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General diffusion equation for light-induced drift

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We derive a general diffusion equation for the density of an atomic vapor immersed in a buffer gas and irradiated by nearly resonant light. The combination of velocity-selective excitation with state-dependent rates of velocity-changing collisions then gives rise to a drift term, and at the same time the diffusion tensor becomes anisotropic and intensity dependent. Both the effect of light-induced drift and diffusive pulling of atoms by the light are described by this diffusion equation in a consistent and correlated fashion.

I. LIGHT-INDUCED DRIFT

Light-induced drift (LID) may occur in an atomic vapor immersed in a buffer gas and illuminated by nearly resonant light, provided that two conditions are fulfilled. Firstly, the radiative excitation must be velocity selective, so that the ground-state atoms and the excited atoms have opposite flows. Secondly, the cross sections for velocity-changing collisions with the buffer-gas particles must be different for atoms in the two states. Then the two opposite flows will suffer different diffusive frictions, and a net flux of active atoms may arise. This LID effect was predicted by Gel'mukhanov and Shalagin,[1,2] and a first experimental observation was reported by Antsigin *et al.*[3] In an optically thick system, this drift effect may act as a semipermeable piston, which pushes the active atoms away and causes large inhomogeneities in the density of active atoms. This effect of the optical piston[2] has been demonstrated experimentally by Werij *et al.*[4,5]

The theoretical description of LID is based upon the evolution equation for the density matrix of the active atoms, as a function of their position and velocity. This evolution equation is a combination of the optical Bloch equations describing the coupling with the radiative field and the Boltzmann equation for velocity-changing collisions.[2] The central result of the theoretical derivation is a diffusion equation for the density n of active atoms. This equation can be expressed in the form of a continuity equation

$$\frac{\partial}{\partial t} n(\mathbf{r},t) = -\nabla\cdot\mathbf{j}(\mathbf{r},t) , \tag{1.1}$$

with the flux of active atoms $\mathbf{j}(\mathbf{r},t)$ given by[6,7]

$$\mathbf{j}(\mathbf{r},t) = -D\nabla n(\mathbf{r},t) + \mathbf{u}(\mathbf{r},t) n(\mathbf{r},t) . \tag{1.2}$$

Here D is the diffusion constant of ground-state atoms in the buffer gas and $\mathbf{u}$ is the drift velocity. The first term on the right-hand side of (1.2) gives the ordinary diffusion equation for the atomic density n, and the second term represents light-induced drift. The drift velocity $\mathbf{u}(\mathbf{r},t)$ is determined by the intensity $I(\mathbf{r},t)$ of the radiation at the same position and time, and it is just the average velocity of the active atoms. Equations (1.1) and (1.2) can be obtained from the evolution equation for the atomic density matrix $\rho(\mathbf{r},\mathbf{v},t)$, by treating the free-flow term $-\mathbf{v}\cdot\nabla\rho$ and the difference of the collisional cross sections for excited atoms and ground-state atoms as a perturbation.[7] It is this perturbation that causes the rate of change of the atomic density n on a slow time scale. Elimination of the rapid variations then leads to Eq. (1.2), with an expression for the drift velocity $\mathbf{u}$ that is valid to first order in the difference in cross section.[7]

For an optically thick system, the intensity I as a function of the position $\mathbf{r}$ at any instant for time t depends upon the density n at the same instant and at all positions between the entrance window and the position $\mathbf{r}$. Generally, the functional dependence of I on n is determined by an absorption equation of the form[7]

$$\hat{\mathbf{k}}\cdot\nabla I(\mathbf{r},t) = -n(\mathbf{r},t)\alpha(\mathbf{I}(\mathbf{r},t)) , \tag{1.3}$$

where $\hat{\mathbf{k}} = \mathbf{k}/|\mathbf{k}|$ denotes the propagation direction of the radiation, and α determines the saturated absorption rate, which depends in turn on the velocity distributions for both atomic states. In the absence of saturation, $\alpha(I)$ would be given by qI with q the absorption cross section. The set of equations (1.1)–(1.3) governs the combined evolution of the density and the intensity. This set provides the basic equations for light-induced drift in general, and for the action of the optical piston in particular.

