Light-induced diffusion pulling of sodium vapor into a light beam

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Light-induced diffusion pulling of sodium vapor into a light beam in an atmosphere of helium, neon, or argon is investigated experimentally. The relative change of the coefficient of diffusion of sodium atoms excited to the $3p$ state in helium, neon, or argon is determined from the experimental data.

1. INTRODUCTION

An interesting effect of radiation on the state of gas particles that absorb it resonantly was predicted in 1970.[1] Its gist is that the absorbing particles can be pulled into or pulled out of the light beam. This effect is described in greater detail in Ref. 2, where it is shown that it takes place only in the presence of a buffer gas, is diffusive, and can lead to appreciable density difference between the absorbing particles. It was shown in Ref. 3 that this effect of light-induced diffusive pulling-pushing (LDP) effect can be comparable with the saturation effect (for the latter see, e.g., Refs. 4 and 5). Observation of the LDP effect was first reported in Ref. 6.

Let us recall the physical basis of the LDP effect. Assume that the cell containing a mixture of two gases has a larger diameter than the light beam, and that the radiation is absorbed by one of the components in a transition from the ground to the first-excited state. The density of the buffer gas is assumed to be high enough to permit the excited particle to undergo several collisions during its lifetime; we can regard its motion as diffusion. On the other hand, we assume that the lifetime of the excited state is considerably shorter than the stay of the particle in the light beam. Under these assumptions, excited particles are present only in the light beam. At the same time, the density of the unexcited particles is lower inside than outside the beam. The spatial inhomogeneity of the beam is known to induce diffusion of the excited and unexcited particles from and into the beam, respectively. In general, the diffusion coefficients of the excited and unexcited particles in the buffer gas are different. Consequently, at equal absolute values of the particle-density gradients in the ground and excited states, i.e., when the gas of the absorbing particles has on the whole a uniform density, the fluxes of the excited and unexcited particles do not balance each other. This means existence of an exciting-particle gas flux as a whole. It is clear that if the particles in the excited state have a smaller diffusion coefficient, the absorbing particles will be pulled into the light beam. In the opposite situation the particles are pushed out of the beam. Under stationary conditions the fluxes balance each other, but the density of the absorbing particles becomes spatially inhomogeneous.

Our present aim was to record and investigate the LDP effect in sodium vapor mixed with noble gases.

2. DERIVATION OF FUNDAMENTAL EQUATIONS

The characteristics of the LDP effect were considered in Refs. 2 and 3 using two- and three-level models of absorbing particles. The interaction of a sodium atom with radiation at resonance with the $3s$–$3p$ transition is generally speaking not described by such models in view of the fine and hyperfine splitting of the states and in view of the degeneracy in the angular-momentum projections. Under the conditions of our experiment (buffer gas pressure several hundred Torr, linearly polarized radiation), however, many of the results of Refs. 2 and 3 can be used, and there is no need for a cumbersome successive solution of the equations for the density matrix.

At a buffer-gas pressure of hundreds of Torr, according to the data on sodium D-line broadening,[7] the homogeneous half-width Γ of the absorption line is several times larger than the Doppler value $k\bar{v}$ or the hyperfine splitting Δ (Ref. 8):

$$\Gamma > k\bar{v},\ \Delta, \tag{1}$$

where $\bar{v}$ is the thermal velocity. Under these conditions the broadening can be regarded as homogeneous, the hyperfine structure of the sodium levels cannot be resolved and can be disregarded if the radiation is linearly polarized.

Figure 1 shows the level and stimulated-transition scheme in the sodium atom following absorption of linearly polarized radiation in the $3s^2S_{1/2}$–$3p^2P_{1/2}$ transition, with the hyperfine structure neglected. Under the conditions of

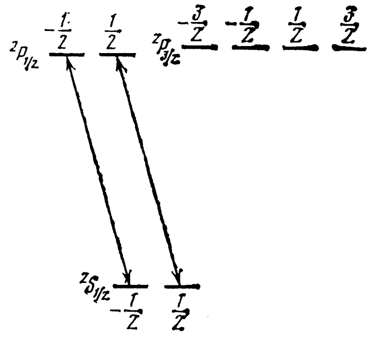

FIG. 1. Structure of levels and of induced transitions of the sodium atom upon absorption of linearly polarized radiation in the $3s^2S_{1/2}$–$3p^2P_{1/2}$ transition.

