatorname{th pert}}[\hat{O}], \qquad (25)$$

i.e.,  $\tilde{P}_c$  factorizes according to

$$\tilde{P}_c = \prod_{j=1}^N \tilde{P}_c^j.$$
(26)

The assumption of statistical independence of the perturbers means that we do neglect superradiance effects that arise because of transfer of coherence between the atoms of the gas. Physically speaking, omitting this term is usually justified by the fact that the phase factors that occur in the terms representing the coupling between dipoles of different atoms in the sample tend to lead to a cancellation of the term when averaging over a wide range of positions. This is sometimes referred to as the random-phase approximation (RPA). The only case for which the RPA is not fulfilled is the case of scattering into the forward direction. In this case the phase factors are all equal to unity and there is no cancellation of the amplitudes in the average over the perturbers. Physically these terms correspond to the refractive index and the absorption coefficient of the medium, i.e., their effect can be incorporated into the driving field as a position dependence of the phase and the Rabi frequency of the laser field. In the low absorption regime this effect would be very small and can be neglected. The effect of superradiance in the problem of N-atom spontaneous emission is, for example, discussed in Chap. 8 of Ref. [19]. In the following we will assume the RPA to be fulfilled for our model. The effect of the refraction

index on the propagation of light in a cloud of cold atoms has recently been discussed by Morice *et al.* [20].

Because we assume the semiclassical approximation to be valid we can also replace the position operators of the perturbers by their mean values  $\mathbf{r}_j$ . In this case the source field due to all the perturbers, Eq. (23), can be written as the sum of the source field contributions of individual perturbers, i.e.,

$$\hat{\mathbf{E}}_{p}^{\pm}(t,\hat{\mathbf{R}}_{a}) = \sum_{j,j\neq a} \left\langle \hat{\mathbf{E}}_{j}^{\pm}(t,\hat{\mathbf{R}}_{a},\mathbf{r}_{j}) \right\rangle_{j}, \qquad (27)$$

where  $\langle \rangle_j$  denotes the average over the *j*th perturber. In Ref. [10] we showed that the interaction of the radiator with the field generated by the second atom induced a momentum change of magnitude  $k\mathbf{e}_r$ , where *k* is the magnitude of the wave vector of the exchanged photon. As a result of the Markov assumption, which has been made in the calculation of interatomic potential [cf. Eq. (A7)], the magnitude of the momentum transfer from the perturber to the radiator is given by the wave vector of the atomic transition frequency, i.e.,  $k = k_0$ . This means that in this approximation even the frequency components centered around  $\omega_L$  and  $2\omega_L - \omega_0$  will give rise to a momentum kick of magnitude  $k_0$  rather than  $k_L$  and  $2k_L - k_0$ , as they should. However, this presents no real problem in our calculations as the difference between  $k_0$  and  $k_L$  is negligible.  $\hat{\mathbf{E}}_j^{\pm}(\hat{\mathbf{R}}_a, \mathbf{r}_j)$  can therefore be written as

$$\hat{\mathbf{E}}^{\pm}(t,\hat{\mathbf{R}}_{a},\mathbf{r}_{j}) = e^{\pm i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\hat{\mathbf{E}}_{j}^{\pm}(\mathbf{r}_{j}), \qquad (28)$$

where  $\mathbf{\hat{E}}_{i}^{\pm}(\mathbf{r}_{i})$  is the standard field amplitude for a radiating dipole given by Eq. (A5) and  $\mathbf{r}_i$  is the position vector of perturber *j* relative to the radiator. The mean dipole of each perturber is dependent on the position of the dipole in the laser field. This position dependence arises because the mean dipole moment is proportional to the optical coherences of the density matrix, which in turn are proportional to the laser polarization at the position of the atom. In laser configurations that lead to sub-Doppler cooling mechanisms the polarization of the laser field varies rapidly as a function of position in the laser beam on a scale of the laser wavelength  $\lambda_L$ . For example, for motion-induced orientation cooling the polarization rotates around the axis of the laser beams with a period of  $\lambda_L$  [15]. In the far-field limit, where we keep only terms to lowest order in  $1/k_0r_j$ , the field amplitude of the source field stays approximately constant over the range of a wavelength. However, the direction of the electric field vector is varying rapidly on this scale due to its proportionality to the orientation of the mean dipole. It is therefore reasonable to assume that for an ensemble of randomly distributed atoms the mean effect of the sum of this rapidly varying field vector is equal to zero, except for a small contribution in the forward direction, giving the refractive index of the cloud of atoms. The mean separation of the atoms can be estimated by  $\bar{r} = (n)^{-1/3}$ . For typical densities in Cs magneto-optical traps where extra heating due to photon exchange processes is observed, the densities are of the order of  $n = 10^{11}$  cm<sup>-3</sup>, giving a mean separation of  $\bar{r} = 15.9 \chi$ , where  $\chi$  has been taken as the corresponding value for Cs. The far-field limit that is necessary for the validity of the theory presented here therefore holds well. For atom numbers of the order of

 $N=10^8$ , this would correspond to trap radii of the order of  $10^{-2} - 10^{-1}$  cm being of the order of magnitude that has been observed in atom traps. We can estimate the contribution of the mean field by replacing the mean dipole of each perturber *i* by  $\chi E_L(\mathbf{r}_i)$ , where  $E_L(\mathbf{r}_i)$  is the laser field at the position of the perturber and  $\chi$  is the polarizability tensor. To lowest order in the atom-atom interactions the polarizability is independent of the atom number and density. The orientation of the mean dipole only depends on the local polarization of the laser field. If we now take the average over a large number of randomly distributed perturbers we find that the mean value of the field produced by all perturbers is proportional to  $n\sigma_L L$ , where  $\sigma_L$  is the scattering cross section for the laser light and L is the trap radius. The mean field is therefore given by the forward-scattering amplitude for the laser light and as such gives rise to the refractive index of the medium. In the low absorption regime this modification of the laser field is small and can be neglected, or as mentioned above, be incorporated into the driving field. In general, for typical trap parameters the low absorption condition is well fulfilled. Hence, in the following we take  $\tilde{P}_c \tilde{V} \tilde{P}_c = 0$ . It should be noted that the above argument depends on the density being sufficiently high that we can take the continuum limit in the average over the perturbers. However, this does not present a problem as in this case the effect of atom-atom interactions is negligible. This picture breaks down if the densities are sufficiently high for binary collisions between individual pairs of atoms to dominate the collision dynamics. These strong binary collisions have been considered by Smith and Burnett [8], but the density dependence of the extra heating in this regime does not agree with experimental observation, thus clearly indicating that strong binary collisions are not important. Hence we do conclude that we can safely proceed in the way we propose. Clearly this picture breaks down in light lattices, where position and polarization are linked. In principle, these effects could be included in an extension of this method.

Substitution of Eq. (21) into Eq. (18) and taking  $\tilde{P}_c \tilde{V} \tilde{P}_c = 0$  yields

$$\tilde{P}_{c}\hat{\sigma}(t) = -\frac{i}{\hbar}\tilde{P}_{c}[\tilde{L}_{a} + \tilde{L}_{a-L}(t) + \tilde{S}]\tilde{P}_{c}\hat{\sigma}(t) + \left(-\frac{i}{\hbar}\right)^{2}\int_{-\infty}^{t} dt_{1}\tilde{K}_{c}(t,t_{1})\tilde{P}_{c}\hat{\sigma}(t_{1}).$$
(29)

As a final step we now have to rewrite the last term in Eq. (29) in terms of a collision operator  $\tilde{\mathscr{C}}(t)$  and the density matrix at time t,

$$-\left(\frac{i}{\hbar}\right)^{2} \int_{-\infty}^{t} dt_{1} \tilde{K}_{c}(t,t_{1}) \tilde{P}_{c} \hat{\sigma}(t_{1}) = -\frac{i}{\hbar} \tilde{\mathscr{E}}(t) \tilde{P}_{c} \hat{\sigma}(t).$$
(30)

It is sufficient to take only terms up to second order in the interaction  $\tilde{V}$  because we restrict our calculation to the farfield limit. This means we can neglect the contribution of  $\tilde{V}$  in the exponent of  $\tilde{U}_c(t,t_1)$ . The collision kernel to second order in  $\tilde{V}$  is then given by

$$\tilde{K}_c^0(t,t_1) = \tilde{P}_c \tilde{V} \tilde{Q}_c \tilde{U}_c^0(t,t_1) \tilde{Q}_c \tilde{V} \tilde{P}_c, \qquad (31)$$

where

$$\tilde{U}_{c}^{0}(t,t_{1}) = T \exp \left[-\frac{i}{\hbar}\tilde{Q}_{c}\int_{t_{1}}^{t}dt'[\tilde{L}_{s}+\tilde{L}_{s-L}(t')+\tilde{S}]\right].$$
(32)

The evolution of the density-matrix  $\hat{\sigma}(t)$  between  $t_1$  and t is given by

$$\hat{\sigma}(t_1) = T \exp\left[-\frac{i}{\hbar} \int_t^{t_1} dt' [\tilde{L}_s + \tilde{L}_{s-L}(t') + \tilde{S} + \tilde{V}]\right] \hat{\sigma}(t).$$
(33)

Again we can neglect the contribution of  $\tilde{V}$  in the exponential because this would lead to contributions that are higher order in  $\tilde{V}$ . Using the relations

$$\tilde{P}_c \tilde{L}_p = \tilde{P}_c \tilde{L}_{p-L}(t) = \tilde{P}_c \tilde{S}_p = 0$$
(34)

and

$$[\tilde{P}_c, \tilde{L}_a] = [\tilde{P}_c, \tilde{L}_{a-L}(t)] = [\tilde{P}_c, \tilde{S}_a] = 0, \qquad (35)$$

it is easy to verify that

$$\tilde{P}_{c} \exp\left[-\frac{i}{\hbar} \int_{t}^{t_{1}} dt' [\tilde{L}_{s} + \tilde{L}_{s-L}(t') + \tilde{S}]\right] \hat{\sigma}(t)$$
$$= \tilde{U}_{c}^{a}(t_{1}, t) \tilde{P}_{c} \hat{\sigma}(t), \qquad (36)$$

where

$$\tilde{U}_{c}^{a}(t_{1},t) = T \exp\left[-\frac{i}{\hbar} \int_{t}^{t_{1}} dt' [\tilde{L}_{a} + \tilde{L}_{a-L}(t') + \tilde{S}_{a}]\right]$$
(37)

is the Green's operator acting only on the radiator. We now define the collision operator  $\tilde{\mathscr{E}}(t)$  by

$$\tilde{\mathscr{C}}(t) \equiv -\frac{i}{\hbar} \int_{-\infty}^{t} dt_1 \tilde{K}_c^0(t,t_1) \tilde{U}_c^a(t_1,t).$$
(38)

The equation of motion for the reduced density operator of the radiator  $\hat{\sigma}_a(t)$  can be obtained by tracing Eq. (29) over all perturber states. Using Eq. (38), we obtain

$$\frac{d}{dt}\hat{\sigma}_{a}(t) = -\frac{i}{\hbar} [\tilde{L}_{a} + \tilde{L}_{a-L}(t) + \tilde{S}_{a} + \tilde{\mathscr{C}}(t)]\hat{\sigma}_{a}(t).$$
(39)

Equation (39) gives the evolution of the reduced density matrix of the radiator. The effect of the perturbers is now included in the operator  $\tilde{\mathscr{E}}(t)$ . It should be noted that  $\tilde{\mathscr{E}}(t)$  includes the evolution of the density matrix due to the driving field to all orders. This means that we do not look in detail at the evolution of the atom due to the driving field during the collision, but we assume that the coupling of the

driving field to the atoms is not modified due to the presence of other atoms. We therefore neglect correlations between the radiator and the perturber, which arise because the shift of the resonance frequency due to the presence of the perturber is large enough that it does alter the interaction of an individual atom with the laser field (see, e.g., [17] and [18] for a discussion on this matter). It was pointed out in the Introduction that these shifts only become important for mean interatomic separations  $r \leq \chi_0$  and that for separations  $r \geq \chi_0$  we can safely neglect these shifts. This also means that we can treat collisions between the radiator and the perturbers as binary events because the only way a third particle could influence the exchange of scattered photons between a pair of atoms is by significantly shifting the emission lines of the colliding atoms.

In the BCA the collision operator can be written as the sum of individual two-body collisions, i.e.,

$$\tilde{\mathscr{C}}(t) = \sum_{j=1}^{N} \tilde{\mathscr{C}}_{j}(t), \qquad (40)$$

where the operator for the collision with the jth particle is equal to

$$\begin{split} \tilde{\mathscr{C}}_{j}(t) &= -\frac{i}{\hbar} \int_{-\infty}^{t} dt_{1} \tilde{P}_{c} \tilde{V}_{aj} \tilde{U}_{c}^{a}(t,t_{1}) \tilde{U}_{c}^{j}(t,t_{1}) \\ & \times \tilde{Q}_{c} \tilde{V}_{aj} \tilde{P}_{c} \tilde{U}_{c}^{a}(t_{1},t). \end{split}$$
(41)

One approximation that is frequently made at this point is to neglect the projection operator  $\tilde{Q}_c$  between the two interactions  $\tilde{V}_{aj}$  [21,17]. We will not make this approximation and show below that this term gives a physically important contribution to the collision operator.

The integrand of the time integral in Eq. (41) can be evaluated using standard Liouville space techniques. We make the *a priori* assumption that the effect of frequency shifts induced by the interaction of the radiator with the medium is small and it is therefore consistent to neglect those shifts in the evaluation of the collision operator. In the RWA we also neglect terms that contain pairs of operators  $\hat{\mathbf{d}}_j^-$  and  $\hat{\mathbf{d}}_j^+$ . In the case of a two-level atom this yields

$$\tilde{\mathscr{C}}_{j}^{2-\text{level}}(t)\hat{\sigma}_{a}(t) = -\frac{i}{\hbar} \int_{-\infty}^{t} dt_{1} [\tilde{R}^{e} \tilde{P}_{c} \hat{\sigma}(t_{1}) \\ + \tilde{R}^{\text{abs}} \tilde{P}_{c} \hat{\sigma}(t_{1}) + \tilde{R}^{\text{ex}} \tilde{P}_{c} \hat{\sigma}(t_{1})], \quad (42)$$

where

$$\tilde{R}^{e}\tilde{P}_{c}\hat{\sigma}(t) = \{ \left[ \left| \tilde{U}_{c}^{a}(t,t_{1})\hat{\sigma}_{a}(t_{1})\hat{d}_{a}^{+}e^{i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}} \right\rangle \right] \hat{d}_{a}^{-}e^{-i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}} \rangle H^{*}(t,t_{1},\mathbf{r}_{j}) + \left| \hat{d}_{a}^{+}e^{i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}} \right\rangle \right] \tilde{U}_{c}^{a}(t,t_{1})\hat{d}_{a}^{-}e^{-i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}} \hat{\sigma}_{a}(t_{1}) \rangle H(t,t_{1},\mathbf{r}_{j}) = \\ - \left[ \left| \hat{d}_{a}^{-}e^{-i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}} \right\rangle \right] \tilde{U}_{c}^{a}(t,t_{1})\hat{\sigma}_{a}(t_{1})\hat{d}_{a}^{+}e^{i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}} \rangle H^{*}(t,t_{1},\mathbf{r}_{j}) + \left| \tilde{U}_{c}^{a}(t,t_{1})\hat{d}_{a}^{-}e^{-i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}} \hat{\sigma}_{a}(t_{1}) \rangle \right] \hat{d}_{a}^{+}e^{i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}} \rangle H(t,t_{1},\mathbf{r}_{j}) ] \},$$

$$(43)$$

$$\tilde{R}^{abs}\tilde{P}_{c}\hat{\sigma}(t_{1}) \equiv \{ [|\tilde{U}_{c}^{a}(t,t_{1})\hat{\sigma}_{a}(t_{1})\hat{d}_{a}^{-}e^{-i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\rangle\rangle|\hat{d}_{a}^{+}e^{i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\rangle\rangle H(t,t_{1},\mathbf{r}_{j}) + |\hat{d}_{a}^{-}e^{-i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\rangle\rangle|\tilde{U}_{c}^{a}(t,t_{1})\hat{d}_{a}^{+} \\
\times e^{i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\hat{\sigma}_{a}(t_{1})\rangle\rangle H^{*}(t,t_{1},\mathbf{r}_{j})] - [|\hat{d}_{a}^{+}e^{i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\rangle\rangle|\tilde{U}_{c}^{a}(t,t_{1})\hat{\sigma}_{a}(t_{1})\hat{d}_{a}^{-}e^{-i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\rangle\rangle H(t,t_{1},\mathbf{r}_{j}) \\
+ |\tilde{U}_{c}^{a}(t,t_{1})\hat{d}_{a}^{+}e^{i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\hat{\sigma}_{a}(t_{1})\rangle\rangle|\hat{d}_{a}^{-}e^{-i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\rangle\rangle H^{*}(t,t_{1},\mathbf{r}_{j})]\}.$$
(44)

and

$$\tilde{\mathsf{R}}^{ex} \tilde{P}_{c} \hat{\sigma}(t_{1}) \equiv (E^{+}(\mathbf{r}_{j}))^{2} [|\tilde{U}_{c}^{a}(t,t_{1})\hat{d}_{a}^{-}e^{-i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}} \hat{\sigma}_{a}(t_{1})\rangle\rangle, |\hat{d}_{a}^{+}e^{i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\rangle\rangle] \frac{H(t,t_{1},\mathbf{r}_{j}) - G^{*}(t,t_{1},\mathbf{r}_{j})}{|E(\mathbf{r}_{j})|^{2}} + (E^{-}(\mathbf{r}_{j}))^{2} [|\tilde{U}_{c}^{a}(t,t_{1})\hat{\sigma}_{a}(t_{1})\hat{d}_{a}^{+}e^{-i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\rangle\rangle, |\hat{d}_{a}^{+}e^{i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\rangle\rangle] \frac{G(t,t_{1},\mathbf{r}_{j}) - H^{*}(t,t_{1},\mathbf{r}_{j})}{|E(\mathbf{r}_{j})|^{2}}.$$
(45)