In the present paper we give a generalization of this result. The main modification is that we treat exclusively the free-flow term $-\mathbf{v}\cdot\nabla$ as a perturbation, while we formally account for velocity-changing collisions to all orders. This conceptually simple generalization leads to an important change of the result for the particle flux $\mathbf{j}$, as given in Eq. (1.2). Firstly, the diffusion constant D must now be replaced by a second-rank tensor, with the propagation direction of the light as a symmetry axis. The strength of this diffusion tensor depends on the light intensity, thereby reflecting the fact that ground-state atoms and excited atoms have different diffusion coefficients. Secondly, the drift velocity $\mathbf{u}$ is supplemented by a term proportional to the intensity gradient. This term basically arises from the variations of the populations of the two atomic states with intensity, and thereby with position. This term gives rise to the effect of diffusive pulling,[8,2,9] which effectively pulls the atoms towards the regions of high intensity. This effect can be qualitatively understood by noting that the higher mobility of ground-state atoms induces a diffusive flux of ground-state atoms towards high-intensity regions that more than counterbalances the opposite diffusive flux of excited atoms. It is important that the two effects of light-induced drift and diffusive pulling are described in a single unified theory, which clearly reveals their interdependence.

II. GENERAL EVOLUTION EQUATION

We describe the distribution of the active atoms over their internal states and over the velocity $\mathbf{v}$ by the density matrix $\rho(\mathbf{r},\mathbf{v},t)$ which is normalized to the atomic density n according to the equality

$$\int d\mathbf{v}\,\mathrm{Tr}\rho(\mathbf{r},\mathbf{v},t)=n(\mathbf{r},t)\ . \tag{2.1}$$

The atoms are driven by a classical radiation field with frequency ω and wave vector $\mathbf{k}$. Then the coupling of the atoms to the radiation field is described by Bloch equations, and the effect of collisions can be expressed by collision kernels, both for excited atoms and ground-state atoms. For two-state atoms in a monochromatic field, suffering only elastic velocity-changing collisions, the evolution equations take the form

$$\begin{aligned}\frac{\partial}{\partial t}\rho_{11}(\mathbf{v})=&-\mathbf{v}\cdot\nabla\rho_{11}(\mathbf{v})-\gamma_1(\mathbf{v})\rho_{11}(\mathbf{v})\\&+\int d\mathbf{v}'\rho_{11}(\mathbf{v}')K_1(\mathbf{v}'\to\mathbf{v})\\&+A\rho_{22}(\mathbf{v})+\tfrac{1}{2}i\Omega[\rho_{21}(\mathbf{v})-\rho_{12}(\mathbf{v})]\ ,\end{aligned} \tag{2.2}$$

$$\begin{aligned}\frac{\partial}{\partial t}\rho_{22}(\mathbf{v})=&-\mathbf{v}\cdot\nabla\rho_{22}(\mathbf{v})-\gamma_2(\mathbf{v})\rho_{22}(\mathbf{v})\\&+\int d\mathbf{v}'\rho_{22}(\mathbf{v}')K_2(\mathbf{v}'\to\mathbf{v})\\&-A\rho_{22}(\mathbf{v})+\tfrac{1}{2}i\Omega[\rho_{12}(\mathbf{v})-\rho_{21}(\mathbf{v})]\ ,\end{aligned} \tag{2.3}$$

$$\begin{aligned}\frac{\partial}{\partial t}\rho_{12}(\mathbf{v})=&-\mathbf{v}\cdot\nabla\rho_{12}(\mathbf{v})-[\tfrac{1}{2}A+\gamma+i(\Delta-\mathbf{k}\cdot\mathbf{v})]\rho_{12}(\mathbf{v})\\&+\tfrac{1}{2}i\Omega[\rho_{22}(\mathbf{v})-\rho_{11}(\mathbf{v})]\ ,\end{aligned} \tag{2.4}$$

