# Light-induced drift of barium atoms

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An observation of light-induced drift (LID) of atoms with an intermediate metastable level between the ground and excited resonance levels is reported. The drift of atoms of barium was investigated. Molecular nitrogen was used as a buffer gas. The $5d\,^1D$ metastable state of Ba atoms was quenched in collisions with $N_2$ molecules. It has been deduced from the experimental results that the collision cross section of Ba atoms with $N_2$ molecules is larger for Ba atoms in the ground state than in the $6p\,^1P$ excited state. The maximal LID velocity was about 250 cm/s.

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Light-induced drift (LID) of atoms and molecules in a buffer gas was predicted by Gel'mukhanov and Shalagin [1]. Many experimental and theoretical papers have been devoted to LID investigations of atoms as well as molecules (see Ref. [2]). A serious problem in LID investigations of atoms is adsorption of the atoms on experimental pipe walls. Up until now, the problem had been solved only for alkali metals by employing suitable coatings which completely eliminate physical adsorption [3]. This is the reason why the LID investigations of atoms have been limited to alkali metals (one exception being the delicate experiment with the LID of Ne atoms in a gas discharge [4] ). This limitation is restricting because LID can be useful in many scientific and practical applications such as in an explanation of chemically peculiar stars [5], radioisotope and isomer separation [6,7], measurements of optical transition frequencies of short-lived isotopes and isomers [8], and traps of radioactive atoms [9]. Recent LID experiments with sodium atoms in a wide cold tube [10,11] show how to avoid the adsorption problem and how to conduct LID investigations of atoms of more refractory elements.

A realization of LID requires multifold (more than $10^3$) photon absorption by one optically active atom. If there is an intermediate metastable level, between the ground and excited resonance atom levels the atoms can be trapped in the metastable state. Without a forced depopulation of the metastable state, most of the atoms will be pumped into the metastable level after several events of photon absorption and emission. As a result, the optically active atoms cease to interact with laser radiation and consequently drift under the light action. The metastable states of atoms can be quenched in collisions with molecules [12].

This paper reports successful investigations of a LID of metallic atoms that do not belong to the alkali-metal group, a LID of Ba atoms. The Ba atom resonance transition between the ground state $6s^2\,^1S_0$ and $6p\,^1P$ level, see Fig. 1, can be easily reached by a cw dye laser ($\lambda = 553.55$ nm). The probability ratio that a barium atom excited into the $6p\,^1P$ state radiates into the ground $6s^2\,^1S_0$ and metastable $5d\,^1D$ state is reported to have a value of 25 [13]. The dipole transition from the $5d\,^1D$ metastable level to the ground state is parity forbidden. Molecular nitrogen quenching the metastable state was used as a buffer gas. $N_2$ is the most suitable buffer gas for a LID study of Ba atoms because it does not react rapidly with barium in the gas phase [12]. Using the example of barium atoms, an experimental LID investigation of atoms with an intermediate metastable level is presented in the paper.

The isotope and hyperfine splittings of the $6s^2\,^1S_0$ – $6p\,^1P$ transition do not exceed the Doppler width of the transition at a 200 °C temperature of Ba atoms [14]. This condition allows us to tune the laser radiation in such a way that all isotopes drift in the same direction. For this reason we could use the natural isotope mixture in the LID investigation of Ba atoms.

The experimental setup was similar to the one used in [10] for an LID study of sodium radioactive isotope atoms. The experimental cell was a glass tube with the inner diameter of 1.5 cm. There was a graphite crucible with barium metal in the center of the tube, see

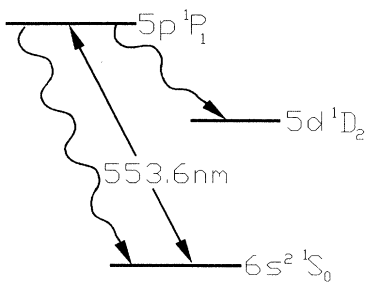


FIG. 1. Energy levels of atomic Ba referred to in the text.

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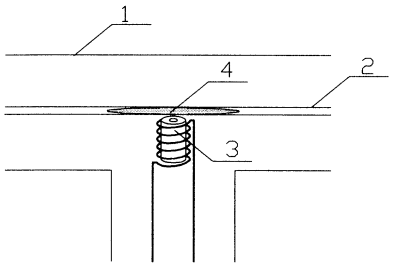


FIG. 2. The experimental cell for investigation of LID of Ba atoms. 1, glass tube; 2, laser beam; 3, graphite crucible with metallic barium; and 4, Ba atom cloud.