Here we have adopted Liouville space notation for Hilbert space operators, i.e., each Hilbert-Schmidt operator is equivalent to a Liouville space vector  $\hat{A} \equiv |A\rangle\rangle$ . The product of two kets in Liouville space is interpreted in terms of a product of two dyadics. For example, for two Liouville space vectors  $|ab\rangle\rangle = |a\rangle\langle b|$  and  $|cd\rangle\rangle = |c\rangle\langle d|$  their product is given by  $|ab\rangle\rangle |cd\rangle\rangle = |a\rangle\langle b|c\rangle\langle d|$ . The functions  $H(t,t_1,\mathbf{r}_j)$  and  $G(t,t_1,\mathbf{r}_j)$  are defined by

$$G(t,t_1,\mathbf{r}_j) \equiv |E(\mathbf{r}_j)|^2 [\langle \hat{d}_j^-(t_1)\hat{d}_j^+(t)\rangle - \langle \hat{d}_j^-(t_1)\rangle \langle \hat{d}_j^+(t)\rangle],$$
(46)

$$H(t,t_{1},\mathbf{r}_{j}) \equiv |E(\mathbf{r}_{j})|^{2} [\langle \hat{d}_{j}^{+}(t_{1}) \hat{d}_{j}^{-}(t) \rangle - \langle \hat{d}_{j}^{+}(t_{1}) \rangle \langle \hat{d}_{j}^{-}(t) \rangle].$$
(47)

 $G(t,t_1,\mathbf{r})$  and  $H(t,t_1,\mathbf{r})$  are the incoherent parts of two-time correlation functions of the dipoles  $\hat{d}_i^-(t)$  and  $\hat{d}_i^+(t)$  [22].

The subtraction of the coherent part of the correlation function is entirely due to maintaining the projection operator  $\tilde{Q}_c$  in the collision operator Eq. (41). The Fourier transform of  $G(t,t_1,\mathbf{r}_j)$  gives the absorption spectrum of a driven atom, the Fourier transform of  $H(t,t_1,\mathbf{r}_j)$  corresponding emission, i.e., the Mollow triplet. The term  $\tilde{R}^{ex}\tilde{P}_c\hat{\sigma}(t_1)$  is proportional to the difference between the absorption and emission spectrum of the driven perturber. This rate is proportional to the amplitude squared rather than the intensity of the scattered field and therefore proportional to a phase factor. It follows that this term is small compared to the first two terms in the collision operator when the average over all perturbers in the trap is taken. We will therefore neglect it in the collision operator in the remainder of this paper.

The equivalent of Eq. (42) for a multilevel atom is obtained by accounting for the vector operator nature of both the dipole operator and the electric field generated by the perturbers. The functions  $G(t,t_1,\mathbf{r}_j)$  and  $H(t,t_1,\mathbf{r}_j)$  have to be replaced by matrices. In a spherical basis with eigenvectors  $\mathbf{u}_q$ , q = +1, -1, 0, their entries are

$$G_{q,q'}(t,t_1,\mathbf{r}_j) \equiv \langle \hat{E}_q^+(\mathbf{r}_j,t_1) \hat{E}_{q'}^-(\mathbf{r}_j,t) \rangle - \langle \hat{E}_q^+(\mathbf{r}_j,t_1) \rangle$$
$$\times \langle \hat{E}_{q'}^-(\mathbf{r}_j,t) \rangle, \qquad (48)$$

$$H_{q,q'}(t,t_1,\mathbf{r}_j) \equiv \langle \hat{E}_q^-(\mathbf{r}_j,t_1) \hat{E}_{q'}^+(\mathbf{r}_j,t) \rangle - \langle \hat{E}_q^-(\mathbf{r}_j,t_1) \rangle \\ \times \langle \hat{E}_{q'}^+(\mathbf{r}_j,t) \rangle.$$
(49)

The indices q and q' denote the possible angular momenta of the photons absorbed and emitted. Matrix elements that are off diagonal in q,q' correspond to processes where the radiator exchanges angular momentum  $\Delta m = q' - q$  with the perturbing atom. The off-diagonal elements therefore stand for Raman processes that change the internal state of the radiator. The diagonal elements, on the other hand, leave the total angular momentum of the radiator conserved.  $\tilde{R}^{e}\tilde{P}_{c}\hat{\sigma}(t_{1})$  and  $\tilde{R}^{abs}\tilde{P}_{c}\hat{\sigma}(t_{1})$  are replaced by

$$\tilde{R}_{q,q'}^{e} \tilde{P}_{c} \hat{\sigma}(t_{1}) \equiv \{ [|\tilde{U}_{c}^{a}(t,t_{1})\hat{\sigma}_{a}(t_{1})(\hat{\mathbf{d}}_{a}^{+} \cdot \mathbf{u}_{q})e^{i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\rangle\rangle |(\hat{\mathbf{d}}_{a}^{-} \cdot \mathbf{u}_{q'}^{*})e^{-i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\rangle\rangle H_{q,q'}^{*}(t,t_{1},\mathbf{r}_{j}) + |(\hat{\mathbf{d}}_{a}^{+} \cdot \mathbf{u}_{q})e^{i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\rangle\rangle |\tilde{U}_{c}^{a}(t,t_{1})(\hat{\mathbf{d}}_{a}^{-} \cdot \mathbf{u}_{q'}^{*})e^{-i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\hat{\sigma}_{a}(t_{1})\rangle\rangle H_{q,q'}(t,t_{1},\mathbf{r}_{j}) - [|(\hat{\mathbf{d}}_{a}^{-} \cdot \mathbf{u}_{q'}^{*})e^{-i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\rangle\rangle |\tilde{U}_{c}^{a}(t,t_{1})\hat{\sigma}_{a}(t_{1})(\hat{\mathbf{d}}_{a}^{+} \cdot \mathbf{u}_{q})e^{i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\rangle\rangle H_{q,q'}^{*}(t,t_{1},\mathbf{r}_{j}) + |\tilde{U}_{c}^{a}(t,t_{1})(\hat{\mathbf{d}}_{a}^{-} \cdot \mathbf{u}_{q'}^{*})e^{-i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\hat{\sigma}_{a}(t_{1})\rangle\rangle |(\hat{\mathbf{d}}_{a}^{+} \cdot \mathbf{u}_{q})e^{i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\rangle\rangle H_{q,q'}(t,t_{1},\mathbf{r}_{j})]\}$$
(50)

and

$$\begin{split} \tilde{R}_{q,q'}^{abs} \tilde{P}_{c} \hat{\sigma}(t_{1}) &\equiv \{ \left[ \left| \tilde{U}_{c}^{a}(t,t_{1}) \hat{\sigma}_{a}(t_{1}) (\hat{\mathbf{d}}_{a}^{-} \cdot \mathbf{u}_{q}) e^{-i\mathbf{k}_{0} \cdot \hat{\mathbf{R}}_{d}} \right\rangle \rangle | (\hat{\mathbf{d}}_{a}^{+} \cdot \mathbf{u}_{q'}^{*}) e^{i\mathbf{k}_{0} \cdot \hat{\mathbf{R}}_{d}} \rangle \rangle H_{q,q'}(t,t_{1},\mathbf{r}_{j}) \\ &+ \left| (\hat{\mathbf{d}}_{a}^{-} \cdot \mathbf{u}_{q}) e^{-i\mathbf{k}_{0} \cdot \hat{\mathbf{R}}_{d}} \right\rangle \rangle | \tilde{U}_{c}^{a}(t,t_{1}) (\hat{\mathbf{d}}_{a}^{+} \cdot \mathbf{u}_{q'}^{*}) e^{i\mathbf{k}_{0} \cdot \hat{\mathbf{R}}_{d}} \hat{\sigma}_{a}(t_{1}) \rangle \rangle H_{q,q'}^{*}(t,t_{1},\mathbf{r}_{j}) \right] \\ &- \left[ \left| (\hat{\mathbf{d}}_{a}^{+} \cdot \mathbf{u}_{q'}^{*}) e^{i\mathbf{k}_{0} \cdot \hat{\mathbf{R}}_{d}} \right\rangle \rangle | \tilde{U}_{c}^{a}(t,t_{1}) \hat{\sigma}(t_{1}) (\hat{\mathbf{d}}_{a}^{-} \cdot \mathbf{u}_{q}) e^{-i\mathbf{k}_{0} \cdot \hat{\mathbf{R}}_{d}} \rangle \rangle H_{q,q'}(t,t_{1},\mathbf{r}_{j}) \right. \\ &+ \left| \tilde{U}_{c}^{a}(t,t_{1}) (\hat{\mathbf{d}}_{a}^{+} \cdot \mathbf{u}_{q'}^{*}) e^{i\mathbf{k}_{0} \cdot \hat{\mathbf{R}}_{a}} \hat{\sigma}_{a}(t_{1}) \rangle \rangle | (\hat{\mathbf{d}}_{a}^{-} \cdot \mathbf{u}_{q}) e^{-i\mathbf{k}_{0} \cdot \hat{\mathbf{R}}_{d}} \rangle \rangle H_{q,q'}^{*}(t,t_{1},\mathbf{r}_{j}) \right] \}. \end{split}$$

Omitting  $\tilde{R}_{q,q'}^{\text{ex}} \tilde{P}_c \hat{\sigma}(t_1)$  the binary-collision operator is equal to

$$\tilde{\mathscr{C}}_{j}(t)\tilde{P}_{c}\hat{\sigma}(t) = -\frac{i}{\hbar}\sum_{q,q'}\int_{-\infty}^{t} dt_{1}[\tilde{R}_{q,q'}^{e},\tilde{P}_{c}\hat{\sigma}(t_{1}) \\ +\tilde{R}_{q,q'}^{abs}\tilde{P}_{c}\hat{\sigma}(t_{1})].$$
(52)

The time integral of the collision operator  $\tilde{\mathscr{C}}_{j}(t)$  is taken over products of two-time correlation functions of the dipole of the radiator and the dipole of the perturber. By virtue of the convolution theorem this product is equal to the Fourier transform of the convolution of the two functions in frequency space. We interpret  $\tilde{R}_{q,q'}^{e}$  as the part of the collision where the perturber stimulates the radiator to emit a photon. In the second term the order of  $\hat{\mathbf{d}}_a^+$  and  $\hat{\mathbf{d}}_a^-$  is reversed.  $\tilde{R}_{q,q'}^{abs}$  therefore can be interpreted in terms of a stimulated absorption process. The interpretation of the two parts of the collision operator in these terms will become more apparent in Sec. IV.

The total effect of the fluctuations generated by the interaction with the perturbers on the evolution of the radiator is obtained by adding the contributions from all perturbers in the cloud. We introduce the particle density of the cloud  $n(\mathbf{r})$ . The sum over all perturbers can then be converted into an integral over the particle density. We obtain

$$\tilde{\mathscr{E}}(t)\tilde{P}_{c}\hat{\sigma}(t) = -\frac{i}{\hbar}\sum_{q,q'}\int_{-\infty}^{t} dt_{1}\int d^{3}rn(\mathbf{r})[\tilde{R}_{q,q'}^{e}(\mathbf{r})\tilde{P}_{c}\hat{\sigma}(t_{1}) + \tilde{R}_{q,q'}^{abs}(\mathbf{r})\tilde{P}_{c}\hat{\sigma}(t_{1})], \qquad (53)$$

where the position dependence of  $\tilde{R}_{q,q'}^{e}(\mathbf{r})$  and  $\tilde{R}_{q,q'}^{abs}(\mathbf{r})$  replaces the particle label *j* in Eq. (52).

Rather than continuing with the evaluation from a formal point of view, we will now use some further approximations, which enable us to obtain a qualitative picture of the effect of radiation trapping on the two sub-Doppler cooling mechanisms: Sisyphus and motion-induced orientation cooling.

# IV. RATE-EQUATION LIMIT OF THE COLLISION OPERATOR

The modification of the equation of motion for the reduced density matrix of a single atom in a laser field has been derived in the preceding section and is given by Eq. (53). The general structure of this equation is very complicated, mainly due to the fact that the memory time of the collisions is of the order of the natural decay time of the colliding atoms. The memory time of the background field generated by the perturbers is in general of the same order as or greater than the time scale for the evolution of the reduced density matrix of the radiator. Therefore the reduced density matrix of the radiator evolves not only due to the free Hamiltonian of the atom but the coupling to the laser field and the vacuum field modes will also contribute appreciably during the memory time of the collision operator. The second-order interaction of a pair will also have an effect on the distribution of the atomic populations in the different Zeeman sublevels. This occurs via second-order exchanges between a pair of atoms where the perturbing atom emits a photon of angular momentum q, which gets absorbed by the radiator. The radiator then emits a photon of different angular momentum q', which is reabsorbed by the perturber. The precise nature of these terms in the binary-collision operator will depend on the relative orientation of the dipoles of the pair of interacting atoms. The total effect of transitions is obtained by averaging over all possible pairs in the cloud. Another complication arises because the source field emitted by each perturber is not spherically symmetric but has the angular dependence of a dipole radiation pattern.

