Light-induced drift dynamics in an optically thin regime: Monochromatic and broadband laser excitations

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Light-induced drift (LID) dynamics are studied in uncoated and silane-coated capillaries containing sodium and a few torr of neon or krypton. The adopted coating reduces the adsorption energy of the atoms at the surface of the cell by permitting the observation of wall frictionless LID. Moreover, it can be heated up to 200 °C, making it possible to work with saturated vapor over a wide range of densities. A measurement method of the drift velocity is proposed for the optically thin regime, and its dependence on the laser linewidth is observed using both a single-mode and a broadband laser. Drift velocities of up to 30 m/s are measured in a cell containing 6 Torr of krypton under broadband excitation.

I. INTRODUCTION

Since light-induced drift (LID) was proposed and observed by Gel'mukhanov and Shalagin,[1,2] many experiments have been performed, mostly with alkali-metal–noble-gas pairs,[3–8] and theoretical models have been developed.[9,10]

LID is due to both laser velocity-selective excitation and different collision cross sections of the ground- and excited-state active atoms with a buffer gas. The two conditions generate two fluxes of atoms moving in opposite directions, which have different transport properties.

Quantitative studies of LID require negligible atom-wall interactions in order to avoid a huge slowing down of the effect, as has been observed by Werij *et al.*[3] The paraffin and the silane coatings, at different temperatures, satisfy this requirement, allowing the observation of LID time evolution.

The paraffin coating works at room temperature, where the saturated vapor density is extremely low and only the very thin optical regime can be studied.[4] The silane coating, which can be heated up to 200 °C, permits the study of LID from the optically thin up to the intermediate-density regime.[6–8]

LID dynamics can be modified by changing the laser linewidth and line shape, as long as the laser velocity selectivity is preserved. A first theoretical approach on this subject has been done by Popov *et al.*,[11] which, for a two-level atom and broadband excitation with a square profile, predicted supersonic LID. Preliminary results have been reported in Ref.[8]

We present our measurements of the drift velocity v_{dr} in an optically thin regime under both broadband (BB) and single-mode (SM) laser excitations. The adopted BB laser has a long resonant cavity which accepts many longitudinal modes inside the atomic Doppler-broadened linewidth, and at the same time has an easily controllable total bandwidth ("lamp laser"[12]).

The drift velocity is measured by fitting the fluorescence signals, recorded as a function of time, with the curves obtained by solving the equations describing the vapor-density time evolution. Under similar conditions, the drift velocities induced by the BB laser are larger than those induced by the SM one, as expected. Drift velocities of the order of 30 m/s have been measured in coated capillaries containing a few torr of krypton, under BB laser excitation.

II. THEORETICAL APPROACH

A. Microscopic description (v_{dr} calculation)

The drift velocity can be derived by solving the set of rate equations describing the excited- and ground-state velocity distributions after a suitable collision model has been adopted. Haverkort[10(a)] has analytically solved them for a two-level atom by assuming a strong collision model. He has calculated v_{dr} under very general conditions, and we refer to that paper for a complete description of this approach. Many of these results also appear in Ref. 10(b). Here the solutions for the SM and BB laser excitations are explicitly given in order to easily introduce all the parameters and to demonstrate the v_{dr} dependence on them. The SM laser excitation induces a v_{dr} given by

$$v_{dr}(\mathrm{SM}) = Y v_L \frac{n_e}{n} , \tag{1}$$

where Y is a factor depending on the collision parameters

$$Y = \left[\frac{\Gamma_g}{\Gamma_g+\Gamma_T}\right]\left[\frac{\Gamma_g-\Gamma_e}{\Gamma_g}\right]\left[\frac{A}{A+\Gamma_e}\right] , \tag{2}$$

and n_e/n gives the excited-atom fraction

$$\frac{n_e}{n} = \frac{1}{2}\frac{A+\Gamma_e}{(1-\beta)A+\Gamma_e}\frac{\xi k_{\mathrm{SM}}}{\sqrt{1+k_{\mathrm{SM}}}+\xi k_{\mathrm{SM}}} , \tag{3}$$

with

$$\xi = \left[\frac{(1-\beta)A+\Gamma_e}{\beta A}\right]\frac{\Gamma\sqrt{\pi}}{k\bar{v}} e^{-(v_L/\bar{v})^2} , \tag{4}$$

$$\beta=\frac{1}{2}\left[1+\frac{\Gamma_e+\Gamma_T}{\Gamma_g+\Gamma_T}\right], \tag{5}$$

where Γ is the homogeneous linewidth, A is the transition probability, $\bar{v}$ is the mean atomic velocity, v_L is the velocity class selected by the laser, Γ_g and Γ_e are the rates for velocity changing collisions in the ground and excited states, respectively, Γ_T is the transverse transit relaxation time, and k_{SM} is the saturation parameter.

