Field variation of the spectrum of light-induced drift of ⁷Li and ²³Na atoms in heavy inert gases

F. Kh. Gel'mukhanov and A. I. Parkhomenko

Institute of Automation and Electrometry, Siberian Branch, Russian Academy of Sciences, 630090 Novosibirsk, Russia (Submitted 16 March 1995) Zh. Éksp. Teor. Fiz. **108**, 1589–1599 (November 1995)

We present a theoretical study of light-induced drift (LID) of light alkali-metal atoms (⁷Li and ²³Na) in an atmosphere of a heavy inert gas (Xe or Kr). We show that as the radiative intensity grows, the LID spectrum may undergo an appreciable variation: the ratio of the peak velocities for positive and negative radiation frequency offset can vary by a factor of 1.3 to 1.5. The characteristic radiative intensity I_0 at which the LID spectrum begins to change depends on the buffer gas pressure and can be arbitrarily low. The effect is caused by the optical pumping to the hyperfine components of the ground states and the large difference between the rates of collisional relaxation in the orientations and absolute value of the velocity v of the resonant particles, provided that $M \ll M_b$, where M and M_b are the masses of the resonant and buffer particles. We also show that at intensities $I > I_0$, an increase in the mass of buffer particles from values $M_b \lesssim M$ to values $M_b \gg M$ may diminish the LID effect (all other conditions being equal) by a factor of 1.2 to 1.3. © 1995 American Institute of Physics.

1. INTRODUCTION

Light-induced drift,¹ or LID, has been studied fairly well both theoretically and experimentally (see, e.g. Refs. 2-8and the references cited therein). The essence of this effect is the emergence of a directional macroscopic flux of absorbing particles that interacts with a traveling light wave and undergoes collisions with the particles of a buffer gas.

The theory predicts the most vivid manifestation of LID in atomic gases in the absorption of radiation from the ground state. Among the existing sources of radiation, vapors of alkali metals are the most suitable objects of investigation. Most experiments in LID of atomic gases have been carried out with alkali-metal vapors in the atmospheres of various inert gases. It is in these experiments that LID shows up most readily.^{9,10} Undoubtedly, alkali-metal vapors will be widely used in future in studies of LID and its applications hence the stimulus for a theoretical analysis of LID of atoms of alkali metals.

The LID phenomenon is a highly nonequilibrium effect, and only by imposing certain restrictions on the parameters of the system is it possible to describe it by rigorous methods. A rigorous solution can be obtained, for instance, in the limit of heavy buffer particles, $M \ll M_b$ (a Lorentzian gas, with M and M_b the masses of the resonant and buffer particles, respectively).^{11,12} For the light atoms of alkali metals (Li and Na) in an atmosphere of heavy inert gases (Xe and Kr), the requirement that the system be described by a Lorentzian gas, $M \ll M_b$, is met fairly well.

In a Lorentzian gas the rate $v_i M/M_b$ of collisional relaxation of isotropic nonequilibrium structures in the population distributions in the velocities **v** of the resonant particles is low compared to the rate v_i of anisotropic nonequilibrium structures, where v_i is the effective transport rate of collisions of particles in the *i*th level with the buffer gas). This fact together with the optical pumping to the hyperfine components of the ground state of atoms of alkali metals can lead to a situation in which, beginning at a certain characteristic radiative intensity I_0 , there emerge sharp isotropic nonequilibrium structures involving the hyperfine components.¹³ The intensity I_0 depends on the buffer gas pressure, and at low pressures can be arbitrarily low. The presence of such structures drastically changes the absorption spectrum of light alkali-metal atoms¹³ and, as a result, leads to an appreciable change in the magnitude of the LID effect and its spectrum.

In this paper we study by theoretical methods the features of LID in a Lorentzian gas, using the example of threelevel particles—with a Λ configuration of the levels—that model the light alkali-metal atoms.

2. KINETIC EQUATION FOR A LORENTZIAN GAS

We examine the interaction of a traveling electromagnetic wave

$$\mathscr{E} = \frac{1}{2} [\mathbf{E} \exp(i\mathbf{k}\mathbf{r} - \omega t) + \text{c.c.}]$$
(2.1)

with three-level absorbing particles in a mixture with buffer particles. The level diagram for the absorbing particles is depicted in Fig. 1. Here the levels n and l are the components of the hyperfine structure of the ground state, and the level m corresponds to the excited state, with g_i the statistical weight of the *i*th level (i=n,l,m). We ignore collisions between the absorbing particles, assuming the buffer gas concentration N_b to be much higher than the absorbing gas concentration N, or $N_b \ge N$.