our experiments, the fraction of the excited sodium atoms did not exceed 20%. This means that the rate w of stimulated transitions is less than the rate Γ_m of the spontaneous decay of the excited state:

$$\Gamma_m > w. \tag{2}$$

In turn, at a buffer-gas pressure of several hundred Torr, the rate of spontaneous decay is lower by more than an order of magnitude than the frequency ν of the gas-kinetic collisions,

$$\nu \gg \Gamma_m, \tag{3}$$

and is lower by more than two orders than the characteristic rate γ of the collisional transitions between the sublevels of the excited state,[7]

$$\gamma \gg \Gamma_m. \tag{4}$$

We note finally that the relation

$$\Gamma_m \gg a^2/D, \tag{5}$$

was satisfied in the experiment (a is the beam radius and D is the sodium-atom diffusion coefficient in the buffer gas). This means that the lifetime of the excited state (~ 16 ns) is much shorter than the time of diffusion of the atom from the light beam ($\gtrsim 10^{-4}$ s).

Under the conditions (1)–(3), the sodium-atom distribution in velocity can be assumed close to Maxwellian. It follows from (4) and (5) that the sodium-atom distribution in the excited-state sublevels is governed mainly by collisions. As a result, regardless of the radiation intensity and of whether the radiation is absorbed in the transition $3s^2S_{1/2}$–$3p^2P_{1/2}$ or $3s^2S_{1/2}$–$3p^2P_{3/2}$, a Boltzmann distribution obtains. Since the fine-splitting energy is low compared with thermal, the sublevels are practically equally populated. Finally, it is obvious that linearly polarized radiation does not alter the equilibrium distribution population over the ground-state levels.

In view of the foregoing, it is easy to obtain a balance equation for the populations of the ground (N_s) and excited (N_p) states:

$$(N_p g - N_s) w + \Gamma_m N_p = 0, \quad N_s + N_p = N. \tag{6}$$

Here N is the density of the sodium vapor, while g is the population ratio of the ground-state sublevels coupled to the ground state by stimulated transitions and of all the sublevels of the excited states. It is easily verified that $g = 1/3$ for radiation absorption from either the $3s^2S_{1/2}$–$3p^2P_{1/2}$ or the $3s^2S_{1/2}$–$3p^2P_{3/2}$ transition.

It follows from (6) that

$$N_p = \frac{3\varkappa}{3+4\varkappa} N, \quad N_s = \frac{3+\varkappa}{3+4\varkappa} N, \quad \varkappa \equiv \frac{w}{\Gamma_m}, \tag{7}$$

where $\varkappa$ is the saturation parameter that characterizes the degree of sublevel-population equalization of the ground and excited states coupled by stimulated transitions. Just as the rates of the stimulated transitions, the saturation parameter is proportional to the radiation intensity.

By virtue of condition (2), the sodium atom undergoes during its excitation lifetime several collisions with strong change of velocity. As shown in Refs. 2 and 3, the diffuse motion of the sodium atoms, both excited and unexcited, can in this case be treated independently, and their diffusion fluxes satisfy the relations

$$\mathbf{j}_p = -D_p \nabla N_p, \quad \mathbf{j}_s = -D_s \nabla N_s, \tag{8}$$

where $\mathbf{j}_p$ and $\mathbf{j}_s$ are the fluxes of the excited and unexcited sodium atoms. Since the radiation does not alter the distribution over the magnetic sublevels of the ground state, D_s is the usual diffusion coefficient of sodium in a buffer gas. According to (3) and (4), D_p is governed only by the collisions of the excited sodium atoms with the buffer particles, so that D_p is the diffusion coefficient of sodium in the $3p$ state.

