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Light-induced vapor jets

S. N. Atutov,^{*} S. Gozzini, C. Gabbanini, and A. Lucchesini Istituto di Fisica Atomica e Molecolare del Consiglio Nazionale delle Ricerche, Via del Giardino, 7-56127 Pisa, Italy

C. Marinelli, E. Mariotti, and L. Moi

Dipartimento di Fisica dell'Universita di Siena, Via Banchi di Sotto, 55 53100 Siena, Italy

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We present experimental evidence of light-induced vapor jets produced by the light-induced-drift effect. They are observed in a cell kept at room temperature, having the active atom source in its central part. The cell has a diameter much larger than that of the laser beam in order to eliminate all interactions with the cell walls. This experiment opens the possibility of studying atomic elements with extremely high temperature of evaporation and/or strong chemical reactivity.

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I. INTRODUCTION

After the light-induced-drift effect (LID) was theoretically predicted [1] and experimentally observed [2,3] much work was devoted to the investigation of this phenomenon (see, for example, Refs. [4,5]). One of the major problems to be solved in order to observe LID is that represented by the atom-wall interaction. In the first LID experiment with sodium [2] and then in the "optical piston" experiment [6], where the LID effect, under optically thick conditions, acts like a piston by pushing the whole vapor to the cell end, a great influence of the capillary cell walls on the vapor drift was found. The adsorption of the atoms on the walls leads both to a very lengthy establishment of a steady-state vapor condition inside the capillary (up to about $10⁵$ times larger then the theoretical predictions [2]) and to a rather small velocity of the optical piston. A detailed analysis of these surface effects has been made both theoretically by Nienhuis [7] and experimentally by Gozzini et al. [8].

These problems have been solved, at least for the alkali-metal atoms with the exception of lithium, by using suitable coatings which permit the friction-free measurements of the drift velocity [9-11]. The thin film of chemical-resistant oil, paraffin or dimethylpolysiloxane deposited on the cell wall, completely eliminates the physical adsorption, making it possible to study LID under ideal conditions. Unfortunately such layers, which cannot be heated up to high temperatures (the maximum is about 200'C for dimethylpolysiloxane), can be used only for elements that have a comparatively low temperature of evaporation and low chemical reactivity. As it is rather difficult to find new layers that are able to resist both high temperatures and strong chemical aggression, any further experimental development of LID was restrained. This limitation is very detrimental, considering that the existing cw laser sources can excite a large part of the alkali metals, alkaline-earth-metal elements, and rare-earth elements, and that new applications of LID seem possible. Recently, for example, an explanation of the "chemical peculiar stars" that show nonhomogeneity in surface composition, by the LID effect produced by the thermal emission from the center of the star spectrally modified by atmospheric Fraunhofer absorption has been suggested [12] and significant experimental evidence supporting this hypothesis has been obtained [13,14].

An attempt to study LID far from the cell walls has been performed by Gozzini et al. [15], who adopted a spherical cell and observed sodium vapor compression at its center. A different approach is presented here which makes use of a large cell that is kept at room temperature and is filled with a buffer gas. The cell has in its middle part an active atom source that, when heated up, produces a spatially well-delimited vapor cloud. When a properly detuned laser beam crosses this cloud, a vapor jet expanding in "the free space" represented by the large buffer gas volume is produced by LID. This is made possible by the fact that the induced-drift velocity can be much higher than the thermal diffusion one [10,16]. Therefore the time the vapor spends diffusing out of the light beam is long enough to allow the vapor to drift along the laser beam, thus creating the vapor jet. This effect has been experimentally demonstrated by us with sodium.

II. THEORY

The vapor density N is ruled by the general diffusion equation [17]:

$$
\frac{\partial N}{\partial t} + \text{div}[UN + \nabla(DN)] + \frac{N}{\tau} = 0, \qquad (1)
$$

where D is the diffusion constant in the ground state, U is the effective drift velocity, and τ is the N decay time due to chemical losses. The detailed solution of this equation, which is valid if the velocity distributions remain approximately Maxwellian and if the light pressure is ignored, is in general very complicated. The vapor is not in a steady-state condition, its geometry is not perfectly defined, and strong temperature gradients are present. Moreover, the problem is no longer one dimensional, as was the case with the capillary cells, and the boundary conditions are more difficult to specify. An effort to