Fig. 2. The crucible was heated up to 600 °C–650 °C by a nichrome spiral. The beam of the single-frequency cw dye laser (a joint product of Inversion LTD and University Laser Center, Novosibirsk) passed through the tube one millimeter above the crucible. The fluorescence of the barium vapor was easily observable with the naked eye.

The barium atoms drifted along or opposite the laser beam until they left the laser beam region as a result of diffusion. The LID of barium atoms shifted the shining barium cloud to the right or to the left according to the LID direction. The cloud movement was registered with a video camera and video records were digitized later with a computer. The transverse size of the shining cloud was determined by the diameter of the laser beam; the length of it dropped from 5 to 1 mm when increasing the $N_2$ pressure (5–200 Torr). The pressure dependency of the barium vapor cloud size is proof that barium atoms react with nitrogen in given experimental conditions. We estimate the order of the magnitude of the barium atom lifetime as $7 \times 10^{-3}$ s.

The LID for tree-level systems has been discussed in [15]. The LID velocity $v$ for Ba atoms can be expressed as

$$
\begin{aligned}
v &= \overline{u}\left[\frac{\nu_s-\nu_p}{\nu_s}\frac{\Gamma_{pd}}{\nu_{ds}+\nu_d}\frac{\nu_s-\nu_d}{\nu_s}\right]\frac{\Gamma_p}{\Gamma_p+\nu_p}w_p\varphi(\Omega),\\
\varphi(\Omega) &= \mathrm{Re}[\xi w(\xi)]/\mathrm{Re}[w(\xi)],\\
w(\xi) &= e^{-\xi^2}\left[1+\frac{2i}{\sqrt{\pi}}\int_0^{\xi}e^{t^2}dt\right],\\
\xi &= \Omega/k\overline{u}+i\Gamma/k\overline{u}.
\end{aligned}
\tag{1}
$$

Here, $\overline{u}$ is the Ba atom average thermal velocity, $k$ is the laser the radiation wave number, $\Omega$ is the radiation frequency detuning from the Ba atom absorption line center, $\nu_{s,p,d}$ is the transport collision rates of Ba atoms in the $6s^2\,^1S_0$, $6p\,^1P$, and $5d\,^1D$ states with $N_2$ molecules, $\nu_{ds}$ is the quenching rate of the barium metastable state, $\Gamma_p$ is the constant of the radiative decay of the $6p\,^1P$ state, $\Gamma_{pd}$ is the constant of the radiative decay of the $6p\,^1P$ state into the $5d\,^1D$ one, $\Gamma$ is the homogeneous width of the barium atom transition, $w_p$ is the fraction of the Ba atoms in the excited $6p\,^1P$ state, and $\varphi(\Omega)$ is the antisymmetric function of $\Omega$ (specific for the LID)

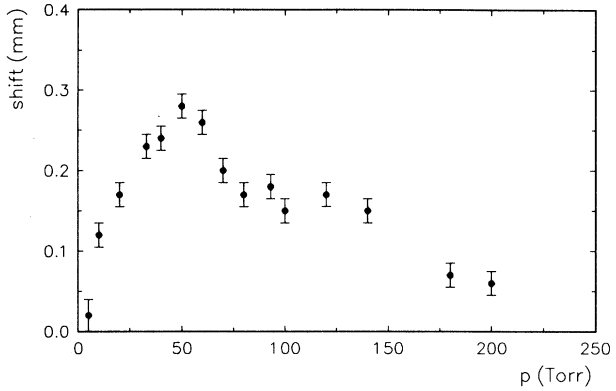


FIG. 3. The dependence of the shift of the Ba atom cloud on nitrogen pressure. Laser power is 45 mW, beam diameter is 0.5 mm.

defined in [15]. In our case, $\Gamma_{pd} \ll \nu_d$, and we can omit the second term in expression (1). The LID velocity is thus defined by the difference of the transport collision rates of the ground $6s^2\,^1S_0$ and $6p\,^1P$ excited states.

In our experiment the barium cloud shift and, correspondingly, also the LID of barium atoms were in the same direction as the movement of the excited barium atoms. This proves that the transport collision cross section of barium atoms with nitrogen molecules is greater for the $6s^2\,^1S_0$ ground state than that for the $6p\,^1P$ excited state of barium atoms. This result came as a surprise to us because the collision cross sections of atoms of alkali metals is greater for the $P$ excited state than for the $S$ ground state. However, a smaller cross section in the $6p\,^1P$ excited state of barium atoms was already observed in an experiment with the scattering of crossing atomic beams [16]. Recently, Red'ko in a theoretical paper [17] on LID of alkaline-earth-metal atoms, predicted that the transport collision cross section of Mg atoms is greater for the $3s^2\,^1S_0$ ground state than that for the $3p\,^1P$ excited state.

