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Spectral anomalies of the light-induced drift effect caused by the velocity dependence of the collision broadening and shift of the absorption line

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Abstract. **We have theoretically investigated the spectral features of the light-induced drift (LID) effect, arising due to the dependence of the collision broadening γ and shift Δ of the absorption line on the velocity of resonance particles, v. It is shown that under certain conditions, account of this dependence can radically change the spectral shape of the LID signal, up to the appearance of additional zeros in the dependence of the drift velocity on the radiation frequency.**

Keywords: *kinetic equation, light-induced drift, collisions, collision broadening, collision shift.*

1. Introduction

Light-induced drift (LID) [1] refers to the number of the most powerful effects of the radiation on the translational motion of particles and has been well studied both theoretically and experimentally (see, for example, [2–9] and references therein). The essence of the effect is the appearance of a macroscopic directional flow of particles that absorb the radiation and are in a mixture with buffer particles.

The magnitude of the LID effect is proportional to the relative difference between the transport collision frequencies of the resonance particles in the ground and excited states with buffer particles. Until recently all the experimental results of the LID study were well described by the theory of light-induced drift with the velocity-independent transport collision frequencies [2–7]. This theory gave a characteristic dispersion-like (tilde-like) frequency dependence of the drift velocity $u_L(\Omega)$ with one zero at zero detuning Ω of the radiation frequency (the so-called normal LID, see below the dashed curves in Figs 2c and 3b–d). In 1992, in studying the LID of C_2H_4 molecules in a Kr buffer gas, Van der Meer et al. [10] found an unexpectedly sharp deviation of the frequency dependence of the drift velocity $u_L(\Omega)$ on the dispersion-like curve: the authors observed an anomalous spectral LID velocity profile with three zeros instead of one. The difference from the predictions of the theory was so strong that the effect was called 'anomalous LID'. By now anomalous LID has been largely studied both experimentally [8, 10–17] (anomalous LID was observed for C_2H_4 molecules in buffer gases Ar, Kr, Xe, SF_6 [10, 12, 13, 15, 17], for HF molecules in buffer gases Ar, Kr, Xe [14, 16], for CH_3F molecules in a buffer gas Kr [11], for potassium atoms in the buffer mixture of neon with other inert gases [8]) and theoretically [9, 13, 15, 17–23]. It was found that this effect is entirely due to the dependence of the transport collision frequencies on the velocity v of resonance particles, the abnormality occurring only when the difference of transport collision frequencies $\Delta\nu(v)$ on the combining (affected by radiation) levels changes sign as a function of v.

In the theory of light-induced drift it is usually assumed that the collision broadening γ and shift Δ of the levels do not depend on the velocity v of resonance particles:

$$\gamma(v) = \gamma_0 = \text{const}, \ \Delta(v) = \Delta_0 = \text{const}. \tag{1}$$

In the case of normal LID [when the difference of transport collision frequencies $\Delta\nu(v)$ does not change its sign], the influence of dependences $\gamma(v)$ and $\Delta(v)$ on the LID line shape is insignificant and can be ignored. This effect is also negligible in the case of anomalous LID if the Doppler width of the absorption line is much greater than the collision broadening of the line (at $\gamma_0 \ll kv_T$, where kv_T is the Doppler width). That is why assumption (1) is used in the theory of anomalous LID. In the case of a significant excess of the collision broadening of the absorption line by the Doppler line width (at $\gamma_0 \gg kv_T$), assumption (1) can lead to severe distortion of the calculated line shape of anomalous LID. This fact was found out in theoretical paper [20] in the case of the Lorentz gas (the limiting case of heavy buffer particles).

Apart from work [20], no other studies of the effect of the dependences $\gamma(v)$ and $\Delta(v)$ on the LID line shape have still been conducted. In this paper, this effect is studied for the general case of an arbitrary ratio of the masses of the absorbing and buffer particles and an arbitrary ratio of the homogeneous and Doppler widths of the absorption line.