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achieve a detailed analysis is actually in progress; nevertheless, a crude but very useful model can be presented here with the aim of qualitatively describing the experimental results. In fact, an estimation of the vapor density N inside the jets and the length of the jets as a function of the laser power and of the laser beam diameter can be attempted once a few approximations are made. First of all, the average drift velocity U , describing the movement of the vapor inside the laser beam, is introduced. Second, when the diameter of the laser beam is considerably smaller than the length of the vapor jet, the vapor drift is assumed to be the same as in a capillary cell where it is pushed or pulled by LID as a whole. In this case U is smaller than that along the beam axis where the power density is maximum and, in the case of small saturation, U is proportional to the average power density. Taking into account the Gaussian profile of the laser beam and assuming that the cell is in the optically thin regime, U can be written in the form

$$
U = U_0 \frac{d_b^2}{d_d^2} \left\{ 1 - \exp[-(d_d/d_b)^2] \right\},\tag{2}
$$

where U_0 is the drift velocity on the beam axis, d_b is the full diameter of the laser beam, and d_d is the diameter of the laser beam inside the cell which is changed by using a diaphragm. Moreover we assume that both D and U do not depend on the cylindrical coordinates r , z, and ϕ , that the chemical losses are negligible, i.e., $1/\tau = 0$, and that

$$
\frac{\partial N}{\partial \phi} = 0.
$$

Then in the stationary regime Eq. (1) becomes

$$
D\left(\frac{\partial^2}{\partial r^2} + \frac{1}{r}\frac{\partial}{\partial r} + \frac{\partial^2}{\partial z^2}\right)N - U\left(\frac{1}{r} + \frac{\partial}{\partial r} + \frac{\partial}{\partial z}\right)N = 0.
$$
\n(3)

It is possible to solve Eq. (3) by assuming $N = A(r)B(z)$, where A and B are only functions of r and z , respectively. This leads to the two equations

$$
\frac{\partial^2 A}{\partial r^2} + \frac{1}{r} \frac{\partial A}{\partial r} - \frac{u}{Dr}A - \frac{u}{D} \frac{\partial A}{\partial r} = -\frac{\gamma}{D}A
$$
 (4)

and

$$
\frac{\partial^2 B}{\partial z^2} - \frac{u}{D} \frac{\partial B}{\partial z} - \frac{\gamma}{D} B = 0,
$$
 (5)

where γ is a constant. We are interested in the solution of Eq. (5), assuming the boundary conditions

$$
B(-0) = B(0) = B_0 \tag{6}
$$

and

$$
B(-\infty) = B(\infty) = 0.
$$
 (7)

The solution of Eq. (5) can be found in an elementary

fashion and is Long cavity dye laser

$$
B = \begin{cases} B_0 \exp(-z/L_1), & z > 0 \\ B_0 \exp(z/L_2), & z < 0 \end{cases}
$$
 (8)

where L_1 and L_2 are characteristic lengths of the vapor iets

$$
(L_{2,1})^{-1} = \left[\left(\frac{U}{2D} \right)^2 + \frac{\gamma}{D} \right]^{1/2} \pm \frac{U}{2D}, \tag{9}
$$

and γ^{-1} plays the role of the lifetime of Na atoms inside the laser beam. This lifetime corresponds to the time necessary for the sodium atoms to diffuse out of the laser beam and is given by

$$
\gamma^{-1} = \frac{d_b^2}{16D} \,. \tag{10}
$$

III. EXPERIMENTAL RESULTS AND DISCUSSION

The experiment outline is sketched in Fig. 1. The main parts of the experimental setup are a cell filled with Kr (pressure 44 Torr) as a buffer gas and a "lamp laser" [18] which, as reported in Ref. [16], impresses drift velocities higher than those induced by a single-mode laser under similar conditions. The cell is a glass tube of 65 mm in diameter and 330 mm long with a Na vapor source in the middle. The vapor source consists of a small glass tube (3 mm in diameter, 10 mm in length), containing metallic Na, placed in a vertical position, and heated up independently of the cell, which is kept at room temperature. The laser has a long cavity configuration (about 5 m), a total bandwidth of the order of 1-2 GHz and a maximum power equal to about 270 mW. This corresponds to a mean power density of about $10-20$ mW/cm² for each longitudinal mode. The laser beam, tuned to the D_2 line

personal computer; PD1,2, photodiodes; M, Mirror; PM, photomultiplier.