Substituting (7) and (8) in the continuity equation

$$\partial N/\partial t + \operatorname{div} \mathbf{j} = 0, \quad \mathbf{j} = \mathbf{j}_p + \mathbf{j}_s, \tag{9}$$

we obtain an equation for the sodium-vapor density in the presence of radiation:

$$\frac{\partial N}{\partial t} = \Delta[D(\varkappa)N], \quad D(\varkappa) \equiv D_s + (D_p - D_s)\frac{3\varkappa}{3+4\varkappa}. \tag{10}$$

Under stationary conditions Eq. (10) has a solution

$$\frac{N-N_0}{N_0} = \frac{3\varkappa(D_s - D_p)}{3D_s + \varkappa(3D_p + D_s)}, \quad N \equiv N(\varkappa), \tag{11}$$

where N_0 is the sodium density outside the light beam. The density is spatially inhomogeneous by virtue of the spatial inhomogeneity of the radiation intensity (of the saturation parameters). Under the assumptions made, the density variation "follows" the local value of the radiation intensity.

Let us compare the magnitude of the LDP effect with that of the known saturation effect determined by the change of the population difference $N - (N_s - N_p/3)$ under the influence of the radiation. From (7) and (11) follows

$$(N-N_0)/[N-(N_s-N_p/3)] = {}^3/_4 (D_s - D_p)/D_s. \tag{12}$$

This shows, first, that if the diffusion coefficients D_s and D_p differ noticeably, and that the LDP effect is comparable with the saturation effect. Second, Eq. (12) does not contain at all the radiation characteristics, is independent of the buffer-gas pressure, and yields directly the relative difference of the diffusion coefficients. This circumstance is very important for the experiment and will be actively used below.

3. DESCRIPTION OF EXPERIMENTAL SETUP AND OF MEASUREMENT METHOD

The experimental setup is illustrated in Fig. 2. A mixture of sodium vapor with a buffer gas filled a glass cell 1 (inside diameter 18 mm) placed in a heater at a temperature 110–120 °C (the length of the heated section of the cell was 140 mm). The cell was thoroughly outgassed and preburned in a glow discharge. The sodium was evaporated in vacuum on the inner surface of the entire heated section. The buffer gas entered through a vacuum exhaust unit and a trap cooled with liquid nitrogen. The buffer gas was additionally purified in a heated absorber taken from a GL-56 laser active element and fused to the cell.

The radiation source was a cw dye laser 2 of up to 80 mW power and emission spectral width ≈ 1 GHz. The laser

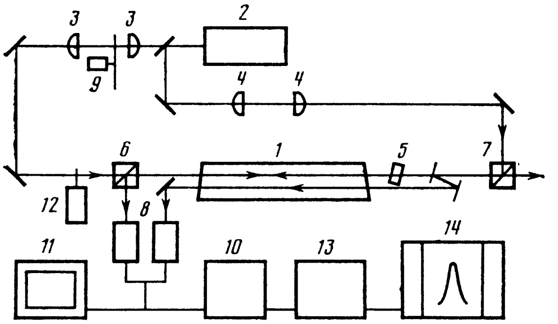

FIG. 2. Diagram of experimental setup (explanations in text).

frequency could be tuned to resonance with both the D_1 and D_2 sodium lines; the intensity distribution in the beam was close to Gaussian.

The density of the irradiated sodium vapor was measured by a test beam taken from the same source and directed perpendicular to the intense beam. Lenses 3 and 4 superimposed the "necks" of the beams on a single section of the heated cell section, where the beams had approximately equal diameters (≈ 0.8 mm). Before entering the cell, the test beam passed through plane-parallel glass plate 5 of 15 mm thickness. Rotation of this plate altered the degree of overlap of the intense and test beams without stopping their axes from remaining parallel. The test-radiation polarization was linear and parallel to that of the strong radiation. The beams were decoupled by Glan prisms 6 and 7. The absorption of the test beam in the sodium vapor was 20–30%, the relative change of its power by the action of the strong radiation did not exceed several percent. It is convenient to measure such small changes by a differential method. To this end, a reference beam with parameters close to those of the test beam passed through the cell at a distance ≈ 6 mm from the intense-beam axis. The test and reference beams were guided to a broadband differential photoreceiver 8. The reference-beam radiation power was adjusted to make the photoreceiver output zero in the absence of the strong radiation.