The BB laser has been schematized as a homogeneous radiation having a Gaussian profile with a linewidth equal to Γ_{BB}. This is correct, in first approximation, if the frequency mode separation is smaller than the homogeneous linewidth of the atoms. The drift velocity is then given by the general formula

$$v_{dr}(BB)=\gamma\bar{v}\Phi_g(\Delta)\frac{n_e}{n}, \tag{6}$$

with

$$\frac{n_e}{n}=\frac{1}{2}\left[\frac{A+\Gamma_e}{A}\right]\frac{k_{BB}\langle G(\Delta,v_z)\rangle}{\beta(1+k_{BB})+(k_{BB}/A)[(1-\beta)A+\Gamma_e]\langle G(\Delta,v_z)\rangle}, \tag{7}$$

$$\Phi_g(\Delta)=\frac{\langle v_z G(\Delta,v_z)\rangle}{\bar{v}\langle G(\Delta,v_z)\rangle}, \tag{8}$$

$$G(\Delta,v_z)=\exp\left[-\left[\frac{\Delta-kv_z}{\Gamma_{BB}\sqrt{1+k_{BB}}}\right]^2\right], \tag{9}$$

$$\langle G(\Delta,v_z)\rangle=\int dv\, G(\Delta,v_z)W(v), \tag{10}$$

where $W(v)$ is the Maxwellian distribution of velocities, Δ is the detuning from resonance, and the other parameters are the same as before. A comparison between the drift velocities achievable for the two laser excitations and under similar conditions is shown in Fig. 1, where v_{dr}(SM) and v_{dr}(BB) are plotted as a function of the laser-power density. The calculations have been made for krypton at 6 Torr and for the same laser detuning and power density. The adopted parameter values are reported in Table I. To compare the two laser intensities k_{BB} is defined as

$$k_{BB}=k_{SM}\frac{\Gamma_{SM}}{\Gamma_{BB}},$$

where Γ_{SM} (the single-mode laser linewidth) is kept equal to the atomic homogeneous linewidth.

The drift velocity induced by the broadband laser is about twice that induced by the single mode one for a larger fraction of the laser intensity range. In Fig. 2, v_{dr} is reported as a function of the laser detuning for SM and BB excitation. For the two lasers the maximum is obtained at about the same detuning, but the BB excitation is still effective at bigger Δ. In Fig. 3 the drift velocity is reported as a function of the BB laser linewidth calculated for a given detuning. v_{dr} increases as long as the velocity selectivity is not compromised by a too large laser linewidth, then it starts decreasing as soon as a larger fraction of atoms with negative velocity component gets on resonance.

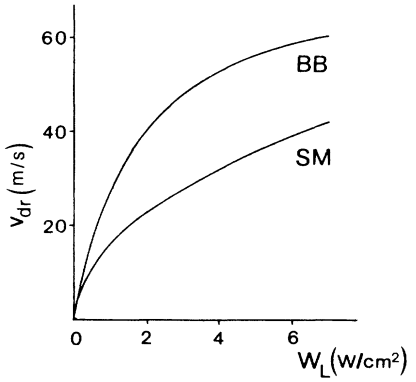

FIG. 1. Plot of the drift velocity v_{dr} vs the laser-power density W_L calculated for SM and BB laser excitation in the cell with 6 Torr of Kr. The adopted parameter values are given in Table I.

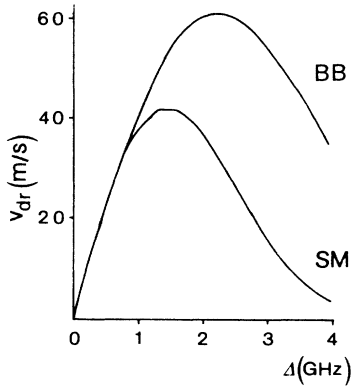

FIG. 2. Plot of v_{dr} vs the laser detuning calculated for SM and BB laser excitations.

TABLE I. Parameter values for the calculation of the drift velocity as derived from Ref. 10(a).