This level diagram gives a good picture of the real structure of the ground and first excited states of light atoms of alkali metals (⁷Li and ²³Na). Indeed the separation of the two



FIG. 1. Energy level diagram. The straight arrows indicate stimulated transitions, wavy arrows indicate spontaneous radiative transitions.

hyperfine components of the ground state is comparable to the Doppler linewidth, with the result that the ground state can be modeled by two levels, n and l (for ⁶Li atoms the ground state is well modeled by a single level in view of the smallness of the hyperfine splitting).

The level *m* models the group of levels that are the components of the hyperfine structure of the excited states, $P_{1/2}$ and $P_{3/2}$. Representing a group of levels by a single level is possible here because of the smallness of the hyperfine splitting in these states in comparison to the Doppler absorption linewidth. Emission of radiation involves only one of the fine components, $P_{1/2}$ or $P_{3/2}$, of the excited state.

The interaction of particles with radiation in steady-state and spatially homogeneous conditions is described by the following equations for the density matrix:¹⁴

$$\Gamma_{m}\rho_{m}(\mathbf{v}) = S_{m}(\mathbf{v}) + N[P_{l}(\mathbf{v}) + P_{n}(\mathbf{v})],$$

$$S_{i}(\mathbf{v}) + \Gamma_{mi}\rho_{m}(\mathbf{v}) = NP_{i}(\mathbf{v}),$$

$$\left[\frac{\Gamma_{m}}{2} - i(\Omega_{0i} - \mathbf{kv})\right]\rho_{mi}(\mathbf{v}) = S_{mi}(\mathbf{v}) + iG\left[\rho_{i}(\mathbf{v}) - \frac{g_{i}}{g_{m}}\rho_{m}(\mathbf{v})\right],$$
where

where

$$NP_{i}(\mathbf{v}) = -2 \operatorname{Re}[iG^{*}\rho_{mi}(\mathbf{v})], \quad |G^{2}| = \frac{BI}{2\pi},$$
$$B = \frac{\lambda^{2}\Gamma_{m}w_{m}}{4\hbar\omega}, \quad w_{m} = \frac{g_{m}}{g_{l}+g_{n}}, \quad \Gamma_{m} = \Gamma_{mn} + \Gamma_{ml}, \quad (2.3)$$
$$\Omega_{0i} = \omega - \omega_{mi} \quad (i = n, l).$$

Here $\rho_i(\mathbf{v})$ is the velocity distribution of particles in the *i*th level; $N = N_n + N_l + N_m$ is the concentration (number density) of the absorbing particles $(N_i = \int \rho_i(\mathbf{v}) d\mathbf{v})$; Γ_{mi} is the rate of spontaneous relaxation of the *m*th level via the $m \rightarrow i$ channel; λ is the radiation wavelength; ω_{mi} is the frequency of the m-i transition; $S_m(\mathbf{v})$, $S_i(\mathbf{v})$, and $S_{mi}(\mathbf{v})$ are the collision integrals; and $I = c^2 E^2 / 8\pi$ is the radiative intensity. In the formula for the Einstein *B* coefficient [see Eqs. (2.3)] we have allowed for the fact that the ratio of the rates of radiative transitions from the *m*th level to the hyperfine components *n* and *l* is determined by the ratio of statistical weights: ¹⁵ $\Gamma_{mn} / \Gamma_{ml} = g_n / g_1$. The probability $P_i(\mathbf{v})$ of radiative absorption per unit time (the absorption rate) in the m-i transition by a particle with fixed velocity **v** is determined by

the off-diagonal matrix element of the density matrix (or coherence) $\rho_{mi}(\mathbf{v})$. Note that the last equation in (2.2) for the off-diagonal element $\rho_{mi}(\mathbf{v})$ holds if one ignores the coherence $\rho_{ln}(\mathbf{v})$ between the hyperfine components *n* and *l*. For ⁷Li and ²³Na atoms such an approximation holds if the radiative intensities are not too high, $I \ll 10 \text{ W cm}^{-2}$ (see Ref. 16).

In the absence of a phase memory involving collisioninduced optical transitions (a customary assumption in nonlinear atomic spectroscopy), the off-diagonal collision integral has the form

$$S_{mi}(\mathbf{v}) = -[\nu_{mi}(v) + i\Delta_{mi}(v)]\rho_{mi}(\mathbf{v}) \quad (i = n, l), \quad (2.4)$$

where $\nu_{mi}(v)$ and $\Delta_{mi}(v)$ are the impact broadening and the collisional shift of levels, respectively. Combining this with the formula in (2.2) for the absorption rate $P_i(v)$, we get

$$NP_{i}(\mathbf{v}) = \frac{BI}{\pi} Y_{i}(\mathbf{v}) \bigg[\rho_{i}(\mathbf{v}) - \frac{g_{i}}{g_{m}} \rho_{m}(\mathbf{v}) \bigg], \qquad (2.5)$$

where

$$Y_{i}(\mathbf{v}) = \frac{\Gamma_{j}(v)}{\Gamma_{j}^{2}(v) + [\Omega_{i}(v) - \mathbf{kv}]^{2}},$$

$$\Omega_{i}(v) = \Omega_{0i} - \Delta_{mi}(v),$$

$$\Gamma_{j}(v) = \frac{\Gamma_{m}}{2} + \nu_{mi}(v) \quad (i = n, j = 1; i = l, j = 2).$$
(2.6)

Here $\Gamma_1(v)$ and $\Gamma_2(v)$ are the homogeneous halfwidths of the absorption lines in the m-n and m-l transitions, respectively.

