Operator description of laser cooling below the Doppler limit

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We give a general theoretical description of radiative forces on atoms in a monochromatic radiation field. The driven transition has a lower level consisting of several substates, and we consider the limit of low velocities and weak intensities. This situation comprises the schemes where cooling to temperatures below the Doppler limit is possible. After expressing the atomic dipole in terms of the polarizability tensor, we obtain an expression of the force as the sum of the radiation pressure, the dipole force, and a contribution from the gradient of the polarization. This latter contribution contains a part resulting from redistribution of photons between the plane waves that compose the field, and a part resulting from fluorescence. We express the average force and the momentum-diffusion tensor in terms of a closed evolution equation for the lower-state density matrix. We show by a scaling argument that in any case of cooling, the final temperature goes down linearly with the intensity, until the recoil limit is approached. The description is valid for a weak and monochromatic, but otherwise arbitrary field, and it allows for the presence of an external magnetic field, and of hyperfine splitting. It is applicable to cooling in one, two, or three dimensions. We give numerical results for the average force in a number of specific cases of two counterrunning plane waves.

I. INTRODUCTION

The simplest model for cooling in an optical molasses is a two-level atom in a standing wave [1,2]. The minimal temperature T_D that can be reached for this configuration occurs at nonsaturating intensities and at a light frequency that is below resonance by about $\Gamma/2$, with Γ the spontaneous decay rate. This minimal value, which is commonly called the Doppler limit, is given by [3,4]

$$kT_D=\hbar\Gamma/2 , \tag{1.1}$$

where k is Boltzmann's constant. Recent experiments have demonstrated that cooling to lower temperatures can occur for systems having a multistate structure in the lower level of the driven transition [5–11]. An essential ingredient for this strong damping force is a variation of the relative orientation of the atomic dipole with respect to the light polarization during the traversal of a wavelength. Such a situation arises when the atom moves through a field with polarization gradients [12,13]. An alternative possibility is that a magnetic field causes Zeeman precession of the atomic dipole [8–11,13]. Theoretical descriptions of laser cooling below the Doppler limit have been given for a few specific polarizations and angular momentum values [12,13].

During the cooling process, the atom suffers momentum jumps of the order of a photon momentum. We assume that the velocity width of the atomic velocity distribution is large compared with the corresponding jumps in velocity $\hbar K/M$, where K is the wave number of the radiation, and M is the atomic mass. This means that the effective temperature is larger than the recoil temperature

$$kT_R=\frac{\hbar^2K^2}{2M} . \tag{1.2}$$

Then a Brownian motion picture holds, and the evolution of the velocity distribution $W(\mathbf{v})$ is governed by a Fokker-Planck equation [14]:

$$\frac{\partial}{\partial t}W(\mathbf{v})=-\frac{1}{M}\frac{\partial}{\partial \mathbf{v}}\cdot\mathbf{F}(\mathbf{v})W(\mathbf{v})+\frac{1}{M^2}\frac{\partial^2}{\partial \mathbf{v}\partial \mathbf{v}}:\overleftrightarrow{D}(\mathbf{v})W(\mathbf{v}) . \tag{1.3}$$

Here

$$\mathbf{F}(\mathbf{v})=\langle \mathbf{f}(\mathbf{v})\rangle \tag{1.4}$$

is the velocity-dependent average force on the atoms, and $\overleftrightarrow{D}$ is the momentum-diffusion tensor, which can be expressed as the integrated autocorrelation function of the force as

$$2\overleftrightarrow{D}(\mathbf{v})=\int_0^\infty d\tau[\langle \mathbf{f}(t)\mathbf{f}(t+\tau)\rangle-\langle \mathbf{f}(t)\rangle\langle \mathbf{f}(t+\tau)\rangle +\langle \mathbf{f}(t+\tau)\mathbf{f}(t)\rangle-\langle \mathbf{f}(t+\tau)\mathbf{f}(t)\rangle] . \tag{1.5}$$

In Eq. (1.5) we allow for the situation that the average force varies with time. The time evolution of the force is governed by the evolution of the internal state of the atom. In a situation where the force as a function of velocity leads to cooling, the limiting temperature of the cooling process is determined by the steady-state solution of (1.3). In the simple case of a friction force

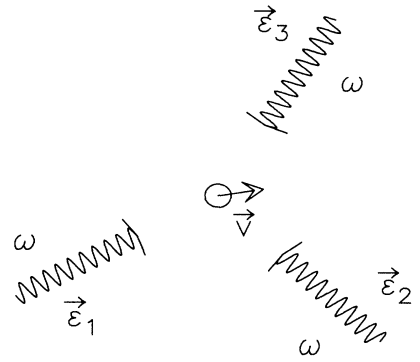

FIG. 1. Atom moving with a given velocity through an arbitrary monochromatic light field.

$$\mathbf{F}(\mathbf{v})=-\gamma\mathbf{v} \tag{1.6}$$

and a velocity-independent isotropic diffusion coefficient D, the obvious solution of (1.3) is a Maxwell distribution with temperature given by

$$kT=D/\gamma\ . \tag{1.7}$$

This result illustrates that the limiting temperature results from the balance between the cooling effect of the friction and the heating due to the force fluctuations. These fluctuations inevitably arise from the random nature of spontaneous decay. In general, the Fokker-Planck equation (1.3) shows that the evolution of the velocity distribution is fully determined by the force and the diffusion tensor as a function of velocity. This equation is no longer reliable when the velocity width of the atoms gets comparable to the recoil velocity $\hbar K/M$.

In the present paper, we give a systematic operator description of the force and the momentum diffusion of an atom with degenerate or nearly degenerate lower levels. We assume that the atom is moving with a given velocity in a weak and monochromatic, but otherwise arbitrary, light field. This situation is depicted in Fig. 1. The velocity-dependent force and diffusion tensor can then be expressed in terms of the lower-state density matrix. We distinguish four different types of contributions to the force. We recover the usual radiation pressure and the dipole force, arising from the gradients of the phase and the intensity of the field, and which exist also for a two-level system. But in addition we obtain two contributions to the force that depend on the gradient of the polarization, and which rely upon the multistate structure of the lower state. One of these latter contributions is determined by the Hermitian part, the other by the anti-Hermitian part of the atomic polarizability. The correlation function determining the momentum diffusion coefficient separates into a rapidly decaying part and a weak but slowly decaying part. These two parts give separate contributions to the diffusion tensor with the same order of magnitude, each having a distinct physical significance.

Since the formalism allows for an arbitrary configuration of light beams, it is well suited to describe cooling configurations in one, two, or three dimensions. We calculate explicitly the velocity-dependent force in several one-dimensional cases, such as a standing wave with a transverse magnetic field, and two counterpropagating waves in orthogonal polarizations, for a number of values of the angular momenta of the states involved.

II. ATOMIC DIPOLE

We consider the situation where an atom moves with a given velocity $\mathbf{v}$ through a monochromatic radiation field with frequency ω. The radiation field is described by the electric field

$$\mathbf{E}(\mathbf{R},t)=\mathbf{E}_+(\mathbf{R})e^{-i\omega t}+\mathbf{E}_-(\mathbf{R})e^{i\omega t}\ . \tag{2.1}$$

The field drives the transition between a lower level indicated by the index g (for ground state) and an excited level indicated by e. Both levels may consist of a manifold of degenerate or nearly degenerate substates, corresponding to Zeeman degeneracy or hyperfine splitting. We wish to derive general expressions for the radiative force on the atom and for the momentum diffusion. The internal state of the atom is described by the density matrix $\sigma(t)$ in the rotating frame. This density matrix has as dimension the sum of the number of substates of the lower level and the excited level. If we separate this matrix into four blocks corresponding to the two levels, we obtain the submatrices σ_{ee} and σ_{gg} for the excited-state and lower-state manifolds, and the submatrices σ_{eg} and σ_{ge} containing the optical coherences. Likewise, we separate the electric-dipole operator of the atom as

$$\boldsymbol{\mu}=\boldsymbol{\mu}_{eg}+\boldsymbol{\mu}_{ge} \tag{2.2}$$

into its raising and its lowering part.

In the rotating-wave approximation, the atom-field coupling is governed by the Rabi operator

$$\mathcal{R}=\boldsymbol{\mu}_{eg}\cdot\mathbf{E}_+/\hbar\ , \tag{2.3}$$

which generalizes the Rabi frequency. Obviously, this operator $\mathcal{R}$ has nonvanishing matrix elements only between a lower and an upper substate. The evolution equations for the submatrices of σ may be put in the form [15]

$$\begin{aligned}
\frac{d\sigma_{ee}}{dt}&=-\Gamma\sigma_{ee}-iL\sigma_{ee}+i\mathcal{R}\sigma_{ge}-i\sigma_{eg}\mathcal{R}^\dagger\ ,\\
\frac{d\sigma_{gg}}{dt}&=\Gamma\sum_\beta Q_\beta^\dagger\sigma_{ee}Q_\beta-iL\sigma_{gg}+i\mathcal{R}^\dagger\sigma_{eg}-i\sigma_{ge}\mathcal{R}\ ,\\
\frac{d\sigma_{eg}}{dt}&=-\left[\frac{\Gamma}{2}-i\Delta+iL\right]\sigma_{eg}+i\mathcal{R}\sigma_{gg}-i\sigma_{ee}\mathcal{R}\ ,\\
\frac{d\sigma_{ge}}{dt}&=-\left[\frac{\Gamma}{2}+i\Delta+iL\right]\sigma_{ge}+i\mathcal{R}^\dagger\sigma_{ee}-i\sigma_{gg}\mathcal{R}^\dagger\ .
\end{aligned} \tag{2.4}$$