For the purpose of the qualitative calculations in this paper we want to take the rate-equation limit of Eq. (53). This limit is obtained by assuming that the memory time of the field generated by the perturbers is short compared with the evolution time of the reduced density matrix of the radiator in the interaction picture. In the Schrödinger picture this means that the density matrix only evolves due to the free Hamiltonian of the radiator. We also neglect all terms in Eq. (53) that are off diagonal in the angular momentum indices qand q' of the exchanged photons. Physically this is equivalent to assuming that there is no net exchange of angular momentum of the radiator with all the perturbers in the mean. We emphasize that this is not the same as assuming that there is no exchange of angular momentum between pairs of atoms in individual exchanges. It just means that on average there is no exchange. In the limit where steady state is reached in the presence of the driving field, the spectral distribution functions for the perturbers  $H_{q,q'}^*(t,t_1,\mathbf{r})$  and  $H_{q,q'}(t,t_1,\mathbf{r})$  depend only on the time difference  $\tau=t-t_1$ [23,22]. We define the Fourier transform thus:

$$H_{q,q'}(t,t_1,\mathbf{r}) = \int d\omega h_{q,q'}(\omega,\mathbf{r})e^{-i\omega\tau}.$$
 (54)

The spectral distribution function  $h_{q,q'}(\omega,\mathbf{r})$  is real. We now make the substitution  $\tau = t - t_1$  in the time integral of the collision operator  $\tilde{\mathscr{E}}(t)$ . In the short memory approximation we can now rewrite the first part of Eq. (53) as

$$\int_{-\infty}^{t} dt_{1} \int d^{3}rn(\mathbf{r}) \tilde{R}_{q,q}^{e}(\mathbf{r}) \tilde{P}_{c} \hat{\sigma}(t_{1}) = \hat{\sigma}_{a}(t) (\hat{\mathbf{d}}_{a}^{+} \cdot \mathbf{u}_{q}) (\hat{\mathbf{d}}_{a}^{-} \cdot \mathbf{u}_{q}^{*}) \int d\omega \int d^{3}rn(\mathbf{r}) \int_{0}^{\infty} d\tau h_{q,q}(\omega, \mathbf{r}) e^{-i(\omega-\omega_{0})\tau} \\ + (\hat{\mathbf{d}}_{a}^{+} \cdot \mathbf{u}_{q}) (\hat{\mathbf{d}}_{a}^{-} \cdot \mathbf{u}_{q}^{*}) \hat{\sigma}_{a}(t) \int d\omega \int d^{3}rn(\mathbf{r}) \int_{0}^{\infty} d\tau h_{q,q}(\omega, \mathbf{r}) e^{i(\omega-\omega_{0})\tau} \\ - (\hat{\mathbf{d}}_{a}^{-} \cdot \mathbf{u}_{q}^{*}) \left( \int d\omega \int d^{3}rn(\mathbf{r}) \int_{0}^{\infty} d\tau h_{q,q}(\omega, \mathbf{r}) e^{-i(\omega-\omega_{0})\tau} e^{-i\mathbf{k}_{0} \cdot \hat{\mathbf{R}}_{a}} \hat{\sigma}_{a}(t) e^{i\mathbf{k}_{0} \cdot \hat{\mathbf{R}}_{a}} \\ + \int d\omega \int d^{3}rn(\mathbf{r}) \int_{0}^{\infty} d\tau h_{q,q}(\omega, \mathbf{r}) e^{i(\omega-\omega_{0})\tau} e^{-i\mathbf{k}_{0} \cdot \hat{\mathbf{R}}_{a}} \hat{\sigma}_{a}(t) e^{i\mathbf{k}_{0} \cdot \hat{\mathbf{R}}_{a}} \right) (\hat{\mathbf{d}}_{a}^{+} \cdot \mathbf{u}_{q}).$$
(55)

If we now use

$$\lim_{\epsilon \searrow 0} \frac{1}{\omega_0 - \omega + i\epsilon} = -i\pi\delta(\omega_0 - \omega) + P \frac{1}{\omega_0 - \omega}$$
(56)

and neglect the terms that arise from its principal parts, the trifferst term of the right-hand side of Eq. (55) may be written as

$$\hat{\tau}_{a}(t)(\hat{\mathbf{d}}_{a}^{+}\cdot\mathbf{u}_{q})(\hat{\mathbf{d}}_{a}^{-}\cdot\mathbf{u}_{q}^{*})\int d\omega\int d^{3}rn(\mathbf{r})\int_{0}^{\infty}d\tau h_{q,q}(\omega,\mathbf{r})$$

$$\times e^{-i(\omega-\omega_{0})\tau} = \hbar^{2}\frac{\Gamma}{2}\hat{\sigma}_{a}(t)(\hat{\mathbf{d}}_{a}^{+}\cdot\mathbf{u}_{q})(\hat{\mathbf{d}}_{a}^{-}\cdot\mathbf{u}_{q}^{*})\xi_{q}.$$
(57)

Here  $\xi_q$  takes the role of a dimensionless parameter giving the coupling strength of the part of the background field having angular momentum q to the radiator.  $\xi_q$  is given by L

$$\xi_q = \left(\frac{2\pi}{\hbar^2 \Gamma}\right) \int d^3 r n(\mathbf{r}) h_{q,q}(\omega_0, \mathbf{r}).$$
(58)

For an atom near the center of a magneto-optical trap it is reasonable to assume that the density distribution of perturbers is isotropic  $n(\mathbf{r}) = n(r)$ . We expect that an isotropic dipole distribution produces a field that also is isotropic. If we average over the radial coordinate r along a fixed distance we therefore expect that the integral

$$\int r^2 dr n(r) h_{q,q}(\omega_0, \mathbf{r})$$
(59)

can be written in the form

$$\int r^2 dr n(r) h_{q,q}(\omega_0, \mathbf{r}) = \sum_{\boldsymbol{\epsilon} \perp \mathbf{e}_r} \epsilon_q \boldsymbol{\epsilon}_q^* \times \text{const.}$$
(60)

The angular integral of the sum over the polarization vectors is of course independent of q,

$$\int d\Omega \sum_{\boldsymbol{\epsilon} \perp \boldsymbol{e}_r} \boldsymbol{\epsilon}_q \boldsymbol{\epsilon}_q^* = \frac{8\pi}{3}.$$
 (61)

Under the assumption of an isotropic distribution of perturbers the coupling strength of the background field to the radiator is independent of q, a result that can be expected for a background field that is unpolarized. Setting  $\xi_q = \xi$  for all q, we find

$$\hat{\sigma}_{a}(t)\sum_{q} (\hat{\mathbf{d}}_{a}^{+} \cdot \mathbf{u}_{q})(\hat{\mathbf{d}}_{a}^{-} \cdot \mathbf{u}_{q}^{*}) \int d\omega \int d^{3}r n(\mathbf{r}) \int_{0}^{\infty} d\tau h_{q,q}(\omega,\mathbf{r}) e^{-i(\omega-\omega_{0})\tau} = \hbar^{2}\Gamma \xi \hat{\sigma}_{a}(t) \hat{\mathbf{d}}_{a}^{+} \cdot \hat{\mathbf{d}}_{a}^{-}.$$
(62)

A similar calculation for the other terms in Eq. (55) and summation over q yields

$$\sum_{q} \int_{-\infty}^{t} dt_{1} \int d^{3}r n(\mathbf{r}) \tilde{R}_{q,q}^{e}(\mathbf{r}) \tilde{P}_{c} \hat{\sigma}(t_{1}) = \hbar^{2} \frac{\Gamma}{2} \xi [(\hat{\mathbf{d}}_{a}^{+} \cdot \hat{\mathbf{d}}_{a}^{-}) \hat{\sigma}_{a}(t) + \hat{\sigma}_{a}(t) (\hat{\mathbf{d}}_{a}^{+} \cdot \hat{\mathbf{d}}_{a}^{-})] - \hbar^{2} \Gamma \xi$$

$$\times \sum_{q} (\hat{\mathbf{d}}_{a}^{-} \cdot \mathbf{u}_{q}^{*}) \bigg( \sum_{\boldsymbol{\epsilon} \perp \mathbf{e}_{r}} \int \frac{d\Omega}{8\pi/3} \epsilon_{q} \epsilon_{q}^{*} e^{-i\mathbf{k}_{0} \cdot \hat{\mathbf{R}}_{a}} \hat{\sigma}_{a}(t) e^{i\mathbf{k}_{0} \cdot \hat{\mathbf{R}}_{a}} \bigg) (\hat{\mathbf{d}}_{a}^{+} \cdot \mathbf{u}_{q}). \quad (63)$$

If we compare Eq. (63) with the term describing the coupling of the atom to the vacuum field (12), we see that both of the terms have the same structure. Equation (12) follows from Eq. (63) in the limit  $\xi = 1$ . We interpret this part of the collision operator as a stimulated emission rate that is induced by the background field. The parameter  $\xi$  gives the mean number of photons in the resonant component of the background field. Only the part of the background field around the atomic resonance frequency contributes because we have made a short memory approximation in our calculation. We note that this result differs from that obtained in Ref. [10], where the diffusion coefficient comprises both the coherent and incoherent components of the scattered spectrum, whereas in the collision operator Eq. (53) only the incoherent part of the Mollow triplet contributes. In the limit  $\Omega \ll \Gamma$  the elastic scatter is entirely due to the coherent part of the scattered spectrum [22] and the second sideband, which is far off resonance, can be safely neglected. The coherent part of the scattered radiation gives the mean interaction between the dipoles of the radiator and the perturbers, which gives a zero average contribution in the RPA.

The second part of the collision operator (53) is evaluated in exactly the same way and yields the result

$$\sum_{q} \int_{-\infty}^{t} dt_{1} \int d^{3}r n(\mathbf{r}) \tilde{R}_{q,q}^{e}(\mathbf{r}) \tilde{P}_{c} \hat{\sigma}(t_{1}) = \hbar^{2} \frac{\Gamma}{2} \xi' [(\hat{\mathbf{d}}_{a}^{-} \cdot \hat{\mathbf{d}}_{a}^{+}) \hat{\sigma}_{a}(t) + \hat{\sigma}_{a}(t) (\hat{\mathbf{d}}_{a}^{-} \cdot \hat{\mathbf{d}}_{a}^{+})] + \hbar^{2} \Gamma \xi' \\ \times \sum_{q} (\hat{\mathbf{d}}_{a}^{+} \cdot \mathbf{u}_{q}^{*}) \bigg( \sum_{\boldsymbol{\epsilon} \perp \mathbf{e}_{r}} \int \frac{d\Omega}{8\pi/3} \boldsymbol{\epsilon}_{q} \boldsymbol{\epsilon}_{q}^{*} \boldsymbol{e}^{-i\mathbf{k}_{0} \cdot \hat{\mathbf{R}}_{a}} \hat{\sigma}_{a}(t) \boldsymbol{e}^{i\mathbf{k}_{0} \cdot \hat{\mathbf{R}}_{a}} \bigg) (\hat{\mathbf{d}}_{a}^{-} \cdot \mathbf{u}_{q}).$$
(64)

The interchanged order of the dipole ladder operators  $\hat{\mathbf{d}}_a^+$  and  $\hat{\mathbf{d}}_a^-$  indicates that Eq. (64) gives the rate of stimulated absorption due to the background field. The normalization convention of the Clebsch-Gordan coefficients implies

$$\langle \boldsymbol{e}_m | \hat{\mathbf{d}}^+ \cdot \hat{\mathbf{d}}^- | \boldsymbol{e}_m \rangle = 1, \tag{65}$$

$$\langle g_m | \hat{\mathbf{d}}^- \cdot \hat{\mathbf{d}}^+ | g_m \rangle = \frac{2J_e + 1}{2J_g + 1}.$$
(66)

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From the relationship between the Einstein coefficients for stimulated absorption and stimulated emission it follows that  $\xi'$  also has to be interpreted as the mean number of photons in the resonance mode of the background field, i.e.,  $\xi = \xi'$ . We can now combine Eqs. (12), (63), and (64) to find the dissipative part of the evolution of the reduced density matrix of the radiator,

$$\left(\frac{d}{dt}\hat{\sigma}_{a}(t)\right)_{R} = -\frac{\Gamma}{2}(1+\xi)\left[\hat{P}_{e}^{a}\hat{\sigma}_{a}(t) + \hat{\sigma}_{a}(t)\hat{P}_{e}^{a}\right] + \Gamma(1+\xi)$$

$$\times \sum_{\lambda} \int \frac{d\Omega}{8\pi/3}(\hat{\mathbf{d}}_{a}^{-}\cdot\boldsymbol{\epsilon}_{\lambda})e^{-i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\hat{\sigma}_{a}(t)e^{i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}(\hat{\mathbf{d}}_{a}^{+}\cdot\boldsymbol{\epsilon}_{\lambda}) - \frac{\Gamma}{2}\frac{2J_{e}+1}{2J_{g}+1}\xi\left[\hat{P}_{g}^{a}\hat{\sigma}_{a}(t) + \hat{\sigma}_{a}(t)\hat{P}_{g}^{a}\right]$$

$$+ \Gamma\xi\sum_{\lambda} \int \frac{d\Omega}{8\pi/3}(\hat{\mathbf{d}}_{a}^{+}\cdot\boldsymbol{\epsilon}_{\lambda})e^{-i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}\hat{\sigma}_{a}(t)e^{i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{a}}(\hat{\mathbf{d}}_{a}^{-}\cdot\boldsymbol{\epsilon}_{\lambda}).$$

$$(67)$$

Equation (67) gives the combined contribution of the fluctuations arising from the coupling of the radiator to the vacuum field and to the source field generated by the surrounding medium of like atoms. We will refer to this term as the atom reservoir coupling.

We note that we can obtain this result also by saying that the perturbers generate a nonzero occupation number in some of the previously empty vacuum field modes. The new ground-state of the vacuum field is then no longer described by a state with zero occupation number in all field modes. Treating the vacuum field as Markovian, the derivation is analogous to the derivation of the dissipative part of the evolution of the reduced density matrix due to coupling to a bath of harmonic oscillators. A description of this method can, for example, be found in Ref. [24]. The expansion of the Liouville equation to second order in the interaction of the vacuum field with the atom then yields nonzero contributions for the normal-ordered and antinormal-ordered expectation values of the pairs of field commutators, thus giving rise to an additional term proportional to the mean photon number in the field mode at the resonance frequency of the atom. This method of derivation also emphasizes the interpretation of  $\xi$  as a mean photon number. In the case of a thermal background field with mean photon number  $\xi$ , the relationship between  $\Gamma$  and  $\Gamma \xi$  is exactly that found between the Einstein A and Einstein B coefficients, further underlining the interpretation of the effect of the medium in terms of stimulated emission and absorption processes.

Equation (67) gives the correct result in the case of a broadband background field where the number of photons per mode is slowly varying as a function of  $\omega_k$  compared to the width of the atomic transition or in the case where its width is extremely small compared to that of the atomic transition. In our case neither of these cases is fulfilled and the rate-equation limit therefore represents a rather crude approximation. We do believe, however, that the rate-equation limit still yields qualitative results especially on the dependence of the extra term on density and number of atoms. This is because the variation with these parameters is independent of the approximations discussed in this section. The rateequation limit primarily changes the outcome of the time integration over the product of the two-time correlation functions of the dipole of a perturber with the dipole of the radiator. In the BCA where each pair is considered independently this integration is independent of the spatial

integration over all the perturbers in the gas. The main effect of the approximations made here will therefore be in the dependence of the collisional term on the detuning and Rabi frequency.

In the remainder of this paper we will drop the label a for quantities referring to the radiator because the influence of the perturbers is now included in the mean photon number  $\xi$ .

## V. ESTIMATION OF THE BACKGROUND FIELD STRENGTH

We now estimate the dimensionless strength of the resonant background field  $\xi$ , using the Mollow formula [22] for the resonant component of the scattered light in the far-field limit, in the case of a weak field  $\Omega \ll \Gamma$ . The ratio of elastically scattered light to the total amount of scattered light is given by

$$\frac{I_{\rm el}}{I_{\rm el} + I_{\rm inel}} = \frac{1}{1 + s_0},\tag{68}$$

where  $s_0$  is the saturation parameter. We find therefore that the fraction of inelastically scattered light is given by

$$I_{\text{inel}} = s_0 I_{\text{scat}} \,. \tag{69}$$

Here  $I_{\text{scat}}$  is the total amount of scattered light that is proportional to the total intensity of the laser light  $I_L$  times the total scattering cross section. To derive an expression for the amount of inelastically scattered light produced by a perturber at a distance r from the radiator we have to multiply the fraction of inelastically scattered light by the cross section for the laser light divided by the surface area of a sphere with radius r. Hence the resonant contribution of the scattered light due to the perturber is simply

$$I_{\rm res}(r) = s_0 \frac{\sigma_L}{4\pi r^2} I_L, \qquad (70)$$

where the scattering cross section for the laser light is taken from Ref. [25] as

$$\sigma_L = \frac{\hbar \,\omega_L \Gamma}{2I_L} s_0. \tag{71}$$



FIG. 1. Mean number of photons on resonance in the background field as a function of the number of atoms N using Eq. (72). The solid line gives the mean number of photons for  $\delta = -6\Gamma$  and the dashed line for  $\delta = -8\Gamma$ . The Rabi frequency in both cases is  $\Omega = 0.5\Gamma$ .