In what follows we ignore the collisional transitions between the hyperfine components of the ground state, which is a valid assumption for alkali-metal atoms in an atmosphere of inert gases.¹⁷ This creates the conditions needed for optical pumping to take place.

For a Lorentzian gas the diagonal collision integrals have the form $^{18}\,$

$$S_{i}(\mathbf{v}) = -\frac{1}{v^{2}} \frac{\partial}{\partial v} [v^{2} s_{i}(\mathbf{v})] + N_{b} v \int d\mathbf{n}' \sigma_{i}(v, \vartheta)$$
$$\times [\rho_{i}(\mathbf{v}') - \rho_{i}(\mathbf{v})], \qquad (2.7)$$

where

$$s_{i}(\mathbf{v}) = -\frac{M}{M_{b}} \nu_{i}(v) v \left[1 + \overline{v}^{2} \frac{\partial}{\partial v^{2}} \right] \rho_{i}(\mathbf{v}), \quad \overline{v}^{2} = \frac{2k_{B}T}{M},$$

$$\nu_{i}(v) = N_{b} v \sigma_{i}(v),$$

$$\sigma_{i}(v) = 2\pi \int_{0}^{\pi} \sigma_{i}(v, \vartheta) (1 - \cos \vartheta) \sin \vartheta d\vartheta, \quad (2.8)$$

$$\cos \vartheta = \mathbf{nn}', \quad \mathbf{n} = \frac{\mathbf{v}}{v}, \quad \mathbf{n}' = \frac{\mathbf{v}'}{v},$$

$$v' = v = |\mathbf{v}| \quad (i = n, l, m).$$

Here v and v' are the velocities of the absorbing particle before and after a collision, $k_{\rm B}$ is the Boltzmann constant, T is the temperature, $\sigma_i(v, \vartheta)$ is the elastic scattering cross section (v'=v) of an absorbing particle in the *i*th state through the angle ϑ , and $\sigma_i(v)$ and $\nu_i(v)$ are the transport cross section and collision rate. For a Lorentzian gas the transport rate $\nu_i(v)$ is responsible for collisions that change only the direction of velocity but not the velocity's magnitude. An appreciable change in the absolute value of the velocity of light absorbing particles occurs only as a result of $M_b/M \ge 1$ collisions, while even one collision is enough to change the direction of velocity. The differential and integral terms on the right-hand side of (27) reflect the changes, respectively, in the absolute value and direction of velocity of the light absorbing particles in collisions.

3. DRIFT VELOCITY

We seek the solution of Eqs. (2.2) in the form

$$\rho_i(\mathbf{v}) = \rho_i(v) + \rho_i(\mathbf{n}) \quad (i = n, l, m), \tag{3.1}$$

where

$$\int \rho_i(\mathbf{n})d\mathbf{n} = 0, \quad \rho_i(v) = \int \rho_i(\mathbf{v}) \frac{d\mathbf{n}}{4\pi}, \quad (3.2)$$

with $\rho_i(v)$ the isotropic part of the distribution function, and $\rho_i(\mathbf{n})$ the anisotropic part. Then the LID velocity is given by the following expression:

$$\mathbf{u} = \frac{1}{N} \int \mathbf{v} \rho(\mathbf{v}) d\mathbf{v} = \frac{\mathbf{k}}{k} \frac{4\pi}{N} \int_0^\infty v^3 \rho^{(1)}(v) dv,$$

$$\rho(\mathbf{v}) = \rho_n(\mathbf{v}) + \rho_l(\mathbf{v}) + \rho_m(\mathbf{v}),$$

$$\rho^{(1)}(v) = \rho_n^{(1)}(v) + \rho_l^{(1)}(v) + \rho_m^{(1)}(v),$$

(3.3)

where

$$\rho_i^{(1)}(v) = \int \frac{\mathbf{n}\mathbf{k}}{k} \rho_i(\mathbf{v}) \frac{d\mathbf{n}}{4\pi} \quad (i=n,l,m).$$
(3.4)