The spontaneous decay rate is indicated by Γ, and $\Delta=\omega-\omega_0$ is the detuning of the light frequency from the average resonance frequency ω_0. The Liouville operator L accounts for the level splitting within the two manifolds e and g, and it is basically the commutator with the hyperfine or the Zeeman Hamiltonian. Hence, in the absence of Zeeman or hyperfine splitting, the operator L

vanishes. Finally, we have introduced the dimensionless dipole operators Q_β to account for the anisotropic repopulation of the lower level by spontaneous decay. These operators are defined by their matrix elements as Clebsch-Gordan coefficients [16]

$$\langle J_e M_e | Q_\beta | J_g M_g \rangle = \langle J_e M_e | J_g M_g; 1\beta \rangle \tag{2.5}$$

for $\beta = -1, 0, 1$, where J_e and J_g are the electronic angular momenta of the two levels. In the case of hyperfine structure, the spontaneous emission does not affect the state of the nuclear spin. The electric fields $\mathbf{E}_\pm$ inherent in the operators $\mathcal{R}$ and $\mathcal{R}^\dagger$ in (2.5) should be taken at the time-dependent position $\mathbf{R}(t)$ of the atom. Since we assume that the atom moves with the given velocity $\mathbf{v}$, this position may be taken as

$$\mathbf{R}(t) = \mathbf{R}_0 + \mathbf{v}t \ . \tag{2.6}$$

This semiclassical picture of a force arising for an atom moving with a given velocity is justified when $\mathbf{v}$ is much larger than the recoil velocity $\hbar K/M$. For a wide velocity distribution, this is the case for a large majority of velocity groups.

We are interested in the limit of low velocities, so that the atomic displacement during a spontaneous lifetime is small compared with the wavelength, or

$$Kv \ll \Gamma \ . \tag{2.7}$$

This implies that the decay of the optical coherences is rapid compared with the rate of change of the field that the atom feels. This allows us to eliminate the coherences adiabatically, and we obtain from the last two equations of (2.4)

$$\begin{aligned} \sigma_{eg} &= i \left[\frac{\Gamma}{2} - i\Delta + iL \right]^{-1} (\mathcal{R}\sigma_{gg} - \sigma_{ee}\mathcal{R}) \ , \\ \sigma_{ge} &= i \left[\frac{\Gamma}{2} + i\Delta + iL \right]^{-1} (\mathcal{R}^\dagger \sigma_{ee} - \sigma_{gg}\mathcal{R}^\dagger) \ . \end{aligned} \tag{2.8}$$

These equations relate the optical coherences at any instant of time to the submatrices σ_{ee} and σ_{gg} at the same instant. If we substitute these equations (2.8) in the first two equations of (2.4), we arrive at a pair of equations for σ_{ee} and σ_{gg} in the form

$$\begin{aligned} \frac{d\sigma_{ee}}{dt} &= -\Gamma\sigma_{ee} - iL\sigma_{ee} \\ &\quad - \mathcal{R} \left[\frac{\Gamma}{2} + i\Delta + iL \right]^{-1} (\mathcal{R}^\dagger \sigma_{ee} - \sigma_{gg}\mathcal{R}^\dagger) \\ &\quad + \left[\left[\frac{\Gamma}{2} - i\Delta + iL \right]^{-1} (\mathcal{R}\sigma_{gg} - \sigma_{ee}\mathcal{R}) \right] \mathcal{R}^\dagger \ , \\ \frac{d\sigma_{gg}}{dt} &= \Gamma \sum_\beta Q_\beta^\dagger \sigma_{ee} Q_\beta \\ &\quad - iL\sigma_{gg} - \mathcal{R}^\dagger \left[\frac{\Gamma}{2} - i\Delta + iL \right]^{-1} (\mathcal{R}\sigma_{gg} - \sigma_{ee}\mathcal{R}) \\ &\quad + \left[\left[\frac{\Gamma}{2} + i\Delta + iL \right]^{-1} (\mathcal{R}^\dagger \sigma_{ee} - \sigma_{gg}\mathcal{R}^\dagger) \right] \mathcal{R} \ . \end{aligned} \tag{2.9}$$

Furthermore, Eqs. (2.8) allow us to express the atomic dipole moment in terms of an effective polarizability tensor. We find

$$\langle \boldsymbol{\mu}(t) \rangle = \boldsymbol{\mu}_+ e^{-i\omega t} + \boldsymbol{\mu}_- e^{i\omega t} \ , \tag{2.10}$$

where the positive-frequency part of the dipole may be expressed by

$$\boldsymbol{\mu}_+ = \mathrm{Tr}\sigma_{eg}\boldsymbol{\mu}_{ge} = \overleftrightarrow{\alpha} \cdot \mathbf{E}_+ \tag{2.11}$$

in terms of the polarizability tensor

$$\overleftrightarrow{\alpha} = \frac{i}{\hbar} \mathrm{Tr}\boldsymbol{\mu}_{ge} \left[\frac{\Gamma}{2} - i\Delta + iL \right]^{-1} (\boldsymbol{\mu}_{eg}\sigma_{gg} - \sigma_{ee}\boldsymbol{\mu}_{eg}) \ . \tag{2.12}$$

Notice that the polarizability $\overleftrightarrow{\alpha}$ depends on the submatrices σ_{gg} and σ_{ee} for the lower state and for the excited state, which are the solutions of the evolution equations (2.9). These solutions, and hence also the polarizability, still depend on the field that the atom has felt during a time in the recent past of the order of the optical-pumping time. Hence the nonadiabatic time lag that is responsible for the force on the atom [12] is implicit in the polarizability. Notice, moreover, that Eq. (2.11) does not imply a linearization in the electric-field amplitude.

III. POLARIZABILITY AND FORCE

For an atom in a radiation field, the Heisenberg operator for the force $\mathbf{f}$ is determined by the commutator of the momentum operator with the Hamiltonian. This gives

$$\mathbf{f} \equiv \nabla(\boldsymbol{\mu} \cdot \mathbf{E}) = \hbar(\nabla\mathcal{R} + \nabla\mathcal{R}^\dagger) \ , \tag{3.1}$$

with $\mathcal{R}$ given in (2.3). For the component j of the force $\mathbf{F}$ this gives

$$F_j = 2\,\mathrm{Re}\boldsymbol{\mu}_+ \cdot \nabla_j \mathbf{E}_- \ . \tag{3.2}$$

Furthermore, we separate the polarizability tensor $\overleftrightarrow{\alpha}$ into its Hermitian and anti-Hermitian parts, according to

$$\overleftrightarrow{\alpha} = \overleftrightarrow{\alpha}_0 + i\overleftrightarrow{\alpha}_1 \ . \tag{3.3}$$

Substituting (2.11) and (3.3) in (3.2) leads to the expression for the force components

$$F_j = 2\,\mathrm{Re}(\mathbf{E}_- \cdot \overleftrightarrow{\alpha}_0 \cdot \nabla_j \mathbf{E}_+ - i\mathbf{E}_- \cdot \overleftrightarrow{\alpha}_1 \cdot \nabla_j \mathbf{E}_+) \ . \tag{3.4}$$

First we separate the positive-frequency part of the field according to

$$\mathbf{E}_+(\mathbf{R}) = \boldsymbol{\epsilon}(\mathbf{R}) A(\mathbf{R}) \ , \tag{3.5}$$

where A is the position-dependent amplitude, and $\boldsymbol{\epsilon}$ is a normalized polarization vector, obeying

$$\boldsymbol{\epsilon}^* \cdot \boldsymbol{\epsilon} = 1 \ . \tag{3.6}$$

If we substitute (3.5) into (3.4), we obtain an expression for the force as a sum of four separate terms

$$\mathbf{F} = \mathbf{F}_1 + \mathbf{F}_2 + \mathbf{F}_3 + \mathbf{F}_4 \tag{3.7}$$

with components

$$F_{1j}=\epsilon^*\cdot\vec{\alpha}_1\cdot\epsilon i[A\nabla_j A^*-(\nabla_j A)A^*] , \quad (3.8)$$

$$F_{2j}=\epsilon^*\cdot\vec{\alpha}_0\cdot\epsilon[A\nabla_j A^*+(\nabla_j A)A^*] , \quad (3.9)$$

$$F_{3j}=AA^*i(\nabla_j\epsilon^*\cdot\vec{\alpha}_1\cdot\epsilon-\epsilon^*\cdot\vec{\alpha}_1\cdot\nabla_j\epsilon) , \quad (3.10)$$

$$F_{4j}=AA^*(\nabla_j\epsilon^*\cdot\vec{\alpha}_0\cdot\epsilon+\epsilon^*\cdot\vec{\alpha}_0\cdot\nabla_j\epsilon) . \quad (3.11)$$

The force $\mathbf{F}_1$ is the common radiation pressure, proportional to the gradient of the phase of the field, and $\mathbf{F}_2$ is the dipole force, determined by the gradient of the local intensity. The forces $\mathbf{F}_3$ and $\mathbf{F}_4$ both arise from the gradient of the polarization direction, and they are determined by the anti-Hermitian and Hermitian parts of the tensor $\epsilon\nabla_j\epsilon^*$. The anti-Hermitian part α_1 of the polarizability, which determines $\mathbf{F}_1$ and $\mathbf{F}_3$, also determines the work done by the field on the atomic dipole, as expressed by

$$W\equiv\left\langle\mathbf{E}\cdot\frac{d\mu}{dt}\right\rangle=2\omega AA^*\epsilon^*\cdot\vec{\alpha}_1\cdot\epsilon . \quad (3.12)$$

The forces $\mathbf{F}_1$ and $\mathbf{F}_3$ arise from absorption of photons that are scattered into the empty modes of the vacuum field. The forces $\mathbf{F}_2$ and $\mathbf{F}_4$ have the dispersive character of $\vec{\alpha}_0$, and arise from redistribution of photons between modes.

