Heating due to long-range photon exchange interactions between cold atoms

G. Hillenbrand, C.J. Foot, and K. Burnett
Clarendon Laboratory, University of Oxford, Parks Road, Oxford OX1 3PU, United Kingdom
(Received 8 February 1994)

We have calculated the long-range part of the momentum diffusion coefficient of two atoms in a radiation field using an S-matrix expansion to first order in the dipole-dipole interaction between the atoms. This perturbative treatment is limited to the low-saturation regime and large detunings ($|\delta| \gg \gamma$). The physical processes arise from the interaction with the coherent and incoherent parts of the scattered spectrum.We find that in the regime of a constant density the extra diffusion term when averaged over the trap volume depends on the total number of trapped atoms to the third power and is independent of the atom laser detuning.

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

I. INTRODUCTION

Measurements by Clairon *et al.* [1] and more recently in this laboratory by Foot *et al.* [2] show that the temperature of Cs atoms in a magneto-optical trap depends on the total number of atoms in the trap, N. For numbers of atoms in the range of $N \sim 10^6$ to $N \sim 10^8$ the temperature varies as the number of trapped atoms to the one-third power.

In this paper we present a model which shows that the extra diffusion caused by the reabsorption of scattered light can give rise to extra heating which scales as number of atoms to the one-third and is independent of the detuning. In a dense cloud of cold atoms the scattered photons from one atom can be reabsorbed by a second atom nearby producing a repulsive force between the two atoms. There is also an attractive force due to the attenuation of the laser beam as it passes through the cloud of atoms [3]. The forces due to these processes have first been discussed by Sesko *et al.* [4] under the assumption of an optically thin medium, i.e., the assumption that a photon is not scattered more than twice on its way through the cloud. In equilibrium the trapping force is then balanced by the repulsive force, leading to a regime of constant density [4,5]. Hence the number of atoms N, the density n, and the trap radius L are related by $L \sim (N/n)^{1/3}$. This approximation breaks down when the cloud is no longer optically thin. Lindquist *et al.* [6] have shown that for larger numbers of atoms the trap radius will go more like the square root of the number of atoms in the cloud. Their data are at rather low detunings. We expect that the regime where the radius depends on the third power of N increases when larger detunings are used.

We show in this paper that these scattering processes also give rise to an extra term in the momentum diffusion coefficient. In our treatment we use an S-matrix expansion in terms of $1/kr$ and take the average separation of the atoms to be much larger than the laser wavelength. We restrict ourselves to the limit of small saturation, i.e., the Rabi frequency Ω is small and the laser detuning $\delta = \omega_L - \omega_0$ is large compared to the natural linewidth γ. In the far-field limit the extra contribution to the diffusion is proportional to the optical thickness of the cloud, i.e.,

$$n\sigma L \sim n^{2/3}\sigma N^{1/3}. \tag{1}$$

Here σ is the scattering cross section for the absorption of light at the laser frequency. If the separation of the atoms becomes comparable to the order of the wavelength higher orders in $1/kr$ have to be taken into account and the dependence on L becomes more complicated. The results presented in this article show qualitatively that as long as the separation of the atoms in the cloud can be considered large compared to the laser wavelength the photon exchange interactions give an excess heating of the cloud which has a functional dependence on the number of trapped atoms which is in agreement with experimental observation. As we restrict ourselves to a two-state atom it is clear that the theory cannot account for effects which the scattered radiation has on the sub-Doppler cooling mechanisms [7] which have been shown to be important in the trap [8]. Nevertheless we believe the formalism and results we describe form a proper basis to examine the more general case.

The paper is organized as follows. In Sec. II A, we describe the mathematical method and the approximations necessary to calculate the momentum diffusion coefficient from an S-matrix expansion. We then calculate the one-atom diffusion term to set up the formalism in Sec. II B. In Sec. II C, we perturbatively calculate the contribution of the photon exchange of the pair of atoms to the diffusion coefficient and finally in Sec. II D we obtain a result for the trap averaged magnitude of this extra term.

II. CALCULATION OF THE DIFFUSION COEFFICIENT USING AN S-MATRIX EXPANSION

A. Mathematical method

In this section we will set up the mathematical formalism for the calculation of the momentum diffusion coefficient in terms of the S matrix. The starting point is the distribution function $f(\vec{p},t)$ of $\vec{p}$ at time t. This probability function can be obtained from the density matrix by adiabatically eliminating the internal fast variables [9,10] and we can then derive a Fokker-Planck equation for $f(\vec{p},t)$. The conditions this imposes on the validity of our treatment are first that the width of the wave packet in momentum space is large compared to a photon momentum $\hbar k$ and secondly that $kv/\Gamma \ll 1$. The first restriction is equivalent to assuming that the atomic wave packet is well localized within a laser wavelength. $W(\vec{p},\Delta\vec{p})d^3(\Delta\vec{p})$ is the probability that an atom initially at momentum $\vec{p}$ after a time τ has a momentum in the interval $\vec{p}+\Delta\vec{p},\ \vec{p}+\Delta\ \vec{p}+d(\Delta\vec{p})$. The momentum distribution after time $t+\tau$ is then given by

$$f(\vec{p},t+\tau)=\int d^3(\Delta\vec{p})f(\vec{p}-\Delta\vec{p},t)W(\vec{p}-\Delta\vec{p},\Delta\vec{p}). \quad (2)$$

We take the time τ to be small enough so that only momentum changes $\Delta\vec{p}$ with $|\Delta\vec{p}| \ll |\vec{p}|$ can occur. This condition will generally be met for $\Delta\vec{p}$ of the order of $\hbar\vec{k}$ as this procedure corresponds to the usual expansion of the optical Bloch equations up to second order in $\hbar k$. From the preceding discussions it becomes clear that the concept of a diffusion coefficient which is part of a Fokker-Planck equation arises from taking the semiclassical limit. We expand $f(\vec{p}-\Delta\vec{p},t)$ and $W(\vec{p}-\Delta\vec{p},\Delta\vec{p})$ to second order in $\Delta\vec{p}$ and keep only the terms up to second order in the expansion parameter to obtain a Fokker-Planck equation for $f(\vec{p},t)$,

$$\frac{\partial f(\vec{p},t)}{\partial t}=-\vec{\nabla}_p\cdot\{\vec{F}(\vec{p})f(\vec{p},t)\}+\sum_{i,j}\frac{\partial^2}{\partial p_i\partial p_j}\{D_{ij}(\vec{p})f(\vec{p},t)\}. \quad (3)$$

The force $\vec{F}(\vec{p})$ and the elements of the momentum diffusion tensor $D_{ij}(\vec{p})$ are defined as

$$\vec{F}(\vec{p})=\int d^3(\Delta\vec{p})\Delta\vec{p}R(\vec{p},\Delta\vec{p}) \quad (4)$$

and

$$D_{ij}(\vec{p})=\frac{1}{2}\int d^3(\Delta\vec{p})\Delta p_i\Delta p_j R(\vec{p},\Delta\vec{p}). \quad (5)$$

$R(\vec{p},\Delta\vec{p})$ is the transition rate for an atom initially in momentum state $\vec{p}$ to experience a momentum change $\Delta\vec{p}$

$$R(\vec{p},\Delta\vec{p})=\frac{W(\vec{p},\Delta\vec{p})}{\tau}. \quad (6)$$

$W(\vec{p},\Delta\vec{p})$ is equal to the modulus squared of the S-matrix element $\langle\vec{p}+\Delta\vec{p}\mid\hat{S}\mid\vec{p}\rangle$. For isotropic momentum distributions the off diagonal elements of the diffusion tensor are zero. We then have

$$D_p(\vec{p})=\sum_i D_{ii}(\vec{p}). \quad (7)$$

The total momentum diffusion coefficient is obtained by averaging $D_p(\vec{p})$ over the distribution of all incoming momenta, $\vec{p}$. Defining $P(\vec{p})$ as the probability distribution for the incoming mean momenta in the cloud of cold atoms we obtain the diffusion coefficient in the form

$$D_p=\frac{1}{2}\int d^3(\Delta\vec{p})\int d^3\vec{p}R(\vec{p},\Delta\vec{p})P(\vec{p})(\Delta\vec{p})^2. \quad (8)$$