By multiplying the first two equations in (2.2) by $\mathbf{n} \cdot \mathbf{k}/k$ and integrating the products over $d\mathbf{n}/4\pi$ we get

$$\nu_{n}(v)\rho_{n}^{(1)}(v) + \nu_{l}(v)\rho_{l}^{(1)}(v) + \nu_{m}(v)\rho_{m}^{(1)}(v) = 0,$$

$$\Gamma_{mi}\rho_{m}^{(1)}(v) = NP_{i}^{(1)}(v) + \nu_{i}(v)\rho_{i}^{(1)}(v) \quad (i=n,l),$$
(3.5)

where $P_i^{(1)}(v)$ can be expressed in terms of $P_i(\mathbf{v})$ in the same way that $\rho_i^{(1)}(v)$ was expressed in terms of $\rho_i(\mathbf{v})$ in Eq. (3.4). For the hyperfine components we can assume with a high degree of accuracy that the transport collision rates are equal:

$$\nu_l(v) = \nu_n(v). \tag{3.6}$$

If we also solve (3.5) for $\rho_i^{(1)}(v)$, we can write the LID velocity (3.3) in the form

$$\mathbf{u} = \frac{\mathbf{k}}{k} 4\pi \int_0^\infty v^3 \tau(v) [P_n^{(1)}(v) + P_i^{(1)}(v)] dv,$$

$$\tau(v) = \frac{\nu_n(v) - \nu_m(v)}{\nu_n(v) [\Gamma_m + \nu_m(v)]}.$$
(3.7)

To find the function $P_i^{(1)}(v)$ in (3.7), we restrict the radiative intensity by the following conditions:

$$\langle P_i \rangle \ll \Gamma_m, \quad \frac{\nu_n y}{1+y},$$
 (3.8)

where $\langle P_i \rangle = \int P_i(\mathbf{v}) d^3 v$ is the total probability of absorption of radiation in the m-i transition; $y = \Gamma(v)/k\overline{v}$, with $\Gamma(v)$ defined in (3.10); and ν_n is the effective transport rate of elastic collisions between particles in level n and the buffer gas. The rate ν_n is related to the coefficient D_n of diffusion of particles in state n by the formula $D_n = \overline{v}^2/2\nu_n$. Analysis shows that if conditions (3.8) are met, in Eq. (2.5) for $P_i(\mathbf{v})$ we can ignore the population $\rho_m(\mathbf{v})$ and the anisotropic part of the distribution function $\rho_i(\mathbf{v})$. Then

$$NP_{i}(\mathbf{v}) = \frac{BI}{\pi} Y_{i}(\mathbf{v})\rho_{i}(v), \qquad (3.9)$$

where $\rho_i(v)$ is the isotropic part of the distribution $\rho_i(v)$. In alkali-metal atoms the homogeneous halfwidths $\Gamma_1(v)$ and $\Gamma_2(v)$ of absorption lines in the m-n and m-l transitions are essentially the same, with the result that in (3.9) we put

$$\Gamma(v) \equiv \Gamma_1(v) = \Gamma_2(v). \tag{3.10}$$

To obtain the final equations describing the LID effect, we need only find $\rho_i(v)$. The sum of equations for the diagonal elements in (2.2) integrated over $d\mathbf{n}/4\pi$ and combined with the conditions (3.6) and (3.8) implies that

$$\rho_n(v) + \rho_l(v) = NW(v), \qquad (3.11)$$

where W(v) is the Maxwellian velocity distribution. Another relationship for $\rho_i(v)$ can be obtained from the second equation in (2.2) integrated over $d\mathbf{n}/4\pi$:

$$\Gamma_{mi}\rho_{m}(v) = NP_{i}(v) - \frac{M}{M_{b}} \frac{1}{v^{2}} \frac{\partial}{\partial v} \times \left[v^{3} \nu_{i}(v) \left(1 + \bar{v}^{2} \frac{\partial}{\partial v^{2}} \right) \rho_{i}(v) \right], \qquad (3.12)$$

where $P_i(v)$ can be expressed in terms of $P_i(v)$ in exactly the same way that $\rho_i(v)$ can be expressed in terms of $\rho_i(v)$ in (3.2).

Analysis of Eq. (3.12) shows that within the conditions (3.8) there exist two drastically different limits, which we will call cases of low and enhanced radiative intensity.