2. Drift velocity

Consider the interaction of a travelling monochromatic electromagnetic wave $\mathcal{E} = [\boldsymbol{E}\exp(\mathrm{i}\boldsymbol{kr} - \mathrm{i}\omega t) + \text{c.c.}]/2$ with two-level absorbing particles that are mixed with the buffer particles. We will neglect the collisions between the absorbing particles, assuming the buffer gas concentration N_b to be much higher than the concentration of the absorbing gas N. The interaction of the particles with the radiation is described by the equations for the density matrix [24]:

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$$\left(\frac{\partial}{\partial t} + \boldsymbol{v}\nabla + \Gamma_m\right)\rho_m(\boldsymbol{v}) = S_m(\boldsymbol{v}) + NP(\boldsymbol{v}),$$

$$\left(\frac{\partial}{\partial t} + \boldsymbol{v}\nabla\right)\rho_n(\boldsymbol{v}) = S_n(\boldsymbol{v}) + \Gamma_m\rho_m(\boldsymbol{v}) - NP(\boldsymbol{v}), \tag{2}$$

$$\left(\frac{\partial}{\partial t} + \boldsymbol{v}\nabla + \frac{\Gamma_m}{2} - \mathrm{i}(\Omega_0 - \boldsymbol{kv})\right)\rho_{mn}(\boldsymbol{v})$$

$$= S_{mn}(\boldsymbol{v}) + \mathrm{i}G[\rho_n(\boldsymbol{v}) - \rho_m(\boldsymbol{v})],$$

where

$$NP(\boldsymbol{v}) = -2\mathrm{Re}[\mathrm{i}G^*\rho_{mn}(\boldsymbol{v})]; \quad G = \frac{Ed_{mn}}{2\hbar}; \quad \Omega_0 = \omega - \omega_{mn}; \tag{3}$$

$\rho_n(\boldsymbol{v})$ and $\rho_m(\boldsymbol{v})$ are the velocity distributions of the particles in the ground (n) and excited (m) levels; $N = N_m + N_n$ is the concentration of absorbing particles ($N_i = \int\rho_i(\boldsymbol{v})\mathrm{d}\boldsymbol{v}$, $i = n,m$); $S_i(\boldsymbol{v})$ and $S_{mn}(\boldsymbol{v})$ are the collision integrals; d_{mn} is the matrix element of the dipole moment of the transition $m-n$; ω and $\boldsymbol{k}$ are the radiation frequency and wave vector; ω_{mn} is the frequency of the transition $m-n$; Γ_m is the velocity of the spontaneous relaxation of the excited level m; and $P(\boldsymbol{v})$ is the probability of radiation absorption by a particle per unit time at a fixed velocity $\boldsymbol{v}$.

In the absence of the phase memory at optical transitions in collisions (natural assumption for atomic spectroscopy) the nondiagonal collision integral has the form

$$S_{mn}(\boldsymbol{v}) = -[\gamma(v) + \mathrm{i}\Delta(v)]\,\rho_{mn}(\boldsymbol{v}). \tag{4}$$

In steady-state and spatially homogeneous conditions, for the absorption probability $P(\boldsymbol{v})$ (3) from the last equation in (2) we find with the help of (4) the expression

$$NP(\boldsymbol{v}) = 2|G|^2Y(\boldsymbol{v})[\rho_n(\boldsymbol{v})-\rho_m(\boldsymbol{v})], \tag{5}$$

where

$$Y(\boldsymbol{v}) = \frac{\Gamma(v)}{\Gamma^2(v) + [\Omega(v) - \boldsymbol{kv}]^2}; \quad \Gamma(v) = \frac{\Gamma_m}{2} + \gamma(v); \tag{6}$$

$$\Omega(v) = \Omega_0 - \Delta(v).$$

For the diagonal collision integrals we will use the model of particle ‘arrival’ that is isotropic in velocity [9, 25]:

$$S_i(\boldsymbol{v}) = -\nu_i(v)\rho_i(\boldsymbol{v}) + S_i^{(2)}(v), \quad i = n,m, \tag{7}$$

where $S_i^{(2)}(v)$ is the function of the velocity modulus $v = |\boldsymbol{v}|$; $\nu_i(v)$ is the transport collision frequency [25]. The collisional model (7) takes into account the dependence of the collision frequency on the velocity and at the same time allows one to obtain an analytical solution of the problem at any ratios of the masses of active and buffer particles.

