Demonstration of a Semipermeable Optical Piston

H. G. C. Werij, J. P. Woerdman, J. J. M. Beenakker, and I. Kuščer[(a)]
Molecular Physics Department, Huygens Laboratory, University of Leiden, 2300 RA Leiden, The Netherlands

(Received 17 April 1984)

Na atoms in a buffer gas are compressed to a steep front by a laser beam tuned into the red Doppler wing of the Na D transition. The Na densities on either side of the front differ by a factor of 500. The front formation is due to a laser-induced drift which is a consequence of the state dependence of the diffusion cross sections involved. The Na drift velocity is deduced to be ~ 4 m/s.

PACS numbers: 42.65.Jx, 32.90.+a, 34.90.+q

Light-induced drift (LID) of optically absorbing atoms in a buffer gas has been theoretically predicted by Gel'mukhanov and Shalagin.[1] A traveling laser beam produces velocity-selective saturation of the absorbing atoms by means of the Doppler effect. This yields opposing fluxes of excited and unexcited atoms. Both fluxes feel different diffusion resistances in the buffer gas since the collision cross section of an excited atom is generally larger than that of a ground-state atom. As a result, the absorbing atoms acquire a drift velocity v_d in the same direction as the laser beam when the latter is tuned into the red Doppler wing. The direction of the drift reverses when the Doppler detuning changes sign. It should be noted that this mechanical action of the light field is completely different from the well-known radiation pressure[2,3]; the LID pressure is predicted to be much larger than the latter since atomic instead of photon momenta are transferred. Experimentally the existence of LID has been verified by Antsygin *et al.* for Na:Ne and Na:He [4]; however, apart from the direction of the drift, no data were reported. Later experimental work has been concentrated on isotope separation by LID using vibrational molecular transitions[5]; here also, quantitative data are scarce. In this Letter we report a new regime of LID: We have observed "piston" action, or front formation, theoretically predicted by Gel'mukhanov and Shalagin as a consequence of v_d being position dependent in an optically dense system.[6] This demonstrates for the first time the large strength of the LID pressure and yields quantitative data on the kinetic aspects of LID.

The experiments have been done with a cell made of sodium-resistant gehlenite glass[7] (Fig. 1). The cell consists of a capillary tube (inside diameter $d = 2$ mm) which connects two reservoirs, I and II. Through another tube, reservoir II communicates with an appendix containing ~ 50 mm^3 Na metal. The appendix and capillary sections of the cell are independently heated to temperatures T_{app} and T_{cap}, respectively, with transparent tin oxide ovens.[7] The cell has been filled with Ar buffer gas (3.2×10^{17} atoms cm^{-3}); we estimate that at this density and at 500 K, Na ^2S atoms have a diffusion coefficient $D \sim 25$ cm^2 s^{-1}.[8] The beam of a single-mode cw dye laser (Spectra Physics 380 D) passes through the capillary, completely filling its cross section. Photodiodes C and D measure the transmission of weak probe beams (~ 4 μW, ~ 1 mm diam) perpendicular to the cell axis; all local Na densities mentioned below have been deduced from these transmission data. We verified from the shape of the absorption spectrum that the probe power was low enough to avoid appreciable optical hyperfine (hf) pumping.[9] For typical experimental conditions, $T_{\text{cap}} = 520$ K and $T_{\text{app}} = 410$ K, the Na density, [Na], in the cell is 1×10^{12} cm^{-3}; this density is steered by T_{cap} (instead of T_{app}) because of the large surface-to-volume ratio of the cell and the noninert character of the glass surface in a sodium atmosphere.[10]

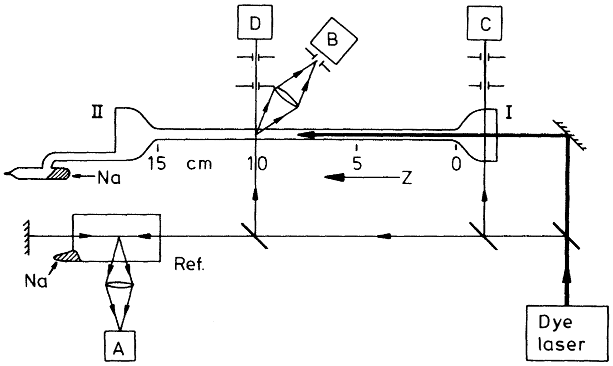

FIG. 1. Experimental setup; the capillary cell has been filled with 10 Torr of Ar (at 300 K). *A, B, C,* and *D* are photodiodes. Lamb dips in the Na *D* fluorescence spectrum from a reference Na cell (without buffer gas) are used for frequency calibration.