The barium cloud shift as a function of nitrogen pressure is shown in Fig. 3. The laser beam diameter was 0.5 mm and the laser power was about 45 mW. The barium cloud shift as a function of laser power is shown in Fig. 4. The measurements were carried out with two

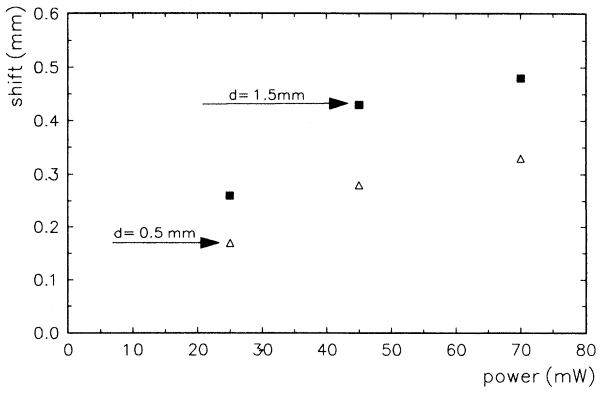


FIG. 4. The dependence of the shift of the Ba atom cloud on laser power. The beam diameters are 0.5 and 1.5 mm.

laser beam diameters, 0.5 and 1.5 mm, at the buffer gas pressure of 40 Torr. The barium cloud shift increases with the laser beam diameter at a constant laser power. This is evidence that a saturation of the transition takes place in the experiment. If saturation was absent, the LID velocity would be proportional to the laser power density depending on the beam radius $r$ as $r^{-2}$ at a constant laser power. The diffusion time for atoms leaving the laser beam depends on the beam radius as $r^2$. As a result, the Ba cloud shift is independent of the laser radius when saturation is absent.

Let us discuss the role of buffer gas convection in the experiment. Convection can accelerate the removal of barium atoms from the laser beam. If the convection flow velocity is higher than 100 cm/s, the time of the atom presence in the laser beam should be determined by the convection flight of barium atoms through the beam. In this case a Ba cloud shift should decrease as the laser beam radius increases, but we observed the reverse tendency in an experiment (see Fig. 3). Furthermore, if convection takes place, the barium atom distribution should be drawn out like a candle flame. But a scanning of the Ba cloud showed that decreasing the Ba vapor density in the upward direction is similar to that in the horizontal direction. The estimation of the Rayleigh number [18] in the experimental conditions $\mathcal{R} < 10$ corroborates the absence of convection also. Convection instability sets in when $\mathcal{R} > 10^3$ [18]. Therefore, we will not take into account convection for these considerations.

To find the connection between the LID velocity $v$ and a cloud shift, it is necessary to consider the diffusion and the drift of barium atoms. The propagation length $l$ of barium atoms is defined by their lifetime $\gamma^{-1}$ as $l = \sqrt{D/\gamma}$, where $D$ is the diffusion coefficient. The experimental value of propagation length $l \simeq 2$ mm is much more than the beam radius $r_0 \simeq 0.25$ mm, while this length is much smaller than tube radius $R \simeq 7.5$ mm. This is why we consider the case of the unlimited medium and ignore the boundary conditions on the cell wall. The stationary barium vapor distribution $n$ obeys the diffusion equation with the drift

$$-D \triangle n(\vec{r}) + \gamma n(\vec{r}) = -v(\vec{r}) \frac{\partial n(\vec{r})}{\partial z} + Q(\vec{r}). \qquad (2)$$

Here, $Q(\vec{r}) = N_0 \delta(\vec{r} - \vec{a})$ is a point source of Ba atoms, which is at a distance of $a \simeq 1$ mm from the beam axis; $\vec{r} = (z, \rho, \varphi)$ is the cylindrical coordinate. The LID velocity is not equal to zero only inside the laser beam. For simplicity we suppose that the LID velocity does not depend on coordinates inside the beam: $v(\rho) = v$ when $\rho \leq r_0$. Let us solve Eq. (2) by assuming that the LID is small and the shift is much less than the propagation length: $\langle z \rangle \ll l/\ln(l/a)$. The average shift of the distribution function under LID in the linear approximation is given by the expression (we omit terms of second order in $r_0/l$):

$$\langle z \rangle = \frac{v\, r_0^2}{2D} \left[ \ln\left(\frac{2l}{r_0}\right) - C + \frac{1}{2} \right], \qquad (3)$$