We note that this formula is equivalent to the definition of the momentum diffusion coefficient as the rate of change of the expectation value of the square of the momentum change

$$D_p=\frac{1}{2}\frac{d}{dt}\langle(\Delta\vec{p})^2\rangle. \quad (9)$$

The S-matrix transition element for the probability of atom A to experience a momentum change of $\Delta\vec{p}$ is given by the sum over all possible interactions which will shift the momentum distribution by the appropriate amount. The first processes are those where the atom interacts only with the laser field, i.e., the absorption of laser photons followed by a stimulated emission of laser photons or a spontaneous emission of fluorescence photons. The net momentum change of such a process is equal to the difference between the total momentum absorbed from the laser field and the sum over the momenta carried away by the fluorescence photons $\Delta\vec{p}=\sum\vec{k}_L-\sum\vec{k}_{\rm fl}$. The second type of process involves the reabsorption of photons scattered from atom B by atom A. The momentum change of atom A due to these processes is given by $\Delta\vec{p}=\sum k_{\rm scat}\vec{e}_r-\sum\vec{k}_{\rm fl}$ where $\vec{e}_r$ is the unit vector point from atom B to atom A and $k_{\rm scat}$ is the magnitude of the k vector of the scattered photon. Figure 1 shows a

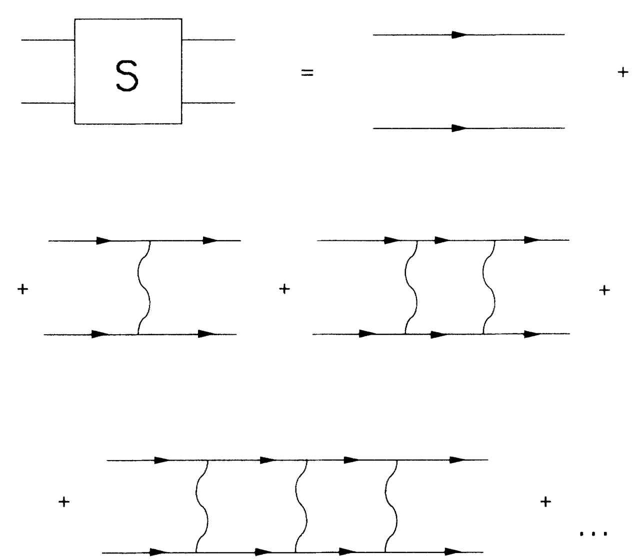

FIG. 1. Diagrammatic representation of the expansion of the S matrix for two particles: the single particle lines include the interaction of the single atom with the electromagnetic field. This includes processes like absorption followed by reemission of a laser photon or by emission of a fluorescent photon. The lines connecting the single particle propagators represent the exchange of scattered photons.

diagrammatic expansion of the S matrix. The important question now is how many of the photon exchange diagrams and the diagrams representing the scattering of photons we have to take into account. Swain [11] and Ballagh and Cooper [12] have shown that in order to obtain the correct spectrum for the scattered light it is necessary to sum over the interaction of the atom with the field to all orders to get the correct width and intensity of the Mollow triplet [13]. Hence for an accurate calculation of the pair diffusion coefficient it would be necessary to take all the diagrams represented in Fig. 1. Taking into account only a single absorption and reemission cycle gives the one photon spectrum with the correct width and line strength within 30%. To establish the number of photon exchanges we need to consider the time scale of the processes involved. The time scale for the scattering of photons is given by the optical pumping rate, which is $\sim \Gamma s_0$, where s_0 is the saturation parameter given by

$$s_0 = \frac{\Omega^2/2}{\delta^2 + \gamma^2} \tag{10}$$

and $\Gamma = 2\gamma$ is the natural linewidth of the transition. The time scale for the photon exchange between the pair of atoms is therefore given by the optical pumping time times the probability to find the photon in the direction of $\vec{r}$ which is approximately given by $\Gamma s_0 (1/kr)^2$.

There are two time scales for the fluctuations of the force between the atoms. The first one is of the order of the natural linewidth corresponding to the exchange of photons which belong to the incoherent part of the scattered spectrum. The second arises from the exchange of coherently scattered photons, i.e., the interaction of the mean dipoles. The correlation time for the latter processes is of the order of the time an atom needs to move a distance of the order of the laser wavelength. For cold atoms this time is comparatively long compared to the time for the exchange of a photon between the atoms and becomes of the order of the time for one photon exchange if the separation of the atoms is of the order of $25\lambda_L$. The amplitude of the nth order photon exchange diagram is proportional to $(1/kr)^n$. This means that the terms contributing to the two-atom part of the momentum diffusion coefficient are at least of the order of $(1/kr)^3$. It is therefore consistent to neglect those terms in the far-field approximation as their contribution is small compared to the lower-order and single-atom terms. To obtain an approximate result of the diffusion coefficient of the pair of atoms it is therefore consistent to take only the lowest-order diagram for the photon exchange and the lowest-order diagrams which give rise to the elastic and inelastic parts of the scattered spectrum.

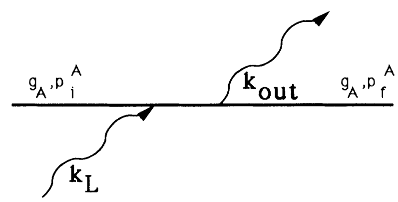

FIG. 2. Lowest-order diagram for the single-atom diffusion coefficient of a ground-state atom. The absorption of a laser photon is followed by emission of a photon with wave vector $\vec{k}_{\text{out}}$.

B. The single-atom diffusion term

To begin with we calculate the single-atom momentum diffusion coefficient to set up the formalism for the calculation of the two-atom contribution to the momentum diffusion coefficient. This result is compared with the one obtained by Gordon and Ashkin [14] for a two-level atom. Perturbative methods for the calculation of diffusion in the low-saturation limit have, for example, been discussed by Cohen-Tannoudji [15].

The momentum diffusion of a single atom in the case of a plane wave arises from the absorption of a laser photon followed by the emission of a photon in a random direction. The basic diagram for this process is given in Fig. 2. Hence the single-atom diffusion rate is easily calculated as follows. Let $\hat{V}_{A\lambda}(\vec{k})$ be the interaction of atom A with a photon with polarization λ and wave vector $\vec{k}$ defined by

$$\hat{V}_{A\lambda}(\vec{k}) = -i\hbar g(k)(\vec{d}_A \cdot \vec{\epsilon}_\lambda) \times [\hat{a}_{\lambda,k} e^{i\vec{k}\cdot\vec{r}_A} - \hat{a}^\dagger_{\lambda,k} e^{-i\vec{k}\cdot\vec{r}_A}], \tag{11}$$

with

$$g(k) \equiv |D| \sqrt{\frac{2\pi\omega_k}{\hbar V}} \tag{12}$$

and

$$g(k_L) \equiv |D| \sqrt{\frac{2\pi n_L \omega_{k_L}}{\hbar V}}. \tag{13}$$

We assume that the laser is sufficiently weak that the system is in the atomic ground state. The initial state of the atom is $|\Psi_A, g\rangle$ where $|\Psi_A\rangle$ describes the external degrees of freedom, i.e., $\langle \vec{p} \mid \Psi_A \rangle$ describes the atomic wave packet in momentum space. The S-matrix element for the scattering from a state with momentum $\vec{p}_i^A$ and n_L laser photons $|\vec{p}_i^A, g\rangle\, |n_L\rangle$ to a state $|\vec{p}_i^A + (\vec{k}_L - \vec{k}_{\text{out}}), g\rangle |n_L - 1, 1_{\lambda, \vec{k}_{\text{out}}}\rangle$ with final momentum $\vec{p}_i^A + \hbar(\vec{k}_L - \vec{k}_{\text{out}})$ and $n_L - 1$ laser photons and a single photon in the mode with wave vector $\vec{k}_{\text{out}}$ and polarization λ is to lowest order given by

$$\langle \vec{p}_f^A, g | \langle n_L - 1, 1_{\lambda,\vec{k}} | \hat{S} | n_L \rangle | g, \vec{p}_i^A \rangle = (-2\pi i)\delta(E_{\text{in}} - E_{\text{out}}) \langle \vec{p}_f^A, g | \langle n_L - 1, 1_{\lambda,\vec{k}} | \hat{V}_{A\lambda}(\vec{k}_{\text{out}}) \hat{G}_{A0}(E_{\text{in}}) \hat{V}_{A\lambda_L}(\vec{k}_L) | g, \vec{p}_i^A \rangle | n_L \rangle. \tag{14}$$