Figure 2 shows an example of optical piston action. The laser beam is linearly polarized, its intensity I_L is 3 W cm^{-2}, which is about 10 times the Na D_2 saturation intensity I_{sat},[11] and its frequency ν_L is tuned to $\nu' = \nu_0 - \frac{1}{2}\Delta\nu_{\text{D}}$, where ν_0 is the Na D_2 $^2S_{1/2}(F=2) \rightarrow {}^2P_{3/2}$ transition frequency and $\Delta\nu_{\text{D}}$ is the 1700-MHz Doppler width. For $t < 0$ the beam is intercepted and capillary and reservoirs are uniformly ($\pm 20\%$) filled with $[\text{Na}] = 1 \times 10^{12}$ cm^{-3} $= n_0$. At $t = 0$ the laser beam is admitted. It penetrates reservoir I which completely lights up as a result of multiple scattering; the beam just reaches the capillary.[12] At $t = 15$ s an intense fluorescent spot starts to travel along the capillary; it reaches reservoir II at $t = 155$ s. Meanwhile, the multiple scattering in reservoir I disappears for $t \geqslant 60$ s. *Apparently the penetrating laser beam acts as a semipermeable optical piston,* sweeping the Na vapor towards reservoir II. From the intensity of the fluorescence in the wake of the piston we conclude that the Na density is uniform ($\pm 20\%$) in the swept volume of the capillary and in reservoir I. Typically, towards the end of the piston run this residual Na density, as measured in reservoir I, is $\sim 0.03 n_0$. The residual density may be due to "geometrical" leaks (the capillary is not a perfectly straight and circular tube). Roughly the same phenomena occur for D_1 and D_2 excitation and for linear and circular laser polarization. Piston action only occurs for excitation in the red Doppler wing, the effect being maximum for $\nu_L \sim \nu'$. For instance, the effect disappears when the laser is tuned to resonance with the $F = 2$ transition, $\nu_L = \nu_0$.[13] This shows that radiation pressure[2,3] is not involved.

If, during a piston run, ν_L is suddenly (0.1 s) tuned from ν' to $\nu_0 - 10$ GHz we find, by observing the fluorescence trail, that the step in the Na density profile smears out over ~ 1 cm within 0.1 s, consistent with D being ~ 25 cm^2/s. If, during a piston run, we try to increase the Na density at the dark side of the piston by rapidly heating the appendix (such that $T_{\text{app}} > T_{\text{cap}}$) we find that the piston stops and travels backward to reservoir I, the width and the peak intensity of the fluorescent spot being unchanged. Apparently there is a maximum partial

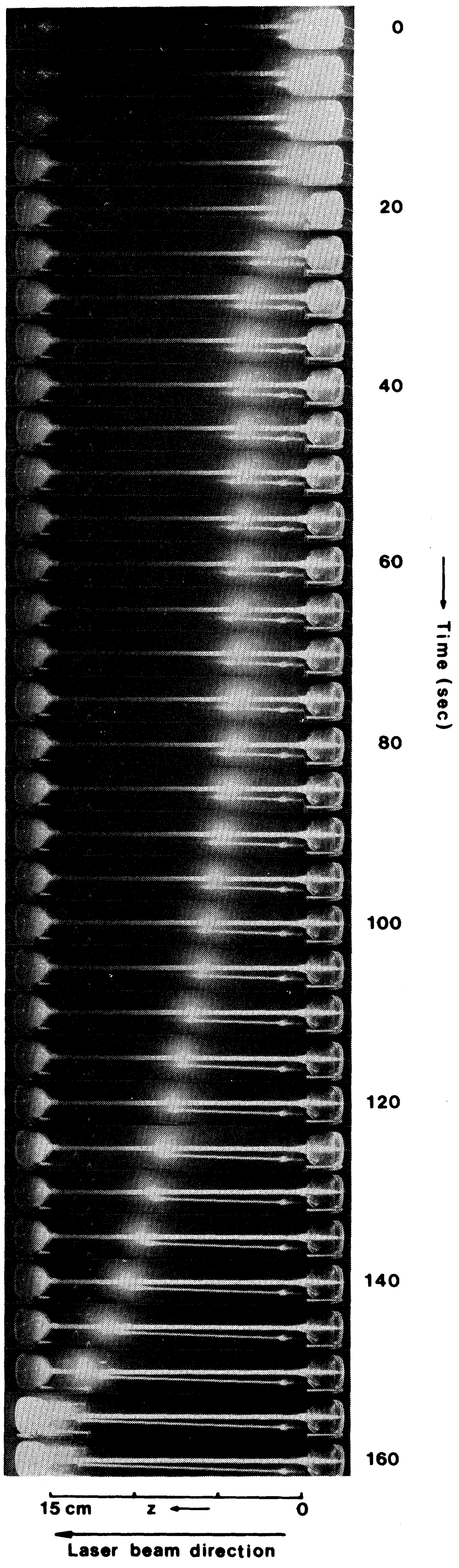

FIG. 2. Example of front propagation through the capillary cell. For $t < 0$ the Na density is uniformly 1×10^{12} cm^{-3}. At $t = 0$ the laser beam is admitted; the laser polarization is linear, the intensity is 3 W cm^{-2}, and the frequency is tuned in the red Na D_2 Doppler wing, $\nu_L = \nu_{F=2} - 0.85$ GHz.