We find the drift velocity of the resonance particles, which by definition is

$$\boldsymbol{u}_\mathrm{L} \equiv \frac{\boldsymbol{j}_m + \boldsymbol{j}_n}{N}, \quad \boldsymbol{j}_i = \int\boldsymbol{v}\rho_i(\boldsymbol{v})\mathrm{d}\boldsymbol{v}, \tag{8}$$

where $\boldsymbol{j}_i$ is the flux of particles in state i. In steady-state and spatially homogeneous conditions, from equations (2) for the drift velocity $\boldsymbol{u}_\mathrm{L}$ we find the expression

$$\boldsymbol{u}_\mathrm{L} = \int\frac{\nu_n(v) - \nu_m(v)}{\nu_n(v)[\Gamma_m + \nu_m(v)]}\boldsymbol{v}P(\boldsymbol{v})\mathrm{d}\boldsymbol{v}. \tag{9}$$

To calculate the drift velocity (9) we restrict our consideration by the condition of weak radiation intensity, assuming that in (5) the population of the excited level $[\rho_m(\boldsymbol{v}) = 0]$ can be neglected, and the population velocity distribution in the ground state can be considered close to the Maxwellian $[\rho_n(\boldsymbol{v}) = NW(\boldsymbol{v})$, where $W(\boldsymbol{v})$ is the Maxwell distribution]. In this case,

$$P(\boldsymbol{v}) = 2|G|^2Y(\boldsymbol{v})W(\boldsymbol{v}). \tag{10}$$

Integrating (9) over the velocity directions, $\boldsymbol{v}$, we obtain the final expression for the drift velocity $\boldsymbol{u}_\mathrm{L}$, which is represented in the form

$$\boldsymbol{u}_\mathrm{L} = \boldsymbol{u}_0 u(x_0), \tag{11}$$

where we have introduced the vector $\boldsymbol{u}_0$ with the dimensions of velocity,

$$\boldsymbol{u}_0 = \frac{\boldsymbol{k}}{k}\frac{4|G|^2}{\sqrt{\pi}\,k\nu_n^\mathrm{tr}}, \tag{12}$$

and the dimensionless velocity u as a function of dimensionless detuning $x_0 = (\Omega_0 - \Delta_0)/(kv_T)$ of the radiation frequency

$$u(x_0) = \int_0^\infty t\tau(t)f(t)\exp(-t^2)\,\mathrm{d}t. \tag{13}$$

Here we have introduced the function of the dimensionless velocity $t = v/v_T$:

$$f(t) = x(t)\psi(t) + \frac{y(t)}{2}\ln\left\{\frac{y^2(t) + [t - x(t)]^2}{y^2(t) + [t + x(t)]^2}\right\},$$

$$\psi(t) = \arctan\frac{t + x(t)}{y(t)} + \arctan\frac{t - x(t)}{y(t)}, \tag{14}$$

$$\tau(t) = \frac{\nu_n(t) - \nu_m(t)}{\nu_n(t)}\frac{\nu_n^\mathrm{tr}}{\Gamma_m + \nu_m(t)},$$

$$y(t) = \frac{\Gamma(t)}{kv_T}, \quad x(t) = \frac{\Omega(t)}{kv_T}, \quad v_T = \sqrt{\frac{2k_\mathrm{B}T}{M}};$$

ν_n^tr is the average transport collision frequency of absorbing particles in state n with buffer particles; Δ_0 is the average value of the collision shift $\Delta(v)$; M is the mass of the absorbing particles; T is the temperature; and k_B is the Boltzmann constant. Thus, the calculation of the drift velocity in the collisional model (7) of the velocity isotropic ‘arrival’ of the particles with account for the dependence of the collision broadening and shift on the velocity v of resonance particles is reduced to calculation of a single integral (13).