Here $\hat{G}_{A0}(E)$ is the resolvent operator given by

$$\hat{G}_{A0}(E) \equiv \frac{1}{E - \hat{H}_{A0} - \hat{\Sigma}}, \tag{15}$$

where $\hat{H}_{A0}$ is the free Hamiltonian of the atom. The diagonal operator $\hat{\Sigma}$ is the self-energy correction to the free propagator which is given by the familiar expression [16]

$$\begin{aligned}\Sigma(e,e) &= -i\hbar\gamma + \hbar\gamma\, P\int d\left(\frac{\omega_k}{\omega_0}\right)\left(\frac{\omega_k}{\omega_0}\right)^3 \frac{1}{1-\left(\frac{\omega_k}{\omega_0}\right)^2},\\ \Sigma(g,g) &= -\hbar\gamma\, P\int d\left(\frac{\omega_k}{\omega_0}\right)\left(\frac{\omega_k}{\omega_0}\right)^3 \frac{1}{1+\left(\frac{\omega_k}{\omega_0}\right)^2}.\end{aligned} \tag{16}$$

In the following calculation we include the Lamb-shift terms arising from the principal parts, which are formally divergent until a proper renormalization is performed, into the transition frequency ω_0. The scattering amplitude is given by the matrix element for the change of momentum from $\vec{p}_i^A$ to the momentum $\vec{p}_f^A$ integrated over all incoming momenta.

$$\langle \vec{p}_f^A, g|\hat{S}|g,\Psi_A\rangle = \int \frac{d^3p_i^A}{(2\pi)^3}\langle \vec{p}_f^A, g|\langle n_L-1, 1_{\lambda,\vec{k}_{\rm out}}|\hat{S}|n_L\rangle|g,\vec{p}_i^A\rangle\langle \vec{p}_i^A|\Psi_A\rangle. \tag{17}$$

Evaluation of the matrix element in formula (14) yields

$$\begin{aligned}&\langle \vec{p}_f^A, g|\langle n_L-1, 1_{\lambda,\vec{k}}|\hat{V}_{A\lambda}(\vec{k}_{\rm out})\hat{G}_{A0}(E_{\rm in})\hat{V}_{A\lambda_L}(\vec{k}_L)|n_L\rangle|g,\vec{p}_i^A\rangle\\ &\quad= \hbar g(k_{\rm out})(\vec{d}_A\cdot\vec{\epsilon}_\lambda)\frac{1}{\left(\omega_L-\omega_0-\vec{v}_i^A\cdot\vec{k}_L-\frac{\hbar k_L^2}{2m}\right)+i\gamma}g(k_L)(\vec{d}_A\cdot\vec{\epsilon}_{\lambda_L})(2\pi)^3\delta^3(\vec{p}_f^A-(\vec{p}_i^A+\vec{k}_L-\vec{k}_{\rm out})).\end{aligned} \tag{18}$$

Finally substituting Eq. (18) into (17) we find for the scattering amplitude

$$\begin{aligned}\langle \vec{p}_f^A, g\mid \hat{S}\mid g,\Psi_A\rangle &= (-2\pi i)\delta(E_{\rm in}-E_{\rm out})\hbar g(k_{\rm out})(\vec{d}_A\cdot\vec{\epsilon}_\lambda)\frac{1}{\left(\omega_L-\omega_0-\vec{v}_i^A\cdot\vec{k}_L-\frac{\hbar k_L^2}{2m}\right)+i\gamma}\\ &\quad\times g(k_L)(\vec{d}_A\cdot\vec{\epsilon}_{\lambda_L})\Psi_A(\vec{p}_f^A-(\vec{k}_L-\vec{k}_{\rm out})).\end{aligned} \tag{19}$$

We now neglect the Doppler and recoil term in the intermediate propagator. This is justified by the fact that for a gas of cold atoms we have very low velocities $|\vec{k}\vec{v}| \ll |\delta|, \delta = \omega_L - \omega_0$. The probability for the atom to undergo a transition into a final momentum state $\vec{p}_f^A$ by absorption of a laser photon followed by the emission of a photon with wave vector $\vec{k}_{\rm out}$ is given by the square of the matrix element in Eq. (19). We then obtain

$$\begin{aligned}dW(\vec{p}_f^A,(\vec{k}_L-\vec{k}_{\rm out})) &= 2\pi\hbar\tau\delta(E_{\rm in}-E_{\rm out})g(k_{\rm out})^2|(\vec{d}_A\cdot\vec{\epsilon}_\lambda)|^2 g(k_L)^2|(\vec{d}_A\cdot\vec{\epsilon}_{\lambda_L})|^2\frac{1}{\delta^2+\gamma^2}\\ &\quad\times|\Psi_A(\vec{p}_f^A-(\vec{k}_L-\vec{k}_{\rm out}))|^2\frac{d^3p_f^A}{(2\pi)^3}\left(V\frac{d^3k_{\rm out}}{(2\pi)^3}\right).\end{aligned} \tag{20}$$

$dW(\vec{p}_f^A,(\vec{k}_L-\vec{k}_{\rm out}))$ is the probability to scatter from an incoming state $\vec{p}_f^A-(\vec{k}_L-\vec{k}_{\rm out})$ into the final state $\vec{p}_f^A$. Note that Eq. (20) already includes a weighting by the probability distribution of the incoming momenta which is given by the factor $|\Psi_A(\vec{p}_f^A-(\vec{k}_L-\vec{k}_{\rm out}))|^2$. The total probability $W(\vec{k}_L-\vec{k}_{\rm out})$ to have momentum change $(\vec{k}_L-\vec{k}_{\rm out})$ is given by integrating Eq. (20) over $d^3p_f^A$ and summing over polarization of the outgoing photon. Using the relations

$$\int \frac{d^3p_f^A}{(2\pi)^3}|\Psi_A(\vec{p}_f^A-(\vec{k}_L-\vec{k}_{\rm out}))|^2 = 1 \tag{21}$$

and

$$\sum_\lambda |(\vec{d}\cdot\vec{\epsilon}_\lambda)|^2 = \vec{d}\cdot(\mathbf{I}-\vec{n}_{k_{\rm out}}\otimes\vec{n}_{k_{\rm out}})\cdot\vec{d} \tag{22}$$

the rate of change of momentum by $\vec{k}_L-\vec{k}_{\rm out}$ is given by

$$R(\vec{k}_l - \vec{k}_{\rm out}) = 2\pi\hbar\delta(E_{\rm in} - E_{\rm out})[\vec{d}_A \cdot (\mathbf{I} - \vec{n}_{k_{\rm out}} \otimes \vec{n}_{k_{\rm out}}) \cdot \vec{d}_A] g(k_{out})^2 |(\vec{d}_A \cdot \vec{\epsilon}_{\lambda_L})|^2 g(k_L)^2 \frac{1}{\delta^2 + \gamma^2} \left(V \frac{d^3 k_{\rm out}}{(2\pi)^3} \right). \quad (23)$$

The Rabi frequency Ω is defined as

$$\Omega = -2\frac{|D| E_{\rm rms}}{\hbar} = -\sqrt{2} g(k_L). \quad (24)$$

We now neglect the contribution of the change in kinetic energy in the δ function for the energy, i.e., $E_{\rm in} - E_{\rm out} \simeq \hbar(\omega_L - \omega_{\rm out})$. Equation (23) can be rewritten as

$$R(\vec{k}_L - \vec{k}_{\rm out}) = 2\pi\delta(\omega_L - \omega_{\rm out})[\vec{d}_A \cdot (\mathbf{I} - \vec{n}_{k_{\rm out}} \otimes \vec{n}_{k_{\rm out}}) \cdot \vec{d}_A] g(k_{\rm out})^2 |(\vec{d}_A \cdot \vec{\epsilon}_{\lambda_L})|^2 \frac{\Omega^2/2}{\delta^2 + \gamma^2}. \quad (25)$$