This picture is confirmed by an alternative separation of the electric field as an expansion in plane waves:

$$\mathbf{E}_+(\mathbf{R})=\sum_n A_n\epsilon_n e^{i\mathbf{K}_n\cdot\mathbf{R}} . \quad (3.13)$$

Substituting (3.13), into (3.4) leads to the expression for the force

$$\mathbf{F}=\sum_n\sum_{n'}A_n^*A_{n'}\times[\epsilon_n^*\cdot\vec{\alpha}_0\cdot\epsilon_{n'}e^{i(\mathbf{K}_{n'}-\mathbf{K}_n)\cdot\mathbf{R}}i(\mathbf{K}_{n'}-\mathbf{K}_n)+\epsilon_n^*\cdot\vec{\alpha}_1\cdot\epsilon_{n'}e^{i(\mathbf{K}_{n'}-\mathbf{K}_n)\cdot\mathbf{R}}(\mathbf{K}_{n'}+\mathbf{K}_n)] . \quad (3.14)$$

This equation is equivalent to the result (3.8)–(3.11) for the total force. Obviously, the first term on the right-hand side of (3.14) is equal to the first term in (3.4). It disappears for $n=n'$, and it describes the redistribution forces $\mathbf{F}_2+\mathbf{F}_4$. This force is proportional to the momentum difference of a pair of modes, which illustrates its redistributive nature. The second term in (3.14) gives the scattering forces $\mathbf{F}_1+\mathbf{F}_3$, and it corresponds to the last term in (3.4). The work done by the field on the atomic dipole, which equals the absorbed power, can likewise be written as

$$W=2\omega\sum_n\sum_{n'}A_n^*A_{n'}\epsilon_n^*\cdot\vec{\alpha}_1\cdot\epsilon_{n'}e^{i(\mathbf{K}_{n'}-\mathbf{K}_n)\cdot\mathbf{R}} . \quad (3.15)$$

IV. SIMPLE EXAMPLES

In order to illustrate the separation (3.7) of the radiative force, we now consider a few simple examples corresponding to one or two plane waves.

A. Plane traveling wave

A single plane wave with wave vector $\mathbf{K}$ is described by the electric field

$$\mathbf{E}_+(\mathbf{R})=A_0\epsilon_0 e^{i\mathbf{K}\cdot\mathbf{R}} , \quad (4.1)$$

so that the amplitude and the polarization vector are

$$\mathbf{A}(\mathbf{R})=A_0e^{i\mathbf{K}\cdot\mathbf{R}},\quad \epsilon(\mathbf{R})=\epsilon_0 . \quad (4.2)$$

One notices that the only nonvanishing force is $\mathbf{F}_1$, and we obtain

$$\mathbf{F}=\mathbf{F}_1=2A_0A_0^*\mathbf{K}(\epsilon_0^*\cdot\vec{\alpha}_1\cdot\epsilon_0) , \quad (4.3)$$

which is the common radiation pressure.

B. Plane standing wave

A plane standing wave is characterized by the field

$$\mathbf{E}_+(\mathbf{R})=2A_0\epsilon_0\sin(\mathbf{K}\cdot\mathbf{R}) , \quad (4.4)$$

so that the amplitude and the polarization are

$$A(\mathbf{R})=2A_0\sin(\mathbf{K}\cdot\mathbf{R}),\quad \epsilon(\mathbf{R})=\epsilon_0 . \quad (4.5)$$

Now the only nonvanishing force is the dipole force $\mathbf{F}_2$, and we find

$$\mathbf{F}=\mathbf{F}_2=4A_0A_0^*\mathbf{K}(\epsilon_0^*\cdot\vec{\alpha}_0\cdot\epsilon_0)\sin(2\mathbf{K}\cdot\mathbf{R}) . \quad (4.6)$$

This result applies in the case of cooling in a standing wave with a transverse magnetic field [10,11]. When we are interested in the force averaged over a wavelength, we obviously need the average of $\vec{\alpha}_0\sin(2\mathbf{K}\cdot\mathbf{R})$. This corresponds to two terms of the Fourier series of the spatially varying polarizability.

C. Counterrunning plane waves with orthogonal polarizations

Our third example is the case of two counterrunning plane waves with equal intensity, and orthogonal polarizations. This is the standard situation for cooling by polarization gradients [12,13]. The field is then described by

$$\mathbf{E}_+(\mathbf{R})=A_0(\epsilon_1e^{i\mathbf{K}\cdot\mathbf{R}}+\epsilon_2e^{-i\mathbf{K}\cdot\mathbf{R}}) , \quad (4.7)$$

with

$$\epsilon_1^*\cdot\epsilon_2=0 , \quad (4.8)$$

so that

$$A(\mathbf{R})=A_0\sqrt{2},\quad \epsilon(\mathbf{R})=\frac{1}{\sqrt{2}}(\epsilon_1e^{i\mathbf{K}\cdot\mathbf{R}}+\epsilon_2e^{-i\mathbf{K}\cdot\mathbf{R}}) . \quad (4.9)$$

In this field the intensity and the phase are uniform, and only the polarization varies with the position. Hence the forces $\mathbf{F}_1$ and $\mathbf{F}_2$ vanish, and we obtain

$$\mathbf{F}_3=2A_0A_0^*\mathbf{K}(\epsilon_1^*\cdot\vec{\alpha}_1\cdot\epsilon_1-\epsilon_2^*\cdot\vec{\alpha}_1\cdot\epsilon_2) \quad (4.10)$$

and

$$\mathbf{F}_4=2A_0A_0^*\mathbf{K}i(\epsilon_2^*\cdot\vec{\alpha}_0\cdot\epsilon_1e^{2i\mathbf{K}\cdot\mathbf{R}}-\epsilon_1^*\cdot\vec{\alpha}_0\cdot\epsilon_2e^{-2i\mathbf{K}\cdot\mathbf{R}}) . \quad (4.11)$$

Equation (4.10) expresses $\mathbf{F}_3$ as the difference in absorption rates of photons from the two counterrunning plane waves. In the special case of two counterrunning waves with opposite circular polarization treated in Ref. [12], this force has been obtained as an unbalanced radiation pressure, resulting from motion-induced atomic orientation. Equation (4.11) is proportional to the probability of coherent photon transfer between the two plane running waves. This illustrates the redistributive nature of $\mathbf{F}_4$. The calculation of the spatial average of $\mathbf{F}_3$ requires the average of $\vec{\vec{\alpha}}_1$ over a wavelength. The average of $\mathbf{F}_4$ is determined by two coefficients of the Fourier expansion of $\vec{\vec{\alpha}}_0$. The Sisyphus-type force found in Ref. [12] in the case of two orthogonal linear polarizations is of the type $\mathbf{F}_4$.

In order to evaluate these forces, we have to calculate the polarizability tensor $\vec{\vec{\alpha}}$, which, according to (2.12), requires knowledge of the submatrices σ_{ee} and σ_{gg}. These submatrices are determined by their evolution equations (2.9).

V. LOW-INTENSITY LIMIT

The effect of optical pumping between the lower states on the cooling process is particularly important at low pumping rates. The low-intensity limit is valid when the Rabi frequencies, determined by the strength of the excitation operator $\mathcal{R}$, are small compared with Γ. Then it is sufficient to calculate the excited-state submatrix σ_{ee} to second order. Furthermore, this submatrix follows adiabatically the evolution of the lower-state submatrix σ_{gg}. We find from (2.9) for the excited-state submatrix

$$\sigma_{ee}=\frac{1}{\Gamma+iL}\left[\mathcal{R}\frac{1}{\Gamma/2+i\Delta+iL}(\sigma_{gg}\mathcal{R}^{\dagger})+\left[\frac{1}{\Gamma/2-i\Delta+iL}(\mathcal{R}\sigma_{gg})\right]\mathcal{R}^{\dagger}\right] . \tag{5.1}$$

If we substitute Eq. (5.1) into the evolution equation (2.9) for σ_{gg}, we obtain a closed evolution equation for the lower-state density matrix σ_{gg}.

For simplicity we make one further restriction for the explicit calculations in this paper. We only consider ground-state level splittings that are small compared with Γ. Then we can omit the Liouville operator L in the denominators in (5.1). In the case of hyperfine splitting, this assumption is usually not justified, but a Zeeman field can always be chosen weak enough to make the assumption valid. In this case the evolution equation for σ_{gg} takes the form

$$\frac{d}{dt}\sigma_{gg}=-iL\sigma_{gg}+\frac{\Gamma}{\Gamma^2/4+\Delta^2}\sum_{\beta}Q_{\beta}^{\dagger}\mathcal{R}\sigma_{gg}\mathcal{R}^{\dagger}Q_{\beta}-(\mathcal{P}+i\mathcal{L})\sigma_{gg}-\sigma_{gg}(\mathcal{P}-i\mathcal{L}) , \tag{5.2}$$

with the Hermitian operators $\mathcal{P}$ and $\mathcal{L}$ defined by

$$\mathcal{P}+i\mathcal{L}=\frac{1}{\Gamma/2-i\Delta}\mathcal{R}^{\dagger}\mathcal{R} . \tag{5.3}$$

Equation (5.2) is a closed evolution equation for the ground-state submatrix alone. It has the form of a generalized master equation. The free-evolution term $-iL$ describes the Zeeman precession, and it disappears in the absence of a magnetic field. The remaining terms describe the effect of optical pumping on the ground-state density matrix. The second term on the right-hand side of (5.2) describes the repopulation of the ground state following spontaneous emission. The operator $\hbar\mathcal{L}$ is an effective Hamiltonian, which has the light shifts as eigenvalues. The operator $\mathcal{P}$ describes the loss due to optical pumping. It is easy to check that $2\,\mathrm{Tr}\sigma_{gg}\mathcal{P}$ is the pumping rate or, equivalently, the fluorescence rate. Sometimes the non-Hermitian operator $\hbar(\mathcal{L}-i\mathcal{P})$ is called the effective Hamiltonian [17].