of sodium, is sent through the cell along its axis and slightly above the vapor source. Its diameter is about 0.5 cm and can be reduced by a diaphragm. In complete contrast to the other LID experiments we use a cell with a diameter much larger (about 13 times) than the diameter of the laser beam. This eliminates the influence of the cell walls. The relatively high pressure of the buffer gas and the diameter of the laser beam were chosen in order to allow a long enough diffusion lifetime of the sodium atoms across the laser beam. For a laser beam diameter larger than 0.5 cm our assumption about the absence of chemical losses becomes incorrect. A good-quality objective at a double focus distance from the laser beam reproduces the sodium vapor distribution on the plane of a differential detector system (DDS) consisting of two photodiodes (PDI, PD2) placed symmetrically with respect to the vapor source position $z = 0$. When the laser is on, the vapor cloud around the source is easily observable by naked eyes. An optical fiber that is connected to a photomultiplier and positioned in the middle of the differential photodetector system gives a signal proportional to the fluorescence in correspondence of the vapor source. The measurements are made in the following way. First the laser frequency is tuned to the fluorescence maximum, then the DDS is carefully positioned in order to have zero signal. The laser is then detuned and the LID effect becomes active. An unbalance between the two photodiodes that is proportional to the induced asymmetry of the vapor cloud is obtained. The signals from the photomultiplier and from the photodiodes are then collected as a function of the laser frequency detuning, of the laser beam diameter, and of the laser power, and are recorded by a digital

FIG. 2. (a) Fluorescence signal I_F and (b) DDS I signal as a function of the laser detuning Δv_L (referred to the central laser frequency).

oscilloscope connected to a computer .

The differential signal, normalized to the signal from the photomultiplier, can be derived from the previous calculations and it is expressed by

$$
I = \frac{B(z_0) - B(-z_0)}{B_0}
$$

= exp(-z₀/L₂) - exp(-z₀/L₁), (11)

where $2z_0$ is the distance from one photodiode to the other.

As expected, a very spectacular manifestation of the vapor jet can be observed easily by naked eyes. When the laser frequency is suddenly detuned from resonance the vapor distribution around the source becomes sharply asymmetrical and a Na steady-state vapor jet appears. It looks like a bright pencil several centimeters long, and is very sensitive to the laser detuning. The vapor jet is, in fact, pulled or pushed depending on the laser frequency. The effect is even more spectacular if the laser beam, after crossing the cell, is reflected back under a very small angle. A second jet is then created which moves in phase opposition with the previous one.

In accordance with the LID features, the jet length depends on the radiation power density and on the laser frequency detuning. In the present case it depends also on the laser beam diameter. Since the diffusion time of the atoms out of the laser beam is proportional to the square of the beam diameter, as it tends to zero the jet disappears. We have verified this by making the laser beam smaller than ¹ mm.

In Fig. 2, two curves as a function of the laser detuning are shown. Figure $2(a)$ represents the sodium fluorescence in correspondence of the vapor source. This curve is symmetrical because the vapor density is kept fixed at that point. Figure 2(b) shows the DDS signal taken at a distance $2z_0$ between photodiodes equal to 3.4 cm. This signal, as expected, has a dispersive behavior and it shows the existence of the vapor jet going from one side to the other of the vapor cloud. The laser frequency is scanned over a few gigahertz by electronically scanning an intracavity etalon.

The amplitude of the differential signal, I , as a function of the laser power and of the diaphragm diameter is

FIG. 3. Plot of the DDS signal I as ^a function of the laser power P_L . The open points represent the experimental measurements and the solid curve is the best fit obtained from Eq. (11) $(d_d = 5 \text{ cm}).$

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FIG. 4. Plots of the DDS signal I as ^a function of the laser beam diameter d_d . The open points represent the experimental measurements and the solid curve the best fit obtained from Eq. (11). (a) $P_L = 116$ mW, $U = 6.4$ m/s; (b) $P_L = 212$ mW, $U = 14.2$ m/s.

'Permanent address: Institute of Automation and Electrometry, Novosibirsk 90, Russia.