By substituting Eq. (25) into Eq. (8) we find the one-atom momentum diffusion coefficient

$$D_p^{\text{1-at}} = \frac{1}{2} V \int \frac{d^3 k_{\rm out}}{(2\pi)^3} R(\vec{k}_L - \vec{k}_{\rm out}) \hbar^2 (k_L^2 + k_{\rm out}^2 - 2k_L k_{\rm out} \cos\theta)$$

$$= \frac{1}{2} |(\vec{d}_A \cdot \vec{\epsilon}_{\lambda_L})|^2 \frac{\Omega^2/2}{\delta^2 + \gamma^2} \gamma \left(\frac{\omega_L}{\omega_0}\right)^3 \int \frac{d\Omega_{\rm out}}{4\pi/3} [\vec{d}_A \cdot (\mathbf{I} - \vec{n}_{k_{\rm out}} \otimes \vec{n}_{k_{\rm out}}) \cdot \vec{d}_A] \hbar^2 (2k_L^2 - 2k_L^2 \cos\theta). \quad (26)$$

γ is half the natural lifetime of the transition defined as

$$\gamma = \frac{2|D|^2 \omega_0^3}{3\hbar c^3}. \quad (27)$$

The angular integral yields

$$\int \frac{d\Omega_{\rm out}}{4\pi} [\vec{d}_A \cdot (\mathbf{I} - \vec{n}_{k_{\rm out}} \otimes \vec{n}_{k_{\rm out}}) \cdot \vec{d}_A]$$

$$\times (2\hbar^2 k_L^2 - 2\hbar^2 k_L^2 \cos\theta) = \frac{4}{3}\hbar^2 k_L^2, \quad (28)$$

where we have used the fact that the contribution of the extra cos term does not contribute in the average and

$$\int d\Omega_{\rm out} [\vec{d} \cdot (\mathbf{I} - \vec{n}_{k_{\rm out}} \otimes \vec{n}_{k_{\rm out}}) \cdot \vec{d}] = \frac{8\pi}{3}. \quad (29)$$

The factor (ω_L/ω_0) is approximately equal to unity in the rotating wave approximation. Substituting Eq. (28) into (26) yields

$$D_p^{\text{1-at}} = \gamma |(\vec{d}_A \cdot \vec{\epsilon}_{\lambda_L})|^2 s_0 (\hbar k_L)^2 \quad (30)$$

for the single-atom diffusion coefficient. s_0 is the saturation parameter for the Rabi frequency at the position of the atom which in the case of a plane wave is position independent as long as shielding effects due to a surrounding medium are not taken into account. For an incident plane wave and low saturation this coincides exactly with the expression calculated by Gordon and Ashkin [14].

C. The two-atom diffusion diagrams

In this section we study the influence of the presence of a second atom at a distance $|\vec{r}|$ on the diffusion rate. The diagrams which we have to include to lowest order are shown in Figs. 3 and 4. Figure 3 gives the part which is due to the interaction with the elastically scattered component and Fig. 4 depicts the lowest-order contributions to the sidebands of the Mollow triplet. These parts also give rise to a δ function contribution for the elastically scattered light. But we do not include this in our calculations as this diagram would be reducible and could be resummed in a different way [11,17]. The particles are described by two Gaussian wave packets at a distance $|\vec{r}|$ apart with mean velocities $\vec{p}_{A0}$ and $\vec{p}_{B0}$, respectively. The wave packets are given in momentum space,

$$\Psi_A(\vec{p}_A) = \left(\frac{a}{\sqrt{2\pi}}\right)^{\frac{3}{2}} \exp\left[-\frac{a^2}{4}(\vec{p}_A - \vec{p}_{A0})^2\right],$$
$$\Psi_B(\vec{p}_B) = \left(\frac{a}{\sqrt{2\pi}}\right)^{\frac{3}{2}} \exp\left[-\frac{a^2}{4}(\vec{p}_B - \vec{p}_{B0})^2\right] \times \exp[-i(\vec{p}_B - \vec{p}_{B0}) \cdot \vec{r}]. \quad (31)$$

To calculate the extra diffusion we assume that the particles are well localized in position space within a wavelength, i.e., $a \ll \lambda_L$. The two particles are treated as

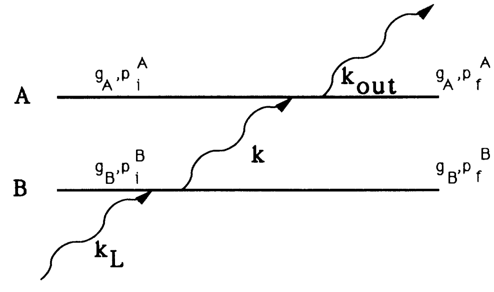

FIG. 3. Lowest-order diagram for the exchange of an elastically scattered photon. Atom B scatters a photon with wave vector $\vec{k}$ which is reabsorbed by atom A. Finally atom A reemits a fluorescent photon with wave vector $\vec{k}_{\rm out}$.

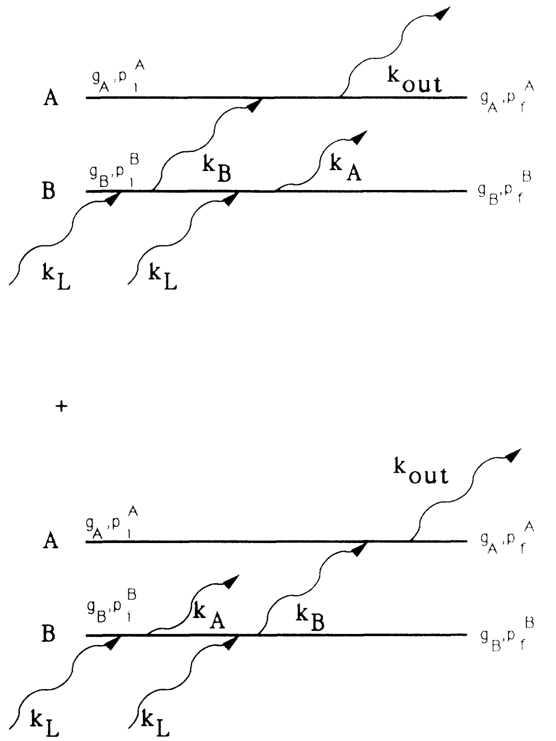

FIG. 4. Diagrams for the sidebands of the Mollow triplet. Atom B undergoes two absorption-emission cycles emitting two photons with different wave vectors k_A and k_B. One of them—here denoted by k_B—is reabsorbed by atom A and rescattered.

independent, that means we do not symmetrize our wave function and the initial state is described by $|\Psi_A, g\rangle \otimes |\Psi_B, g\rangle$. For spatial separations much larger than the spread of the wave packet the exchange integral due to the symmetrization gives a zero contribution. We regard the second atom as a bath, i.e., in order to get the total transition matrix element we take the average over the second particle. The S-matrix element for the extra diffusion on atom A due to the surrounding cloud of cold atoms is hence given by the relation

$$\langle \vec{p}_f^A, g|\langle \Psi_B, g|\hat{S}(\vec{r})|g, \Psi_B\rangle|g, \Psi_A\rangle = \int d^3p_f^B \langle \Psi_B|\vec{p}_f^B\rangle\langle \vec{p}_f^A, g|\langle \vec{p}_f^B, g|\hat{S}(\vec{r})|g, \Psi_B\rangle|g, \Psi_A\rangle. \tag{32}$$

Again we neglect the kinetic energy part in the intermediate propagators.