This formula is valid only in the far-field approximation. For separations of the order of  $\chi_0$  higher-order terms in 1/r must be taken into account. The exact expression for the photon exchange interaction to all orders in 1/r can, for example, be found in Hillenbrand *et al.* [10]. If we now average over all atoms in a trap of radius *L* and constant density *n* and substitute Eq. (71) for  $\sigma_L$ , Eq. (70) yields the intensity of resonant radiation (in the limit  $s_0 \ll 1$ )

$$\bar{I}_{\rm res} = nLs_0^2 \left(\frac{\hbar\,\omega_L\Gamma}{2}\right). \tag{72}$$

The number of resonant photons per unit area is then given by  $\bar{I}_{\rm res}/\hbar\omega_L$  times the rate of spontaneous emission  $\Gamma/2$ . The mean number of photons that is actually reabsorbed on resonance is found by multiplication with the resonance cross section  $\sigma_{\rm res} \approx 4 \pi k_0^{-2}$ . Under the assumption of a constant density  $L \propto (N/n)^{1/3}$ , we approximate the mean number of photons on resonance by

$$\xi \approx \frac{4\,\pi}{k_0^2} n^{2/3} s_0^2 N^{1/3}.\tag{73}$$

We note that this result is consistent with the definition of  $\xi$  in Eq. (58) when the low-intensity result Eq. (4.30) in Ref. [22] for the spectral distribution function is used.

We can use Eq. (73) to find an estimate of the mean number of photons in the redistributed field, which is reabsorbed on resonance. Thus  $\xi$  defines a mean coupling strength of the background field to the radiator. Typical constant densities in the magneto-optical trap are of the order of  $n = 10^{11}$  cm<sup>-3</sup>. For cesium this corresponds to densities of 0.0618 atom per  $\lambda_0^3$ ,  $\lambda_0$  being the optical wavelength of the resonance transition. Figure 1 shows a plot of the mean number of photons in the background field as a function of the number of atoms *N* for different detunings. The mean number of photons is always considerably smaller than unity. The steady-state solutions for the excited-state populations in the absence of the laser field predict an increase of the population of the excited state, which initially is linear in  $\xi$ . For these small values of  $\xi$ , the low saturation approximation, which we will use in the subsequent calculations, and the factorization assumption Eq. (24) therefore hold well.

# VI. SUB-DOPPLER COOLING IN THE lin Lin CONFIGURATION

In this section we will discuss the modifications of the Sisyphus cooling mechanism by the presence of a background field of strength  $\xi$  for a  $J_g = 1/2$  to  $J_e = 3/2$  transition. The principle of Sisyphus cooling has, for example, been discussed in detail in Refs. [15,26]. The efficiency of this cooling mechanism relies on the correlation between optical pumping, which transfers population between the different atomic ground-state sublevels and the position of the atom in the laser beam. The presence of a fluctuating background field with a random polarization will weaken this correlation, therefore leading to a diminished position dependence of the populations in the atomic ground-state sublevels. We will now calculate the modified friction coefficient and diffusion coefficient for Sisyphus cooling as a function of the parameter  $\xi$  introduced earlier.

## A. Optical Bloch equations for a $J_g = 1/2$ to $J_e = 3/2$ transition

In this subsection we derive the optical Bloch equations for a  $J_g = 1/2$  to  $J_e = 3/2$  transition. We assume that the driving laser field is weak so that we can neglect the excited state in the equations of motion for the optical coherences. As usual, we transform the density matrix into a rotating reference frame, so that

$$\tilde{\sigma}(e_{\mu},g_{\nu}) = \sigma(e_{\mu},g_{\nu})e^{+i\omega_{L}t},$$
  
$$\tilde{\sigma}(g_{\mu},e_{\nu}) = \sigma(g_{\mu},e_{\nu})e^{-i\omega_{L}t},$$
 (74)

and make the rotating-wave approximation. The atom-laser interaction is given by

$$\hat{G}^{+}(\mathbf{r}) = \frac{\Omega}{\sqrt{2}} \operatorname{sink} z \left[ \left| e_{3/2} \right\rangle \left\langle g_{1/2} \right| + \frac{1}{\sqrt{3}} \left| e_{1/2} \right\rangle \left\langle g_{-1/2} \right| \right] + \frac{\Omega}{\sqrt{2}} \operatorname{cosk} z \left[ \left| e_{-3/2} \right\rangle \times \left\langle g_{-1/2} \right| + \frac{1}{\sqrt{3}} \left| e_{-1/2} \right\rangle \left\langle g_{1/2} \right| \right].$$
(75)

The equation for the optical coherences is

$$\frac{d}{dt}\tilde{\sigma}(g_{\pm 1/2}, e_{\nu}) = -\left(\frac{\Gamma}{2}(3\xi + 1) + i\delta\right)\tilde{\sigma}(g_{\pm 1/2}, e_{\nu})$$
$$+i\sum_{\mu} \tilde{\sigma}(g_{\pm 1/2}, g_{\mu})\langle g_{\mu}|\hat{G}^{-}(\mathbf{r})|e_{\nu}\rangle,$$
(76)

where we have already made the approximation  $\tilde{\sigma}(e_{\mu}, e_{\nu}) \approx 0$ . The optical Bloch equation for the excited state is given by

$$\frac{d}{dt}\tilde{\sigma}(e_{\mu},e_{\nu}) = \left(\frac{d}{dt}\tilde{\sigma}(e_{\mu},e_{\nu})\right)_{R}$$
$$-i\sum_{j} \left\{ \langle e_{\mu} | \hat{G}^{+}(\mathbf{r}) | g_{j} \rangle \tilde{\sigma}(g_{j},e_{\nu}) - \tilde{\sigma}(e_{\mu},g_{j}) \right.$$
$$\times \left\langle g_{j} | \hat{G}^{-}(\mathbf{r}) | e_{\nu} \right\rangle \right\}.$$
(77)

The part of the equation that is due to the coupling to the reservoir is

$$\left( \frac{d}{dt} \tilde{\sigma}(e_{\mu}, e_{\nu}) \right)_{R} = -\Gamma(\xi + 1) \tilde{\sigma}(e_{\mu}, e_{\nu})$$

$$+ \Gamma \xi \sum_{q,q'} \int \frac{d\Omega}{8 \pi/3} [\mathbf{d} \cdot (\mathbf{I} - \mathbf{n} \otimes \mathbf{n}) \cdot \mathbf{d}]_{q,q'}$$

$$\times (C_{\mu}^{\mu+q})^{*} C_{\nu}^{\nu+q'} \tilde{\sigma}(g_{\mu+q}, g_{\nu+q'}).$$
(78)

The coefficients  $C^{\mu+q}_{\mu}$  are the Clebsch-Gordan coefficients for the transition. The optical Bloch equations describing the evolution of the ground state are given by

$$\frac{d}{dt}\tilde{\sigma}(g_{\mu},g_{\nu}) = \left(\frac{d}{dt}\tilde{\sigma}(g_{\mu},g_{\nu})\right)_{R}$$
$$-i\sum_{j}\left\{\langle g_{\mu}|\hat{G}^{-}(\mathbf{r})|e_{j}\rangle\tilde{\sigma}(e_{j},g_{\nu}) - \tilde{\sigma}(g_{\mu},e_{j})\right.$$
$$\times \left\langle e_{j}|\hat{G}^{+}(\mathbf{r})|g_{\nu}\rangle\right\}, \tag{79}$$

where the coupling of the ground-state matrix elements to the reservoir is equal to

$$\left(\frac{d}{dt}\tilde{\sigma}(g_{\mu},g_{\nu})\right)_{R} = -2\Gamma\xi\tilde{\sigma}(g_{\mu},g_{\nu})$$
$$+\Gamma(\xi+1)\sum_{q,q'}\int\frac{d\Omega}{8\pi/3}$$
$$\times [\mathbf{d}\cdot(\mathbf{I}-\mathbf{n}\otimes\mathbf{n})\cdot\mathbf{d}]_{q,q'}$$
$$\times C_{\mu}^{\mu+q}(C_{\nu}^{\nu+q'})^{*}\tilde{\sigma}(e_{\mu+q},e_{\nu+q'}). \tag{80}$$

#### **B.** Steady-state solutions

By adiabatically eliminating the optical coherences and the excited-state in the equations for the ground-state matrix elements as in Ref. [15], we can use Eqs. (76), (77), and (79) to find the steady-state solutions of the density matrix. This procedure is justified in the low saturation approximation because the ground-state matrix elements evolve on a time scale that is much longer than the natural lifetime. The steady-state solution for the optical coherences is given by

$$\tilde{\sigma}(g_{\pm 1/2}, e_{\nu}) = \frac{\sum_{\mu} \tilde{\sigma}(g_{\pm 1/2}, g_{\mu}) \langle g_{\mu} | \hat{G}^{-}(\mathbf{r}) | e_{\nu} \rangle}{\delta - i \frac{\Gamma}{2} (3\xi + 1)}.$$
 (81)

We now substitute this solution into the equations for the excited-state density-matrix elements. This yields

$$\tilde{\sigma}(e_{\mu}, e_{\nu}) = \frac{1}{\Gamma(\xi+1)} \left\{ \xi \sum_{q} |C_{\mu}^{\mu+q}|^{2} \tilde{\sigma}(g_{\mu+q}, g_{\nu+q}) - i \sum_{j=\pm 1/2} [\langle e_{\mu} | \hat{G}^{+}(\mathbf{r}) | g_{j} \rangle \tilde{\sigma}(g_{j}, e_{\nu}) - \tilde{\sigma}(e_{\mu}, g_{j}) \langle g_{j} | \hat{G}^{-}(\mathbf{r}) | e_{\nu} \rangle ] \right\}.$$
(82)

Substituting this expressions together with the expressions for the optical coherences into the ground-state equations gives two differential equations of the form

$$\frac{d}{dt}\Pi_{1/2} = a_1 + a_2 \Pi_{1/2},$$

$$\frac{d}{dt}\Pi_{-1/2} = b_1 + b_2 \Pi_{-1/2}.$$
(83)

 $\Pi_{1/2}$  and  $\Pi_{-1/2}$  are the ground-state population for  $m_j = +1/2$  and  $m_j = -1/2$ , respectively, and the coefficients  $a_i, b_i$ , which are dependent on detuning, Rabi frequency, and the strength of the background field, are given by

$$a_{1} = \frac{\Gamma}{9} s_{0}(\xi) \left\{ 8 \left( \frac{\delta}{\Omega} \right)^{2} \xi + 2 \left( \frac{\Gamma}{\Omega} \right)^{2} \xi (1+3\xi)^{2} + (1+3\xi)(1-\cos 2kz) \right\},$$
(84)

$$a_{2} = -\frac{2\Gamma}{9}s_{0}(\xi) \left\{ 8 \left(\frac{\delta}{\Omega}\right)^{2} \xi + 2 \left(\frac{\Gamma}{\Omega}\right)^{2} \xi (1+3\xi)^{2} + (1+3\xi) \right\},$$
(85)

$$b_{1} = \frac{\Gamma}{9} s_{0}(\xi) \left\{ 8 \left( \frac{\delta}{\Omega} \right)^{2} \xi + 2 \left( \frac{\Gamma}{\Omega} \right)^{2} \xi (1+3\xi)^{2} + (1+3\xi)(1+\cos 2kz) \right\}.$$
(86)

We also have assumed that to first order in  $\Omega$  the population is completely in the atomic ground state. The coefficient  $b_2$ is equal to  $a_2$ .  $s_0(\xi)$  is defined as

$$s_0(\xi) = \frac{\Omega^2/2}{\delta^2 + \Gamma^2/4(1+3\xi)^2}.$$
 (87)

The steady-state solutions for the ground-state populations are calculated from formulas (83) by setting the left-hand side equal to zero. We hence find for the steady state of the ground-state populations

$$\Pi_{1/2}^{\text{st}} = \frac{1}{2} \left( 1 - \frac{s_0(\xi)(1+3\xi)}{4\xi + s_0(\xi)(1+3\xi)} \cos 2kz \right), \quad (88)$$

$$\Pi_{-1/2}^{\text{st}} = \frac{1}{2} \left( 1 + \frac{s_0(\xi)(1+3\xi)}{4\xi + s_0(\xi)(1+3\xi)} \cos 2kz \right).$$
(89)

## EFFECT OF SCATTERED RADIATION ON SUB-DOPPLER COOLING



FIG. 2. Variation of  $\Pi_{1/2}^{\text{st}}$  as a function of position in the beam for  $\Omega = 0.5\Gamma$  and  $\delta = -6.0\Gamma$ , corresponding to a saturation parameter  $s_0 = 0.0034$ . The solid line shows the oscillation of the population for zero background field, the dashed line for a background field  $\xi = 1.8 \times 10^{-4}$ , and the dot-dashed line is equivalent to  $\xi = 8.6 \times 10^{-4}$ . At a density of  $10^{11}$  cm<sup>-3</sup> this corresponds to  $10^{6}$ and  $10^{8}$  atoms, respectively. The dotted line shows that at a background field strength  $\xi = 0.01$  larger than the saturation parameter  $s_0$  the population depends only weakly on  $z/\lambda$ ,  $\lambda$  being the laser wavelength.

For  $\xi = 0$  we retrieve the familiar expressions for the steadystate ground-state populations found in Ref. [15],

$$\Pi_{1/2}^{\text{st}} = \sin^2 kz,$$

$$\Pi_{-1/2}^{\text{st}} = \cos^2 kz.$$
(90)

To obtain a better physical grasp of the effect of the background field, we first discuss the steady-state solutions for the ground-state populations.

# C. Dependence of the steady-state ground-state populations $\Pi^{st}_{\pm 1/2}$ on the background field

Figure 2 shows a plot of the steady-state population  $\Pi_{1/2}^{\text{st}}$  as a function of z over the laser wavelength  $\lambda$  for different  $\xi$ . The dependence of the population in the groundstate sublevels  $m_g = \pm 1/2$  on the position in the beam diminishes very rapidly, and in the case where the background field strength is significantly larger than the saturation parameter the population depends only weakly on the position. For example, a background field of strength  $\xi = 0.01$ , a Rabi frequency of  $0.5\Gamma$ , and a detuning of  $-6\Gamma$  (corresponding to  $s_0 = 0.0034$ ) will lead to an oscillation of only  $\pm 0.04$  around the mean value of 0.5, whereas for  $\xi = 0$  we have a variation between the values 0 and 1. The background field of strength  $\xi$  introduces transition rates between the two ground-state sublevels that are independent of the position of the atom in the laser beam. If these processes are strong enough to compete with the optical pumping processes induced by the laser photons, the efficiency of the latter processes is reduced.

The peak to peak amplitude of the position-dependent oscillations of the two ground-state populations given by Eqs. (88) and (89) can be characterized by the parameter

$$f = \frac{s_0(\xi)(1+3\xi)}{4\xi + s_0(\xi)(1+3\xi)}.$$
(91)

This factor can be interpreted in the following way. The steady-state solution for the excited-state population  $\Pi_m^e$  in the presence of a background field inducing transitions at a rate  $\Gamma \xi$  and without the laser field is given by

$$\Pi_m^e = \frac{1}{2} \frac{\xi}{1+3\xi}.$$
 (92)

The total excited-state population induced by the background field is obtained by summing over all the excited-state sublevels. As the population in Eq. (92) is independent of the magnetic quantum number, the total excited-state population is the number of sublevels times Eq. (92). For a  $J_e = 3/2$  excited state the total population is therefore  $4\Pi_m^e$ . At the same time the total excited-state population induced by a weak laser field is approximately given by  $\frac{1}{2}s_0$ . Rearranging Eq. (91) slightly, we find that f can be written as the ratio of the fraction of the excited-state population induced by the laser field over the total excited-state population induced by the background and the laser field, i.e.,

$$f = \frac{\frac{1}{2}s_0(\xi)}{4\frac{\xi}{2(1+3\xi)} + \frac{1}{2}s_0(\xi)}.$$
 (93)

If the number of transitions induced by the laser field is much less than the number of transitions induced by the background field, the ground-state populations will be almost independent of z.