where $C = 0.577\ldots$ is Euler's constant. The LID velocity can be evaluated from the average shift as

$$v = \langle z \rangle \frac{2D}{r_0^2 \ln(2l/r_0)}. \qquad (4)$$

To obtain an estimate of the unknown diffusion coefficient $D$ of Ba atoms in $N_2$, we used the known one of Xe in $N_2$ [19] (because Xe is the nearest Ba element with a known $D$). An additional uncertainty of the diffusion coefficient is caused by a change of temperature from 650 °C on the crucible to room temperature on the cell wall. We estimate that the characteristic gas temperature in the region of Ba atom interaction with the laser beam (3 mm away from crucible) was about 200 °C. Assuming this, the diffusion coefficient is estimated as $D \simeq 180 \times P^{-1}$ cm$^2$/s, where $P$ is the nitrogen pressure expressed in units of Torr [19].

The LID velocity of the Ba atoms, calculated according to (4) as a function of nitrogen pressure under 45 mW laser power, is shown in Fig. 5. There is a maximum LID velocity at a pressure of about 10 Torr. In comparison with the same dependence for Na atoms [3], the maximum LID velocity is shifted into the high pressure region because a quenching rate is proportional to a buffer gas pressure and the rapid quenching of the metastable $5d\,^1D$ state of Ba atoms is necessary for an effective LID action. The $5d\,^1D$ state lifetime is about $2 \times 10^{-7}$ s at a nitrogen pressure of 10 Torr [12]. This is about 20 times greater than the radiative lifetime of Ba atoms in the $6p\,^1P$ excited state ($\simeq 1.2 \times 10^{-8}$ s). It means that the population of the $5d\,^1D$ metastable state is not greater than the resonance for the $6p\,^1P$ state. The maximum of the LID velocity achieved in our experiments was about 250 cm/s. It may be noted that the large uncertainty in the diffusion coefficient can result in a systematic error of an LID velocity calculation. We therefore conclude that the LID velocity of Ba atoms is calculated within a constant factor of order of magnitude 1.

When we substitute in (1) the values of the experimental parameters at the buffer gas pressure of 40 Torr $v \sim 2$ m/s, $\overline{u} \simeq 150$ cm/s, $\Gamma_p/(\Gamma_p + \nu_p) \sim 0.2$, $w_p \sim 0.1$, and $\varphi(\Omega) \simeq 0.5$ [15], we can estimate the value of the collision rates relative difference as $(\nu_s - \nu_p)/\nu_s \sim 1$. This agrees with the value calculated in [17] for the Mg atoms.

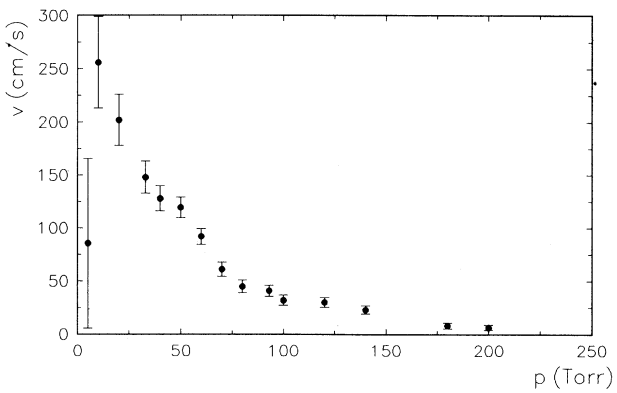


FIG. 5. The dependence of the LID velocity on nitrogen pressure. Laser power is 45 mW, beam diameter is 0.5 mm.

It should be noted that the metastable state can be depopulated by an excitation of an additional laser radiation, for example, with a laser wavelength of $\lambda = 0.5829$ $\mu$m into the $6p5d\,^1P_1$ state or $\lambda = 1.5$ $\mu$m into the $6s6p\,^1P_1$ state [14]. This would enable us to use a noble buffer gas with a significantly lower pressure. The LID velocity of Ba atoms should then be considerably higher [3].

We conclude that the results obtained are proof that LID can be realized with atoms with intermediate metastable states if the states are effectively quenched. For effective quenching an inert molecular gas can be applied. The maximum of the LID velocity achieved in our experiments was about 250 cm/s. The LID velocity can be sufficiently higher if the metastable state is deexcited with an additional tunable laser radiation. In such a case a noble buffer gas can be used and consequently the barium atom lifetime can be sufficiently prolonged. That should play a key role in future isotope separation investigations.

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