In the case of low-intensity radiation, i.e.,

$$\langle P_i \rangle \ll \Gamma_m, \quad \frac{\nu_n y}{1+y} \frac{M}{M_b},$$
 (3.13)

the differential term in Eq. (3.12) vanishes, which yields

$$\rho_i(v) = N_i W(v). \tag{3.14}$$

By integrating the second equation in (2.2) with respect to v we obtain $(\int S_i(\mathbf{v})d\mathbf{v}=0$ in elastic collisions)

$$\Gamma_{mi}N_m = N\langle P_i \rangle \quad (i = n, l). \tag{3.15}$$

Combining (3.9), (3.14), and (3.15) and applying the normalization condition $N_n + N_l \approx N$, where we have allowed for the fact that in conditions (3.8) $N_m \ll N$, we obtain

$$\rho_{i}(v) = \frac{NW(v)w_{i}\langle Y_{j}\rangle}{w_{n}\langle Y_{l}\rangle + w_{l}\langle Y_{n}\rangle}, \quad w_{i} = \frac{g_{i}}{g_{n} + g_{l}},$$
(3.16)

E. Kh. Gol'mukhapov and A. J. Barkhamarka

$$\langle Y_i \rangle = \int Y_i(\mathbf{v}) W(v) d\mathbf{v} \quad (i=n, j=l; i=l, j=n).$$

In the case of enhanced radiative intensity, i.e.,

$$\frac{\nu_n y}{1+y} \frac{M}{M_b} \ll \langle P_i \rangle \ll \Gamma_m, \quad \frac{\nu_n y}{1+y}, \tag{3.17}$$

the differential term in (3.12) can be neglected, and from (3.9), (3.11), and (3.12) we obtain

$$\rho_i(v) = \frac{NW(v)w_i\Psi_j(t)}{w_n\Psi_l(t) + w_l\Psi_n(t)} \quad (i=n, \ j=l; \ i=l, \ j=n),$$
(3.18)

where

$$\Psi_{i}(t) = \tan^{-1} \left(\frac{t + x_{i}}{y} \right) + \tan^{-1} \left(\frac{t - x_{i}}{y} \right),$$

$$t = \frac{v}{v}, \quad y = \frac{\Gamma(v)}{k\overline{v}}, \quad x_{i} = \frac{\Omega_{i}(v)}{k\overline{v}} \quad (i = n, l).$$
(3.19)

The case (3.17) can be implemented only in a Lorentzian gas $(M \ll M_b)$, in which two scales of collisional relaxation rates can be singled out: in the orientations (ν_n) and absolute values $(\nu_n M/M_b)$ of the velocity **v** of resonant particles. If conditions (3.17) are met, the anisotropic part $\rho_i(\mathbf{n})$ of the distribution $\rho_i(\mathbf{v})$ is negligible, but the stimulated transition rate is still high enough for sharp nonequilibrium structures to develop in the isotropic part $\rho_i(\mathbf{v})$ of the distribution.¹³

Substituting (3.9) into (3.7) and allowing for (3.16) and (3.18), we arrive at the following expression for the drift velocity:

$$\mathbf{u} = \frac{\mathbf{k}}{k} \,\overline{v} \, \frac{2 \kappa \Gamma_m}{\sqrt{\pi}} \int_0^\infty \exp(-t^2) t \,\tau(v) \\ \times \left[\frac{w_n f_n(t) A_l + w_l f_l(t) A_n}{w_n A_l + w_l A_n} \right] dt, \qquad (3.20)$$

where

$$\kappa = \frac{BI}{\pi \Gamma_m k \overline{v}}, \quad f_i(t) = x_i \Psi_i(t) + \frac{y}{2} \ln \left(\frac{y^2 + (t - x_i)^2}{y^2 + (t + x_i)^2} \right).$$
(3.21)

In (3.20),

$$A_i = \langle Y_i \rangle \tag{3.22}$$

in the case of low-intensity radiation [conditions (3.13)] and

$$A_i = \Psi_i(t) \tag{3.23}$$

in the case of enhanced radiative intensity (3.17).

The effect of field variation of the LID spectrum is caused by the change that the function A_i undergoes as the radiative intensity grows. When the hyperfine components nand l formally merge (the transition frequency between them tends to zero, or $\omega_{ln} \rightarrow 0$), Eq. (3.20) for **u** transforms into the formula for a two-level system with allowance for level degeneracy $(f_n(t)=f_l(t)\equiv f(t))$:

$$\mathbf{u} = \frac{\mathbf{k}}{k} \,\overline{v} \, \frac{2\kappa\Gamma_m}{\sqrt{\pi}} \int_0^\infty \exp(-t^2) t \,\tau(v) f(t) dt. \tag{3.24}$$

This formula does not depend on A_i , which means that the effect does not emerge in a two-level system and is a specific feature of a three-level Λ -system.