1. The S-matrix element for the elastic component of the scattered spectrum

The S-matrix element for the elastic component is of fourth order in the atom field interaction and is shown in Fig. 3. We have to sum over the intermediate photon to get the total contribution. Denoting the diagram for the exchange of a particular momentum $\vec{k}$ by $\hat{S}_{\vec{k}}(\vec{r})$ the total contribution of the photon exchange can be written as

$$\hat{S}(\vec{r}) = V \int \frac{d^3k}{(2\pi)^3} \hat{S}_{\vec{k}}(\vec{r}), \tag{33}$$

with

$$\hat{S}_{\vec{k}}(\vec{r}) \equiv \hat{V}_{B\lambda_L}(\vec{k}_L)\hat{G}_{B0}(E_{\rm in}) \sum_\lambda \hat{V}_{B\lambda}(\vec{k})\ \hat{D}_F(\omega_k)\hat{V}_{A\lambda}(\vec{k}) \times \hat{G}_{A0}(E_{\rm in})\hat{V}_{A\lambda_{\rm out}}(\vec{k}_{\rm out}) \times(-2\pi i)\delta(E_{\rm in} - E_{\rm out}). \tag{34}$$

This corresponds to atom B absorbing a laser photon after which it propagates freely until it emits a photon of wave vector $\vec{k}$. $\hat{D}_F$ is the free relativistic photon propagator for the electric field and we neglect the Doppler and recoil shift in the propagator as it is negligible compared to the laser detuning. After absorption of the intermediate photon by atom A the latter propagates freely until it emits a photon of wave vector $\vec{k}_{\rm out}$. The matrix element for the exchange of a photon of wave vector $\vec{k}$, $\langle \vec{p}_f^B, g|\langle \vec{p}_f^A, g|\hat{S}_{\vec{k}}(\vec{r})|g, \vec{p}_i^A\rangle|g, \vec{p}_i^B\rangle$, is proportional to the δ functions $(2\pi)^3\delta^3(\vec{p}_f^B - [\vec{p}_i^B - \hbar(\vec{k}_{\rm out} - \vec{k})])$ and $(2\pi)^3\delta^3(\vec{p}_f^B - [\vec{p}_i^B - \hbar(\vec{k} - \vec{k}_L)])$. The matrix element $\langle \vec{p}_f^A, g|\langle \Psi_B, g|\hat{S}(\vec{r})|g, \Psi_B\rangle|g, \Psi_A\rangle$ is therefore equal to

$$\langle \vec{p}_f^A, g|\langle \Psi_B, g|\hat{S}(\vec{r})|g, \Psi_B\rangle|g, \Psi_A\rangle = V\int \frac{d^3k}{(2\pi)^3}\int \frac{d^3p_f^B}{(2\pi)^3}\Psi_B^*(\vec{p}_f^B)\Psi_B(\vec{p}_f^B + \hbar(\vec{k} - \vec{k}_L)) \times\langle \vec{p}_f^A, g|\langle \vec{p}_f^B, g|\hat{S}_{\vec{k}}(\vec{r})|g, \vec{p}_i^B + \hbar(\vec{k} - \vec{k}_L)\rangle|g, \vec{p}_i^A + \hbar(\vec{k}_{\rm out} - \vec{k})\rangle \times\Psi_A(\vec{p}_f^A + \hbar(\vec{k}_{\rm out} - \vec{k})). \tag{35}$$

The matrix element $\langle \vec{p}_f^B, g|\langle \vec{p}_f^A, g|\hat{S}_{\vec{k}}(\vec{r})|g, \vec{p}_f^B + \hbar(\vec{k} - \vec{k}_L)\rangle|g, \vec{p}_f^A + \hbar(\vec{k}_{\rm out} - \vec{k})\rangle$ for the scattering between different momentum states is given by

$$\langle \vec{p}_f^B, g|\langle \vec{p}_f^A, g|\hat{S}_{\vec{k}}(\vec{r})|g, \vec{p}_f^B + \hbar(\vec{k} - \vec{k}_L)\rangle|g, \vec{p}_f^A + \hbar(\vec{k}_{\rm out} - \vec{k})\rangle = (2\pi)^6\hbar g(k_{\rm out})(\vec{d}_A\cdot\vec{\epsilon}_{\lambda_{\rm out}})\left(\frac{1}{\delta + i\gamma}\right)^2 g(k_L)(\vec{d}_B\cdot\vec{\epsilon}_{\lambda_L})g^2(k)[\vec{d}_B\cdot(\mathbf{I} - \vec{n}_k\otimes\vec{n}_k)\cdot\vec{d}_A] \times\left[\frac{1}{\omega_L - \omega_k + i\epsilon} - \frac{1}{\omega_L + \omega_k - i\epsilon}\right](-2\pi i)\delta(E_{\rm in} - E_{\rm out}). \tag{36}$$

It is worth noting that $\langle \vec{p}_f^A, g|\langle \vec{p}_f^B, g|\hat{S}_{\vec{k}}(\vec{r})|g, \vec{p}_f^B + \hbar(\vec{k}-\vec{k}_L)\rangle|g, \vec{p}_f^A + \hbar(\vec{k}_{\text{out}}-\vec{k})\rangle$ is independent of $\vec{p}_f^B$ and $\vec{p}_f^A$ in the low-velocity approximation. The assumption of good localization in position space means that

$$\frac{a}{2}\left(\vec{p}_f^B + \hbar(\vec{k}-\vec{k}_L) - \vec{p}_{B0}\right) = \frac{a}{2}\left(\vec{p}_f^B - \vec{p}_{B0}\right) + O\left(\frac{a}{\lambda}\right). \tag{37}$$

The terms of the order of $\frac{a}{\lambda}$ are small so that we can neglect them in the Gaussian. Hence using Eq. (31)

$$\int d^3p_f^B \Psi_B^*(\vec{p}_f^B)\Psi_B(\vec{p}_f^B + \hbar(\vec{k}-\vec{k}_L)) \approx \exp[-i(\vec{k}-\vec{k}_L)\cdot\vec{r}]. \tag{38}$$

The integration over the intermediate photon momentum is demonstrated in detail in the Appendix. In the far-field approximation we get the result

$$\begin{aligned}\langle \vec{p}_f^A, g|\langle \Psi_B, g|\hat{S}(\vec{r})|g, \Psi_B\rangle|g, \Psi_A\rangle = {}& \hbar g(k_{\text{out}})(\vec{d}_A\cdot\vec{\epsilon}_{\lambda_{\text{out}}})\left(\frac{1}{\delta+i\gamma}\right)^2 g(k_L)(\vec{d}_B\cdot\vec{\epsilon}_{\lambda_L})\frac{3}{2}\gamma\left(\frac{\omega_L}{\omega_0}\right)^3 \\ &\times[\vec{d}_A\cdot(\mathbf{I}-\vec{e}_r\otimes\vec{e}_r)\cdot\vec{d}_B]\frac{\exp[+ik_Lr]}{k_Lr} \\ &\times\Psi_A(\vec{p}_f^A + (\vec{k}_{\text{out}} - k_L\vec{e}_r))\exp[+i\vec{k}_L\cdot\vec{r}]2\pi i\delta(E_{\text{in}}-E_{\text{out}}).\end{aligned} \tag{39}$$

The net effect of the average over all intermediate photons is an extra momentum kick of magnitude k_L in the direction of the unit vector of $\vec{r}$. The change in momentum of atom A is equal to $\Delta\vec{p} = (\vec{k}_{\text{out}} - k_L\vec{e}_r)$. In the low-velocity approximation (39) is independent of $\vec{p}_f^A$ and is interpreted as the transition amplitude for a ground-state atom A with final velocity $\vec{p}_f^A$ to have experienced a momentum change $\Delta\vec{p}$ from scattering of a photon emitted by a second ground-state atom driven by a laser at a distance $|\vec{r}|$ apart. Taking the modulus squared and summing over all possible final momentum states gives the total probability to have a momentum change $\Delta\vec{p}$. The probability for this process can be written as

$$\begin{aligned}dW_{\lambda_{\text{out}}}(\vec{r},\vec{\Delta p}) = {}& \left|\hbar g(k_{\text{out}})(\vec{d}_A\cdot\vec{\epsilon}_{\lambda_{\text{out}}})\left(\frac{1}{\delta+i\gamma}\right)^2 g(k_L)(\vec{d}_B\cdot\vec{\epsilon}_{\lambda_L})\right|^2 \\ &\times\left|\frac{3}{2}\gamma\left(\frac{\omega_L}{\omega_0}\right)^3[\vec{d}_B\cdot(\mathbf{I}-\vec{e}_r\otimes\vec{e}_r)\cdot\vec{d}_A]\frac{\exp[+ik_Lr]}{k_Lr}\right|^2 \\ &\times\int d^3p_f^A|\Psi_A(\vec{p}_f^A-\Delta\vec{p})|^2 2\pi\frac{\tau}{\hbar}\delta(E_{\text{in}}-E_{\text{out}})\left(V\frac{d^3\vec{k}_{\text{out}}}{(2\pi)^3}\right).\end{aligned} \tag{40}$$

The index λ_{out} indicates that the polarization of the outgoing photon is fixed. Summing over λ_{out} and using Eqs. (21) and (22) the total probability is given by

$$\begin{aligned}dW(\vec{r},\vec{\Delta p}) = {}& \frac{\tau}{2\pi}\delta(k_L-k_{\text{out}})dk_{\text{out}}d\Omega_{\text{out}}[\vec{d}_A\cdot(\mathbf{I}-\vec{n}_{k_{\text{out}}}\otimes\vec{n}_{k_{\text{out}}})\cdot\vec{d}_A]\left[\left(\frac{\omega_L}{\hbar c}\right)|D|^2\frac{\gamma}{\delta^2+\gamma^2}\right] \\ &\times\frac{3}{4}\gamma|(\vec{d}_B\cdot\vec{\epsilon}_{\lambda_L})|^2\frac{\Omega^2/2}{\delta^2+\gamma^2}\times\left|\frac{[\vec{d}_A\cdot(\mathbf{I}-\vec{e}_r\otimes\vec{e}_r)\cdot\vec{d}_B]}{r}\right|^2.\end{aligned} \tag{41}$$

Again we have replaced factors of the order (ω_L/ω_0) by unity and the weighting by the probability distribution of the incoming momenta is already included.