We have to keep in mind that in any case we have to restrict ourselves to  $\xi \ll 1$ , so that we can still assume that the ground-state population is equal to unity. In this case it is valid to set  $1+3\xi\approx 1$ . It is interesting to consider the two limiting cases of  $4\xi \gg s_0$  and  $4\xi \ll s_0$ . For  $4\xi \gg s_0$  we can expand f in terms of the small parameter  $\epsilon_1 = s_0/(4\xi)$ . To first order in  $\epsilon_1$  we find

$$f = \frac{k_0^2}{16\pi n^{2/3}} s_0^{-1} N^{-1/3}.$$
 (94)

Here we have already substituted the expression for  $\xi$ , Eq. (73), into Eq. (91). We can rewrite the condition  $\epsilon_1 \ll 1$  as

$$4\Gamma\sigma_{\rm res}nL \gg \Gamma s_0^{-1}.$$
(95)

In the second case we expand in the small parameter  $\epsilon_2 = 4\xi/s_0$ . To first order this yields

$$f = 1 - (16\pi k_0^{-2} n^{2/3}) s_0 N^{1/3}, \tag{96}$$

with the condition

$$4\Gamma\sigma_{\rm res}nL \ll \Gamma s_0^{-1}.$$
 (97)

We interpret the conditions (95) and (97) as follows. Condition (95) states that the pumping rate due to the source field is larger than the optical pumping rate induced by the laser. Since the source field is unpolarized, the stimulated emission and absorption processes caused by the source field are independent of position and tend to distribute the population equally among the sublevels. In the limit of condition (95) this process is dominant over the optical pumping processes caused by the laser field and the Sisyphus cooling is destroyed. In the limit of Eq. (97) optical pumping between the ground-state sublevels is still stronger than the pumping processes induced by the source field. Equation (96) shows that there is, however, a decrease of the peak to peak amplitude of the oscillations that is proportional to the density, the saturation parameter, and the number of atoms to the one-third power.

#### **D.** Force in the $lin \perp lin$ configuration

We will now discuss the dependence of the friction coefficient on the background field. The equation for the force in the  $lin \perp lin$  configuration is given by

$$F(\xi) = -\frac{2}{3}\hbar k \,\delta s_0(\xi) \sin(2kz) [\Pi_{+1/2}(\xi) - \Pi_{-1/2}(\xi)].$$
(98)

In order to find the friction coefficient we have to solve the set of differential equations given by Eq. (83). We rewrite the differential equations in the form

$$\frac{d\Pi_{\pm 1/2}(\xi)}{dt} = -\Gamma_p(\xi) [\Pi_{\pm 1/2}(\xi) - \Pi_{\pm 1/2}^{\text{st}}(\xi)].$$
(99)

Here we have defined the optical pumping rate as a function of the background field strength

$$\Gamma_{p}(\xi) = \frac{2}{9} \Gamma s_{0}(\xi) (1+3\xi) f^{-1}.$$
(100)

 $s_0(\xi)$  is the saturation parameter defined in Eq. (87) and *f* is defined by Eq. (91).  $\Pi_{\pm 1/2}^{\text{st}}(\xi)$  are the steady-state solutions for the ground-state populations defined in Eqs. (88) and (89). The differential equations for the ground-state populations can be solved analytically by assuming that the ground-state populations contain no explicit time dependence. Their solutions are found to be

$$\Pi_{\pm 1/2}(z,\xi) = \frac{1}{2} \mp \frac{1}{2} \frac{\cos 2kz + [v/v_c(\xi)]\sin 2kz}{1 + [v/v_c(\xi)]^2} f.$$
(101)

The critical velocity  $v_c(\xi)$  gives the capture range of the Sisyphus cooling force. It is defined similar to Ref. [15] in terms of the optical pumping rate (100) as

$$v_c(\xi) = \frac{\Gamma_p(\xi)}{2k}.$$
 (102)

It is thus dependent on the background field through the  $\xi$  dependence of  $\Gamma_p(\xi)$ . The expression for the force in the lin  $\perp$  lin configuration as a function of the background field is then given by

$$F(\xi) = -\frac{2}{3} \hbar k \,\delta s_0(\xi) \sin 2kz \frac{\cos 2kz + [v/v_c(\xi)] \sin 2kz}{1 + [v/v_c(\xi)]^2} f.$$
(103)



FIG. 3. Sisyphus friction coefficient as a function of  $N^{1/3}$ , normalized with respect to its value at  $\xi=0$ . The nonlinear behavior of  $\alpha$  as a function of  $N^{1/3}$  is clearly visible. Note also the strong decrease of  $\alpha$  from about 67% of its value for  $\xi=0$  to only about 25% at  $N=10^8$ . The dashed line shows the diffusion coefficient Eq. (112). The friction decreases at a faster rate than the diffusion coefficient leading to a net increase of the ratio of the two. The parameters chosen were  $\Omega=0.5\Gamma$  and  $\delta=-6\Gamma$  at a density of  $n=10^{11}$  cm<sup>-3</sup>.

If we average the force over a wavelength we find the friction force to be given by

$$\bar{F}(\xi) = -\frac{\alpha(\xi)v}{1 + [v/v_c(\xi)]^2}$$
(104)

and the corresponding friction coefficient  $\alpha(\xi)$  is

$$\alpha(\xi) = -3\hbar k^2 \frac{\delta}{(1+3\xi)\Gamma} f^2.$$
(105)

If we now expand f in terms of the parameters  $\epsilon_1$  and  $\epsilon_2$ , respectively, and keep only the lowest-order terms in those parameters, we find that in the case described by condition (95) the lowest-order term is quadratic in the expansion parameter  $\epsilon_1$ , whereas in the case of condition (97) the friction coefficient can be written as

$$\alpha \approx -3\hbar k^2 \frac{\delta}{\Gamma} [1 - 2(16\pi k_0^{-2} n^{2/3}) s_0 N^{1/3}].$$
 (106)

In the case of large detuning the reduction of the friction coefficient due to the background field scales as  $(\Omega^2/\delta)N^{1/3}$ . However, the region where a linear expansion in terms of  $4\xi/s_0$  is accurate is quite small. For example, for a Rabi frequency  $\Omega = 0.5\Gamma$  and  $\delta = -6\Gamma$ ,  $s_0 = 0.0034$ , and  $n = 10^{11}$  cm<sup>-3</sup> we find that the expansion parameter  $\epsilon_2 = 4\xi/s_0$  is in the range 0.2–1 for numbers of atoms between  $N = 10^6$  and  $10^8$ . In this regime the first-order expansion in  $\epsilon_2$  is no longer valid. Figure 3 shows the friction coefficient for these parameters as a function of  $N^{1/3}$  for atom numbers between  $10^6$  and  $10^8$ . The nonlinear behavior with respect to  $\xi$  is clearly visible. The friction coefficient is nor-

malized with respect to its value at  $\xi = 0$ . It drops from about 67% of its initial value (for  $\xi = 0$ ) at  $N = 10^6$  to only about 25% at  $N = 10^8$ .

#### E. The equilibrium temperature in the lin $\perp$ lin configuration

To calculate the equilibrium temperature we have to work out the momentum diffusion coefficient for this configuration. The method used is similar to Ref. [27]. The main part of the momentum diffusion coefficient arises from the fluctuations of the dipole force Eq. (98) due to the atom jumping back and forth between the two ground-state sublevels. Its contribution can be calculated using the formula

$$D_{p} = \int_{0}^{\infty} d\tau \{ \bar{F}(t+\tau)F(t) - \bar{F}(t)^{2} \}, \qquad (107)$$

where F(t) denotes the dipole force at time t. The two-time correlation function can be evaluated using [15,27]

$$\bar{F}(t+\tau)F(t) = \sum_{i=\pm 1/2} \sum_{j=\pm 1/2} F_i F_j P(i,t;j,t+\tau).$$
(108)

Here  $P(i,t;j,t+\tau)$  represents the probability of being in the state *i* at time *t* and in state *j* at time  $t+\tau$ .  $F_{\pm 1/2}$  are the force contributions acting on the individual sublevels  $|m_{\pm 1/2}\rangle$ . From Eq. (98) we find

$$F_{\pm 1/2} = \mp \frac{2}{3} \hbar k \, \delta s_0(\xi) \sin 2kz. \tag{109}$$

Assuming that the system is invariant under time translation, we get

$$\overline{F(\tau)F(0)} = 4F_{1/2}^2 \Pi_{1/2}^{\text{st}}(\xi) \Pi_{-1/2}^{\text{st}}(\xi) e^{-\Gamma_p(\xi)\tau} + 2F_{1/2}^2 \{ [(\Pi_{1/2}^{\text{st}}(\xi))^2 + [\Pi_{-1/2}^{\text{st}}(\xi)]^2 - \frac{1}{2} \}.$$
(110)

This yields the momentum diffusion coefficient as

$$D_{p}(z) = \left[\frac{2}{3}\hbar k^{2} \delta s_{0}(\xi)\right]^{2} \Gamma_{p}(\xi)^{-1} \left[(1-f^{2})\sin^{2}2kz + f^{2}\sin^{4}2kz\right].$$
(111)

We now have to average this term over the distance of a laser wavelength to get the average diffusion coefficient, i.e.,

$$\overline{D_p}(\xi) = (\hbar k)^2 \frac{\delta^2}{(1+3\xi)\Gamma} s_0(\xi) [f - \frac{1}{4}f^3].$$
(112)

 $D_p(\xi)$  is the part of the momentum diffusion due to the dipole force fluctuations only. The dashed line in Fig. 3 shows the scaling of  $\overline{D_p}$  with N, for  $\delta = -6\Gamma$  and  $\Omega = 0.5\Gamma$ . The diffusion coefficient decreases at a slower rate than the friction coefficient, leading to a net increase of the ratio of the two. The total momentum diffusion coefficient is obtained by adding the contributions due to fluctuations in the momentum carried away by the fluorescent photons and the fluctuations in the number of photons absorbed from each laser beam. It was pointed out that in the case  $\xi = 0$  this term can be neglected as it is much smaller than  $\overline{D_p}(\xi)$  in the regime where  $|\delta| \ll \Gamma$  [15]. Apart from numerical factors due



FIG. 4. Plot of temperature vs number of atoms to the one-third power in the case of Sisyphus cooling. The temperature has been normalized with respect to the temperature that would have been obtained for  $\xi=0$ . The dotted line corresponds to  $\Omega=0.5\Gamma$  and  $\delta=-6\Gamma$ , and the dashed line to  $\Omega=0.5\Gamma$  and  $\delta=-8\Gamma$ , when only  $\overline{D_p}/\alpha(\xi)$  is used to determine the temperature. The solid line shows the temperature increase when  $(\overline{D_p}+D_p^{\rm sp})/\alpha(\xi)$  is used instead. The parameters for the solid line are the same as for the dotted line.

to the angular momentum of the atomic transition this term can be modeled by the diffusion coefficient obtained from a two-level model. A simple estimate gives

$$D_{p}^{\rm sp}(\xi) = \frac{2}{5}\hbar^{2}\mathbf{k}^{2}\Gamma[s_{0}(\xi) + \xi]$$
(113)

[the contribution due to spontaneous emission].

The equilibrium temperature is given by the ratio of the averaged momentum diffusion coefficient and the friction coefficient. Taking only the induced part of the momentum diffusion coefficient, we find in the limit of large detuning

$$k_B T = \frac{\hbar \Omega^2}{6|\delta|} \left\{ \frac{1}{f} - \frac{1}{4}f \right\}.$$
 (114)

The variation of the temperature given by Eq. (114) as a function of  $N^{1/3}$  is shown in Fig. 4. The parameters for the density and the Rabi frequency are the same as in Fig. 3. The dotted line corresponds to  $D_p(\xi)/\alpha(\xi)$  at a detuning of  $\delta = -6\Gamma$  and the dashed line gives the same quantity but for a detuning of  $\delta = -8\Gamma$ . The effect of including  $D_p^{\rm sp}(\xi)$  is depicted by the solid curve. The parameters are the same as for the dotted line. The maximum error that is made by neglecting  $D_p^{\rm sp}(\xi)$  is about 5%. The temperature varies by a factor 2 or more even for very small values of  $\xi$ ; see Fig. 1 for the magnitude of  $\xi$ .

From Eq. (91) we obtain, for 1/f,

$$\frac{1}{f} = 1 + \frac{4\xi}{s_0(\xi)(1+3\xi)}$$
(115)

and by substituting Eq. (73) for the background field

$$\frac{1}{f} = 1 + (16\pi k_0^{-2} n^{2/3}) s_0 N^{1/3}.$$
 (116)

In the limit of Eq. (97) we can expand f according to (96). Also, in the large detuning limit  $s_0 \approx \Omega^2/(2\delta^2)$ . Combining Eqs. (96) and (116) with Eq. (114) yields

$$k_B T = \frac{\hbar \Omega^2}{8|\delta|} \left\{ 1 + \frac{17}{12} (16\pi k_0^{-2} n^{2/3}) \left( \frac{\Omega^2}{2\delta^2} \right) N^{1/3} \right\}.$$
 (117)

We have noted above that the expansion of f in terms of  $\xi/s_0$  breaks down very quickly in the parameter range investigated. This is not so for the temperature because 1/f increases linearly with  $\xi/s_0$ . For small numbers of N, where the expansion is valid, the temperature will increase according to Eq. (117). It will then go through a small transition region where it is slightly nonlinear. For large numbers of atoms f tends to zero and the dominant term is 1/f so that the temperature again behaves linearly with respect to  $N^{1/3}$ . A comparison of Eqs. (114) and (117) shows that in the parameter range investigated the difference between the two expressions never exceeds 2%. Equation (117) suggests that the excess temperature depends on the detuning approximately as  $1/\delta^{-3}$ , assuming that the density is independent of detuning (which is not physical [14]).