$$\begin{aligned}\frac{\partial}{\partial t}\rho_{21}(\mathbf{v})=&-\mathbf{v}\cdot\nabla\rho_{21}(\mathbf{v})-[\tfrac{1}{2}A+\gamma-i(\Delta-\mathbf{k}\cdot\mathbf{v})]\rho_{21}(\mathbf{v})\\&+\tfrac{1}{2}i\Omega[\rho_{11}(\mathbf{v})-\rho_{22}(\mathbf{v})]\ ,\end{aligned} \tag{2.5}$$

with A the spontaneous decay rate, γ the collisional homogeneous linewidth, Ω the Rabi frequency, and $\Delta=\omega-\omega_0$ the detuning from resonance. The elastic collisions are described by the collision rates γ_n and the collision kernels K_n. These expressions can be generalized for the case of a transition between two degenerate states with angular momenta J_1 and J_2.[10] Formal expressions for the effect of velocity-changing collisions on degenerate atomic levels can be found in the literature.[11–13]

In the present paper the detailed form of the evolution equations is of little concern. We simply summarize the total evolution of ρ in the formal evolution equation

$$\frac{\partial\rho}{dt}=(L_0+L_1)\rho\ , \tag{2.6}$$

where

$$L_1\rho=-\mathbf{v}\cdot\nabla\rho \tag{2.7}$$

gives the free-flow term, and L_0 describes the evolution due to radiative transitions and velocity-changing collisions with buffer-gas particles. We shall make use of the fact that L_0 is a linear operator that may depend parametrically on the position $\mathbf{r}$, due to the variation of the intensity (and therefore the Rabi frequency) with $\mathbf{r}$, but it contains no derivative with respect to $\mathbf{r}$. Hence we may introduce the steady-state solution $\bar{\rho}(\mathbf{v})$ of the approximate evolution equation

$$\frac{d\rho}{dt}=L_0\rho\ , \tag{2.8}$$

by solving the equation

$$L_0\bar{\rho}(\mathbf{v})=0\ . \tag{2.9}$$

This steady-state density matrix with respect to L_0 depends parametrically on $\mathbf{r}$, and we can normalize it for each value of $\mathbf{r}$ by imposing the condition

$$\mathrm{Tr}\int d\mathbf{v}\,\bar{\rho}(\mathbf{v})=1\ . \tag{2.10}$$

Now collisions and radiative transitions modify the state of the atoms on a rapid time scale of the order of a few radiative lifetimes, whereas the free-flow term L_1 gives rise to slow diffusive flows between macroscopically separated positions. Hence it is reasonable to assume that the collisions and radiative transitions, as described by L_0, drive the density matrix at any position to its steady state $\bar{\rho}$, before the diffusive flow, as expressed by L_1, has had a chance to produce appreciable changes of the local density. On the other hand, it is just the evolution of the density n that we are interested in. Therefore we wish to eliminate the rapid variations due to L_0. This can be done by standard techniques, which we briefly indicate in Sec. III.

III. ELIMINATION OF RAPID EVOLUTION

On the rapid time scale determined by the rate of collisional thermalization and radiative transitions, the density matrix ρ at each position $\mathbf{r}$ is expected to approach the steady state, which is proportional to $\bar{\rho}$. On this rapid time scale the density n is not affected. However, on the slower time scale as determined by the free-flow term L_1, the atoms can drift away over macroscopic distances, as a result of a nonvanishing average velocity or of diffusive fluxes. The local density matrix deviates only slightly from the steady state $\bar{\rho}$ during this slow evolution of the density.

These general considerations are put on a mathematical footing by using a projector-operator formalism. We introduce the projector P in such a way that $P\rho$ is proportional to the steady-state solution $\bar{\rho}$ with respect to L_0.