The Rayleigh cross section $\sigma_{\text{Ray}}(\delta)$ for a two-level atom in the low-saturation approximation is given by

$$\sigma_{\text{Ray}}(\delta) = 4\pi|D|^2\frac{\omega_L}{\hbar c}\frac{\gamma}{\delta^2+\gamma^2} \tag{42}$$

and the transition rate is defined by $R(\vec{r},\Delta\vec{p}) = \frac{dW(\vec{r},\vec{\Delta p})}{\tau}$. Using Eq. (42), Eq. (41), and Eq. (30) for the single-atom diffusion coefficient gives the transition rate for the photon exchange interaction,

$$\begin{aligned}R(\vec{r},\vec{\Delta p}) = {}& dk_{\text{out}}d\Omega_{\text{out}}\delta(k_L-k_{\text{out}}) \\ &\times[\vec{d}_A\cdot(\mathbf{I}-\vec{n}_{k_{\text{out}}}\otimes\vec{n}_{k_{\text{out}}})\cdot\vec{d}_A] \\ &\times\frac{3^2}{2(8\pi)^2}\sigma_{\text{Ray}}(\delta)\frac{D_p^{1-\text{at}}}{(\hbar k_L)^2} \\ &\times\left|\frac{[\vec{d}_A\cdot(\mathbf{I}-\vec{e}_r\otimes\vec{e}_r)\cdot\vec{d}_B]}{r}\right|^2.\end{aligned} \tag{43}$$

Substitution of the transition rate in Eq. (43) into Eq. (8) gives the contribution of the elastically scattered light to the momentum diffusion coefficient which is equal to

$$D_p^{\mathrm{el}}(\vec{r}) = D_P^{1-\mathrm{at}} \frac{3}{8\pi} \sigma_{\mathrm{Ray}}(\delta) \left| \frac{[\vec{d}_A \cdot (\mathbf{I} - \vec{e}_r \otimes \vec{e}_r) \cdot \vec{d}_B]}{r} \right|^2 . \tag{44}$$

2. The S-matrix element for the inelastic component of the scattered spectrum

The diagrams for the sideband of the triplet are given in Fig. 4. For the evaluation of the contribution of the side bands to the two-atom momentum diffusion it is sufficient to consider only the resonance component of the scattered light where the reabsorbed photon $\vec{k}_B$ has the frequency $\omega_B = \omega_L - \delta$ and the second photon has the frequency $\omega_L + \delta$ which is far off resonance for large detunings δ. The contribution is of the same order as the contribution of the elastically scattered light due to the fact that the reabsorption process is proportional to the resonance cross section rather than the elastic cross section for large detuning. The two diagrams corresponding to the two possible ways of emitting the resonance photon are evaluated the same way as before and we find for the sum of the amplitudes

$$\begin{aligned} \langle \vec{p}_f^A, g | \langle \Psi_B, g | \hat{S}(\vec{r}) | g, \Psi_B \rangle | g, \Psi_A \rangle = & \frac{-2\pi i}{\hbar} \delta(\omega_0 - \omega_{\mathrm{out}}) \frac{\Omega^2/2}{\delta + i\gamma} \frac{g(k_{\mathrm{out}}) g(k_A)}{\delta} \left(\frac{1}{i\gamma}\right)^2 (\vec{d}_A \cdot \vec{\epsilon}_{\lambda_{\mathrm{out}}})(\vec{d}_B \cdot \vec{\epsilon}_{\lambda_{k_A}}) \\ & \times (\vec{d}_B \cdot \vec{\epsilon}_{\lambda_L})^2 \frac{3}{2} \gamma [\vec{d}_A \cdot (\mathbf{I} - \vec{e}_r \otimes \vec{e}_r) \cdot \vec{d}_B] \frac{\exp[+ik_0 r]}{k_0 r} \exp[i\vec{k}_0 \cdot \vec{r}] \\ & \times \left[1 + \frac{i\gamma}{2\delta + i\gamma}\right] \Psi_A(\vec{p}_f^A + \vec{k}_{\mathrm{out}} - k_0 \vec{e}_r). \end{aligned} \tag{45}$$

In the limit of $|\delta| \gg \gamma$ the second term in the square brackets is a lot smaller than 1 and we can neglect it. To obtain the probability for this process we have to sum the modulus squared of the matrix element in Eq. (45) over all possible polarizations of the two outgoing photon lines, integrate over the solid angle of the second photon scattered by atom B and multiply by a factor $\omega_A^2 \Delta\omega_A / (2\pi^2)$ for the width of the energy interval of photon k_A. ω_A is equal to $\omega_L + \delta$ and the width of the energy interval is given by the width of the sideband which is approximately equal to the natural linewidth 2γ. The contribution of the resonantly scattered light to the momentum diffusion coefficient is then given by

$$\begin{aligned} D_p^{\mathrm{inel}}(\vec{r}) = & D_P^{1-\mathrm{at}} |\vec{d}_B \cdot \vec{\epsilon}_{\lambda_L}|^2 \frac{6}{8\pi^2} \frac{\Omega^2}{2\gamma^2} \sigma_{\mathrm{Ray}}(\delta) \\ & \times \left| \frac{[\vec{d}_A \cdot (\mathbf{I} - \vec{e}_r \otimes \vec{e}_r) \cdot \vec{d}_B]}{r} \right|^2 . \end{aligned} \tag{46}$$

The numerical factor for the extra diffusion due to an atom a distance r away can be estimated by integrating over $d\Omega_r$ to get the contribution of all the amplitudes of particles located in a spherical shell of width dr. Performing the integration over the solid angle $d\Omega_r$ gives a factor of $\frac{8\pi}{3}$. The integral over the solid angle of the scattered photon gives another factor of $\frac{8\pi}{3}$. We therefore have

$$D_p^{\mathrm{el}}(r) = \frac{8\pi}{3} |(\vec{d}_A . \vec{d}_B)|^2 \left(\frac{\omega_L}{\omega_0}\right)^3 \frac{\sigma_{\mathrm{Ray}}(\delta)}{r^2} D_p^{1\text{-at}}. \tag{47}$$

If we now assume that the atomic dipole is oriented along the local laser polarization which varies across the trap and take into account a further factor of 2 which arises in the averaging over the change in momentum the factor multiplying the extra diffusion term is given by $2 \times \frac{4\pi}{3} \times \frac{1}{3} = 2.8$.

$$D_p^{\mathrm{el}}(r) = 2.8 \left(\frac{\omega_L}{\omega_0}\right)^3 \frac{\sigma_{\mathrm{Ray}}(\delta)}{r^2} D_p^{1\text{-at}}. \tag{48}$$

In a similar way we obtain for the contribution of the inelastically scattered light

$$D_p^{\mathrm{inel}}(r) = 1.7 \frac{\Omega^2}{2\gamma^2} \frac{\sigma_{\mathrm{Ray}}(\delta)}{r^2} D_p^{1\text{-at}}. \tag{49}$$