# VII. THE $\sigma_+$ - $\sigma_-$ CONFIGURATION

In this section we discuss the influence of the background field on the motion-induced orientation mechanism [15]. As in Ref. [15], we solve the optical Bloch equations (OBE's) by transformation into a reference frame rotating with a frequency kv. As usual, we make the rotating-wave approximation and transform the density matrix according to Eq. (74). In the low saturation approximation we also take  $\sigma(e_{\mu}, e_{\nu})$  to be zero to first order in the Rabi frequency. The OBE's for the optical coherences, the excited-state matrix elements, and the ground-state matrix elements are hence given by

$$\frac{d}{dt}\tilde{\sigma}(g_{\mu},e_{\nu}) = -\left[\frac{\Gamma}{2}(1+\frac{8}{3}\xi) + i[\delta + kv(\mu-\nu)]\right]\tilde{\sigma}(g_{\mu},e_{\nu}) + i\sum_{j}\tilde{\sigma}(g_{\mu},g_{j})\langle g_{j}|\hat{\mathscr{G}}^{-}(\mathbf{r})|e_{\nu}\rangle, \qquad (118)$$

$$\frac{d}{dt}\tilde{\sigma}(e_{\mu},e_{\nu}) = -ikv(\mu-\nu)\tilde{\sigma}(e_{\mu},e_{\nu}) + \left(\frac{d}{dt}\tilde{\sigma}(e_{\mu},e_{\nu})\right)_{R}$$
$$-i\sum_{j}\left\{\langle e_{\mu}|\hat{\mathscr{G}}^{+}(\mathbf{r})|g_{j}\rangle\tilde{\sigma}(g_{j},e_{\nu}) - \tilde{\sigma}(e_{\mu},g_{j})\right\}$$
$$\times \left\langle g_{j}|\hat{\mathscr{G}}^{-}(\mathbf{r})|e_{\nu}\rangle\right\}, \tag{119}$$

$$\frac{d}{dt}\tilde{\sigma}(g_{\mu},g_{\nu}) = -ikv(\mu-\nu)\tilde{\sigma}(g_{\mu},g_{\nu}) + \left(\frac{d}{dt}\tilde{\sigma}(g_{\mu},g_{\nu})\right)_{R}$$
$$-i\sum_{j} \left\{ \langle g_{\mu} | \hat{\mathscr{G}}^{-}(\mathbf{r}) | e_{j} \rangle \tilde{\sigma}(e_{j},g_{\nu}) - \tilde{\sigma}(g_{\mu},e_{j}) \right.$$
$$\times \left\langle e_{j} | \hat{\mathscr{G}}^{+}(\mathbf{r}) | g_{\nu} \rangle \right\}.$$
(120)

Here  $\hat{\mathscr{G}}^{\pm}(\mathbf{r})$  denotes the atom-laser coupling. The coupling to the vacuum field and the background field is found from Eq. (67) as

$$\left(\frac{d}{dt}\tilde{\sigma}(e_{\mu},e_{\nu})\right)_{R} = -\Gamma(\xi+1)\tilde{\sigma}(e_{\mu},e_{\nu}) + \Gamma\xi\sum_{q,q'}\int \frac{d\Omega}{8\pi/3} \\ \times [\mathbf{d}\cdot(\mathbf{I}-\mathbf{n}\otimes\mathbf{n})\cdot\mathbf{d}]_{q,q'}(C_{\mu}^{\mu+q})^{*} \\ \times C_{\nu}^{\nu+q'}\tilde{\sigma}(g_{\mu+q},g_{\nu+q'}), \qquad (121)$$

$$\left(\frac{d}{dt}\tilde{\sigma}(g_{\mu},g_{\nu})\right)_{R} = -\frac{5}{3}\Gamma\xi\tilde{\sigma}(g_{\mu},g_{\nu}) + \Gamma(\xi+1)\sum_{q,q'}\int\frac{d\Omega}{8\pi/3}$$
$$\times [\mathbf{d}\cdot(\mathbf{I}-\mathbf{n}\otimes\mathbf{n})\cdot\mathbf{d}]_{q,q'}C_{\mu}^{\mu+q}$$
$$\times (C_{\nu}^{\nu+q'})^{*}\tilde{\sigma}(e_{\mu+q},e_{\nu+q'}).$$

In the low saturation limit the optical coherences and the excited-state matrix elements can be eliminated adiabatically from the ground-state equations and we obtain a set of four coupled equations for the ground-state matrix elements.

# A. Steady-state solutions for the ground-state elements in the low-velocity limit

The ground-state equations in their general form are still too complicated to allow an analytical solution of the problem. In the remainder of this paper we want to restrict ourselves to the low-velocity limit  $kv \ll \Gamma$ . In this limit, we can obtain analytical solutions for the ground-state densitymatrix elements. Neglecting terms of order 2 in  $kv/\Gamma$  and of order  $s_0kv/\Gamma$ , we find

$$0 = -\left(\frac{14\xi}{3(1+\frac{8}{3}\xi)s_{0}(\xi)} + \frac{5}{6}\right)\Pi_{-1}^{g} + \left(\frac{13\xi}{3(1+\frac{8}{3}\xi)s_{0}(\xi)} + \frac{3}{2}\right)\Pi_{0}^{g} + \left(\frac{\xi}{3(1+\frac{8}{3}\xi)s_{0}(\xi)} + \frac{1}{6}\right)\Pi_{1}^{g} - \frac{2}{3}C_{r} - \frac{2\delta}{\Gamma(1+\frac{8}{3}\xi)}C_{i},$$
(122)

$$0 = \left(\frac{\xi}{3(1+\frac{8}{3}\xi)s_{0}(\xi)} + \frac{1}{6}\right)\Pi_{-1}^{g} + \left(\frac{13\xi}{3(1+\frac{8}{3}\xi)s_{0}(\xi)} + \frac{3}{2}\right)\Pi_{0}^{g} - \left(\frac{14\xi}{3(1+\frac{8}{3}\xi)s_{0}(\xi)} + \frac{5}{6}\right)\Pi_{1}^{g} - \frac{2}{3}C_{r} + \frac{2\delta}{\Gamma(1+\frac{8}{3}\xi)}C_{i},$$
(123)

$$1 = \Pi_{-1}^{g} + \Pi_{0}^{g} + \Pi_{1}^{g}, \qquad (124)$$

$$0 = \frac{1}{8} \Pi_{-1}^{g} + \frac{3}{8} \Pi_{0}^{g} + \frac{1}{8} \Pi_{1}^{g} - \frac{6kv}{\Gamma(1 + \frac{8}{3}\xi)s_{0}(\xi)} \bigg[ 1 + \frac{\xi}{1 + \xi} \bigg] C_{i}$$
$$- \bigg[ \frac{5}{4} + \frac{13\xi}{4(1 + \frac{8}{3}\xi)s_{0}(\xi)} \bigg] C_{r}, \qquad (125)$$

$$0 = \frac{\delta}{4\Gamma(1+\frac{8}{3}\xi)s_{0}(\xi)}\Pi_{-1}^{g} - \frac{\delta}{4\Gamma(1+\frac{8}{3}\xi)s_{0}(\xi)}\Pi_{1}^{g} - \left[\frac{5}{4} + \frac{13\xi}{4(1+\frac{8}{3}\xi)s_{0}(\xi)}\right]C_{i} + \frac{6kv}{\Gamma(1+\frac{8}{3}\xi)s_{0}(\xi)}\left[1 + \frac{\xi}{1+\xi}\right]C_{r}, \qquad (126)$$

where  $C_r$  and  $C_i$  are the real and imaginary parts of the ground-state coherence  $\tilde{\sigma}(g_{-1},g_1)$ , defined by

$$\tilde{\sigma}(g_{-1},g_1) \equiv C_r + iC_i, \qquad (127)$$

and the background-field-dependent saturation parameter is given by

$$s_0(\xi) = \frac{\Omega^2/2}{\delta^2 + \Gamma^2/4(1 + \frac{8}{3}\xi)^2}.$$
 (128)

The solution of this closed set of linear equations is straightforward and we obtain the steady-state solutions for the ground-state matrix elements as

$$\Pi_1^g(\xi) = \frac{1}{2} \left[ 1 - \Pi_0^g(\xi) \right] + \frac{1}{2} \Delta_{\Pi} , \qquad (129)$$

$$\Pi_{-1}^{g}(\xi) = \frac{1}{2} \left[ 1 - \Pi_{0}^{g}(\xi) \right] - \frac{1}{2} \Delta_{\Pi}, \qquad (130)$$

$$\Pi_0^g(\xi) = \frac{13\xi + 4(1 + \frac{3}{3}\xi)s_0(\xi)}{39\xi + 17(1 + \frac{8}{3}\xi)s_0(\xi)},\tag{131}$$

$$C_r(\xi) = \frac{5(1+\frac{8}{3}\xi)s_0(\xi)}{78\xi+34(1+\frac{8}{3}\xi)s_0(\xi)},$$
(132)

$$C_{i}(\xi) = \frac{60\Gamma kv (1 + \frac{8}{3}\xi)s_{0}(\xi)}{39\xi + 17(1 + \frac{8}{3}\xi)s_{0}(\xi)} \left[1 + \frac{\xi}{1 + \xi}\right] \frac{5\xi + (1 + \frac{8}{3}\xi)s_{0}(\xi)}{\xi\Gamma^{2}[65\xi + 38(1 + \frac{8}{3}\xi)s_{0}(\xi)] + s_{0}^{2}(\xi)[4\delta^{2} + 5\Gamma^{2}(1 + \frac{8}{3}\xi)^{2}]},$$
(133)

where  $\Delta_{\Pi}(\xi)$  is the population imbalance between the two stretched states

$$\Delta_{\Pi}(\xi) = \frac{24kv}{s_0(\xi)\delta} \bigg[ 1 + \frac{\xi}{1+\xi} \bigg] C_r(\xi) - \frac{5\Gamma}{\delta} \bigg[ (1 + \frac{8}{3}\xi) + \frac{13\xi}{5s_0(\xi)} \bigg] C_i(\xi).$$
(134)

In the following subsections we use this set of solutions to find analytical expressions for the low-velocity friction force, the momentum diffusion coefficient, and the temperature as a function of the mean number of background photons  $\xi$ .

#### B. The mean force in the low-velocity approximation

The mean force is calculated from the gradient of the atom-laser interaction by taking the average over the internal atomic states. In the low-velocity approximation, where we can neglect the Doppler term in the saturation parameters of the beam, we find

$$F(\xi) = \frac{\hbar k \Gamma}{2} (1 + \frac{8}{3}\xi) s_0(\xi) \Biggl\{ \frac{5}{6} [\Pi_1^g(\xi) - \Pi_{-1}^g(\xi)] + \frac{2\delta}{3\Gamma(1 + \frac{8}{3}\xi)} C_i(\xi) \Biggr\}.$$
 (135)

Using the solutions for the ground-state density-matrix elements, Eqs. (129)–(133), we obtain a force that is linear in the atomic velocity. For  $\xi=0$  formula (135) coincides with the expression of the motion-induced orientation force de-



FIG. 5. Dependence of the friction coefficient on the number of atoms N to the one-third power. The Rabi frequency is equal to  $\Omega = 0.5\Gamma$  and the detuning to  $\delta = -8\Gamma$ . The solid curve shows a plot of the friction coefficient for the total force Eq. (135), the dashed line shows the friction coefficient obtained from perturbation theory, and the dotted curve gives only the contribution of the dissipative part of the force.

rived in Ref. [15]. Figure 5 shows the dependence of the friction coefficient for motion-induced orientation cooling on the number of atoms to the one-third power. The parameters chosen are  $n=10^{11}$  cm<sup>-3</sup>,  $\delta=-8\Gamma$ , and  $\Omega=0.5\Gamma$ . Each curve is normalized with respect to its value at  $\xi=0$ . The solid line represents a plot of the normalized friction coefficient,  $\alpha(\xi)/\alpha(0)$ ; the dotted line shows the variation of the friction coefficient, which is due to the dissipative force only. It is noticeable that the dissipative force decreases stronger than the total force because the reactive part of the force initially increases as a function of  $\xi$ .

The decrease of the force as a function of the number of photons in the background field can be understood as follows. For an atom at rest the coupling of the atom to the laser field leads to the production of an alignment in the atomic ground state and it is the coupling of this alignment to the fictitious magnetic field that produces the force. The background field introduces additional transition rates between the different Zeeman sublevels, which reduces the atomic alignment and hence leads to a corresponding reduction of the motion-induced orientation force. In order to verify this argument we have calculated the force using perturbation theory as described by Dalibard and Cohen-Tannoudji [15] and Steane et al. [28]. For an atom at rest the steady-state populations in a basis set quantized along the direction of the local laser field polarization (the y basis) and a basis set quantized along the direction of propagation of the laser beams (the z basis) are related by a unitary transformation U. We can therefore use the ground-state solutions for kv = 0 and retransform into the y basis to get the populations in the states  $|+1_{\nu}\rangle$ ,  $|-1_{\nu}\rangle$ , and  $|0_{\nu}\rangle$  as a function of  $\xi$ . The calculations show that for the parameters used in Fig. 5 the population in the central state  $m_{y}=0$  decreases from 9/17 = 0.53 at N = 0 to 0.48 at  $N = 10^8$  and the population in the stretched states  $m_y = \pm 1$  increases from 4/17 = 0.235 to 0.26, clearly showing the weakening of the alignment by the background field. We can use the populations in the y basis to estimate the force in the perturbative limit assuming that the only effect of the background field is a change in the equilibrium populations for an atom at rest. In particular, this means the light shifts of the ground state remain unaffected by the background field. The effect of the atomic motion on the populations in the y basis can then be included in exactly the same way as in Refs. [15,28] using a perturbation expansion in terms of  $kv/\Delta$ , where  $\Delta$  again denotes the light shift. For the motion-induced population imbalance in the z basis this yields

$$(\Pi_1 - \Pi_{-1})(\xi) = \frac{6kv}{\Delta} [2\Pi_0^{(y)}(\xi) - \Pi_1^{(y)}(\xi) - \Pi_{-1}^{(y)}(\xi)].$$
(136)

The normalized friction coefficient obtained from the perturbation treatment is shown in Fig. 5 by the dashed curve. In Ref. [28] we noted that the perturbation calculation only gives the dissipative part of the motion-induced orientation force. The variation of the perturbative result and the dissipative part of the force with  $\xi$  are in agreement with each other to within a few percent, indicating that the decrease in the motion-induced orientation force is indeed due to the destruction of the alignment by the background field.

#### C. The momentum diffusion coefficient

The momentum diffusion coefficient is calculated for an atom with zero velocity that still exchanges momentum with the laser field. For a sample of laser-cooled atoms the mean momentum is equal to zero and the momentum diffusion coefficient can be found from

$$D_p = \frac{1}{2} \frac{d}{dt} \langle p^2 \rangle. \tag{137}$$

The mean of the momentum squared is given by the sum over the expectation values of  $p^2$  for each individual internal atomic state so that we can rewrite its rate of change as

$$\frac{d}{dt}\langle p^{2}\rangle = \langle p^{2}\dot{\Pi}_{1}^{g}(p)\rangle + \langle p^{2}\dot{\Pi}_{0}^{g}(p)\rangle + \langle p^{2}\dot{\Pi}_{-1}^{g}(p)\rangle + \langle p^{2}\dot{\Pi}_{2}^{e}(p)\rangle + \langle p^{2}\dot{\Pi}_{1}^{e}(p)\rangle + \langle p^{2}\dot{\Pi}_{0}^{e}(p)\rangle + \langle p^{2}\dot{\Pi}_{-1}^{e}(p)\rangle + \langle p^{2}\dot{\Pi}_{-2}^{e}(p)\rangle.$$
(138)

To calculate the rates of change for the second moments of the momentum distribution we start by setting up the equations of motion for the density matrix including photon recoil using the momentum family approach [29,30]. The only part of the optical Bloch equations that can redistribute atoms among different momentum families is the dissipative part of the optical Bloch equations describing the coupling of the atom to the vacuum field and the background field Eq. (67). The coupling of different momentum families by spontaneous emission in the limit of zero background field has, for example, been discussed by Castin *et al.* [29] for the case of a  $J=0 \rightarrow J=1$  transition. We now introduce the notation

$$\tilde{\sigma}(i_{\mu}, j_{\nu}, p) = \langle i_{\mu}, p + \mu \hbar k | \tilde{\sigma} | j_{\nu}, p + \nu \hbar k \rangle, \quad (139)$$

$$\Pi^{e}_{\mu}(p) = \langle e_{\mu}, p + \mu \hbar k | \tilde{\sigma} | e_{\mu}, p + \mu \hbar k \rangle, \qquad (140)$$

$$\Pi^{g}_{\mu}(p) = \langle g_{\mu}, p + \mu \hbar k | \tilde{\sigma} | g_{\mu}, p + \mu \hbar k \rangle, \qquad (141)$$

$$\tilde{\sigma}(g_{-1},g_1,p) = C_r(p) + iC_i(p),$$
 (142)

where  $\mu \neq \nu$  and i, j = g, e. For conciseness we have omitted the argument  $\xi$  indicating the dependence of these elements on the background field. As before we adiabatically eliminate the optical coherences in the equation for the excited- and ground-state density-matrix elements. We then find, for example, for the equation of motion for  $\Pi_{i}^{g}(p)$ ,

$$\begin{split} \dot{\Pi}_{1}^{g}(p) &= \frac{\Gamma}{2} (1 + \frac{8}{3}\xi) s_{0}(\xi) \Biggl\{ -\frac{7}{6} \Pi_{1}^{g}(p) - \frac{1}{6} C_{r}(p) \\ &+ \frac{\delta}{3\Gamma(1 + \frac{8}{3}\xi)} C_{i}(p) \Biggr\} \\ &+ \Gamma(1 + \xi) \Biggl\{ \frac{1}{6} \overline{\Pi_{0}^{e}(p + \hbar k)} + \frac{1}{2} \overline{\Pi_{1}^{e}(p)} \\ &+ \overline{\Pi_{2}^{e}(p - \hbar k)} \Biggr\} - \frac{5}{3} \xi \Gamma \Pi_{1}^{g}(p). \end{split}$$
(143)