To record the temporal properties of the response of the absorbing medium to the action of the intense beam, the latter was modulated with a shutter 9, and the photoreceiver, output signal was fed to the input of the analog-signal sampling and storage unit (SSU) 10. The durations of radiation pulses formed by the shutter and of the pauses between them were equal, usually 5–6 ms. The rise times did not exceed 8 μs.

The time-locking pulse was applied to the SSU unit in synchronism with the radiation pulse from the shutter. The SSU sampling interval was 8 μs and could be set in any position relative to the trailing edge of the pulse. To monitor its position, the sampling-interval pulse and the signal from the photoreceiver were simultaneously observed on the screen of the oscilloscope 11.

To eliminate the effect of the differential-photoreceiver unbalance, and intense beam was additionally modulated by a mechanical chopper 12 at a frequency ≈ 7 Hz, while the signal from the SSU output was detected by phase-sensitive detector (PD) 13. The PD output signal was recorded with an x-y plotter.

Thus, the output signal of the PD was proportional to the test-beam-power change $\delta W(\tau,s)$ due to the strong radiation. Here τ is the delay or lead time of the sampling interval relative to the trailing edge of the pulse, and s is the shift of the trial-beam axis relative to that of strong signal. The described procedure yielded reliable records of change of order 10^{-4} in the test-radiation signal power.

The quantity $\delta W(\tau,s)$ can be related to the change of the population difference between the ground and excited states of sodium, due to the strong radiation. If the relative change of the test-beam is small, the given-field approximation[9] can be used, and then

$$\delta W(\tau, s) = \int dx\,dy\,dz\,I(x, y, z)\,\delta\alpha(x, y, z; \tau). \qquad (13)$$

Here $I(x,y,z)$ is the spatial distribution of the test-radiation intensity in the absence of the strong radiation, and $\delta\alpha(x,y,z;\tau)$ is the local change produced in the test-signal absorption coefficient by the strong radiation. If the test signal is linearly polarized and Eq. (6) holds, we have

$$\begin{gathered}\delta\alpha(x, y, z; \tau) \propto N_0 - (N_s - N_p/3) = N_0 - 3N/(3+4\varkappa), \\ N = N(x, y, z; \tau), \quad \varkappa = \varkappa(x, y, z).\end{gathered} \qquad (14)$$

Thus, $\delta W(\tau,s)$ differs from zero because some of the atoms become excited (saturation effect) and the total density of the sodium atoms changes (in particular, because of the LDP effect).

4. MEASUREMENT RESULTS AND THEIR ANALYSIS

We recorded in the experiments $\delta W(\tau,s)$ as functions of s at fixed τ and of τ at $s = 0$ (complete spatial congruence of the test and strong beams). In the former case the signal to the plotter X coordinate came from the device that rotates the plate 5, and was proportional to the displacement (s) of the test-beam axis. In the second case, the X signal came from the SSU time-interval sensor and was proportional to τ.

We recorded also simultaneously two signals, $\delta W(\tau_1,s)$ and $\delta W(\tau_2,s)$, with different positions of the sampling interval relative to the trailing edge of the strong-radiation pulse. Additional SSU and PD were used for this purpose. One signal was fed to the X terminal of the plotter, and the other to the Y terminal. Plots were obtained as functions of the power and frequency of the radiation, as well as of the distance between the beam axis.

Figure 3 shows a typical plot of $\delta W(\tau,0)$ in response to a rectangular strong-radiation pulse. Turning on the strong radiation causes a rapid (almost instantaneous in the scale of Fig. 3) growth of δW, corresponding to an increase of the intensity of the test beam at the exit from the cell. This is followed by a relatively slow decrease of δW to a certain stationary value. When the strong radiation is turned off, δW decreases abruptly and becomes negative, after which it approaches zero slowly.