The expression (2.12) for the polarizability in the present limit of low intensity and low precession frequency takes the simplified form

$$\vec{\vec{\alpha}}=\frac{i}{\hbar}\frac{1}{\Gamma/2-i\Delta}\mathrm{Tr}\boldsymbol{\mu}_{ge}\boldsymbol{\mu}_{eg}\bar{\sigma}_{gg} . \tag{5.4}$$

The density matrix $\bar{\sigma}_{gg}$ refers to the steady-state situation. One should notice that even in the steady state, the density matrix, and therefore the polarizability tensor (5.4), depends on time, corresponding with the motion of the atom through the field. Alternatively, we can look upon the steady-state density matrix for a given velocity as a function of the position of the atom. The total force (3.2) or (3.4) on the atoms in this limit may be written as an average over the lower-state density matrix in the form

$$\mathbf{F}(t)=\mathrm{Tr}\bar{\sigma}_{gg}(t)\mathbf{f}_{\mathrm{eff}}(t) , \tag{5.5}$$

where

$$\mathbf{f}_{\mathrm{eff}}=\frac{-i\hbar}{\Gamma/2+i\Delta}\mathcal{R}^{\dagger}\nabla\mathcal{R}+\frac{i\hbar}{\Gamma/2-i\Delta}(\nabla\mathcal{R}^{\dagger})\mathcal{R} \tag{5.6}$$

serves as an effective force operator. The time dependence of this operator in (5.5) is determined by the local field at the position of the moving atom. It is easily shown that the redistribution part of the force can be expressed in the form of an average light-shift gradient in the form

$$\mathbf{F}_2+\mathbf{F}_4=-\hbar\,\mathrm{Tr}\bar{\sigma}_{gg}\nabla\mathcal{L} . \tag{5.7}$$

The scattering force $\mathbf{F}_1+\mathbf{F}_3$ cannot generally be described as the average over the ground state of a gradient.

The force $\mathbf{F}$ is generally a function of the velocity and the position of the atom. In practice, it is usually sufficient to substitute into the Fokker-Planck equation (1.3) the average of the force over a wavelength.

The absorbed power (3.12) can likewise be expressed as an expectation value over the ground-state density matrix. If we substitute (5.4) we obtain

$$W=\hbar\omega\frac{\Gamma}{\Gamma^2/4+\Delta^2}\mathrm{Tr}\bar{\sigma}_{gg}\mathcal{R}^{\dagger}\mathcal{R} . \tag{5.8}$$

VI. DIFFUSION TENSOR

In this section we wish to derive a complete expression for the diffusion tensor in the limit of weak velocity and

weak-field intensity. The diffusion tensor is defined by (1.5) as the integrated force autocorrelation function. The force operator arising from the external driving field is given in (3.1). Furthermore, there is a contribution from the coupling of the atomic dipole to the vacuum field. The corresponding term in the diffusion tensor reflects the random angular distribution of spontaneous emission [18,19]. Hence we separate the total diffusion tensor

$$\overleftrightarrow{D}=\overleftrightarrow{D}_{sp}+\overleftrightarrow{D}_{st} \tag{6.1}$$

into a spontaneous and a stimulated term. The first term takes the form [16]

$$\overleftrightarrow{D}_{sp}=\tfrac{1}{2}\int d\hat{\mathbf{n}}\, g(\hat{\mathbf{n}})(\hbar K)^2\hat{\mathbf{n}}\hat{\mathbf{n}} , \tag{6.2}$$

where

$$g(\hat{\mathbf{n}})=\frac{1}{\Gamma^2/4+\Delta^2}\frac{3\Gamma}{8\pi}\sum_{\mathbf{u}\perp\hat{\mathbf{n}}}\mathrm{Tr}\mathbf{u}^*\cdot\mathbf{Q}^\dagger\mathcal{R}\bar{\sigma}_{gg}\mathcal{R}^\dagger\mathbf{Q}\cdot\mathbf{u} \tag{6.3}$$

is the rate of spontaneous emission per unit solid angle in the direction of the unit vector $\hat{\mathbf{n}}$. The summation in (6.3) runs over two independent polarization directions orthogonal to the emission direction $\hat{\mathbf{n}}$. The vector operator $\mathbf{Q}$ has the operators Q_β as its spherical components, so that its Cartesian components are [16]

$$Q_x=\frac{-1}{\sqrt{2}}(Q_1-Q_{-1}),\quad Q_y=\frac{-1}{i\sqrt{2}}(Q_1+Q_{-1}),\quad Q_z=Q_0 . \tag{6.4}$$

In order to evaluate the stimulated contribution to (6.1), we need to determine the autocorrelation functions of the force operator (3.1), substitute the result into (1.5), and calculate the integral to second order in the field amplitude. The correlation functions can be expressed in terms of an evolution operator U, which expresses the solution of the evolution equations (2.4) as

$$\sigma(t+\tau)=U(t+\tau,t)\sigma(t) . \tag{6.5}$$

For convenience, we introduce an abbreviated notation for the correlation functions in the form

$$\begin{aligned}\overleftrightarrow{C}_+(\tau)&\equiv\langle\mathbf{f}(t)\mathbf{f}(t+\tau)\rangle=\mathrm{Tr}\,\mathbf{A}_+(\tau)\mathbf{f}(t+\tau) ,\\ \overleftrightarrow{C}_-(\tau)&\equiv\langle\mathbf{f}(t+\tau)\mathbf{f}(t)\rangle=\mathrm{Tr}\mathbf{f}(t+\tau)\mathbf{A}_-(\tau) ,\end{aligned} \tag{6.6}$$

where the vector operators $\mathbf{A}_\pm$ are defined by the relations

$$\begin{aligned}\mathbf{A}_+(\tau)&=U(t+\tau,t)[\bar{\sigma}(t)\mathbf{f}(t)] ,\\ \mathbf{A}_-(\tau)&=U(t+\tau,t)[\mathbf{f}(t)\bar{\sigma}(t)] .\end{aligned} \tag{6.7}$$

The force operators $\mathbf{f}$ in (6.6) and (6.7) are defined in (3.1), and they have an explicit time dependence that is imposed by the motion of the atom as given by (2.6). We have suppressed the dependence of the correlation functions $\overleftrightarrow{C}_\pm$ and the operators $\mathbf{A}_\pm$ on the time t.

We wish to emphasize that in order to calculate the diffusion tensor to second order in the field amplitude, it is not sufficient to evaluate the correlation functions themselves to second order. This is obvious once we distinguish the rapid decay of the correlation function, which is governed by the spontaneous-decay rate Γ, from the slow decay determined by optical pumping. This slow decay time is of the order of the inverse of the pumping rate, which itself is of second order. Hence we must calculate the long-time tail of the correlation function to fourth order. Therefore we have to evaluate $\mathbf{A}_\pm(\tau)$ to first order for times τ of the order of Γ^{-1}, and to third order for larger values of τ. The separation of time scales of the operators $\mathbf{A}_\pm$ leads to a separation of the correlation functions (6.6) into a short-time and a long-time term, according to

$$\overleftrightarrow{C}_\pm(\tau)=\overleftrightarrow{C}_\pm^{(s)}(\tau)+\overleftrightarrow{C}_\pm^{(1)}(\tau) . \tag{6.8}$$

The first term on the right-hand side of (6.8) is of second order in the field amplitude, and decays to zero on the rapid time scale Γ^{-1}. The last term in (6.8) is of fourth order, and its decay rate is the pumping rate. The separation of the correlation function into a rapidly decaying part of second order and a slowly decaying part of fourth order is shown in Fig. 2. After substitution into (1.5), this separation of the correlation function gives rise to a corresponding separation of the diffusion tensor in the form

$$\overleftrightarrow{D}_{st}=\overleftrightarrow{D}_{st}^{(s)}+\overleftrightarrow{D}_{st}^{(1)} , \tag{6.9}$$

where both terms are of second order.