This is accomplished by the definition

$$P\rho(\mathbf{r},\mathbf{v},t)=\bar{\rho}(\mathbf{v})n(\mathbf{r},t)\ , \tag{3.1}$$

where the density n is expressed in terms of ρ by Eq. (2.1). One readily checks that P obeys the operator equalities

$$PL_0=0,\quad L_0P=0 \tag{3.2}$$

which follow from that fact that collisions and radiative transitions do not remove atoms from the local position $\mathbf{r}$, and from the defining relation (2.9). A reduced evolution equation for the density n alone follows by deriving an evolution equation for $P\rho$ with

$$Q=1-P\ . \tag{3.3}$$

By using the obvious projector equalities

$$P^2=P,\quad Q^2=Q \tag{3.4}$$

combined with (3.2), one obtains from (2.6) the evolution equations

$$\frac{d}{dt}P\rho=PL_1P\rho+PL_1Q\rho\ , \tag{3.5a}$$

$$\frac{d}{dt}Q\rho=QL_1P\rho+Q(L_0+L_1)Q\rho\ . \tag{3.5b}$$

A closed equation for $P\rho$ alone, which is valid to second order in L_1, follows after solving (3.5b) for QP to first order in L_1 and substituting the result in the last term of (3.5a).[14,7] The final result takes the form

$$\frac{d}{dt}P\rho(t)=PL_1P\rho(t)+\int_0^\infty d\tau\, PL_1e^{L_0\tau}QL_1P\rho(t)\ . \tag{3.6}$$

The integrand in the last term of (3.6) decays to zero as a function of τ on the rapid time scale, so that the total evolution of $P\rho$ remains slow, as expected.

Each term in (3.6) is proportional to the steady-state density matrix $\bar{\rho}(\mathbf{v})$, which can therefore be divided out. This produces then an evolution equation for n. It is important to note that $\bar{\rho}$ depends on the local intensity, so that $\bar{\rho}$ also has an intrinsic parametric dependence on the position $\mathbf{r}$.

IV. GENERAL DIFFUSION EQUATION

The reduced evolution equation (3.6) yields an equation for n when we substitute the definition (3.1) for the projection operator P, and the definition (2.7) for the evolution operator L_1. The evaluation of the terms on the right-hand side of (3.6) is straightforward. The resulting equation for the density n takes the form of the continuity equation (1.1), with the particle flux given by

$$\mathbf{j}=-\mathbf{D}\cdot\nabla n+\mathbf{u}_0n+\mathbf{u}_1n\ . \tag{4.1}$$

The term $\mathbf{u}_0$ in (4.1) originates from the first term on the right-hand side of (3.6), and the time integral in (3.6) gives rise to the other two terms in (4.1). The velocity $\mathbf{u}_0$ is the drift velocity, which is related to the normalized steady-state density matrix $\bar{\rho}$ by

$$\mathbf{u}_0=\int d\mathbf{v}\,\mathbf{v}\,\mathrm{Tr}\,\bar{\rho}(\mathbf{v})\ , \tag{4.2}$$

which is just the average velocity of the atoms in the steady state with respect to L_0. This drift velocity is usually nonzero due to the combination of velocity-selective excitation, as expressed by the Doppler shift $\mathbf{k}\cdot\mathbf{v}$ in (2.4) and (2.5), and the state dependence of the collision rates. Hence $\mathbf{u}_0$ is directed along the wave vector $\mathbf{k}$. The diffusion tensor $\overleftrightarrow{\mathrm{D}}$ in (4.1) is given by the formal expression

$$\overleftrightarrow{\mathrm{D}}=\int_0^\infty d\tau\left[\int d\mathbf{v}\,\mathbf{v}\,\mathrm{Tr}e^{L_0\tau}\mathbf{v}\bar{\rho}-\mathbf{u}_0\mathbf{u}_0\right]\ , \tag{4.3}$$

which has the significance of the time integral of the velocity autocorrelation function of atoms in the steady state. The integrand of the time integral in (4.3) approaches zero on the rapid time scale. This diffusion tensor depends on the local intensity, and it is therefore an implicit function of $\mathbf{r}$. Furthermore, since the evolution operator L_0 is anisotropic, due to its dependence on the wave vector $\mathbf{k}$ of the light, the diffusion tensor $\overleftrightarrow{\mathrm{D}}$ will also be anisotropic, and it must have cylindrical symmetry with the $\mathbf{k}$ vector as a symmetry axis. Therefore, the diffusion tensor $\overleftrightarrow{\mathrm{D}}$ is determined by two constants, a transverse diffusion constant D_t and a longitudinal one D_l, so that