Figure 3 was obtained for a mixture of sodium vapor with argon, but a similar picture is obtained also when helium or neon is the buffer gas. The only difference is that the

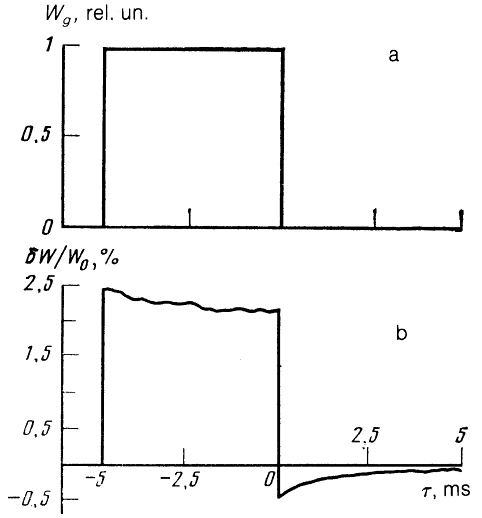

FIG. 3. Strong-radiation pulse (a) and dependence of δW on τ (b) for congruent test and intense beams ($s = 0$), normalized to the test-beam power in the absence of intense radiation.

slow component of δW has a smaller amplitude. Another feature is that the plots of $\delta W(\tau)$ are perfectly identical for radiation absorption on either the $3s^2S_{1/2}$–$3p^2P_{1/2}$ or the $3s^2S_{1/2}$–$3p^2P_{3/2}$ transition.

Obviously, the rapid change of δW is connected with the saturation effect. Indeed, even the rise and fall times of the strong-radiation pulse are substantially longer than the excited-state lifetime, so that the population change due to saturation can follow the variation of the strong-radiation intensity. The characteristic time of the slow δW change is of the order of a^2/D (a is the beam radius and D the diffusion coefficient), i.e., of the order of the diffusion time from the light beam. We therefore attribute the slow change of δW to the change of the sodium-vapor density in the light beam.

It is thus natural to interpret the $\delta W(\tau,0)$ plot in Fig. 3 in the following manner. When the strong radiation is turned on, δW increases almost instantaneously to a certain value governed by the saturation. This is followed by slow increases, at the diffusion rate, of the sodium-vapor density in the light beam, causing a certain decrease of δW. The stationary value of δW during the action of the strong radiation is due both to saturation and to the increase of the sodium density in the beam. After the strong radiation is turned off, all the sodium atoms go just as rapidly to the ground state. The saturation effect vanishes, but the vapor density does not have time to change and remains too high, meaning a negative δW. The subsequent behavior of $\delta W(\tau,0)$ is governed entirely by the diffusive equalization of the sodium-vapor density.

Additional important information is provided by a 3D picture, i.e., plots of δW vs the displacement s of the test-beam axis relative to that of the intense one, plotted at different values of τ. An example of such plots is shown in Fig. 4. Curve 1 was plotted with the sampling interval ahead of the trailing edge ($\tau = -40$ μs) of the strong-radiation pulse. For the remaining curves the sampling interval was past the trailing edge. In the case of curve 2 it can be assumed that the diffusive spreading has not yet come into play.

We call attention to the fact that the shape of curve 2 practically duplicates that of curve 1 which is governed by the saturation. Since the saturation parameter was not large in our case ($\varkappa \lesssim 0.1$), the spatial distribution of the excited atoms is close to the spatial distribution of the strong-radiation intensity. Starting from Eq. (11) and recognizing that the beams have parallel axes and that their diameters change little along the heated zone of the cell, we find that curve 1 can be approximated by a Gaussian, i.e.,

$$\delta W(\tau, s)=\delta W(\tau, 0)\exp[-s^2/(a_g^2+a_\mu^2)], \quad \tau<0, \quad (15)$$

where a_μ and a_g are the Gaussian parameters of the trial and strong beams.

Starting from the assumption that after the strong radiation is turned off δW is governed by the change of the sodium density, and also from the experimental fact that curves 1 and 2 are of like shape, we conclude that the behavior of curves 2–5 in Fig. 4 should be described by the equation

$$\delta W(\tau, s)=\delta W(0, 0)(1+\tau/\tau_d)^{-1}\exp[-s^2/(a_g^2+a_\mu^2+4D_s\tau)], \quad (16)$$

$$\tau>0, \quad \tau_d=(a_\mu^2+a_g^2)/4D_s,$$

which reflects the diffusive spreading of the initial ($\tau = +0$) Gaussian distribution of the excess of the density above equilibrium.