Na pressure which can be sustained by the piston (cf. osmotic pressure). If, after the piston run, the laser beam is intercepted it takes ~ 1 h before the initial uniform Na distribution has been reestablished in the whole cell. We believe that this is mainly determined by the recovery time of the outer glass layer which has been depleted of Na after passage of the piston.

Figure 3 shows the Na density measured at $z = 10$ cm, as a function of time during a typical piston run. The density increases from n_0 to $15n_0$ when the front approaches and decreases, as discussed above, to $\sim 0.03n_0$ when it has passed. Figure 3 also shows the fluorescence from a 0.4-mm section of the capillary at $z = 10$ cm, induced by the main beam and detected by photodiode B (Fig. 1). When passing $z = 10$ cm the fluorescent spot has a velocity v_f of 1.5 mm/s and a width Δz of 2.5–3.0 mm. In the wake of the piston the fluorescence drops to $\sim \frac{1}{40}$ of the peak value; this ratio is not easily interpreted in terms of [Na] because of effects of saturation and radiation trapping.

Simple kinetic considerations show that for an optically dense system LID should manifest itself as front propagation if $D/v_d < (n_0\sigma_a)^{-1} << L$, where σ_a is the optical absorption cross section, L the capillary length, and v_d the drift velocity at the entrance of the system. For a stationary front the absorbers are compressed to a density $n_{\max} \sim v_d/\sigma_a D$. The width of the front $\Delta z \sim 4D/v_d$. Substitution of $D \sim 25$ cm^2/s, $\sigma_a \sim 10^{-12}$ cm^2,[12] and of the experimental results $n_{\max} = 1.5 \times 10^{13}$ cm^{-3}, $\Delta z = 2.5$–3.0 mm yields $v_d \sim 4$ m/s. Since this value is much larger than the front velocity v_f ($\sim$ mm/s) the front may indeed be considered as stationary. In fact, v_f is 10–100 times smaller than diffusive flow speeds which one would expect; we believe that v_f is mainly limited by surface effects. For the case of weakly saturated two-level absorbers LID theory[6] gives, in the limit $\gamma^*_{\text{vcc}} >> \gamma_{\text{rad}}$,

$$v_d \sim u_L \eta \gamma_{\text{rad}}[(\gamma_{\text{vcc}})^{-1} - (\gamma^*_{\text{vcc}})^{-1}], \qquad (1)$$

with u_L the axial thermal velocity of the resonant atoms, γ_{vcc} and γ^*_{vcc} the rates of velocity-changing collisions of Na ^2S and Na ^2P atoms with Ar atoms, γ_{rad} the Na ^2P decay rate, and $\eta = [\text{Na}\,^2P]/[\text{Na}]$. Since fine-structure–changing collisions proceed at rates which are a few times larger than γ^*_{vcc},[14] we assume for simplicity complete mixing of the ^2P sublevels.[15] We incorporate effects of strong saturation and of hf pumping in η. A rough estimate, based on the shape of the fluorescence-excitation spectrum from the capillary, yields $\eta = 0.01$–0.1 for $I_L = 3$ W cm^{-2} and $\nu_L = \nu'$.[9] Substituting this value in Eq. (1), together with $u_L = 5.3 \times 10^2$ m/s, $\gamma_{\text{rad}} = 62$ MHz, $\gamma_{\text{vcc}} = 83$ MHz,[8] and $\gamma^*_{\text{vcc}} = 118$ MHz,[8] yields $v_d = 1$–10 m/s, in order-of-magnitude agreement with experiment.

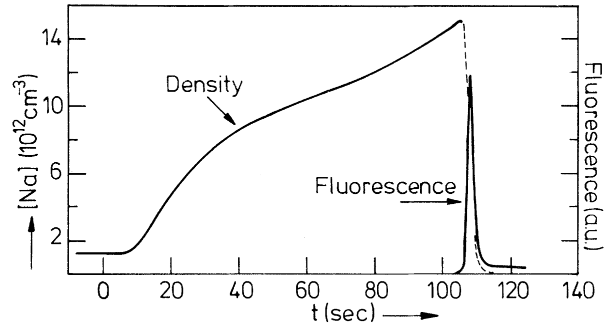

FIG. 3. Na density in the capillary cell and Na fluorescence from the capillary cell, both at $z = 10$ cm (see Fig. 1), as a function of time during propagation of the front. The laser beam is admitted at $t = 0$. The Na density could not be measured during passage of the fluorescent spot; a dashed line has been drawn to guide the eye. Laser polarization, intensity, and frequency are as in Fig. 2.