3. Functions $\gamma(t)$ and $\Delta(t)$

To calculate the drift velocity, it is necessary to know the dependences $\gamma(t)$ and $\Delta(t)$. For the power potential of interaction between the particles

$$U(r) \propto r^{-p}, \tag{15}$$

the dependences of the collision broadening γ and shift Δ of the levels on the velocity t can be calculated explicitly [26]:

$$\frac{\gamma(t)}{\gamma_0} = \frac{\Delta(t)}{\Delta_0} = (1+\beta)^a {}_1F_1\left(a; \frac{3}{2}; -\beta t^2\right), \tag{16}$$

$$a \equiv \frac{1}{p-1} - \frac{1}{2}, \quad p \geqslant 2,$$

where $\beta = M_b/M$ is the mass ratio of the particles of buffer (M_b) and absorbing (M) gases; ${}_1F_1(a;3/2;-\beta t^2)$ is the Kummer confluent hypergeometric function; γ_0 and Δ_0 are the average values (in terms of the Maxwell distribution) of $\gamma(t)$ and $\Delta(t)$ [$\gamma_0 = \int\gamma(v)W(\mathbf{v})d\mathbf{v}$ and $\Delta_0 = \int\Delta(v)W(\mathbf{v})d\mathbf{v}$]. The sign of the shift of the absorption line centre can be either negative ($\Delta_0 < 0$, the red shift), or positive ($\Delta_0 > 0$, the blue shift). It follows from (16) that the values of $\gamma(t)$ and $|\Delta(t)|$ increase with increasing t at $p > 3$ and decrease with increasing t at $p < 3$. At $p = 3$, relation (1) is fulfilled: the values of γ and Δ do not depend on the velocity. The dependence of γ and Δ on t can also be neglected in the case of light buffer particles ($\beta \ll 1$). With increasing β the dependence of γ and Δ on t increases and reaches a maximum in the case of heavy buffer particles ($\beta \gg 1$). Collision broadening and shift are related by the expression [26]

$$\frac{|\Delta(t)|}{\gamma(t)} = \frac{|\Delta_0|}{\gamma_0} = \tan\left(\frac{\pi}{p-1}\right). \tag{17}$$

This formula is valid at $p > 3$.

4. Analysis of results

As was noted above, anomalous manifestation of the LID is entirely due to the sign-alternating dependence of the factor τ on the velocity t. Let the factor $\tau(t)$ change sufficiently smoothly and vanish at $t = t_0$. Then the function $\tau(t)$ can be written in the form

$$\tau(t) = B(t - t_0), \quad B \equiv \left.\frac{d\tau(t)}{dt}\right|_{t=t_0}. \tag{18}$$

The coefficient B does not affect the shape of the LID line. Next, in the calculations we assume for definiteness that $B = 0.1$.

The strong influence of the dependences $\gamma(v)$ and $\Delta(v)$ on the LID line shape is illustrated in Figs 1–4. The calculations of the drift velocity $u(x_0)$ by (13) were performed for the broadening and shift of the absorption line, corresponding to the power potential of interaction between particles with $p = 6$ (van der Waals interaction of neutral atoms at large distances). The sign of the shift of the absorption line centre was assumed positive ($\Delta_0 > 0$).

The sensitivity of the LID line shape to the mass ratio β of buffer and resonance particles is shown in Fig. 1. The influence of the velocity dependence of the broadening and shift on the LID line shape is minimal for light ($\beta \ll 1$) buffer particles [in this case, $\gamma(v)$ and $\Delta(v)$ weakly depend on v] and maximal for heavy ($\beta \gg 1$) buffer particles. The numerical analysis shows that for the problem under study the limit $\beta \gg 1$ is reached, starting with $\beta \approx 3$. In other words, the value $\beta = 3$ is actually equivalent to the condition $\beta \gg 1$. The influence of the dependences $\gamma(v)$ and $\Delta(v)$ on the LID can significantly manifest itself even in the case of light buffer particles, which can be clearly seen from the comparison of curve (*1*) ($\beta = 0.1$) with the dashed curve in Fig. 1.