Reduction of curves of the type shown in Fig. 4, plotted at different buffer-gas pressures, has shown that their behavior indeed agrees with (16). Figure 5 shows the τ-dependence of the parameter η (the square of the width at half-maximum of each curve of Fig. 4, normalized to the squared width of curve 1). According to (16), we should have

$$\eta(\tau)=1+\tau/\tau_d, \quad (17)$$

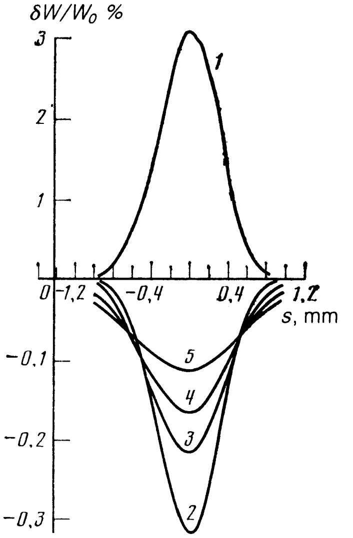

FIG. 4. Dependence of $\delta W/W_0$ on the displacement s of the test beam relative to the position of the maximum overlap of the beams, for different values of τ (in μs): 1—(−40), 2—20, 3—200, 4—400, 5—800. Helium pressure 420 Torr.

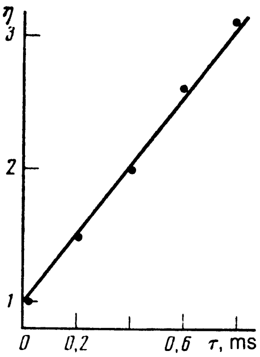

FIG. 5. Relative change, with s, of the width of the $\delta W(\tau,s)$ curves at half-maximum, as a function of the delay time τ.

i.e., a linear dependence on τ, as was indeed observed in experiment. From the slope of the $\eta(\tau)$ line we can get the value of τ_d. This coefficient turned out to be, as it should, proportional to the buffer-gas pressure (the dependence of τ_d on the helium pressure is shown in Fig. 6), i.e., τ_d is indeed inversely proportional to the sodium diffusion coefficient. The diffusion coefficient of sodium in the ground state can therefore be determined from the plots of τ_d vs pressure.

For the diffusion coefficients of sodium at $T = 120$ °C we obtained for D_s the values 0.7 ± 0.15, 0.34 ± 0.06, and 0.24 ± 0.06 cm^2/s · atm in helium, neon, and argon, respectively. They agree well with the values given in Refs. 10 and 11.

The described experimental results are undeniable evidence that the sodium-vapor density is increased inside the strong light beam. From among the known effects, this result can be due only to LDP.

Slow changes of δW are made possible in principle by thermal effects. A change of δT of the temperature in the light beam leads, with allowance for the constancy of the mixture of the sodium vapor with the buffer gas and for the diffusion effect, to a relative change of the bulk density of the sodium atoms

$$\delta N/N \simeq -(1+\alpha_T)\delta T/T.$$

The absolute value of the thermodiffusion constant α_T is less than unity,[12] therefore $\delta N/N \approx -\delta T/T$. Since we are observing pulling of the sodium vapor into the beam, we must have $\delta T < 0$. Quenching of the excited sodium atoms by the buffer gas heats the mixture. Cooling is possible by collisional transitions between the fine-structure components, but only if the radiation frequency is in tune with the D_1 line of sodium. In our experiments the sodium-vapor density increases when the radiation frequency is tuned both to the D_1 and D_2 absorption lines.

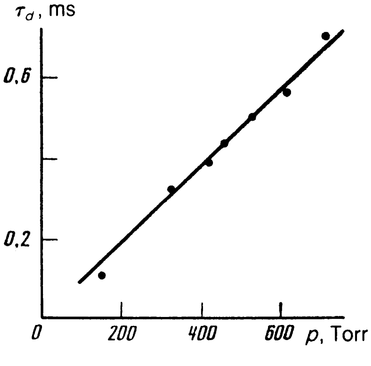

FIG. 6. Dependence of τ_d on the helium pressure.