$$D_{ij}=D_t\delta_{ij}+(D_l-D_t)k_ik_j/k^2\ . \tag{4.4}$$

For zero intensity, the diffusion tensor is isotropic and has the strength of the diffusion constant of ground-state atoms in the buffer gas. Finally, the velocity $\mathbf{u}_1$ occurring in (4.1) is a correction on the effective drift velocity that arises from the variation of the steady-state density matrix $\bar{\rho}$ with position. We obtain

$$\mathbf{u}_1=-\int_0^\infty d\tau\left[\int d\mathbf{v}\,\mathbf{v}\,\mathrm{Tr}e^{L_0\tau}\mathbf{v}\cdot\nabla\bar{\rho}-\mathbf{u}_0\nabla\cdot\mathbf{u}_0\right]\ . \tag{4.5}$$

Again, the integrand of the time interval in (4.5) approaches zero on the rapid time scale. Since $\bar{\rho}$ and $\mathbf{u}_0$ depend exclusively on the position $\mathbf{r}$ through the $\mathbf{r}$-dependent intensity or Rabi frequency, we may express $\mathbf{u}_1$ in the form

$$\mathbf{u}_1=\overleftrightarrow{\mathrm{C}}\cdot\nabla\Omega\ , \tag{4.6}$$

with $\overleftrightarrow{\mathrm{C}}$ the cylindrically symmetric second-rank tensor

$$\overleftrightarrow{\mathrm{C}}=-\int_0^\infty d\tau\left[\int d\mathbf{v}\,\mathbf{v}\,\mathrm{Tr}e^{L_0\tau}\mathbf{v}\frac{\partial\bar{\rho}}{\partial\Omega}-\mathbf{u}_0\frac{\partial\mathbf{u}_0}{\partial\Omega}\right]\ . \tag{4.7}$$

For excitation on resonance, the velocity distribution $\mathrm{Tr}\bar{\rho}(\mathbf{v})$ is symmetric, and the drift velocity $\mathbf{u}_0$ disappears. The gradient velocity $\mathbf{u}_1$, however, will be nonzero even in this case, provided that the intensity varies with position.

In summary, we conclude that the atomic flux takes the form (4.1), with $\mathbf{u}_0$ a velocity in the direction of the wave vector $\mathbf{k}$, with $\mathbf{u}_1$ expressed as in (4.6) as a cylindrically symmetric tensor acting on the gradient of the Rabi frequency, and with $\overleftrightarrow{\mathrm{D}}$ a cylindrically symmetric tensor. This result is fully general in that it is based upon nothing else but the assumption that the macroscopic modifications in the density n occur on a time scale that is slow compared with the rate of collisional thermalization and radiative transitions. In particular, this result is not dependent on restrictive assumptions concerning the multiplet structure of the atomic levels, and it allows for the inclusion of Zeeman degeneracy as well as fine structure or hyperfine structure. On the other hand, the result is rather formal, and explicit expressions for the drift velocity $\mathbf{u}_0$, the gradient velocity $\mathbf{u}_1$, and the diffusion tensor $\overleftrightarrow{\mathbf{D}}$ can only be obtained by adopting specific models for the atomic levels and for the collision kernels. Nevertheless, the general structure of Eqs. (4.1)–(4.5) allows us to draw general conclusions on the interdependence of the various physical effects, in particular, the light-induced drift and diffusive pulling. This we shall do in Sec. V.