The first-order term in $\mathbf{A}_+$ arises from the zeroth-order evolution operator U. Hence the only nonvanishing submatrix is $\mathbf{A}_{+ge}$, which decays by spontaneous emission only. On this rapid time scale, the atomic motion is negligible because of the low-velocity limit. It is this term which gives the rapidly decaying part of the correlation function in the form

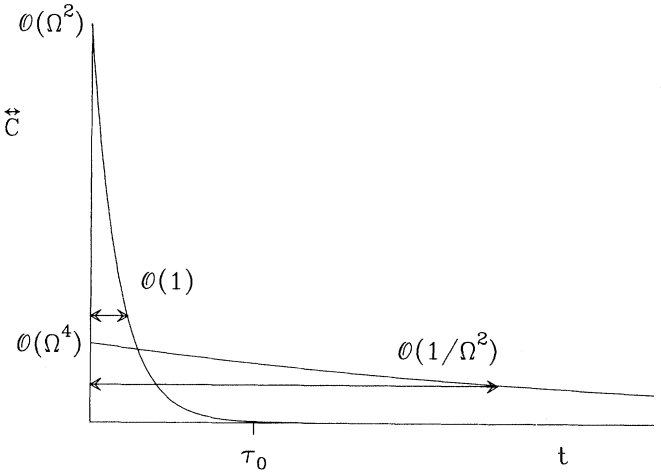

FIG. 2. Sketch of the separation of the force correlation function. The rapidly decaying part is of second order in the Rabi frequency, and the slowly decaying part is of fourth order. The integral of both parts is of second order. The length of the time tail of the rapidly decaying part is of zeroth order, and is indicated by the short arrow. The slowly decaying part has a time tail of order Ω^{-2}, which is represented by the long arrow.

$$\overleftrightarrow{C}_{+}^{(s)}(\tau)=\exp\left[-\left[\frac{\Gamma}{2}+i\Delta\right]\tau\right]\mathrm{Tr}\bar{\sigma}_{gg}(t)\nabla\mathcal{R}^{\dagger}(t)\nabla\mathcal{R}(t+\tau)\ , \tag{6.10}$$

Likewise, we obtain

$$\overleftrightarrow{C}_{-}^{(s)}(\tau)=\exp\left[-\left[\frac{\Gamma}{2}-i\Delta\right]\tau\right]\mathrm{Tr}\nabla\mathcal{R}^{\dagger}(t+\tau)\nabla\mathcal{R}(t)\bar{\sigma}_{gg}(t)\ . \tag{6.11}$$

The corresponding contribution to the stimulated diffusion tensor is found to be

$$\overleftrightarrow{D}_{\mathrm{st}}^{(s)}=\frac{\hbar^2\Gamma/2}{\Gamma^2/4+\Delta^2}\mathrm{Tr}\bar{\sigma}_{gg}(t)\nabla\mathcal{R}^{\dagger}(t)\nabla\mathcal{R}(t)\ . \tag{6.12}$$

This contribution to the momentum diffusion arises from the fluctuations in the number of spontaneous emissions. It is fully determined by the steady-state ground-state density matrix at the position of the atom.

The evaluation of the long-time contribution to $\overleftrightarrow{D}_{\mathrm{st}}$ is more delicate. From Eq. (6.6) for the correlation function $\overleftrightarrow{C}_{+}$ and Eq. (3.1) for the force operator, it is obvious that we only need the *eg* and *ge* submatrix of $\mathbf{A}_{+}$. We consider a time τ_0 that is long compared with the spontaneous lifetime, but still short compared with the pumping time. Then these submatrices already follow the submatrix $\mathbf{A}_{+gg}$ adiabatically, in the same way as the submatrices σ_{eg} and σ_{ge} follow σ_{gg}, as discussed in Sec. V. It is therefore sufficient to evaluate $\mathbf{A}_{+gg}$. But since τ_0 is still short compared with the pumping time, the only evolution of $\mathbf{A}_{+}$ that has taken place since $\tau=0$ is spontaneous decay. Therefore, at this intermediate time τ_0, the submatrix $\mathbf{A}_{+gg}$ is given by

$$\mathbf{A}_{+gg}(\tau_0)=\mathbf{A}_{+gg}(0)+\sum_{\beta}Q_{\beta}^{\dagger}\mathbf{A}_{+ee}(0)Q_{\beta}\ , \tag{6.13}$$

where the last term is the contribution that the ground state has gained since $\tau=0$ from spontaneous decay. The left-hand side of (6.13) is evaluated explicitly if we substitute

$$\begin{aligned}\mathbf{A}_{+gg}(0)&=\bar{\sigma}_{ge}(t)\mathbf{f}_{eg}(t)\\&=\frac{-i\hbar}{\Gamma/2+i\Delta}\bar{\sigma}_{gg}(t)\mathcal{R}^{\dagger}(t)\nabla\mathcal{R}(t)\ ,\\\mathbf{A}_{+ee}(0)&=\bar{\sigma}_{eg}(t)\mathbf{f}_{ge}(t)\\&=\frac{i\hbar}{\Gamma/2-i\Delta}\mathcal{R}(t)\bar{\sigma}_{gg}(t)\nabla\mathcal{R}^{\dagger}(t)\ .\end{aligned} \tag{6.14}$$

On the slow time scale, the evolution of $\mathbf{A}_{+gg}$ is determined by the same evolution equation (5.2) as σ_{gg}. Hence we introduce the evolution operator G, which describes the solution of (5.2) as

$$\sigma_{gg}(t+\tau)=G(t+\tau,t)\sigma_{gg}(t)\ . \tag{6.15}$$

Then for values of τ that are large compared to Γ^{-1}, we obtain to a good approximation

$$\mathbf{A}_{+gg}(\tau)=G(t+\tau,t)\left[\mathbf{A}_{+gg}(0)+\sum_{\beta}Q_{\beta}^{\dagger}\mathbf{A}_{+ee}(0)Q_{\beta}\right]\ , \tag{6.16}$$

since the value (6.13) for $\mathbf{A}_{+gg}(\tau_0)$ may serve as the initial condition for the evolution on the slow time scale. The *eg* and *ge* submatrices of $\mathbf{A}_{+}(\tau)$ follow $\mathbf{A}_{+gg}(\tau)$ adiabatically, so that

$$\begin{aligned}\mathbf{A}_{+eg}(\tau)&=\frac{i}{\Gamma/2-i\Delta}\mathcal{R}(t+\tau)\mathbf{A}_{+gg}(\tau)\ ,\\\mathbf{A}_{+ge}(\tau)&=\frac{-i}{\Gamma/2+i\Delta}\mathbf{A}_{+gg}(\tau)\mathcal{R}^{\dagger}(t+\tau)\ .\end{aligned} \tag{6.17}$$

These submatrices are of third order in the field amplitude. If we substitute successively (6.17), (6.16), and (6.14) into (6.6), we obtain the lengthy but explicit expression for the long-time behavior of the correlation function $\mathbf{C}_{+}(\tau)$ for $\tau\gg\Gamma^{-1}$:

$$\overleftrightarrow{C}_{+}^{(1)}(\tau)=\hbar\mathrm{Tr}\left[G(t+\tau,t)\left[\frac{-i}{\Gamma/2+i\Delta}\bar{\sigma}_{gg}(t)\mathcal{R}^{\dagger}(t)\nabla\mathcal{R}(t)+\frac{i}{\Gamma/2-i\Delta}\sum_{\beta}Q_{\beta}^{\dagger}\mathcal{R}(t)\bar{\sigma}_{gg}(t)\nabla\mathcal{R}^{\dagger}(t)Q_{\beta}\right]\right]\mathbf{f}_{\mathrm{eff}}(t+\tau)\ , \tag{6.18}$$

where the effective force operator is defined in (5.6). This is a correlation function that is fully governed by the ground-state evolution operator G.

A similar treatment leads to an explicit result for the long-time behavior of the correlation function $\overleftrightarrow{C}_{-}(\tau)$ in the form

$$\overleftrightarrow{C}_{-}^{(1)}(\tau)=\hbar\mathrm{Tr}\mathbf{f}_{\mathrm{eff}}(t+\tau)G(t+\tau,t)\left[\frac{i}{\Gamma/2-i\Delta}\nabla\mathcal{R}^{\dagger}(t)\mathcal{R}(t)\bar{\sigma}_{gg}(t)+\frac{-i}{\Gamma/2+i\Delta}\sum_{\beta}Q_{\beta}^{\dagger}\nabla\mathcal{R}(t)\bar{\sigma}_{gg}(t)\mathcal{R}^{\dagger}(t)Q_{\beta}\right]\ . \tag{6.19}$$

The long-time correlation functions (6.18) and (6.19) are not quite identical to autocorrelation functions of the effective force operator $\mathbf{f}_{\mathrm{eff}}$, defined in Eq. (5.6), in terms of the pure ground-state evolution operator G. This is basically due to the fact that the actual force operator $\mathbf{f}$ is not restricted to the ground state. Physically speaking, this implies that the long-time correlation function of the force due to the external field is interrupted by spontaneous emission. However, it is possible to express (6.18) and (6.19) in a form that looks like a standard correlation

function if we introduce the vector operators $\mathbf{\Phi}_{\pm}$, which operate on ground-state density matrices, and which are defined by the equalities

$$\mathbf{\Phi}_{+}\sigma_{gg}=\hbar\left[\frac{-i}{\Gamma/2+i\Delta}\sigma_{gg}\mathcal{R}^{\dagger}\nabla\mathcal{R}+\frac{i}{\Gamma/2-i\Delta}\sum_{\beta}Q_{\beta}^{\dagger}\mathcal{R}\sigma_{gg}\nabla\mathcal{R}^{\dagger}Q_{\beta}\right],$$
$$\mathbf{\Phi}_{-}\sigma_{gg}=\hbar\left[\frac{i}{\Gamma/2-i\Delta}\nabla\mathcal{R}^{\dagger}\mathcal{R}\sigma_{gg}+\frac{-i}{\Gamma/2+i\Delta}\sum_{\beta}Q_{\beta}^{\dagger}\nabla\mathcal{R}\sigma_{gg}\mathcal{R}^{\dagger}Q_{\beta}\right]. \tag{6.20}$$

These operators reproduce the average force by the identities

$$\mathrm{Tr}\mathbf{\Phi}_{+}\sigma_{gg}=\mathrm{Tr}\mathbf{\Phi}_{-}\sigma_{gg}=\mathrm{Tr}\sigma_{gg}\mathbf{f}_{\mathrm{eff}}\ . \tag{6.21}$$

Then the correlation functions (6.18) and (6.19) may be expressed in terms of their matrix elements as

$$C_{+ij}^{(1)}(\tau)=\mathrm{Tr}\mathbf{\Phi}_{+j}(t+\tau)G(t+\tau,t)[\mathbf{\Phi}_{+i}(t)\bar{\sigma}_{gg}(t)]\ ,$$
$$C_{-ij}^{(1)}(\tau)=\mathrm{Tr}\mathbf{\Phi}_{-i}(t+\tau)G(t+\tau,t)[\mathbf{\Phi}_{-j}(t)\bar{\sigma}_{gg}(t)]\ . \tag{6.22}$$

The combined mechanism of adiabatic following on the slow time scale and rapid spontaneous decay mixes the order of multiplication in the evaluation of the long-time correlation function. The complete separation of the diffusion coefficient into a contribution exclusively determined by spontaneous decay and a term resulting from the correlation function of the effective force in the ground state [12] seems not to be justified in general.