The overlined quantities give rise to coupling between different momentum families by spontaneous emission, and absorption and emission of background photons. They are defined in a similar way as in [29] by

$$\overline{\Pi_{m}^{e}(p-q\hbar k)} = \int_{-\hbar k_{0}}^{\hbar k_{0}} dp' N_{q}(p') \Pi_{m}^{e}(p-q\hbar k+p').$$
(144)

The factors  $N_q(p')$  are the projections of the photon momentum  $p' = \hbar k \cos \theta$  on the z axis for the three possible polarizations  $q = \pm 1,0$  [29]

$$N_{\pm 1}(p') = \frac{3}{8\hbar k_0} \left[ 1 + \left(\frac{p'}{\hbar k_0}\right)^2 \right],$$

$$N_0(p') = \frac{3}{4\hbar k_0} \left[ 1 - \left(\frac{p'}{\hbar k_0}\right)^2 \right].$$
(145)

The equations of motion for the first and second moments of p are found by multiplying Eq. (143) and the respective equations for the other density-matrix elements by p and  $p^2$ , respectively, and averaging over p. Using Eq. (143) we find for  $\langle p^2 \Pi_i^g(p) \rangle$ ,

$$\langle p^{2}\dot{\Pi}_{1}^{g}(p)\rangle = \frac{\Gamma}{2} (1 + \frac{8}{3}\xi)s_{0}(\xi) \left\{ -\frac{7}{6} \langle p^{2}\Pi_{1}^{g}(p)\rangle - \frac{1}{6} \langle p^{2}C_{r}(p)\rangle + \frac{\delta}{3\Gamma(1 + \frac{8}{3}\xi)} \langle p^{2}C_{i}(p)\rangle \right\}$$

$$+ \Gamma(1 + \xi) \left\{ \frac{1}{6} \langle p^{2}\overline{\Pi}_{0}^{e}(p + \hbar k)\rangle + \frac{1}{2} \langle p^{2}\overline{\Pi_{1}^{e}(p)}\rangle + \langle p^{2}\overline{\Pi_{2}^{e}(p - \hbar k)}\rangle \right\} - \frac{5}{3}\xi\Gamma \langle p^{2}\Pi_{1}^{g}(p)\rangle.$$

$$(146)$$

The terms containing the coupling between the different momentum families can be simplified by using the formulas

$$\langle p^2 \overline{\Pi_m^i(p)} \rangle = \frac{\hbar^2 k_0^2}{5} \langle \Pi_m^i(p) \rangle + \langle p^2 \Pi_m^i(p) \rangle, \qquad (147)$$

$$\langle p^2 \overline{\Pi_m^i(p-\hbar k)} \rangle = \left( \hbar^2 k^2 + \frac{2\hbar^2 k_0^2}{5} \right) \langle \Pi_m^i(p) \rangle + 2\hbar k \langle p \Pi_m^i(p) \rangle + \langle p^2 \Pi_m^i(p) \rangle,$$
(148)

$$\langle p^2 \overline{\Pi_m^i(p-\hbar k)} \rangle = \left( \hbar^2 k^2 + \frac{2\hbar^2 k_0^2}{5} \right) \langle \Pi_m^i(p) \rangle - 2\hbar k \langle p \Pi_m^i(p) \rangle + \langle p^2 \Pi_m^i(p) \rangle, \tag{149}$$

with i=e,g and m=0,1,2. Similar relations apply for the excited- and ground-state coherences. We can now simplify Eq. (138) by substituting the equations of motion for the second moments of p. The idealized steady-state momentum distribution of laser-cooled atoms, which is a Gaussian, is independent of the direction of p and we therefore require the first and second moments to be invariant under inversion of the z axis. This symmetry immediately implies

$$\langle p \Pi_0^i \rangle = 0, \tag{150}$$

$$\langle pC_r \rangle = 0, \tag{151}$$

$$\langle p\Pi_m^i \rangle = -\langle p\Pi_{-m}^i \rangle \tag{152}$$

and after some cumbersome but straightforward algebra we find

$$\frac{d}{dt} \langle p^{2} \rangle = \Gamma \xi \{ k^{2} [\frac{7}{3} \langle \Pi_{1}^{g}(p) \rangle + \langle \Pi_{0}^{g}(p) \rangle ] - \frac{10}{3} k \langle \Pi_{1}^{g}(p) \rangle \} + \Gamma (1 + \xi) \{ k^{2} [\frac{1}{3} \langle \Pi_{0}^{e}(p) \rangle + \langle \Pi_{1}^{e}(p) \rangle + 2 \langle \Pi_{2}^{e}(p) \rangle ]$$

$$+ 2k [\langle p \Pi_{1}^{e}(p) \rangle + 2 \langle p \Pi_{2}^{e}(p) \rangle ] \} + \Gamma (1 + \xi) \frac{k_{0}^{2}}{5} [\frac{4}{3} \langle \Pi_{0}^{e}(p) \rangle + 3 \langle \Pi_{1}^{e}(p) \rangle + 4 \langle \Pi_{2}^{e}(p) \rangle ]$$

$$+ \Gamma \xi \frac{k_{0}^{2}}{15} [17 \langle \Pi_{1}^{g}(p) \rangle + 8 \langle \Pi_{0}^{g}(p) \rangle ].$$
(153)

Here we have deliberately kept apart terms proportional to k,  $k^2$ , and  $k_0^2$ . The terms proportional to k and  $k^2$  give the contribution of the fluctuations in the number of photons absorbed from each laser beam; the terms proportional to  $k_0^2$  give the contribution from spontaneous emission. This is a slight improvement over the method suggested in Ref. [15] to calculate the

two contributions to the momentum diffusion coefficient. We also note that  $d\langle p^2 \rangle/dt$  is independent of the second moments of p and it is sufficient to solve for the first moments and the momentum averaged populations only.

For a stationary distribution of  $\langle p \rangle$  we can solve for the first moments in steady state. Multiplying Eq. (143) and the corresponding equations for the other density-matrix elements by p and taking the average we get, for example,

$$\langle p\dot{\Pi}_{1}^{g}(p)\rangle = \frac{\Gamma}{2} (1 + \frac{8}{3}\xi)s_{0}(\xi) \left\{ -\frac{7}{6} \langle p\Pi_{1}^{g}(p)\rangle - \frac{1}{6} \langle pC_{r}(p)\rangle + \frac{\delta}{3\Gamma(1 + \frac{8}{3}\xi)} \langle pC_{i}(p)\rangle \right\}$$
$$+ \Gamma(1 + \xi) \left\{ \frac{1}{6} \langle p\overline{\Pi_{0}^{e}(p + \hbar k)}\rangle + \frac{1}{2} \langle p\overline{\Pi_{1}^{e}(p)}\rangle + \langle p\overline{\Pi_{2}^{e}(p - \hbar k)}\rangle \right\} - \frac{5}{3}\xi \Gamma \langle p\Pi_{1}^{g}(p)\rangle.$$
(154)

The p averages containing overlined quantities again can be simplified using

$$\langle p \overline{\Pi}_{m}^{i}(p \pm \hbar k) \rangle = \langle \Pi_{m}^{i}(p) \rangle \mp \hbar k \langle \Pi_{m}^{i}(p) \rangle, \qquad (155)$$

$$\langle p \overline{\Pi_m^i(p)} \rangle = \langle \Pi_m^i(p) \rangle.$$
 (156)

The solution of the set of equations for the first moments of p is lengthy, but poses no particular problem. We obtain

$$\langle p\Pi_{2}^{e}(p)\rangle = \frac{\xi}{1+\xi} [\langle p\Pi_{1}^{g}(p)\rangle - k\langle \Pi_{1}^{g}(p)\rangle] + \frac{1+\frac{8}{3}\xi}{2(1+\xi)} s_{0}(\xi)\langle p\Pi_{1}^{g}(p)\rangle,$$
(157)

$$\langle p\Pi_1^e(p)\rangle = \frac{\xi}{2(1+\xi)} [\langle p\Pi_1^g(p)\rangle - k\langle \Pi_0^g(p)\rangle], \tag{158}$$

$$\langle pC_i(p) \rangle = \frac{-2 \, \delta s_0(\xi)}{\Gamma[13\xi + (1 + \frac{8}{3}\xi)s_0(\xi)]} \langle p\Pi_1^g(p) \rangle.$$
 (159)

 $\langle p \Pi_1^g(p) \rangle$  is equal to

$$\langle p \Pi_{1}^{g}(p) \rangle = \frac{\hbar k}{5\xi + (1 + \frac{8}{3}\xi)s_{0}(\xi)} \left/ \left\{ 1 + \frac{[2\,\delta\Gamma^{-1}s_{0}(\xi)]^{2}}{[13\xi + 5(1 + \frac{8}{3}\xi)s_{0}(\xi)][5\xi + (1 + \frac{8}{3}\xi)s_{0}(\xi)]} \right\} \times \{ (1 + \xi)[12\langle \Pi_{2}^{e}(p) \rangle - 2\langle \Pi_{0}^{e}(p) \rangle] - \xi[12\langle \Pi_{1}^{g}(p) \rangle + 3\langle \Pi_{0}^{g}(p) \rangle] \}.$$

$$(160)$$

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The *p*-averaged density-matrix elements are the solutions of the optical Bloch equations (118)–(120) for the case kv = 0. The excited-state elements can be found by substituting the steady-state results for the optical coherences into the solutions for the excited-state matrix elements. The two excited-state populations  $\langle \Pi_2^e(p) \rangle$  and  $\langle \Pi_2^e(p) \rangle$  that are needed in Eq. (160) are

$$\langle \Pi_2^e(p) \rangle = \left[ \frac{1 + \frac{8}{3}\xi}{2(1+\xi)} s_0(\xi) + \frac{\xi}{1+\xi} \right] \langle \Pi_1^e(p) \rangle, \quad (161)$$

$$\langle \Pi_{0}^{e}(p) \rangle = \frac{1}{6} \left[ \frac{1 + \frac{8}{3}\xi}{1 + \xi} s_{0}(\xi) + \frac{2\xi}{1 + \xi} \right] \langle \Pi_{1}^{g}(p) \rangle + \frac{1}{6} \frac{1 + \frac{8}{3}\xi}{1 + \xi} s_{0}(\xi)$$
$$\times \langle C_{r}(p) \rangle + \frac{2\xi}{3(1 + \xi)} \langle \Pi_{0}^{g}(p) \rangle.$$
(162)

The ground-state elements are found by setting kv = 0 in Eqs. (129)–(133).

The laser-induced part of the momentum diffusion coefficient in terms of the ground-state matrix elements is equal to

$$D_{p}^{\text{ind}}(\xi) = \frac{\Gamma(\hbar k)^{2}}{2} \xi(\frac{2}{9} \langle \Pi_{0}^{g}(p) \rangle - \frac{7}{6} \langle \Pi_{1}^{g}(p) \rangle - \frac{1}{9} \langle C_{r}(p) \rangle) + \frac{\Gamma(\hbar k)^{2}}{2} [(1 + \frac{8}{3}\xi)s_{0}(\xi) + 2\xi](\frac{19}{18} \langle \Pi_{1}^{g}(p) \rangle + \frac{1}{4} \langle \Pi_{0}^{g}(p) \rangle + \frac{1}{18} \langle C_{r}(p) \rangle) + \hbar k \Gamma[(1 + \frac{8}{3}\xi)s_{0}(\xi) + 2\xi] \langle p \Pi_{1}^{g}(p) \rangle$$
(163)

and the contribution due to spontaneous emission is

$$D_{p}^{\rm sp}(\xi) = \frac{\Gamma}{2} \frac{(\hbar k_{0})^{2}}{5} \xi \left[\frac{43}{6} \langle \Pi_{1}^{g}(p) \rangle + \frac{32}{9} \langle \Pi_{0}^{g}(p) \rangle - \frac{4}{9} \langle C_{r}(p) \rangle \right] \\ + \left[ (1 + \frac{8}{3} \xi) s_{0}(\xi) + 2 \xi \right] \left[\frac{20}{9} \langle \Pi_{1}^{g}(p) \rangle + \frac{3}{4} \langle \Pi_{0}^{g}(p) \rangle \\ + \frac{2}{9} \langle C_{r}(p) \rangle \right].$$
(164)

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In the limit of no background field we retrieve the results obtained by Dalibard and Cohen-Tannoudji [15] for the induced and the spontaneous part of the momentum diffusion coefficient.

In the limit of  $\xi = 0$  the laser-induced diffusion coefficient has been discussed in detail in Refs. [15] and [31]. The laserinduced induced momentum diffusion is due to the fluctuation in the number of photons absorbed from each laser beam. For transitions with ground-state angular momenta  $J \ge 1$  this part of the diffusion coefficient is strongly peaked around  $\delta = 0$ . Castin and Mølmer [31] attribute this increase to a correlation between successive steps in the random walk of the atom in momentum space. This correlation is destroyed when processes are present that redistribute the population between different ground-state sublevels at a rate faster than the optical pumping rate. When no background field is present this redistribution happens through temporal oscillations at a frequency equal to the light shift between adjacent m states, which is of the order of  $\delta s_0$ . Hence these oscillations are of the order of the optical pumping rate when  $\delta \approx \Gamma$ . With a background field present there are additional processes that can destroy the correlations between successive steps in the random walk. Consider, for example, an atom in the state  $m_z = 1$  of a J = 1 ground state. The probability of absorbing a  $\sigma_+$  photon is six times larger than the probability to absorb a  $\sigma_{-}$  photon, which for  $\xi = 0$  gives rise to a strong correlation between successive steps in the random walk. In the case  $\xi \neq 0$ , excitation can occur not only by absorption of a laser photon but also by a background photon. This has two effects, both of which lead to a weakening of the correlation in successive steps of the random walk. The first one is an absorption-emission cycle, which puts the atom back into the initial state, and the second possibility is a redistribution process, which actually changes the initial state. Since the momentum transferred to the atoms by the background photons is of a random nature, these processes destroy the correlation between successive random walk steps and lead to a decrease of the laser-induced diffusion for small detunings. This is shown in Fig. 6, where we have plotted  $D_p^{\text{ind}}(\xi)$  as a function of detuning for several values of N. The Rabi frequency was  $\Omega = 0.1\overline{\Gamma}$  to ensure that the low saturation approximation was fulfilled even at zero detuning. In this case the maximum number of photons in the background field was  $\xi = 0.03$ , corresponding to  $N = 10^8$  and  $\delta = 0$ . The proportion of atoms in the excited state was less than 3.5% so that the low saturation approximation was well satisfied. We see that the laser-induced momentum diffusion is decreasing sharply as a function of the number of atoms.

Most experiments on magneto-optical traps and optical molasses are performed in the limit of large detuning  $|\delta| \ge \Gamma$  [14]. In this regime there is no enhancement of the laser-induced momentum diffusion coefficient. The value for  $\xi=0$  zero is then of the order of the value calculated for a  $J=0\rightarrow 1$  transition [31]. The presence of the background field then leads to an increase of the momentum diffusion coefficient. This is shown in Fig. 7. The parameters chosen were  $\Omega=0.5\Gamma$  and  $\delta=-6\Gamma$  for an estimated atomic density  $n=10^{11}$  cm<sup>-3</sup>. This increase is due to the extra fluctuations introduced into the evolution of the momentum distribution by the random nature of the background field. The relative increase of the laser-induced momentum diffusion coefficient



FIG. 6. Plot of the laser-induced momentum diffusion coefficient  $D_p^{\text{ind}}(\xi)$  as a function of  $\delta$  for different values of N in units of  $(\hbar k)^2 \Gamma$ . The solid line is the diffusion coefficient for N=0, the dashed line corresponds to  $N=10^6$ , the dotted line to  $N=10^7$ , and the dash-dotted line to  $N=10^8$ . The Rabi frequency in all cases was  $\Omega=0.1\Gamma$ .

with respect to  $N^{1/3}$  (dashed line) is smaller than the increase of the momentum diffusion coefficient due to spontaneous emission (dotted line). The change of  $D_p$  with respect to  $N^{1/3}$  is linear in the regime of experimental interest.