Equation (3.20) can be simplified in the case of lowintensity radiation [conditions (3.13)], with $A_i = \langle Y_i \rangle$, if the collision characteristics are assumed to be velocityindependent, or

$$\nu_i(v), \quad \Gamma(v), \quad \Delta_{mi}(v) = \text{const.}$$
 (3.25)

Then the drift velocity is given by the expression $(\nu_i(v) \equiv \nu_i = \text{const})$

$$\mathbf{u} = \frac{\mathbf{k}}{k} \overline{v} \frac{\nu_n - \nu_m}{\nu_n} \frac{\langle P \rangle}{\Gamma_m + \nu_m} \bigg[w_n \frac{\phi(x_n)}{g(x_n)} + w_l \frac{\phi(x_l)}{g(x_l)} \bigg],$$
(3.26)

where

$$\langle P \rangle = \sqrt{\pi} \kappa \Gamma_m \frac{g(x_l)g(x_n)}{w_n g(x_l) + w_l g(x_n)}, \quad g(x_i) = \operatorname{Re}[w(z_i)],$$

$$\phi(x_i) = \operatorname{Re}[z_i w(z_i)], \quad z_i = x_i + iy \quad (i = n, l).$$

$$(3.27)$$

Here $\langle P \rangle \equiv \langle P_n \rangle + \langle P_l \rangle$ is the total cumulative probability of radiative absorption, and w(z) is the error function of a complex-valued argument, which is tabulated in Ref. 19. Equation (3.26) coincides with the one obtained for the LID velocity of three-level particles with the Λ -configuration of levels in the strong-collision model.²⁰

4. DISCUSSION

To identify the effect of field variation of the LID spectrum in "pure" form, we assume from now on that the collision characteristics are independent of velocity [see Eq. (3.25)]. Maximum manifestation of the effect should be expected in the Doppler broadening limit ($y \ll 1$). If the conditions (3.25) are met and

$$y \ll 1, \quad \sqrt{\pi} x_i^2 \exp(-x_i^2),$$
 (4.1)

Eq. (20) assumes the form

$$\mathbf{u} = \frac{\mathbf{k}}{k} \,\overline{v} \, \frac{\nu_n - \nu_m}{\nu_n} \, \frac{\langle P \rangle}{\Gamma_m + \nu_m} \bigg[x_0 + \frac{\delta}{2} (w_l - w_n) \bigg], \qquad (4.2)$$

where

$$\delta = \frac{\omega_{ln}}{k\bar{v}}, \quad x_0 = \frac{\omega - \omega_0}{k\bar{v}}, \quad \omega_0 = \frac{\omega_{mn} + \omega_{ml}}{2}.$$
 (4.3)

Here δ is the dimensionless separation of the components of the hyperfine structure, and x_0 is the dimensionless offset of the radiation frequency from the arithmetic-mean frequency ω_0 of the *n*-*n* and *m*-*l* transitions. The absorption probability $\langle P \rangle$ is given by the expression

$$\langle P \rangle = \sqrt{\pi} \kappa \Gamma_m \phi, \quad \phi = \frac{\exp[-\delta^2/4 - x_0^2]}{w_l \exp(\delta x_0) + w_n \exp(-\delta x_0)}$$
(4.4)

in the case of low-intensity radiation (3.13) and by the expression

$$\langle P \rangle = \sqrt{\pi} \kappa \Gamma_m \phi_L, \quad \phi_L = \exp\left[-\left(\frac{\delta}{2} + |x_0|\right)^2\right]$$
(4.5)



FIG. 2. Drift velocity of ⁷Li atoms as a function of the radiation frequency offset at T=300 K ($\omega_{ln}=5.049\times10^9$ s⁻¹, according to Ref. 21, and $\delta=0.636$). Here $\nu_m > \nu_n$. The solid vertical lines F=1 and F=2 designate resonant frequencies of the m-n and m-l transitions. The curves 1 and 2 correspond, respectively, to the strong-collision model and a Lorentzian gas at enhanced radiative intensity [conditions (3.17)]. Curve 1 also describes the LID spectrum in a Lorentzian gas with low-intensity radiation [conditions (3.13)].

in the case of enhanced radiative intensity (3.17).

As Eq. (4.2) shows, the effect of variation of the LID spectrum with the increase in the radiative intensity is entirely due to the variation in the spectrum of the cumulative probability of radiative absorption $\langle P \rangle$. A detailed analysis of the variation of the spectrum of $\langle P \rangle$ was carried out in Ref. 13.

Note that in the strong-collision model with Doppler broadening, the LID velocity is described by Eq. (4.2) with the absorption probability $\langle P \rangle$ specified by Eq. (4.4) for both low-intensity radiation [conditions (3.13)] and enhanced radiative intensity [conditions (3.17)] (see Ref. 20). Since $\phi > \phi_L$ (only at point $x_0=0$ is ϕ equal to ϕ_L), in the event of enhanced radiative intensity [conditions (3.17)] the calculation of LID done in the strong-collision model yields an overestimated value for the drift velocity. This fact is illustrated by Figs. 2 and 3, which depict the spectral dependence of the drift velocity of ⁷Li and ²³Na atoms in an atmosphere of heavy inert buffer gases.