The rate of heat release is proportional to the density of the excited sodium atoms. In view of the Gaussian profiles of their distribution, it is easy to determine the temperature change in the region of the beam towards the end of the next radiation pulse:

$$\delta T \approx \frac{\beta W_p}{4\pi L\lambda_T}\ln\frac{d}{a_g} + \frac{\beta W_p}{4\pi L\lambda_T}\int_1^{1+\tau_{\text{pul}}/\tau_T} \exp\left(-\frac{\rho^2}{a_g{}^2\xi}\right)\frac{d\xi}{\xi},$$

$$\tau_T = \frac{a_g{}^2}{4\chi}, \tag{18}$$

where β is the coefficient of conversion of the absorbed radiation power W_p into heat, L is the length of the heated section of the cell, λ_T and χ are the thermal conductivity and the thermal diffusivity of the buffer gas, d is the distance from the beam axis to the cell walls, and τ_{pul} is the duration of the radiation pulses. The second term in (18), which describes the temperature change due to the next radiation pulse, is of the same order as the first ($d/a_g \approx 20$, $\tau_{\text{pul}}/\tau_T \approx 10$–$30$), but is more important, since it depends on the coordinates in the beam region. This dependence, however, differs substantially from that for the density of the excited sodium atoms. As a result, the width $\delta W_T(s)$ of the distribution due to the temperature change is approximately double the width observed by us.

Let us estimate the temperature change on the beam axis for the same conditions as for the $\delta W(\tau,s)$ plots in Fig. 4, viz., radiation power 40 mW, absorption 20%, helium buffer gas. Assume that the entire radiation power is converted into heat ($\beta = 1$). Using the given values, we find that during the time of action of the radiation beam the relative change of the temperature reaches a value $2 \cdot 10^{-4}$. This leads to a relative change of the test-beam power $\delta W_T/W_0 \approx 2 \cdot 10^{-5}$, or smaller by two orders than the change observed by us. For neon and argon buffers, similar estimates lead to $\delta W_T/W_0$ likewise smaller by an order than observed by us. Note that when the mixture is cooled by collisional transitions between the fine-structure components the coefficient β is equal to the ratio of the fine-splitting interval to the energy of the emitted photon, i.e., 10^{-3} (Ref. 8). It follows from the foregoing that no thermal effects are observed in our experiments.

One can imagine that the excited sodium atoms enter into a chemical bond (short- or long-lived) with some uncontrollable impurity. This would also lead to a slow change of δW. The sign and the characteristic dependence of δW on τ and s, however, would also be different in this case. Note that we have actually observed an effect of this kind during the first stages of the experiment. It became clear that it was due to contamination by hydrogen, led to an increase of δW, the characteristic times of its establishment and relaxation were substantially longer than τ_d, and the characteristic spa-

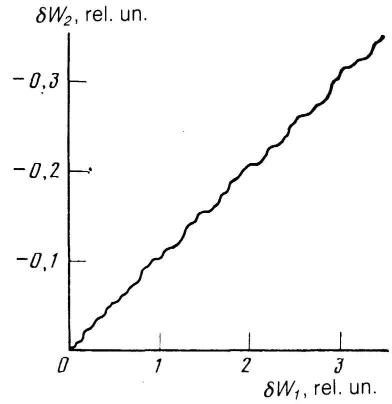

FIG. 7. Plot describing the connection between δW_1 and δW_2 as the intense-radiation power is varied.

tial scale was several light-beam radii. This effect is undoubtedly of interest in itself and is worthy of a special investigation, but in our study it was a disturbance and we got rid of it by thoroughly outgassing of the discharge cell and by purifying the buffer gas.

A diffusion-governed change of δW can be caused also by optical pumping of the hyperfine components of the ground state of the sodium atoms. Notwithstanding the large impact broadening compared with the hyperfine splitting of the ground state, some asymmetry in the level population of the hyperfine structure can still occur when the radiation frequency is detuned from the center of the absorption line. This effect was also observed by us. It was noted, however, against an LDP background and only when the buffer gas was neon, in which the LDP effect is itself quite small. The effect connected with optical pumping led to a wider spatial distribution of δW than the LDP, and had a specific dependence on the frequency detuning. The last circumstance enabled us to ensure its absence by tuning the radiation frequency to the center of the absorption line.