#### D. The equilibrium temperature

The equilibrium temperature as a function of  $\xi$  is given by the ratio of  $\alpha(\xi) = F(\xi)/kv$  and  $D_p(\xi) = D_p^{\text{ind}}(\xi) + D_p^{\text{sp}}(\xi)$ , but because of its complexity we do not give an analytical expression in this section. Instead we write the temperature as a function of  $\xi$  in the form



FIG. 7. Variation of the momentum diffusion coefficient for  $\sigma_+ - \sigma_-$  molasses as a function of  $N^{1/3}$  in units of  $(\hbar k)^2 \Gamma$ . The parameters chosen were  $\Omega = 0.5\Gamma$  and  $\delta = -6\Gamma$  at a density of atoms  $n = 10^{11}$  cm<sup>-3</sup>. The solid line gives the total momentum diffusion coefficient, the dashed line the laser-induced part  $D_p^{\text{ind}}(\xi)$ , and the dotted line represents  $D_p^{\text{sp}}(\xi)$ .



FIG. 8. Temperature for motion-induced orientation cooling as a function of  $N^{1/3}$ . The Rabi frequency was  $\Omega = 0.5\Gamma$ , the detuning  $\delta = -6\Gamma$ , and the constant atomic density in the trap was  $n = 10^{11}$  cm<sup>-3</sup>. The increase is almost linear in  $N^{1/3}$ .

$$k_B T(\xi) = k_B T_0 [1 + \Theta(\xi)], \qquad (165)$$

where  $k_B T_0$  represents the temperature for motion-induced orientation cooling in the absence of any background photons, given by [15]

$$k_B T_0 = \frac{\hbar \Omega^2}{|\delta|} \left[ \frac{29}{300} + \frac{254}{75} \frac{\Gamma^2/4}{\delta^2 + \Gamma^2/4} \right].$$
(166)

The function  $\Theta(\xi)$  gives the excess temperature due to the background field and varies with detuning as approximately  $1/\delta^2$ . The dependence of the temperature on detuning is therefore proportional to approximately  $\delta^{-3}$  for a fixed density (which is unphysical). Figure 8 shows the variation of T with respect to  $N^{1/3}$ . We have normalized the graph with respect to  $k_B T_0$ . The parameters chosen were  $\delta = -6\Gamma$  and  $\Omega = -0.5\Gamma$  and the density in the trap was  $n = 10^{11}$  cm<sup>-3</sup>. We again see that for numbers of atoms in the range  $10^6 - 10^8$  the temperature increases almost linearly with  $N^{1/3}$ . For the parameters used here the interaction with the background field leads to a temperature increase by a factor of about 2.

### VIII. CONCLUSION

In this paper we have used projection operators to derive a master equation for the reduced density matrix of an atom that is driven by a laser and interacts with a gas of similar atoms through the exchange of scattered photons. This interaction is included into a collision operator that under the assumption of a zero mean of the fluctuating field produced by the medium is of second and higher order in the photon exchange interaction. In the BCA the collision operator is a superposition of two-body collisions. In the far-field limit this is justified because the presence of a third or more atoms becomes important only if the frequency shifts due to the perturbers significantly alter the evolution of the radiator in the laser field. The collision operator has a memory time that is of the order of the natural lifetime of the colliding atoms.

To obtain some analytical results we have taken the rateequation limit of the collision operator. We emphasize, however, that this is an approximation that is not strictly valid because of the similar time scales for the evolution of the perturbers and the radiator. Although taking this limit does affect the scaling of the collision strength with respect to the detuning and the Rabi frequency, it does not change the dependence on the density and the number of atoms. Keeping those restrictions in mind, we have shown that the Sisyphus and motion-induced orientation cooling mechanisms are very sensitive to the effects of a fluctuating unpolarized background field. In both cases we found that even a very small background field arising from photons scattered out of the laser beams by the cold atoms can lead to a significant rise in temperature. Therefore this effect is important even when the cloud is optically thin. The temperature increase predicted by our model is of the same order of magnitude as observed experimentally and the nearly linear dependence on  $N^{1/3}$  was verified. Within the present accuracy of the experimental data any nonlinearity would not be detectable. The dependence of the temperature rise on detuning can be described by a factor  $1/\delta^{-\beta}$ , where  $\beta$  is a positive number. In the present model  $\beta \approx 3$  for both polarization gradient cooling mechanisms when a constant density is assumed. Experimentally the density was found to increase slightly with increasing detuning (approximately  $\delta^{1/2}$ ), but even taking this into account the measured dependence of the temperature increase on detuning (approximately  $\delta^{-1.5}$  for onedimensional  $\sigma^+$ - $\sigma^-$  molasses and approximately  $\delta^{-1}$  for one-dimensional Sisyphus cooling) was much less than predicted [14]. Measurements in three-dimensional sub-Doppler molasses give a temperature increase proportional to  $\delta^{-1}$ . This disagreement is hardly surprising, bearing in mind the simplifications made at the onset of the calculations. However, by making those simplifications we were able to gain some physical insight into the effect of the background field on the two cooling mechanisms.

In the case of Sisyphus cooling the presence of the background field leads to a position-independent transition rate between the different ground-state sublevels. This transition rate causes the position dependence of the population in the ground-state sublevels to decrease. This position dependence is fundamental for the functioning of the cooling mechanism and the background field therefore destroys the cooling. In the case of motion-induced orientation cooling it is the alignment in the atomic ground state that is reduced by the background field. Although this reduction is small, it is sufficient to lower the efficiency of the cooling mechanism. An additional source of heating common to both cooling mechanisms is the extra heating because of the random direction of the momentum of the background photons.

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#### **APPENDIX: INTERATOMIC POTENTIALS**

In this appendix we briefly discuss the expressions for the atom-atom interaction that arise from the elimination of the radiation field from the full master equation. The interaction of a pair of atoms can in principle be calculated in the same way as the single-atom spontaneous decay term Eq. (12), as discussed in detail in Ref. [24]. An alternative approach, which starts from the equations of motion for Heisenberg

operators, can be found, for example, in [9]. We note, however, that the result derived therein is valid only in the special case of an operator that acts on a single atom. In the more general case we have to use operators that act on all particles simultaneously. The final step consists of converting the operator optical Bloch equations into equations of motion for the density-matrix elements. The part of the interaction that is due to photons scattered by atom j being reabsorbed by atom i is given by the expression

$$-\frac{i}{\hbar} \tilde{V}_{ij}^{(1)} \hat{\sigma}(t) = -\frac{i}{\hbar} \hat{\mathbf{d}}_{i} \cdot \hat{\mathbf{E}}^{+} (\hat{\mathbf{R}}_{i}, \hat{\mathbf{R}}_{j}) \hat{\sigma}(t) + \frac{i}{\hbar} \hat{\sigma}(t) \hat{\mathbf{d}}_{i} \cdot \hat{\mathbf{E}}^{-} (\hat{\mathbf{R}}_{i}, \hat{\mathbf{R}}_{j})$$

$$+ \left\{ \sum_{\lambda} \left[ \frac{\Gamma}{2} \int \frac{d\Omega}{8\pi/3} (\hat{\mathbf{d}}_{j}^{-} \cdot \boldsymbol{\epsilon}_{\lambda}) e^{-i\mathbf{k}_{0} \cdot \hat{\mathbf{R}}_{j}} \hat{\sigma}(t) e^{i\mathbf{k}_{0} \cdot \hat{\mathbf{R}}_{i}} (\hat{\mathbf{d}}_{i} \cdot \boldsymbol{\epsilon}_{\lambda}) \right.$$

$$+ i \left( -\frac{3\pi\Gamma}{2k_{0}^{3}} \right) \mathbf{P} \int \frac{d^{3}k}{(2\pi)^{3}} \left( \frac{k}{k-k_{0}} + \frac{k}{k+k_{0}} \right) (\hat{\mathbf{d}}_{j}^{-} \cdot \boldsymbol{\epsilon}_{\lambda}) e^{-i\mathbf{k} \cdot \hat{\mathbf{R}}_{j}} \hat{\sigma}(t) e^{i\mathbf{k} \cdot \hat{\mathbf{R}}_{i}} (\hat{\mathbf{d}}_{i} \cdot \boldsymbol{\epsilon}_{\lambda}) \right] + \mathrm{H.c.} \right\}, \qquad (A1)$$

where H.c. denotes the Hermitian conjugate of the first term in the curly brackets. The expression for the reverse process  $\tilde{V}_{ij}^{(2)}$ , the absorption of photons scattered by atom *i* by atom *j*, is found from Eq. (A1) by exchanging the indices *i* and *j*.  $\hat{\mathbf{E}}^+(\hat{\mathbf{R}}_i, \hat{\mathbf{R}}_j)$  and  $\hat{\mathbf{E}}^-(\hat{\mathbf{R}}_i, \hat{\mathbf{R}}_j)$  are the quantized field amplitudes for the photons scattered by atom *j*. They are given by the relation

$$\hat{\mathbf{E}}^{\pm}(\hat{\mathbf{R}}_{i},\hat{\mathbf{R}}_{j}) = \left( \mp i \frac{\hbar\Gamma}{2} \Phi(\hat{\mathbf{R}}_{a},\hat{\mathbf{R}}_{j}) + V(\hat{\mathbf{R}}_{i},\hat{\mathbf{R}}_{j}) \right) \hat{\mathbf{d}}_{j}^{\mp},$$
(A2)

where  $\Phi(\hat{\mathbf{R}}_i, \hat{\mathbf{R}}_j)$  and  $V(\hat{\mathbf{R}}_i, \hat{\mathbf{R}}_j)$  are defined as

$$\Phi(\hat{\mathbf{R}}_{i},\hat{\mathbf{R}}_{j}) \equiv \int \frac{d\Omega}{8\pi/3} (\mathbf{I} - \mathbf{n} \otimes \mathbf{n}) e^{ik_{0}\mathbf{n}\cdot\hat{\mathbf{R}}_{j}} e^{-ik_{0}\mathbf{n}\cdot\hat{\mathbf{R}}_{j}} \quad (A3)$$

and

$$V(\hat{\mathbf{R}}_{i},\hat{\mathbf{R}}_{j}) \equiv \left(-\frac{3\pi\hbar\Gamma}{2k_{0}^{3}}\right) \mathbf{P} \int \frac{d^{3}k}{(2\pi)^{3}} \left(\frac{k}{k-k_{0}} + \frac{k}{k+k_{0}}\right)$$
$$\times (\mathbf{I} - \mathbf{n} \otimes \mathbf{n}) e^{i\mathbf{k}\cdot\hat{\mathbf{R}}_{i}} e^{-i\mathbf{k}\cdot\hat{\mathbf{R}}_{j}}. \tag{A4}$$

**n** is the unit vector along the direction of the wave vector **k**. Making the substitutions  $\hat{\mathbf{R}}_i \rightarrow \hat{\mathbf{R}}_j$  and  $\hat{\mathbf{R}}_i \rightarrow \mathbf{r}_i$ , we find the semiclassical limit for Eq. (A2),

$$\hat{\mathbf{E}}_{j}^{\pm}(\mathbf{r}) = -\frac{3\hbar\Gamma}{4} \left\{ (\mathbf{I} - \mathbf{e}_{r} \otimes \mathbf{e}_{r}) \frac{1}{k_{0}|\mathbf{r}|} - (\mathbf{I} - 3\mathbf{e}_{r} \otimes \mathbf{e}_{r}) \left[ \frac{i}{k_{0}^{2}|\mathbf{r}|^{2}} + \frac{1}{k_{0}^{3}|\mathbf{r}|^{3}} \right] \right\}$$
$$\times \exp[\pm ik_{0}|\mathbf{r}|] \hat{\mathbf{d}}_{j}^{\pm}, \qquad (A5)$$

where  $\mathbf{r} = \mathbf{r}_i - \mathbf{r}_j$ . In the far-field limit we restrict ourselves to the lowest-order term in  $1/|\mathbf{r}|$ . When the expectation value for the reduced dipole operator  $\hat{\mathbf{d}}_j^{\pm}$  is taken we find that  $\langle \hat{\mathbf{E}}^{\pm}(\mathbf{r}) \rangle$  resembles exactly the field produced by a classical oscillating dipole [33].  $\hat{\mathbf{E}}^+(\hat{\mathbf{R}}_i, \hat{\mathbf{R}}_j)$  corresponds to a spherical wave moving away from its scattering center, atom *j*, and  $\hat{\mathbf{E}}^-(\hat{\mathbf{R}}_i, \hat{\mathbf{R}}_j)$  is the corresponding incoming wave. In the case where we can factorize the density matrix  $\hat{\sigma}(t)$  into a part  $\hat{\sigma}_j(t)$ , which commutes with all operators referring to atom *i*, and another part  $\hat{\sigma}_i(t)$ , which commutes with all operators referring to atom *j*, we can rewrite  $\tilde{V}_{ij}^{(1)}$  in the simpler form

$$\tilde{V}_{ij}^{(1)} = [\hat{\mathbf{d}}_i \cdot \hat{\mathbf{E}}^+ (\hat{\mathbf{R}}_i, \hat{\mathbf{R}}_j) \hat{\sigma}_j(t), \hat{\sigma}_i(t)] \\ + [\hat{\sigma}_j(t) \hat{\mathbf{d}}_i \cdot \hat{\mathbf{E}}^- (\hat{\mathbf{R}}_i, \hat{\mathbf{R}}_j), \hat{\sigma}_i(t)]$$
(A6)

and a corresponding term for the case where atom i acts as the source of the photons. We will return to the issue of factorization when we separate the atomic system into a radiator and a bath of statistically independent perturbers, which acts as the source of the background field.

In the more general case when a factorization is not possible,  $\tilde{V}_{ij} = \tilde{V}_{ij}^{(1)} + \tilde{V}_{ij}^{(2)}$  can be written in the form

$$-\frac{i}{\hbar}\tilde{V}_{ij}\hat{\sigma}(t) = -\frac{\Gamma}{2}\{[\hat{\sigma}(t),\hat{\mathbf{d}}_{i}^{+}\cdot\Phi(\hat{\mathbf{R}}_{i},\hat{\mathbf{R}}_{j})\cdot\hat{\mathbf{d}}_{j}^{-}]_{+}+[\hat{\sigma}(t),\hat{\mathbf{d}}_{j}^{+}\cdot\Phi(\hat{\mathbf{R}}_{i},\hat{\mathbf{R}}_{j})\cdot\hat{\mathbf{d}}_{i}^{-}]_{+}\}$$

$$+\frac{i}{\hbar}\{[\hat{\sigma}(t),\hat{\mathbf{d}}_{i}^{+}\cdot V(\hat{\mathbf{R}}_{i},\hat{\mathbf{R}}_{j})\cdot\hat{\mathbf{d}}_{j}^{-}]+[\hat{\sigma}(t),\hat{\mathbf{d}}_{j}^{+}\cdot V(\hat{\mathbf{R}}_{i},\hat{\mathbf{R}}_{j})\cdot\hat{\mathbf{d}}_{i}^{-}]\}$$

$$+\Gamma\sum_{\lambda}\int\frac{d\Omega}{8\pi/3}\{(\hat{\mathbf{d}}_{i}^{-}\cdot\boldsymbol{\epsilon}_{\lambda})e^{-i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{i}}\hat{\sigma}(t)e^{i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{j}}(\hat{\mathbf{d}}_{j}^{+}\cdot\boldsymbol{\epsilon}_{\lambda})+(\hat{\mathbf{d}}_{j}^{-}\cdot\boldsymbol{\epsilon}_{\lambda})e^{-i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{j}}\hat{\sigma}(t)e^{i\mathbf{k}_{0}\cdot\hat{\mathbf{R}}_{i}}(\hat{\mathbf{d}}_{i}^{+}\cdot\boldsymbol{\epsilon}_{\lambda})\},\quad(A7)$$

where  $[\hat{\sigma}(t)]_+$  denotes the anticommutator. The rotating-wave approximation has been made in this equation.

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