The most suitable inert buffer gas from the viewpoint of registering the effect of LID spectrum variations caused by an increasing radiative intensity is xenon. In the ⁷Li-Xe system the mass ratio M_b/M is 18.9 and in the ²³Na-Xe system this ratio is 5.7, so that a Lorentzian gas $(M_b/M \ge 1)$ describes both systems fairly well. As Figs. 2 and 3 show, in the strong-collision model the height of the left peak (near F=2) in the LID velocity is overestimated by a factor of 1.55 for ⁷Li atoms and by a factor of 1.95 for ²³Na atoms, while the height of the right maximum (which is greater in amplitude) is overestimated by a factor of 1.22 and 1.30, respectively. In other words, for enhanced radiative intensity [conditions (3.17)] an increase in the mass of buffer particles from $M_b \le M$ to $M_b \ge M$ can decrease the LID effect (all other things being equal) by a factor of 1.2 to 1.3 (in the



FIG. 3. Drift velocity of ²³Na atoms as a function of the radiation frequency offset at T = 1000 K ($\omega_{ln} = 1.113 \times 10^9$ s⁻¹, according to Ref. 21, and $\delta = 1.227$). Here $\nu_m > \nu_n$. Curves *l* and 2 correspond to the same cases as in Fig. 2.

strong-collision model the rates of collisional relaxation in the orientations and magnitude of the velocity v of the resonant particles are the same, which is equivalent to the condition that $M_b \leq M$.

Measuring the ratio of the heights of the right and left peaks in the drift velocity is a convenient way of registering variations in the LID spectrum in experiments. For instance, for ²³Na atoms this ratio is 1.30 for low-intensity radiation [conditions (3.13); curve *1* in Fig. 3] and 1.95 for enhanced radiative intensity [conditions (3.17); curve 2 in Fig. 3].

The extent to which the LID spectrum varies as the radiative intensity grows depends on the separation of the hyperfine components and manifests itself most clearly at $\delta \sim 1$. Since the parameter δ is temperature-dependent, so is the size of the effect. In view of this, the variation of the LID spectrum for ⁷Li atoms manifests itself more strongly at low temperatures ($T \approx 300$ K), while for ²³Na atoms it manifests itself more strongly at elevated temperatures ($T \sim 1000$ K).

Let us estimate the radiative intensity I_0 at which the LID spectrum begins to change. In the case of Doppler broadening ($y \ll 1$), the boundary between the cases of low-intensity radiation [conditions (3.13)] and enhanced radiative intensity [conditions (3.17)] can be specified by the condition $\langle P_i \rangle \sim y \nu_n M/M_b$. This yields

$$I_0 \sim \frac{\pi \Gamma \nu_n}{B} \frac{M}{M_b}.$$
(4.6)

For ⁷Li and ²³Na, the Einstein *B* coefficient is, respectively, $1.39 \times 10^{17} w_m$ [cm⁻² W⁻¹ s⁻²] and $1.57 \times 10^{17} w_m$ [cm² W⁻¹ s⁻²]. When the buffer gas pressure is roughly 1 Torr, we have $\nu_n \sim 10^7$ s⁻¹ and $\Gamma \sim 5 \times 10^7$ s⁻¹. Assuming that $w_m \sim 1$, from (4.6) we arrive at the following estimate of the radiative intensity: $I_0 \sim 0.5 \times 10^{-3}$ W cm⁻² for the ²³Na-Xe mixture and $I_0 \sim 2 \times 10^{-3}$ W cm⁻² for the characteristic intensity I_0 is much lower than 10^{-3} W cm⁻².

In the limit $|z_i| \ge 1$, which is implemented in the case of homogeneous broadening $y \ge 1$ or at large values of the radiation frequency offset, $|x_i| \ge 1$, the drift velocity is specified by a single expression,

$$\mathbf{u} = \frac{\mathbf{k}}{k} \overline{\nu} \frac{\nu_n - \nu_m}{\nu_n} \frac{\langle P \rangle}{\Gamma_m + \nu_m} \left[\frac{w_n x_n}{y^2 + x_n^2} + \frac{w_l x_l}{y^2 + x_l^2} \right],$$

$$\langle P \rangle = \frac{\kappa y \Gamma_m}{y^2 + \delta^2 / 4 + x_0^2 + (w_l - w_n) \delta x_0},$$
(4.7)

in the cases of low-intensity radiation [conditions (3.13)] and enhanced radiative intensity [conditions (3.17)]. Thus, in the event of homogeneous broadening, $y \ge 1$, the LID spectrum does not change as the radiative intensity grows from values specified by (3.13) to values specified by (3.17).