$A=10$ MHz
$\Gamma=55$ MHz
$\Gamma_g=3.1$ MHz
$\Gamma_e=4.4$ MHz
$\Gamma_T=2.7$ kHz
$\Delta\omega_{hfs}=1.77$ GHz
$k_{SM}(D_2)=9.4$ mW/cm^2
$\Gamma_{SM}=\Gamma$
$\Gamma_{BB}=2$ GHz

B. Macroscopic description (v_{dr} measurement)

The time evolution of the vapor density is described, in the general case, by two coupled differential equations.[9] The first describes the diffusion of the vapor in the presence of a laser-dependent drift term and the second one the dependence of the laser intensity both on the vapor density and on the position z in the cell. The situation simplifies significantly in the optically thin regime. In this case the laser is assumed to be only slightly attenuated by the vapor so that $I(z,t)=I$ and the drift velocity can be assumed uniform along the cell. Moreover, the fluorescence intensity is directly proportional to the vapor density n.

The diffusion equation becomes

$$\frac{\partial n}{\partial t}=D_g\frac{\partial^2 n}{\partial z^2}-v_{dr}\frac{\partial n}{\partial z}\ , \tag{11}$$

where D_g is the diffusion coefficient of the ground-state atoms. The solution for a semi-infinite cell ($z>0$) with the initial condition of a uniform distribution

$$n(z,0)=n_0\ , \tag{12}$$

and the boundary condition of a vanishing flux at the cell entrance

$$\frac{\partial n}{\partial z}_{z=0}-\frac{v_{dr}}{D_g}n(0,t)=0 \tag{13}$$

becomes, by defining $x=(v_{dr}/D_g)z$ and $\tau=(v_{dr}^2/D_g)t$,

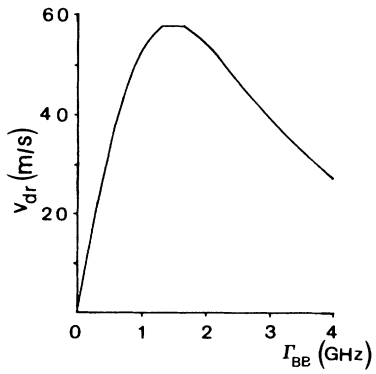

FIG. 3. Plot of v_{dr} vs the BB laser linewidth Γ_{BB}.

$$\frac{n(x,\tau)}{n_0}=A+B\ , \tag{14}$$

where

$$A=1-\int_{(x-\tau)/2\sqrt{\tau}}^{\infty}e^{-s^2}\frac{ds}{\sqrt{\pi}}\ , \tag{15}$$

$$B=(1+x+\tau)e^x\int_{(x+\tau)/2\sqrt{\tau}}^{\infty}e^{-s^2}\frac{ds}{\sqrt{\pi}}-\left[\frac{\tau}{\pi}\right]^{1/2}e^{-(x-\tau)^2/4\tau}\ . \tag{16}$$

The term A has the property of going to $\frac{1}{2}$ after a time t^* such that $t^*=z_0/v_{dr}$, where z_0 is the observation point. The term B gives a vanishing contribution depending on the value of the parameter $S=z_0v_{dr}/D$. For $S=1$, B represents about 8% of the signal, while it has a decreasing weight for increasing S values. Therefore, when $B=0$, v_{dr} can be directly derived by measuring z_0 and t^*.

III. EXPERIMENTAL APPARATUS AND RESULTS

The experimental apparatus is essentially the same as described in Refs. 7 and 8. The reported measurements have been obtained from four capillary cells (internal diameter $\cong 0.2$ cm; length $\cong 15$ cm) in order to study the LID dependence on the kind of buffer gas and on the interaction with the cell walls. Two cells, one coated and one uncoated, have been filled with 10 Torr of neon. The two others, both coated, contain 3 and 6 Torr of krypton, respectively. The coating, as described in detail in Ref. 7, is obtained from a dimethylpolysiloxane ether solution.

The dependence of the drift velocity on the laser radiation bandwidth has been studied by using two different lasers: a single mode actively stabilized dye laser, and a broadband one. This last is a first-generation standing-wave Coherent Radiation dye laser, whose output mirror has been moved far away (up to 15 m) to obtain a long cavity configuration. This gives, on the one hand, many longitudinal modes inside the Doppler linewidth and, on the other hand, a total bandwidth of the same order.[12] This kind of laser has already been used to demonstrate the radiation pressure on sodium vapor[13] and to decelerate a sodium beam.[14] In both cases the results indicate that all the modes oscillate during the time determined by the excited-level lifetime.