The long-time correlation functions (6.18) and (6.19) give rise to a contribution to the momentum diffusion tensor to second order in the field amplitudes, even though these correlation functions themselves are of fourth order. The result for the long-time part of the diffusion tensor is

$$\overleftrightarrow{\mathbf{D}}_{\mathrm{st}}^{(1)}=\tfrac{1}{2}\int_{0}^{\infty}d\tau[\overleftrightarrow{\mathbf{C}}_{+}^{(1)}(\tau)-\mathbf{F}(t)\mathbf{F}(t+\tau)+\overleftrightarrow{\mathbf{C}}_{-}^{(1)}(\tau)-\mathbf{F}(t+\tau)\mathbf{F}(t)]\ , \tag{6.23}$$

where the one-time averages $\mathbf{F}=\langle\mathbf{f}\rangle$ are defined as in (5.5). It is easy to check that the integrand in (6.23) decays to zero for large values of τ. This results from the fact that the effective evolution operator G drives an arbitrary operator to the steady-state density matrix, so that

$$\lim_{\tau\to\infty}G(t+\tau,t)B=\bar{\sigma}_{gg}(t+\tau)\mathrm{Tr}B\ . \tag{6.24}$$

The contribution (6.23) has the significance of the momentum fluctuations arising from the state fluctuations over the ground-state sublevels.

The total stimulated diffusion tensor is now obtained by substituting (6.12) and (6.23) into (6.9). Together with the spontaneous contribution (6.2), this specifies the total diffusion tensor (6.1). Each one of these terms is of second order in the field amplitude. The terms (6.2) and (6.12) are fully determined by the instantaneous ground-state density matrix together with the local field. The term (6.23) contains an integration over the slow decay to the steady state, and a typical decay time is of the order of the pumping time.

These terms have a clear physical significance. The part (6.2) describes the momentum fluctuations corresponding to the random direction of spontaneous emission. The term (6.12) corresponds to the fluctuations in the number of absorptions (or the number of spontaneous emissions). The term (6.23) describes the force fluctuations arising from the state fluctuations within the ground-state manifold.

Each term in the diffusion tensor is a function of the velocity and the position of the atom. The position dependence may be eliminated by averaging each term over a wavelength, and the result can be substituted in the Fokker-Planck equation (1.3).

VII. INTENSITY SCALING

The evolution equation (5.2) for the ground-state density matrix in the low-intensity limit has the formal structure

$$\frac{d\sigma_{gg}}{dt}=W(\mathbf{B},I,\mathbf{R}(t))\sigma_{gg}(t)\ , \tag{7.1}$$

where the evolution operator W is defined by the right-hand side of (5.2). In (7.1) we explicitly indicate the dependence of the evolution on the magnetic field $\mathbf{B}$ and the intensity I. The Zeeman evolution operator L in (5.2) is simply proportional to $\mathbf{B}$, and the remaining terms on the right-hand side are proportional to the field intensity. Multiplying the intensity I and the magnetic field $\mathbf{B}$ by a factor of ζ has the effect that the total evolution operator is multiplied by ζ. Furthermore, we emphasize that the operator W depends on time only due to the motion of the atom with velocity $\mathbf{v}$. This has the result that scaling down the magnetic field, the intensity, and the velocity by a factor ζ implies that the entire time evolution of σ_{gg} is scaled down by the same factor, so that

$$\sigma_{gg}(\zeta\mathbf{B},\zeta I,\zeta\mathbf{v},t)=\sigma_{gg}(\mathbf{B},I,\mathbf{v},\zeta t)\ . \tag{7.2}$$

This is directly checked by taking the time derivative of the left-hand side of (7.2), while using

$$W(\zeta\mathbf{B},\zeta I,\mathbf{R})=\zeta W(\mathbf{B},I,\mathbf{R})\ . \tag{7.3}$$

Equation (7.2) implies that the atomic evolution for a scaled-down intensity, magnetic field, and velocity is the original evolution in slow motion.

From (7.2) it is easy to show that the average force and diffusion tensor obey the scaling laws

$$\mathbf{F}(\zeta\mathbf{B},\zeta I,\zeta\mathbf{v})=\zeta\mathbf{F}(\mathbf{B},I,\mathbf{v}) \tag{7.4}$$

and

$$\overleftrightarrow{D}(\zeta\mathbf{B},\zeta I,\zeta\mathbf{v})=\zeta\overleftrightarrow{D}(\mathbf{B},I,\mathbf{v})\ . \tag{7.5}$$

These equations indicate that the steady-state distribution function gets narrower by a factor of the order of $\sqrt{\zeta}$ if we lower the intensity. This is easily illustrated in the case where the velocity width is sufficiently low, so that over this width the force obeys the simple damping law (1.6), and the diffusion tensor varies negligibly with velocity. Then it follows from (7.4) that the damping constant γ is independent of the scaling parameter ζ, whereas the diffusion coefficient D is proportional to ζ. Then Eq. (1.7) proves that the steady-state temperature T is proportional to ζ, so that lowering the intensity and the external field leads to a correspondingly lower temperature. Obviously, the region of validity of the damping law scales down with ζ as well. Furthermore, this argument loses its validity for temperatures of the order of the recoil limit (1.2), where the Fokker-Planck equation (1.3) is no longer valid.

The fact that the steady-state temperature goes down linearly with the intensity has been shown before in special cases [12,10]. The present argument shows the general validity of this conclusion. Obviously, this mechanism for reaching temperatures below the Doppler limit requires that there are values of the velocity where the force vanishes, and where the derivative of the force with respect to the velocity is negative. These conditions require either polarization gradients or ground-state level splitting. In the absence of level splitting we can omit the operator L in (5.2). For a uniform polarization, the remaining terms on the right-hand side of (5.2) depend on time only through their proportionality with the local intensity at the position of the moving atom. Then the steady-state density matrix which solves (5.2) is independent of the position. Then the average force as evaluated according to (5.5) varies with the local intensity, but it does not depend on velocity, so that cooling cannot occur.

We conclude that cooling below the Doppler limit requires a steady-state solution of (5.2) that depends on position. This in turn requires either ground-state level splitting or polarization gradients. A level splitting causes a precession of the coherences between ground-state sublevels, whereas the optical-pumping terms in (5.2) try to restore a steady state. The balance between these two effects depends in a sensitive way on the ratio of the precession time and the time needed to travel a wavelength, and this corresponds to a sensitive dependence of the position-dependent density matrix on the velocity. The resulting velocity dependence of the average force then leads to cooling around velocity values where the force is zero.

On the other hand, when the polarization varies over a wavelength, the local field tends to drive the density matrix to a steady state that is different for different positions. The effective time variation of the evolution operator due to the atomic motion now induces a nonadiabatic coupling. The ground-state density matrix and the average force will therefore depend in a sensitive way on the atomic velocity, which again will lead to cooling around velocity values where the force vanishes. In both cases it is essential that the ground state consists of more than one substate.

VIII. TWO COUNTERPROPAGATING WAVES

We wish to present the results of numerical calculations of the force in the one-dimensional case of two counterpropagating plane waves with the same amplitude. In the case of a standing wave, where the two waves have the same polarization, sub-Doppler cooling can only occur in the presence of a level splitting. In this case we include a transverse magnetic field along the X direction, so that the Liouville operator L is defined by

$$-iL\sigma_{gg}=-\frac{i}{\hbar}\omega_L[J_X,\sigma_{gg}]\ , \tag{8.1}$$

where ω_L is the Larmor precession frequency. In the case of a standing wave with a transverse magnetic field, parity invariance shows that the average force is an odd function of the velocity. This implies that the force passes zero for zero velocity.

In the complementary case of two counterrunning waves with equal amplitudes and orthogonal polarizations, and in the absence of external dc fields, the radiation field is invariant for rotation over π around an axis normal to the propagation direction. This demonstrates again that the average force is an odd function of the velocity.