- [1] F. Kh. Gel'mukhanov and A. M. Shalagin, Pis'ma Zh. Eksp. Teor. Fiz. 29, 773 (1979) [JETP Lett. 29, 711 (1979)].
- [2] V. D. Antsigin, S. N. Atutov, F. Kh. Gel'mukhanov, G. G. Telegin, and A. M. Shalagin, Pis'ma Zh. Eksp. Teor. Fiz. 30, 262 (1979) [JETP Lett. 30, 243 (1979)].
- [3] V. N. Panfilov, V. P. Strumin, P. L. Chapovsky, and A. M. Shalagin, Pis'ma Zh. Eksp. Teor. Fiz. 33, 52 (1981) [JETP Lett. 33, 48 (1981)].
- [4] Light-Induced Kinetic Effects on Atoms, Ions and Molecules, edited by L. Moi, S. Gozzini, C. Gabbanini, E. Arimondo, and F. Strumia (ETS Press, Pisa, 1991).
- [5] E. R. Eliel, Adv. At. Mol. Opt. Phys. (to be published).
- [6] H. G. C. Werij, J. P. Woerdman, J. J. M. Beenakker, and I. Kuscer, Phys. Rev. Lett. 52, 2237 (1984).
- [7] G. Nienhuis, Opt. Commun. 62, 81 (1987).
- [8l S. Gozzini, G. Nienhuis, E. Mariotti, G. Paffuti, C. Gabbanini, and L. Moi, Opt. Commun. (to be published).

shown in Figs. 3 and 4, respectively. As can be seen, all curves show a nonlinear dependence as predicted. In Fig. 3 the signal is almost zero for laser power lower than 25 mW, it has a roughly linear dependence up to about 150 mW and then begins to saturate. The behavior of I in Figs. $4(a)$ and $4(b)$ looks similar, even if in Fig. $4(b)$ the saturation is more evident. The continuous curves show the best fit of Eq. (11) with U as a free parameter. In Figs. 3 and 4 the other parameter values are z_0 =1.7 cm, $D = 3$ cm²/sec [17]. The effective drift velocities obtained from the fitting result are in agreement with those obtained under similar conditions in coated capillary cells.

IV. CONCLUSIONS

The obtained results clearly show the effectiveness of this approach to LID experiments. This method can be used successfully any time atoms with very low vapor densities and high chemical reactivity have to be studied. The method also permits isotope separation and very thin layer deposition on prepared surfaces. Finally, it can contribute to the understanding of transport phenomena in open spaces or far from equilibrium situations.

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[9] S. N. Atutov, Phys. Lett. 119, 121 (1986).

- [10]S. N. Atutov, St. Lesjak, S. P. Pod'yachev, and A. M. Shalagin, Opt. Commun. 60, 41 (1986).
- [11]J. H. Xu, M. Allegrini, S. Gozzini, E. Mariotti, and L. Moi, Opt. Commun. 63, 43 (1987).
- [12] S. N. Atutov and A. M. Shalagin, Pis'ma Astron. Zh. 14, 664 (1988) [Sov. Astron. Lett. 14, 284 (1988)].
- [13]S. Gozzini, C. Marinelli, E. Mariotti, C. Gabbanini, A. Lucchesini, and L. Moi, Europhys. Lett. 17, 309 (1992).
- [14] F. Wittgrefe, J. L. C. van Saarlos, S. N. Atutov, and E. R. Eliel, J. Phys. B 24, 145 (1991).
- [15] S. Gozzini, D. Zuppini, C. Gabbanini, and L. Moi, Europhys. Lett. 11, 207 (1990).
- [16]S. Gozzini, J. H. Xu, C. Gabbanini, G. Palfuti, and L. Moi, Phys. Rev. A 40, 6349 (1989).
- [17] S. A. Atutov, I. M. Ermolaev, and A. M. Shalagin, Zh. Eksp. Teor. Fiz. 92, 1215 (1986) [Sov. Phys. JETP 65, 679 (1987)].
- [18]J. Liang, L. Moi, and C. Fabre, Opt. Commun. 52, 131 (1984).

FIG. 1. Sketch of the experimental apparatus. PC denotes personal computer; PD1,2, photodiodes; M, Mirror; PM, photomultiplier.