The most convincing evidence that we are indeed dealing with LDP is the result of a check on a specific property of the LDP, based on Eq. (12). We denote by δW_1 the value of δW at the instant just before the strong radiation is turned off, and by δW_2 the value of δW immediately after it is turned off, when the saturation has vanished and the LDP remains in pure form. We use (13) and (14) to calculate the difference

$$\delta W_1-\delta W_2=\int dx\,dy\,dz I(x,y,z) \times[\delta\alpha_1/\delta\alpha_2-1]\delta\alpha_2, \tag{19}$$

$$\delta\alpha_1\infty N_0-3N/(3+4\varkappa),\quad \delta\alpha_2\infty N_0-N,$$

where N is the stationary density value corresponding to the local intensity (to the saturation parameter $\varkappa$). According to (12),

$$\delta\alpha_1/\delta\alpha_2-1={}^4/_3 D_s/(D_s-D_p) \tag{20}$$

and is independent of the coordinates, so that we get from (18) (cf. Ref. 3)

$$\delta W_2/(\delta W_1-\delta W_2)={}^3/_4(D_s-D_p)/D_s. \tag{21}$$

This relation is of interest from two standpoints. First, it indicates that the ratio of the easily measured δW_1 and δW_2 is independent of a number of experimental conditions: of the intensity and frequency of the radiation, of the buffer-gas pressure, of the degree of overlap of the test and strong beams, of their diameters, or even of the precision of the adjustment. Second, a definite combination of δW_1 and δW_2 yields directly the value of an important physical property—the relative change of the diffusion coefficient of optically excited sodium.

An experimental check has shown that the ratio $\delta W_1/\delta W_2$ for all three buffer gases is indeed independent of the radiation intensity (Fig. 7 shows by way of example the linear relation between δW_1 and δW_2 in the case of a helium buffer as the strong-radiation power is varied from 0 to 40 mW), of the buffer-gas pressure in the entire measurement range, of the degree of beam overlap (of s), and of the beam adjustment. For argon and helium as buffers, $\delta W_1/\delta W_2$ is independent of the radiation frequency. (This experiment yields no information for a mixture with neon, in view of the aforementioned influence of optical pumping).

From the aggregate of the measured $\delta W_1/\delta W_2$ we determined using (21) the relative changes of the diffusion coefficients of sodium excited into the states $3p$. For the three buffer gases used in the experiment the result was:

$$\frac{D_s-D_p}{D_s}=\begin{cases}0{,}22\pm0{,}02 & \text{(argon)},\\ 0{,}12\pm0{,}02 & \text{(helium)},\\ 0{,}03\pm0{,}005 & \text{(neon)}.\end{cases}$$

In all three cases the diffusion coefficient was smaller in the $3p$ state than in the ground state.

Note that the value we obtained for the relative change of the diffusion coefficient of sodium in helium agrees with the value $(D_s-D_p)/D_s\approx0.15$, calculated from the data of Ref. 10, and also with the estimate (≈0.1) obtained for this parameter by investigating light-induced drift of sodium vapor in helium.[13]

5. CONCLUSION

The experiments described in the present paper prove convincingly the existence of light-induced diffusive pulling (in and out)—LDP. The LDP effect accounts for an appreciable fraction of the saturation. Under the suitable experimental conditions the LDP is not masked by other effects, and can be reliably recorded and quantitatively investigated. It was shown by experiment that a definite combination of measured quantities is insensitive to a large number of characteristics of the medium and of the radiation, and is free by the same token of many sources of systematic errors. In addition, the very same combination of measured quantities is directly connected with an important physical property, viz., the relative change of the diffusion coefficient of the particle when it is optically excited. We regard the latter circumstance as particularly important, since it suggests that the LDP effect can be the basis of a promising method of measuring the diffusion coefficients of short-lived excited states. That this method is realistic was demonstrated above with a mixture of sodium vapor and noble gases as an example. To our knowledge, we have obtained here the first data on the diffusion coefficients of short-lived atomic states.

It must be noted that the LDP effect permits also a simple and reliable measurement of the diffusion coefficients of unexcited particles (ordinary diffusion coefficients) by producing a controllable spatial inhomogeneity of the density.

Measurement of the diffusion coefficients of excited particles on the basis of the LDP effect, as an independent method, is an apt supplement and control for a method based on the effect of light-induced drift.[14]

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