We take the Z axis along the propagating direction, so that the atom-field coupling operator (2.3) takes the form

$$\mathcal{R}(Z)=\mathcal{R}_1 e^{iKZ}+\mathcal{R}_2 e^{-iKZ}\ , \tag{8.2}$$

where the Rabi operators $\mathcal{R}_1$ and $\mathcal{R}_2$ do not depend on position. The force (5.5) is then

$$F=\frac{\hbar K}{\Gamma^2/4+\Delta^2}\mathrm{Tr}\bar{\sigma}_{gg}[\Gamma(\mathcal{R}_1^\dagger\mathcal{R}_1-\mathcal{R}_2^\dagger\mathcal{R}_2)+2i\Delta(\mathcal{R}_1^\dagger\mathcal{R}_2 e^{-2iKZ}-\mathcal{R}_2^\dagger\mathcal{R}_1 e^{2iKZ})]\ , \tag{8.3}$$

and the absorbed field power is

$$W=\frac{\hbar\omega\Gamma}{\Gamma^2/4+\Delta^2}\mathrm{Tr}\bar{\sigma}_{gg}(\mathcal{R}_1^\dagger\mathcal{R}_1+\mathcal{R}_2^\dagger\mathcal{R}_2+\mathcal{R}_1^\dagger\mathcal{R}_2 e^{-2iKZ}+\mathcal{R}_2^\dagger\mathcal{R}_1 e^{2iKZ})\ . \tag{8.4}$$

Since the intensities of the two plane waves are equal, we can write

$$\mathcal{R}_1=\tfrac{1}{2}\Omega\boldsymbol{\epsilon}_1\cdot\mathbf{Q},\quad \mathcal{R}_2=\tfrac{1}{2}\Omega\boldsymbol{\epsilon}_2\cdot\mathbf{Q}\ , \tag{8.5}$$

where $\boldsymbol{\epsilon}_1$ and $\boldsymbol{\epsilon}_2$ are the polarization vectors, and Ω is the effective Rabi frequency, which measures the strength of the atom-field coupling. In (8.3) one notices the distinction between the scattering and the redistribution contribution to the force. The scattering part is the photon momentum times the difference in absorption rate for the two beams. The redistribution part of (8.3) in units of the photon momentum is twice the net redistribution rate of photons from beam 1 to beam 2. For a standing wave, where the two polarization vectors are equal, and $\mathcal{R}_1=\mathcal{R}_2$, the scattering term vanishes, and (8.3) is a special case of (3.9). For two orthogonal polarizations, (8.3) corresponds to the sum of (3.10) and (3.11).

The symmetry of the force for reversal of the detuning Δ can be studied by applying the time-reversal invariance to the evolution equations (5.2). The antiunitary time-reversal operators θ is defined as usual by [20]

$$\theta|JM\rangle=(-1)^{J-M}|J-M\rangle \ . \tag{8.6}$$

Then for a steady-state solution $\bar{\sigma}_{gg}(t)$ of (5.2) with $\mathcal{R}$ defined by (8.2) and (8.5), the time-reversed density matrix $\theta^{\dagger}\bar{\sigma}_{gg}(t)\theta$ is the solution for the situation with the velocity and a magnetic field reversed, with the polarization vectors replaced by their complex conjugates, and with opposite detuning. This gives for the average force the general relation

$$\bar{F}(\mathbf{B},v,\boldsymbol{\epsilon}_1,\boldsymbol{\epsilon}_2,\Delta)=\bar{F}(-\mathbf{B},-v,\boldsymbol{\epsilon}_1^*,\boldsymbol{\epsilon}_2^*,-\Delta) \ . \tag{8.7}$$

In the two cases of a standing wave with a transverse magnetic field, and of two counterrunning waves with equal amplitude and orthogonal polarizations, we conclude that the force is an odd function both of the velocity and the detuning. Hence a positive derivative of the force with respect to the velocity can always be made negative by inverting the detuning.

In the steady state in the periodic radiation field, the density matrix can be expanded in a Fourier series:

$$\bar{\sigma}_{gg}=\sum_{n=-\infty}^{\infty}\sigma_n e^{2inKZ} \ , \tag{8.8}$$

where the matrices σ_n obey a set of coupled equations that can be derived by substituting (8.8) into (5.2), with $Z=vt$. The force (8.3) averaged over a wavelength is then found to be

$$\bar{F}=\frac{\hbar K}{\Gamma^2/4+\Delta^2}[\Gamma \,\mathrm{Tr}\sigma_0(\mathcal{R}_1^{\dagger}\mathcal{R}_1-\mathcal{R}_2^{\dagger}\mathcal{R}_2) +2i\Delta(\mathrm{Tr}\sigma_1\mathcal{R}_1^{\dagger}\mathcal{R}_2-\mathrm{Tr}\sigma_{-1}\mathcal{R}_2^{\dagger}\mathcal{R}_1)] \ . \tag{8.9}$$

Explicit expressions for the diffusion tensor (6.1) follow by substituting (8.2) into the equations (6.3), (6.12), (6.18), and (6.19). The stimulated contribution has $D_{\mathrm{sp},zz}$ as the only nonvanishing component, which corresponds to broadening of the distribution of the velocity component in the propagation direction. The spontaneous contribution to the diffusion tensor leads to broadening of all velocity components. Hence transverse heating is exclusively due to the random directions of spontaneous emission.

We have numerically solved the coupled equations for the Fourier components σ_n of the steady-state density matrix, in various specific cases of polarization directions and values of the atomic angular momenta, with or without an external static transverse magnetic field, and as a function of the velocity v in the Z direction. The Fourier components allow us to evaluate the average force. For convenience we introduce the dimensionless force

$$\phi=\frac{\Gamma\bar{F}}{\hbar K\Omega^2} \tag{8.10}$$

and the dimensionless velocity

$$w=\frac{\Gamma K v}{\Omega^2} \ . \tag{8.11}$$

The strength of the magnetic field is described by

$$b=\frac{\Gamma\omega_L}{\Omega^2} \ . \tag{8.12}$$

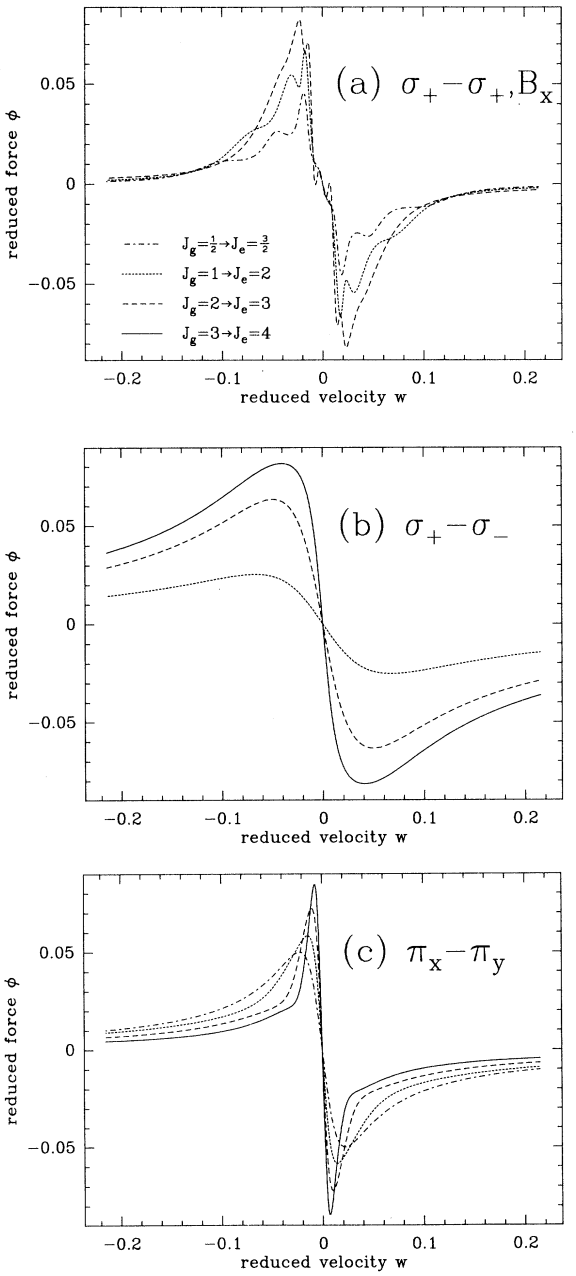

FIG. 3. Reduced force as a function of the reduced velocity for three different configurations and four values of the lower-state angular momentum J_g, and with the excited-state angular momentum $J_e=J_g+1$. The detuning is $\Delta=-1.5\Gamma$ in these (and all subsequent) curves. (a) Circularly polarized standing wave (σ_+) with transverse magnetic field with reduced strength $b=0.05$; (b) counterpropagating waves with opposite circular polarization ($\sigma_+-\sigma_-$); (c) counterpropagating waves with orthogonal linear polarization ($\pi_X-\pi_Y$).

The use of these quantities is that the reduced force ϕ as a function of the reduced velocity w (and the reduced magnetic field b, if present) is independent of the intensity and of the oscillator strength of the atomic transition.