The lasers are switched on or off via a chopper driven by a function generator. The laser-induced fluorescence is collected by an optical fiber movable along the capillary. The signals, monitored via a fast transient digitizer, can be averaged over many samplings or directly recorded.

When the vapor is optically thin, it is a good approximation to assume the density to be directly proportional to the laser-induced fluorescence and hence to take its temporal evolution to be that of the vapor density itself.

In Fig. 4 two typical signals for uncoated and coated cells, respectively, are reported with the fitting curves obtained from Eq. (14). They correspond to two very different drift velocities, 20±2 cm/sec [Fig. 4(a)] and

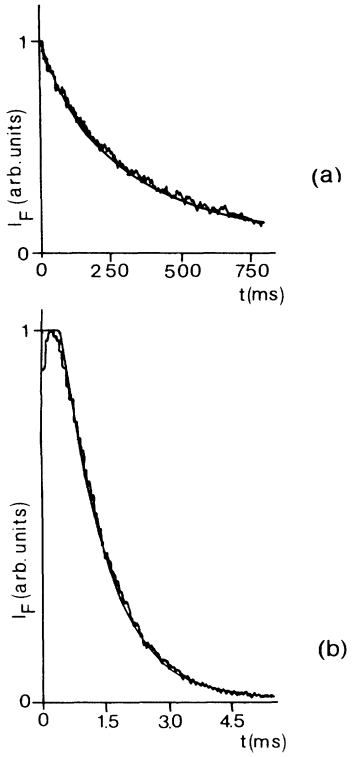

FIG. 4. Two typical fluorescence signals: (a) uncoated cell $z=3$ cm, $D=40$ cm^2/s, $v_{dr}=20$ cm/s; (b) coated cell $z=8$ cm, $D=31$ cm^2/s, $v_{dr}=800$ cm/s. In both cases the excitation is broadband. The curves represent the fittings obtained from Eq. (13).

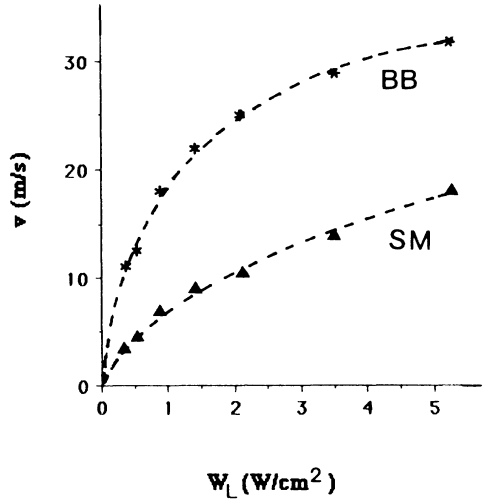

FIG. 5. Measurements of v_{dr} plotted as a function of the laser-power density of W_L. The detunings of the two lasers have been adjusted to get the maximum speed.

800 ± 100 cm/sec [Fig. 4(b)]; in the two cases the parameter S changes from 1.5 to 200. In both cases the agreement between the experimental signals and the theoretical calculations is very good.

The effective stationary density profile along the capillary has been taken into consideration. This profile is experimentally determined by moving the fiber along z. This precaution is necessary because, inside the capillary, the density may not be truly uniform depending on whether or not it has reached its equilibrium. This correction, which does not make a noticeable difference when v_{dr} is small, becomes important in the other cases. When the drift velocity is small the diffusion process is fast enough to hide the variation of the density profile. If, on the contrary, v_{dr} is big the density profile modifies the fluorescence signal and thus the measurement of v_{dr} itself.

As discussed in the Introduction, LID is affected, among the other parameters, by the atom-wall interaction, by the kind of buffer gas, and by the laser linewidth. The influence of the atom-wall interaction has been already observed and studied.[3,4,6] We have evidence of it in Fig. 4 where the fittings of the curves give different drift velocities under comparable laser excitations. This difference, which cannot be totally ascribed to the different reported diffusion coefficients, is essentially due to the presence or absence of the coating, respectively. A discussion of the influence of the other parameters is reported as follows.

(a) a detailed comparison between the v_{dr} induced by the SM and the BB laser excitations has been made and the results obtained as a function of the laser-power density are shown in Fig. 5 for the coated cell filled with 6 Torr of krypton. Analogous results have been obtained for the other coated cells.