In summary, we have disclosed a new regime of light-induced drift by demonstrating a semipermeable optical piston which separates partial densities differing by a factor of 500. The drift velocity obtained at present is ~ 4 m/s; we expect that this value can be increased by an order of magnitude by using two-frequency excitation in order to suppress optical hf pumping.[3] Surface effects, which clearly play an important role in our experiment, might be eliminated by use of sapphire capillaries. Obviously, the effects of strong saturation and multilevel absorber structure should be properly incorporated into the theory. A direct application of LID is elimination of trace impurities and isotope separation.[5] In this respect the insight gained in LID piston action for the model system Na:Ar may have bearing on other systems which are less easily diagnosed. In particular, for Na–noble-gas systems knowledge of γ_{vcc} and γ^*_{vcc} from available potentials facilitates comparison with theory.

We thank B. I. M. ten Bosch and J. E. M. Haverkort for theoretical support and Philips Research Laboratories for supplying the Na cell. This work is part of the research program of the Foundation for Fundamental Research on Matter (FOM) and was made possible by financial support from the Netherlands Organization for the Advancement of Pure Research (ZWO).

[a] Visiting scientist from Department of Physics, University of Ljubljana, Ljubljana, Yugoslavia.

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[11] $I_{\text{sat}} \sim 0.3$ W cm^{-2} for $[\text{Ar}] = 3.2 \times 10^{17}$ cm^{-3}.

[12] We estimate that the saturated Na D_2 absorption cross section at $\nu = \nu'$ and $I_L = 3$ W cm^{-2}, taking into account Doppler broadening, collisional broadening, and population partitioning over the ^2S $(F = 1, 2)$ levels.

[13] For $\nu_L = \nu_0$ also the $F = 1$ transition is excited, at 1772 MHz below its resonance. This excitation, however, is hardly velocity selective since $\Delta\nu_\text{D} = 1700$ MHz (full width at half maximum) and $\Delta\nu_c = 225$ MHz (full width at half maximum), where $\Delta\nu_c$ is the collisional width due to 3.2×10^{17} Ar atoms cm^{-3}.

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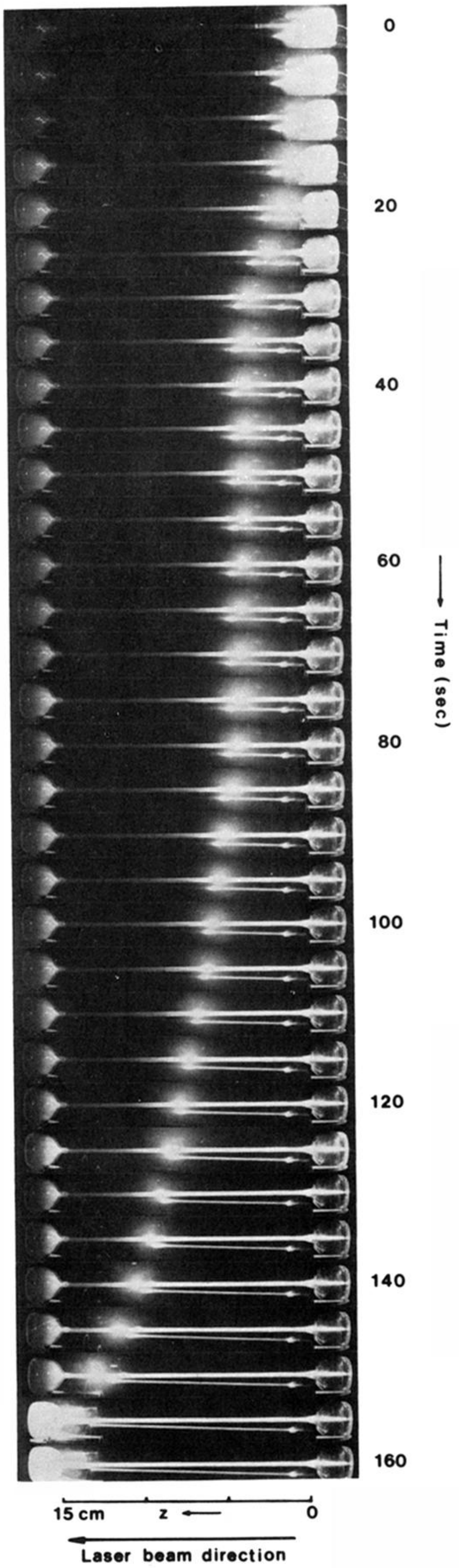

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