5. CONCLUSION

We have described the effect of field variation of the LID spectrum of light alkali-metal atoms (⁷Li and ²³Na) in an atmosphere of heavy buffer particles of an inert gas (Xe and Kr). For the effect to manifest itself, it is important that there be optical pumping to the hyperfine components of the ground state and a large difference between the masses of the resonant and buffer particles, $M/M_b \ll 1$ (a Lorentzian gas). These factors make it possible to identify two scales of the rate of collisional relaxation in the orientations (ν_n) and absolute value ($\nu_n M/M_b$) of the velocity **v** of the resonant particles. The effect is present at all radiative intensities, and depends on the buffer gas pressure.

As is known, one of the main scientific applications of LID is the measurement of the relative variation $(\nu_m - \nu_n)/\nu_n$ of the classical transport collision rates when a particle is excited from state *n* to state *m* (see Refs. 1, 5, and 6). We have found that calculations of the LID velocity of ⁷Li and ²³Na atoms in the strong-collision model, which is usually employed for processing the experimental data, can overestimate the peak velocity value by a factor of 1.2 to 1.3. In other words, if the experimental data are processed by the strong-collision model instead of the Lorentzian gas model considered here, the measured value of $(\nu_m - \nu_n)/\nu_n$ may be

smaller than the true value by a factor of 1.2 to 1.3.

We thank A. M. Shalagin for fruitful discussion of the results and valuable advice. This research was partially sponsored by the International Science Foundation (Grant No. RCM000) and the Russian Fund for Fundamental Research (Grant No. 93-02-03567) and was also supported by the Netherlands Organization for Scientific Research and the Universities of Russia Program.

- ¹F. Kh. Gel'mukhanov and A. M. Shalagin, Pis'ma Zh. Eksp. Teor. Fiz. **29**, 773 (1979) [JETP Lett. **29**, 711 (1979)].
- ²F. Kh. Gel'mukhanov and A. M. Shalagin, Zh. Eksp. Teor. Fiz. 78, 1674 (1980) [Sov. Phys. JETP 51, 839 (1980)].
- ³F. Kh. Gel'mukhanov, Avtometriya No. 1, 49 (1985).
- ⁴G. Nienhuis, Phys. Rep. **138**, 151 (1986).
- ⁵H. G. C. Werij and J. P. Woerdman, Phys. Rep. 169, 145 (1988).
- ⁶S. G. Rautian and A. M. Shalagin, *Kinetic Problems in Nonlinear Spectroscopy*, North-Holland, Amsterdam (1991).
- ⁷E. R. Eliel, Ad. At. Mol. Opt. Phys. 30, 199 (1992).
- ⁸K. A. Nasyrov and A. M. Shalagin, Astron. Astrophys. 268, 201 (1993).
- ⁹H. G. C. Werij, J. P. Woerdman, J. J. M. Beenakker, and I. Kuščer, Phys. Rev. Lett. **52**, 2237 (1984).
- ¹⁰S. N. Atutov, St. Lesyak, S. P. Pod'yachev, and A. M. Shalagin, Opt. Commun. **60**, 41 (1986).
- ¹¹B. Ya. Dubetskiĭ, Zh. Éksp. Teor. Fiz. **88**, 1586 (1985) [Sov. Phys. JETP **61**, 945 (1985)].
- ¹²F. Kh. Gel'mukhanov, L. V. Il'ichov, and A. M. Shalagin, Physica A 137, 502 (1986).
- ¹³F. Kh. Gel'mukhanov and A. O. Parkhomenko, Zh. Éksp. Teor. Fiz. **107**, 1853 (1995) [JETP **80**, 1029 (1995)].
- ¹⁴S. G. Rautian, G. I. Smirnov, and A. M. Shalagin, Nonlinear Resonances in the Spectra of Atoms and Molecules [in Russian], Nauka, Novosibirsk (1979).
- ¹⁵I. I. Sobelman, Atomic Spectra and Radiative Transitions, Springer-Verlag, Berlin (1979).
- ¹⁶A. D. Streater and J. P. Woerdman, J. Phys. B 22, 677 (1989).
- ¹⁷W. Happer, Rev. Mod. Phys. 44, 169 (1972).
- ¹⁸ E. M. Lifshitz and L. P. Pitaevskiĭ, *Physical Kinetics*, Pergamon Press, Oxford (1983).
- ¹⁹M. Abramowitz and I. A. Stegun (eds.), Handbook of Mathematical Functions, Dover, New York (1965).
- ²⁰ S. N. Atutov, A. I. Parkhomenko, S. P. Pod'yachev, and A. M. Shalagin, J. Phys. B **25**, 2943 (1992).
- ²¹A. A. Radtsig and B. M. Smirnov, *Reference Data on Atoms, Molecules, and Ions*, Springer-Verlag, Berlin (1985).

Translated by Eugene Yankovsky