The factor of $\Omega^2/2\gamma^2$ comes from the resonance cross section which has been taken in the limit of $\Omega^2 \ll \gamma^2$. For larger Rabi frequencies the strong dependence of the inelastic two-atom diffusion coefficient is unrealistic. The cross section for the reabsorption of the inelastically scattered light can be approximated by [5]

$$\sigma_{\mathrm{inel}} = \left(1 - \frac{1}{1 + s_0}\right) \frac{s_0 + 1}{s_0 + \gamma^2/(\delta^2 + \gamma^2)} \sigma_{\mathrm{Ray}}, \tag{50}$$

where the factor $\left(1 - \frac{1}{1+s_0}\right)$ indicates the relative strength of the inelastic component of the scattered light. The total intensity of the scattered light is given by $I_{\mathrm{scat}} = \sigma_{\mathrm{Ray}}(\delta) I$, I being the total laser intensity. The total contribution of the inelastic scatter to the diffusion is proportional to the total scattered intensity times the cross section for the reabsorption of the inelastic part which is given by

$$\begin{aligned} & \left(1 - \frac{1}{1 + s_0}\right) \frac{s_0 + 1}{s_0 + \gamma^2/(\delta^2 + \gamma^2)} \sigma_{\mathrm{Ray}}^2 I \\ & = \frac{s_0}{s_0 + \gamma^2/(\delta^2 + \gamma^2)} \sigma_{\mathrm{Ray}}^2 I = \frac{\Omega^2/2}{\Omega^2/2 + \gamma^2} \sigma_{\mathrm{Ray}}^2 I. \end{aligned} \tag{51}$$

The resonance cross section for Rabi frequencies of the order of the natural linewidth gives rise to a factor of

$\Omega^2/(\Omega^2/2+\gamma^2)$ which is of the order of unity for large Rabi frequencies. The perturbative treatment for low saturation does not exhibit this dependence because it is equivalent to making the approximation $s_0/(1+s_0)\approx s_0$ in the expression for the inelastic cross section. Making this approximation in Eq. (50) yields a factor of $\Omega^2/(2\gamma^2)$. This illustrates the limitations of the diagrammatic treatment. Its virtue lies, however, in clearly showing the physical processes which contribute to the diffusion coefficient of the pair.

D. The trap averaged momentum diffusion coefficient

Under the assumption of a constant atomic density n throughout the trap the total contribution from the cloud can be calculated by integrating over the trap size. This yields

$$\langle D_p^{\rm el}+D_P^{\rm inel}\rangle_{\rm Av}=n\int drr^2[D_p^{\rm el}(r)+D_p^{\rm inel}(r)]$$
$$=\left(2.8+1.7\frac{\Omega^2}{2\gamma^2}\right)D_p^{\text{1-at}}\;n\;\sigma_{\rm Ray}(\delta)L, \tag{52}$$

where L is the trap radius. In the regime where radiation trapping forces become important L scales as $N^{1/3}$, with N the number of trapped atoms. With the aid of formula (1) the diffusion coefficient for a two-level atom including the effects of reabsorption of scattered radiation can be written as

$$D_p=D_p^{\text{1-at}}\left\{1+\left(2.8+1.7\frac{\Omega^2}{2\gamma^2}\right)n^{2/3}\sigma_{\rm Ray}(\delta)N^{1/3}\right\}. \tag{53}$$

Formulas for the limiting constant density expected from the model developed by Sesko *et al.* can be found in Refs. [4,5]. According to these expressions the density has a dependence on detuning which is proportional to δ^3. The scattering cross section $\sigma_{\rm Ray}$ in the large detuning limit scales as $1/\delta^2$ and the density n to the power of two-thirds is proportional to δ^2. The extra term in the diffusion coefficient in Eq. (53), $n^{2/3}\sigma_{\rm Ray}(\delta)N^{1/3}$, is therefore independent of detuning as was observed in Refs. [1,2]. We have to point out, however, that these density formulas are derived under the assumption of a spring constant which is independent of the density. This in fact is not true as the sub-Doppler cooling mechanisms are very sensitive to the interaction with the scattered radiation [18].

Equation (A3) clearly shows that this result is only valid in a density regime where the average separation of the atoms is large compared to the wavelength. This is also true for the theory developed by Sesko, Walker, and Wieman [4] as they assume that the radiation trapping forces have a dependence on the atomic separation which is proportional to $1/r^2$. Our calculation shows that this assumption is only valid for the far-field regime. We therefore expect that the condition of a constant density is violated if the spacing of the atoms in the trap is comparable to the laser wavelength. We also note that the validity of the S-matrix expansion only depends on the condition $kr\gg 1$. This condition is not equivalent to the approximation that the cloud is optically thin and our treatment does not exclude the possibility that $n\sigma L$ is of the order of unity. In the average over all pairs in the trap we still have to assume the cloud to be optically thin in order to exclude processes which are due to photon exchanges which involve three or more atoms. There will also be an effect due to the attenuation of the incoming laser beam. In the regime where the cloud is considered to be optically thin and a photon is rescattered not more than once the attenuation of the laser will only affect the one-atom diffusion coefficient. The effect can be included by a reduction of the local laser intensity due to scattering out of the beam. This attenuation will not affect the two-atom part as a shielding of the incoming laser photon will in this case correspond to a higher-order effect. For optically thick clouds the attenuation is included by replacing the incoming photon line by a shielded photon line. This can be done, for example, by performing a ring summation over an infinite sequence of bubble diagrams. As this effect would affect the single-atom diffusion the same way as the two-atom diffusion this term will not show in the extra dependence of the temperature on the number of trapped atoms. The problem in the summation for optically thick clouds lies in the summation for multiple scattering of the intermediate photon between atoms A and B via a third or more atoms.

III. CONCLUSION

We have calculated the momentum diffusion for a pair of atoms in a laser beam. In the low-saturation regime the total momentum diffusion is calculated by adding the contributions of the individual beams. The Feynman diagram technique used allows us to identify the physical processes which contribute to the pair diffusion coefficient. The results obtained are only giving a qualitative picture and do not allow an accurate prediction within more than a factor of 2. In our treatment we take the cloud to be optically thin, which means the photon does not get scattered more than twice on its way out of the cloud. This is the same regime used by Sesko *et al.* [4] to derive their expression for a constant density. Our calculations show that one additional assumption has to be made for the validity of their discussion. If the spacing between the atoms becomes too small the $1/r$ dependence of the radiation trapping forces becomes invalid and one has to take higher orders in $1/r$ of the dipole-dipole interaction into account. The expressions for the limiting constant density derived in [4,5] are no longer correct in this case. As Sesko, Walker, and Wieman [4] pointed out, there is also an attenuation of the laser beam as it propagates through the trap. As the intensity used in computing the diagrams is the local intensity the attenuation of the beam leads to a reduction of the one-atom diffusion which also has to be taken into account as its magnitude is of the same order as the contribution due to the radiation trapping force. This contribution reduces the effect of the elastically scattered light. Due to the stronger resonant cross section the resonantly scattered light gives a contribution which is of the same order of magnitude as the contribution of the elastic light. This stronger interaction will also have a strong effect on the sub-Doppler cooling mechanisms [7] as it will shorten the optical pumping time and will also lead to an increase of the excited-state population hence again decreasing the efficiency of the sub-Doppler cooling mechanisms. We shall address these points in a forthcoming paper.

ACKNOWLEDGMENTS

We wish to acknowledge many helpful discussions with Professor J. Cooper. G.H. acknowledges support by the U.K. Science and Engineering Research Council.