V. DISCUSSION AND CONCLUSIONS

The origin of the various terms in the atomic flux (4.1) is rather evident. The term proportional to $\mathbf{u}_0$ simply accounts for the fact that in the steady state $\bar{\rho}$, the atoms have a nonzero average velocity. It is this velocity $\mathbf{u}_0$ that has been numerically or analytically calculated by solving the evolution equations without the free-flow term.[15–17] The diffusion tensor has the standard form of the integrated autocorrelation function of the velocity. In the present system this correlation function is anisotropic, and it depends on the light intensity. This simply reflects the anisotropy and the intensity dependence of the evolution operator L_0. The term proportional to $\mathbf{u}_1$ arises from the fact that the diffusion tensor is basically a weighted average of the diffusion of atoms in the ground state and atoms in the excited state. However, the atomic motion due to the net diffusive flux brings the atoms to a region with a different distribution over the atomic states, which results in a modified diffusion tensor. This change in the distribution over the atomic states then leads to a correction in the diffusive flux, which is expressed by the velocity $\mathbf{u}_1$.

These considerations may be illustrated by adopting a simplifying approximation in Eqs. (4.4) and (4.5), by supposing that the velocity autocorrelation function for atoms in either one of the two states depends negligibly on the shape of the steady-state velocity distributions. On the other hand, we allow these correlation functions to be different for ground-state atoms and for excited atoms. This means that the diffusion tensor becomes isotropic, since the anisotropy of the steady-state distributions can no longer matter, and the diffusion constants can be expressed as a weighted average of the diffusion constant D_1 and D_2 corresponding to atoms in the two states. Hence we write

$$D_{ij}=\delta_{ij}(D_1p_1+D_2p_2)\ , \tag{5.1}$$

with

$$P_n=\int d\mathbf{v}\,\bar{\rho}_{nn}(\mathbf{v}) \tag{5.2}$$

the fraction of the atoms in the state n $(n=1,2)$. The same approximation applied to (4.5) yields the result

$$\mathbf{u}_1=-D_1\nabla p_1-D_2\nabla p_2\ , \tag{5.3}$$

where the gradients of the steady-state populations p_1 and p_2 are due to the intensity variation with the position $\mathbf{r}$. Substituting the results (5.1) and (5.3) into (4.1) gives the very simple result

$$\mathbf{j}=-\nabla(D_1p_1n+D_2p_2n)+\mathbf{u}_0n\ . \tag{5.4}$$

This equation expresses the atomic flux as the sum of a diffusive flux and a light-induced drift. The diffusive flux is simply the sum of two fluxes, one for each atomic state.

It is obvious that this simple expression (5.4) contains light-induced drift as well as diffusive pulling. In particular, when we assume that the light frequency is at resonance, the drift velocity $\mathbf{u}_0$ must disappear, and only the diffusive flux remains in (5.4). In the stationary state in a closed cell, the total flux must vanish. When we use $p_1+p_2=1$, this leads to the result

$$[D_1+p_2(D_2-D_1)]n=\text{const}\ . \tag{5.5}$$

If the excited-state diffusion constant D_2 is the smaller one, we find that in regions of high intensity, where p_2 is appreciable the density n must be larger than in the darker regions. This is the effect of diffusive pulling.[8] Obviously, the term $\mathbf{u}_1$ is essential for the description of this effect.

Apart from the simplified result discussed in this section, the derivation of the general result (4.1) demonstrates the connection between light-induced drift and diffusive pulling. In particular it is obvious that it would not be consistent to evaluate the drift velocity $\mathbf{u}$ from the steady-state distribution $\bar{\rho}$ with respect to L_0, and substitute it in (1.2), while retaining for D simply the gas-kinetic diffusion coefficient pertaining to a gas mixture without radiation. Inclusion of the difference in cross section in $\mathbf{u}$ to higher order than the first one implies that also the diffusion tensor is affected by the presence of excited atoms. Similarly, the combined description of light-induced drift and diffusive pulling demands the inclusion of a term with $\mathbf{u}_1$, which only arises when the free-flow term is included to second order and the difference in cross sections at least to first order. This explains why diffusive pulling did not appear in our earlier derivation of a diffusion equation.[6,7]

Obviously, it would be interesting to obtain explicit results for the intensity dependence and the anisotropy of the diffusion tensor. This is possible for specific simple collision models, as we intend to demonstrate in a subsequent paper.

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