The dependence is the same as in Fig. 1 even if the absolute v_{dr} values are about a factor of 2 lower. This difference is due to the limited validity of the two-level theory when applied to alkali-metal atoms, as checked by Haverkort.[10(a)]

The BB laser is very effective and the maximum speed obtained in the coated cell with 6 Torr of krypton and 6 W/cm^2 has been $v_{dr}(\max)=(30\pm3)$ m/s. This velocity seems to disagree with the predictions made by the four-level theory which, on the other hand, has been successfully tested in other experiments.[10] This seems to indicate that, by working on the laser line shape, it is possible to get very high drift velocities according to the predictions made by Popov *et al.*[11] It is interesting to note that, in our conditions, the drift velocities obtained with the SM excitation also seem to be larger than they would be after the four-level calculations. In fact, from Fig. 9(a) of Ref. 10(b) we obtain that v_{dr} is of the order of 1 m/s for Na/Kr with 5 W/cm^2 laser power density. Only at much higher laser-power densities ($W_L=5\times10^2$ W/cm^2) does v_{dr} become comparable to that which we measured. The origin of this disagreement is, at present, unclear to us.

The drift velocity is not critical with the laser length when L becomes larger than 5 m, although the best results have been obtained for $L=10$ m. The difference is not due, in this case, to the closer spacing between the laser modes, but probably to the different total laser linewidth. The 10-m cavity laser had a bandwidth, as measured with a Fabry Pérot interferometer, of 2 GHz,

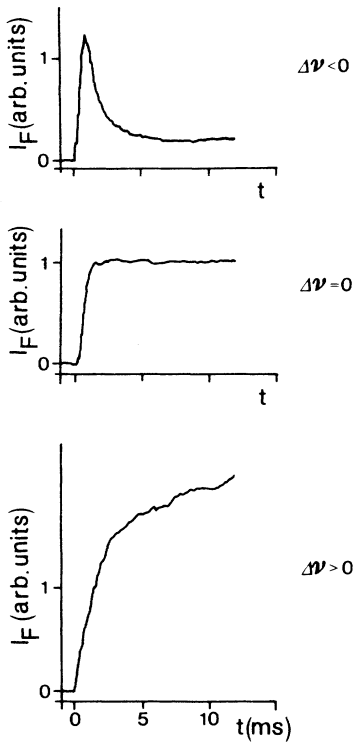

FIG. 6. Fluorescence signals obtained from the 6-Torr krypton cell under BB laser excitation. The curves from (a) to (c) correspond to negative, zero, and positive detuning, respectively.

which corresponds more or less to the maximum of Fig. 3.

(b) The dependence on the buffer-gas pressure and species is as predicted. The drift velocity obtained by shifting from neon to krypton is about ten times faster, according to the ratio $D_g - D_e / D_g$ between the diffusion coefficients for the two gases (0.42 for krypton and 0.041 for neon[10(a)]). There is about a factor of 2 between the velocities measured with the 3 and 6 Torr of krypton, which is the difference calculated by Haverkort.[10(a)]

(c) Our experimental apparatus permits the observation of the drift in both directions. In one case the vapor is confined to the end of the cell, while in the other it is pulled against the entrance window, as can be seen by looking at the fluorescence. In the optically thin regime the density variation is very fast in both cases.

It is interesting to observe that it is possible to tune the broadband laser continuously across the resonance, as is shown in Fig. 6. The fine tuning is obtained by tilting the intracavity étalon with a piezoelectric ceramics. This simple device permits frequency scanning over a few gigahertz and switching from positive to negative LID directions.

The increase in fluorescence due to the drift of the vapor in the direction of the entrance window is evident from the figure. The time scale is the same as for "positive" detuning, indicating comparable drift velocities. In this case no fitting has been done.

IV. CONCLUSIONS

Experimental observations of LID dynamics in the optically thin regime have been done in coated and uncoated capillary cells. We have shown that the BB laser excitation can be very effective by inducing faster drift velocities than the SM one. In particular, we have obtained high v_{dr} with relatively low laser-power densities, in apparent disagreement with the predictions made by the four-level theory.[10(a),10(b)] This circumstance has also been observed for SM excitation in the coated cells, where the absolute value of the drift velocity does not exceed the calculated limits, but these values were obtained with very low laser-power densities.

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