In Fig. 3 we display the reduced force for various configurations and for a few values of J_g and $J_e = J_g + 1$. One notices that the force tends to be larger for larger angular momenta. A standing wave with a transverse magnetic field gives a much richer structure than does the case of counterpropagating waves with orthogonal polarizations. Cooling to zero velocity occurs in all displayed cases. Figure 4 shows the force for various

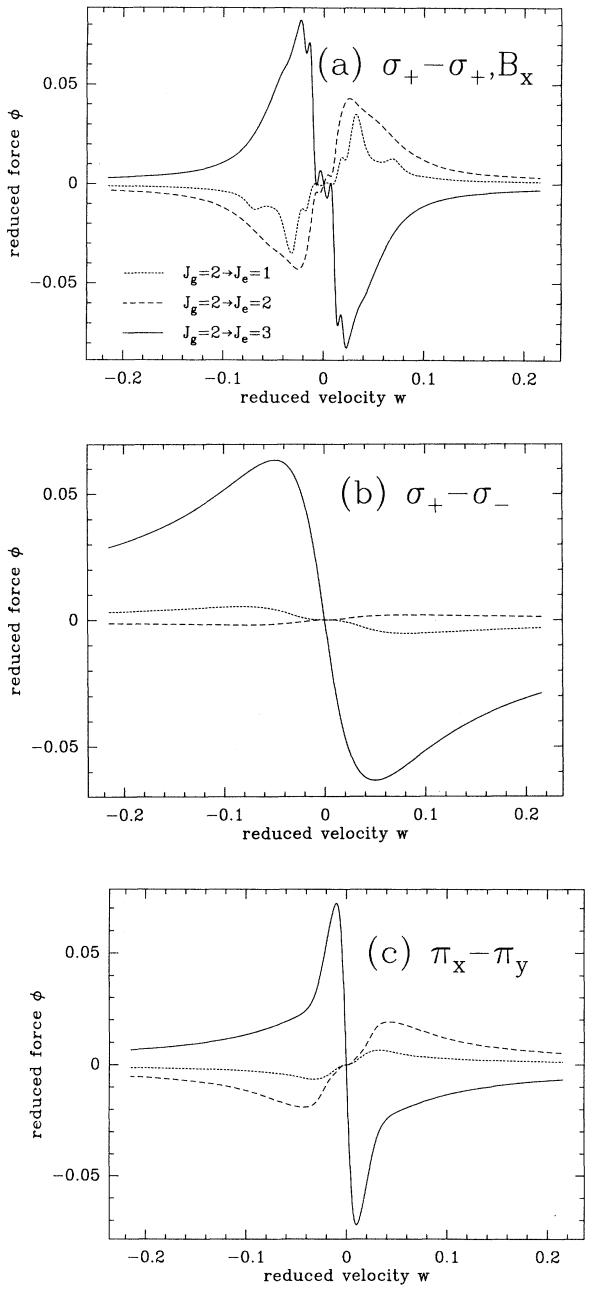

FIG. 4. Reduced force as a function of the reduced velocity for three different configurations, for detuning $\Delta = -1.5\Gamma$, and for a fixed value of the lower-state angular momentum $J_g = 2$. The configurations in (a)–(c) are the same as in Fig. 3.

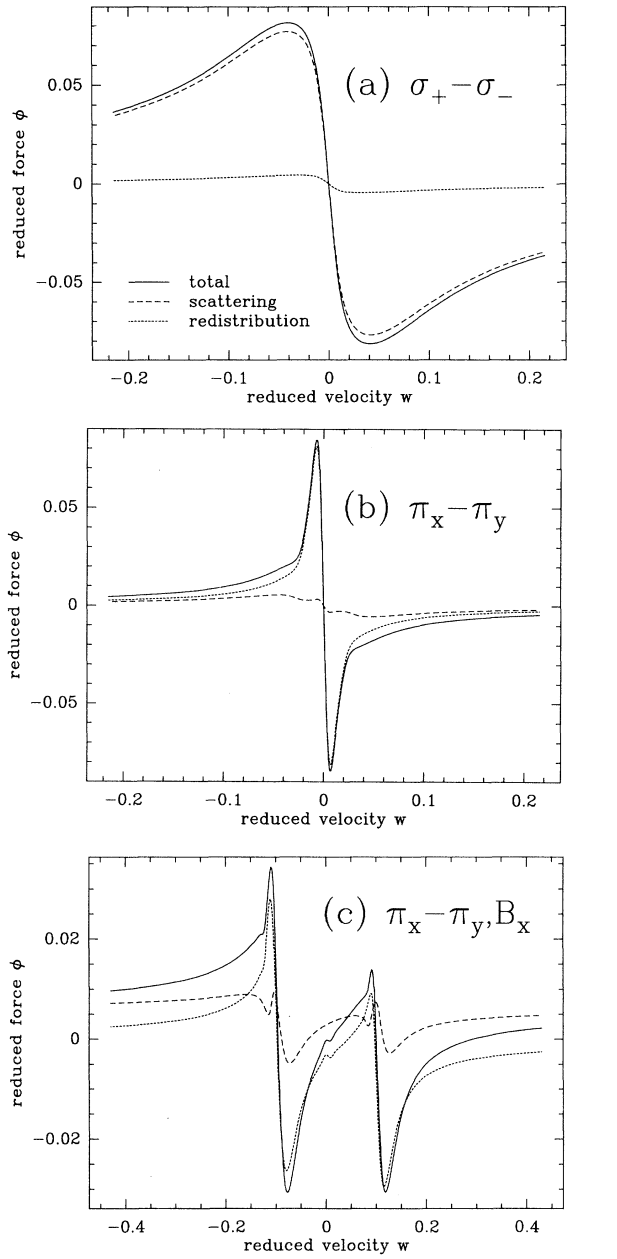

FIG. 5. Reduced force as a function of the reduced velocity for three different configurations for the angular momentum values $J_g = 3$ and $J_e = 4$, and for detuning $\Delta = -1.5\Gamma$. The total force is shown along with the scattering force F_3 and the redistribution force F_4. (a) Counterpropagating waves with opposite circular polarization ($\sigma_+ - \sigma_-$); (b) counterpropagating waves with orthogonal linear polarization ($\pi_X - \pi_Y$); (c) counterpropagating waves with orthogonal linear polarization ($\pi_X - \pi_Y$), and a transverse magnetic field in the X direction, with reduced strength $b = 0.2$.

configurations for a fixed value of $J_g=2$ and different values of J_e. The sign of the derivative of the force at zero velocity tends to reverse with J_e-J_g. For the cases (b) and (c) of counterrunning waves with orthogonal polarizations without a magnetic field, this derivative is appreciably smaller for $J_e=J_g$ or J_g-1 than in the case where $J_e=J_g+1$. Figure 5 presents the total force along with the scattering part F_3 and the redistribution part F_4 in a few cases of counterrunning waves with orthogonal polarizations for angular momenta $J_g=3$ and $J_e=4$. Figure 5(a) shows that for opposite circular polarizations, the force is mainly due to scattering, whereas it is obvious from Fig. 5(b) that orthogonal linear polarizations create dominantly a redistribution force. Figure 5(c) shows the case of orthogonal linear polarizations and a magnetic field along one of the polarization directions. This is the only displayed case where the force is not an odd function of the detuning and the velocity. Still it follows from Eq. (8.7) that the force is invariant for inversion of both Δ and w.

Calculations of the average force as a function of velocity were presented for a few specific configurations and intensities in Refs. [12] and [13]. Our results demonstrate the general applicability of the formalism presented in this paper, and the scaling properties of the force. Moreover, the composition of the total force as resulting from a combination of scattering and redistribution of photons is clearly displayed by our results.

IX. CONCLUSIONS

When an atom moves with a low velocity through an arbitrary monochromatic radiation field, the instantaneous atomic dipole is related to the local field by an effective polarizability tensor. This result, which is expressed in Eqs. (2.11) and (2.12), is valid when the time needed to traverse a wavelength is long compared with the atomic lifetime, so that the optical coherences can be adiabatically eliminated. The atomic levels coupled by the monochromatic field may consist of an arbitrary number of degenerate or nearly degenerate substates. The total radiative force on the atom may be generally separated into a sum of four terms, as shown in (3.7)–(3.11). Apart from the radiation pressure and the dipole force, we obtain two terms that depend on the gradient of the polarization. One of these latter terms contains the anti-Hermitian part of the polarizability and corresponds to fluorescent scattering of absorbed photons. The other term arises from the redistribution of photons between the plane waves that compose the radiation field, and it depends on the Hermitian part of the polarizability.

Several recent methods for cooling below the Doppler limit require a radiation field at low intensity [8–13]. In this case the atomic polarizability depends exclusively on the density matrix for the lower state of the atom. We derive a general closed evolution equation (5.2) for this density matrix. The force can be written as an average of an effective force operator (5.6) over the lower-state density matrix. The diffusive heating of the velocity distribution is determined by a diffusion tensor, which we represent as a sum of three terms. The first term (6.2) expresses the force fluctuations due to the random directions of spontaneous emission, and the second term (6.12) results from the fluctuations in the number of absorbed photons. Both terms are determined by the instantaneous lower-state density matrix. The third term (6.23) contains the slow evolution of the force correlation function, and it arises from the fluctuations within the substates of the lower state. The evolution of the atomic velocity distribution in the present semiclassical approximation is fully determined by the average force and the diffusion tensor, which specify the Fokker-Planck equation (1.3).

By inspection of the scaling properties of the evolution equation (5.2) we show that for a transition between two states with given angular momenta, and for any configuration of polarized beams that gives cooling, the limiting temperature can be scaled down by lowering the intensity. All results mentioned so far are valid for an arbitrary monochromatic field. It may be composed of any number of plane waves with arbitrary polarizations, and we allow for an arbitrary magnetic field, which adds Zeeman precession to the atomic evolution. Hence cooling schemes in one, two, or three dimensions are described by our formalism.

In the special case of two counterrunning plane waves, we calculate numerically the velocity-dependent force, averaged over a wavelength, both for a standing wave with a transverse magnetic field, and for two orthogonal polarizations. Here we have assumed that the velocity of the atom is constant during the passage of a wavelength. This assumption is justified provided that a typical light shift is small compared with the atomic kinetic energy, so that

$$\frac{\hbar\Delta\Omega^2}{\Gamma^2/4+\Delta^2} < Mv^2/2 \ . \tag{9.1}$$

If this inequality is not valid, the variation of the velocity over a wavelength could be appreciable, and spatial localization could occur.

The results for the calculated average forces are shown in Figs. 3–5. The force depends strongly on the polarization and on the values of the atomic angular momenta. However, since in a large class of cases the force is an odd function both of the velocity and of the detuning, cooling is possible in many cases for an appropriate choice of the detuning. One should be careful, however, in predicting the cooling behavior for two- or three-dimensional schemes for the results for waves propagating in a single direction, since the effects of various beam directions in the evolution equation (5.2) are highly nonadditive.

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