APPENDIX

In this appendix we show how to perform the summation over the intermediate photon momentum for the exchange of elastic scattered photons between the pair of atoms. The summations for the inelastic contributions are done in a similar fashion. To carry out the $d^3\vec{k}$ integration we use the Fourier expansion of the Gaussian $\Psi_A(\vec{p}_f^A+\hbar(\vec{k}_{\rm out}-\vec{k}))$. Substituting Eq. (38) into Eq. (35) yields

$$\begin{aligned}\langle \vec{p}_f^A, g|\langle\Psi_B, g|\hat{S}(\vec{r})|g,\Psi_B\rangle|g,\Psi_A\rangle &= \int d^3\vec{r}_A \exp\left[-\frac{i}{\hbar}(\vec{p}_f^A+\vec{k}_{\rm out})\cdot\vec{r}_A\right]\Psi_A(\vec{r}_A)\exp[i\vec{k}_L\cdot\vec{r}]\\ &\quad\times V\int\frac{d^3k}{(2\pi)^3}\exp[-i\vec{k}\cdot(\vec{r}-\vec{r}_A)]\\ &\quad\times\langle\vec{p}_f^A,g|\langle\vec{p}_f^B,g|\hat{S}_{\vec{k}}(\vec{r})|g,\vec{p}_f^B+\hbar(\vec{k}-\vec{k}_L)\rangle|g,\vec{p}_f^A+\hbar(\vec{k}_{\rm out}-\vec{k})\rangle. \end{aligned}\tag{A1}$$

The integration over the intermediate photon momentum can now be carried out without further ado. If we use the relations

$$\lim_{\epsilon\searrow 0}\frac{1}{(\omega_L-\omega_k)+i\epsilon} = -i\pi\delta(\omega_L-\omega_k)+\mathrm{P}\frac{1}{\omega_L-\omega_k}\tag{A2}$$

and [19]

$$\begin{aligned}\int\frac{d\Omega_k}{4\pi}(\mathbf{I}-\vec{n}_k\otimes\vec{n}_k)\exp[-i\vec{k}\cdot(\vec{r}-\vec{r}_A)] &= (\mathbf{I}-\vec{e}_{|\vec{r}-\vec{r}_A|}\otimes\vec{e}_{|\vec{r}-\vec{r}_A|})\frac{\sin k|\vec{r}-\vec{r}_A|}{k|\vec{r}-\vec{r}_A|}\\ &\quad+(\mathbf{I}-3\vec{e}_{|\vec{r}-\vec{r}_A|}\otimes\vec{e}_{|\vec{r}-\vec{r}_A|})\\ &\quad\times\left(\frac{\cos k|\vec{r}-\vec{r}_A|}{k^2|\vec{r}-\vec{r}_A|^2}-\frac{\sin k|\vec{r}-\vec{r}_A|}{k^3|\vec{r}-\vec{r}_A|^3}\right)\end{aligned}\tag{A3}$$

and neglect terms of the order $\frac{1}{k^2|\vec{r}-\vec{r}_A|^2}$ or higher, the integral over the intermediate photon can be evaluated to give

$$\begin{aligned}V\int\frac{d^3k}{(2\pi)^3}g^2(k)[\vec{d}_A\cdot(\mathbf{I}-\vec{n}_k\otimes\vec{n}_k)\cdot\vec{d}_B]\left[\frac{1}{\omega_L-\omega_k+i\epsilon}-\frac{1}{\omega_L+\omega_k-i\epsilon}\right]\exp[-i\vec{k}\cdot(\vec{r}-\vec{r}_A)]\\ =\frac{-3}{2}\gamma\left(\frac{\omega_L}{\omega_0}\right)^3[\vec{d}_A\cdot(\mathbf{I}-\vec{e}_{|\vec{r}-\vec{r}_A|}\otimes\vec{e}_{|\vec{r}-\vec{r}_A|})\cdot\vec{d}_B]\frac{\exp[-ik_L|\vec{r}-\vec{r}_A|]}{k_L|\vec{r}-\vec{r}_A|}.\end{aligned}\tag{A4}$$

The principal part of the integral over dk is equal to [20]

$$\mathrm{P}\int dk\left[\frac{k^3}{k-k_L}+\frac{k^3}{k_L+k}\right]\times\frac{\sin kR}{kR}=\pi\frac{k_L^2}{R}\cos k_LR.\tag{A5}$$

Here $\vec{r}_A$ represents the vector from the center of the wave packet representing particle A in position space. The width of this wave packet is much smaller than a wavelength so that in the far-field limit we always have $|\vec{r}|\gg|\vec{r}_A|$. Expanding Eq. (A4) to first order in $\frac{|\vec{r}_A|}{|\vec{r}|}$ yields

$$\begin{aligned}V\int\frac{d^3k}{(2\pi)^3}g^2(k)[\vec{d}_A\cdot(\mathbf{I}-\vec{n}_k\otimes\vec{n}_k)\cdot\vec{d}_B]\left[\frac{1}{\omega_L-\omega_k+i\epsilon}-\frac{1}{\omega_L+\omega_k-i\epsilon}\right]\exp[-i\vec{k}\cdot(\vec{r}-\vec{r}_A)]\\ =\exp[ik_L\vec{e}_r\cdot\vec{r}_A]\frac{-3}{2}\gamma\left(\frac{\omega_L}{\omega_0}\right)^3[\vec{d}_A\cdot(\mathbf{I}-\vec{e}_r\otimes\vec{e}_r)\cdot\vec{d}_B]\frac{\exp[+ik_Lr]}{k_Lr}.\end{aligned}\tag{A6}$$

It should be noted that the expansion of the tensor part also yields terms of the order one in $\frac{|\vec{r}_A|}{|\vec{r}|}$ but their contribution averages to zero when the integral over $d^3\vec{r}_A$ is done. Combining Eqs. (36), (A1), and (A6) and integrating over $d^3\vec{r}_A$ gives

$$\begin{aligned}\langle \vec{p}_f^{\,A}, g|\langle \Psi_B, g|\hat{S}(\vec{r})|g, \Psi_B\rangle|g, \Psi_A\rangle = \hbar g(k_{\text{out}})(\vec{d}_A \cdot \vec{\epsilon}_{\lambda_{\text{out}}})\left(\frac{1}{\delta + i\gamma}\right)^2 g(k_L)(\vec{d}_B \cdot \vec{\epsilon}_{\lambda_L})\frac{3}{2}\gamma\left(\frac{\omega_L}{\omega_0}\right)^3 \\ \times[\vec{d}_A \cdot (\mathbf{I} - \vec{e}_r \otimes \vec{e}_r) \cdot \vec{d}_B]\frac{\exp[+ik_L r]}{k_L r}\Psi_A(\vec{p}_f^{\,A} + (\vec{k}_{\text{out}} - k_L \vec{e}_r)) \\ \times \exp[+i\vec{k}_L \cdot \vec{r}](2\pi i)\delta(E_{\text{in}} - E_{\text{out}}),\end{aligned} \tag{A7}$$

which proves Eq. (39).

[1] A. Clairon, Ph. Laurent, A. Nadir, M. Drewsen, D. Grison, B. Lounis, and C. Salomon (unpublished).

[2] C. J. Foot, C. J. Cooper, J. Rink, C. Townsend, and K. P. Zetie (unpublished).

[3] J. Dalibard, Opt. Commun. **68,** 203 (1988).

[4] D. Sesko, T. Walker, and C. Wieman, J. Opt. Soc. Am. B **8,** 946 (1991).

[5] A. M. Steane, M. Chowdhury, and C. J. Foot, J. Opt. Soc. Am. B **9,** 2142 (1992).

[6] K. Lindquist, M. Stephens, and C. Wieman, Phys. Rev. A **46,** 4082 (1992).

[7] J. Dalibard and C. Cohen-Tannoudji, J. Opt. Soc. Am. B **6,** 2023 (1989).

[8] A. M. Steane and C. J. Foot, Europhys. Lett. **14,** 231 (1991).

[9] M. Walhout, J. Dalibard, S. L. Rolston, and W. D. Phillips, J. Opt. Soc. Am. B **9,** 1997 (1992).

[10] J. Dalibard and C. Cohen-Tannoudji, J. Phys. B **18,** 1661 (1985).

[11] S. Swain, J. Phys. A **8,** 1277 (1975).

[12] R. J. Ballagh and J. Cooper, J. Quant. Spectrosc. Radiat. Transfer **23,** 535 (1980).

[13] B. Mollow, Phys. Rev. A **188,** 1969 (1969).

[14] J. P. Gordon and A. Ashkin, Phys. Rev. A **21,** 1606 (1980).

[15] C. Cohen-Tannoudji, in *Fundamental Systems in Quantum Optics,* 1990 Les Houches Session LIII, edited by J. Dalibard, J. Raimond, and J. Zinn-Justin (Elsevier, New York, 1992), p. 1.

[16] C. R. Stroud, Jr., Phys. Rev. A **3,** 1044 (1971).

[17] See, e.g., J. M. Ziman, *Elements of Advanced Quantum Theory* (Cambridge University Press, Cambridge, England, 1969).

[18] G. Hillenbrand, K. Burnett, and C. J. Foot (unpublished).

[19] R. R. McLone and E. A. Power, Proc. R. Soc. London **286,** 573 (1965).

[20] This formula follows directly from Eqs. (A7) and (A8) in the Appendix of M. Trippenbach, B. Gao, and J. Cooper, Phys. Rev. A **45,** 6539 (1992).