Under condition of large Doppler broadening of the absorption line (at $\gamma_0 \ll kv_T$) the influence of the dependences $\gamma(v)$ and $\Delta(v)$ on the LID line shape is small and can be neglected (Fig. 2a). With increasing homogeneous width of the absorption line (with increasing γ_0) the influence of the dependences $\gamma(v)$ and $\Delta(v)$ on the LID line shape increases and can be significant even when the homogeneous width and Doppler width are equal: $\gamma_0 = kv_T$ (Fig. 2b). Under conditions of large homogeneous broadening (at $\gamma_0 \gg kv_T$) the influence of the dependences $\gamma(v)$ and $\Delta(v)$ on the LID line shape is maximal (Fig. 2c).

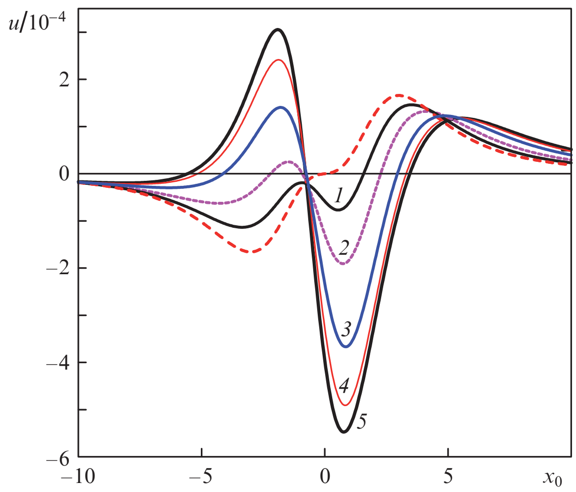

Figure 1. Dimensionless drift velocity u vs. dimensionless detuning of the radiation frequency x_0 at mass ratios of buffer and resonance particles $\beta = 0.1$ (*1*), 0.3 (*2*), 1 (*3*), 3 (*4*) and 10 (*5*), $\gamma_0/(kv_T) = 3$, $t_0 = 1.43$, $p = 6$, $\Delta_0 > 0$, $(\Gamma_m/2)/(kv_T) = 0.01$. The dashed curve is given for $\gamma(v) = \gamma_0 = \text{const}$, $\Delta(v) = \Delta_0 = \text{const}$, solid curves are calculated taking into account the effect of the dependences $\gamma(v)$ and $\Delta(v)$ on the LID line shape.

Anomalous LID appears only when the difference of transport collision frequencies $\Delta\nu(v)$ at the combining levels changes its sign near the average thermal velocity v_T of resonance particles (at $t \sim 1$). Strong sensitivity of the LID line shape to the value of the dimensionless velocity t_0, at which the difference between the transport collision frequency [or a factor of $\tau(t)$] is zero, is presented in Fig. 3. If $t_0 \sim 1$, then there appears anomalous LID and the dependences $\gamma(v)$ and $\Delta(v)$ strongly influence the LID line shape (Figs 3a and b). If t_0 is several times less than or greater than unity, then there appears normal LID, at which the dependences $\gamma(v)$ and $\Delta(v)$ have little effect on the LID line shape (Figs 3c and d).

The fact that the calculation of anomalous LED requires joint consideration of dependences of collision broadening γ and shift Δ of the absorption on the resonance particle velocity v is illustrated in Fig. 4. Each of these dependences equally strongly affects the shape of the LID line.

5. Conclusions

The presented results show that in calculating the LID the account for the dependence of collision broadening γ and shift Δ of the absorption line on the resonance particle velocity v can radically change the spectral shape of the LID signal, up to the appearance of additional zeros in the dependence of the drift velocity on the radiation frequency [see the dashed curve with curves (*2–5*) in Fig. 1 and the curves in Fig. 3b]. The strong influence of the dependences $\gamma(v)$ and $\Delta(v)$ on the

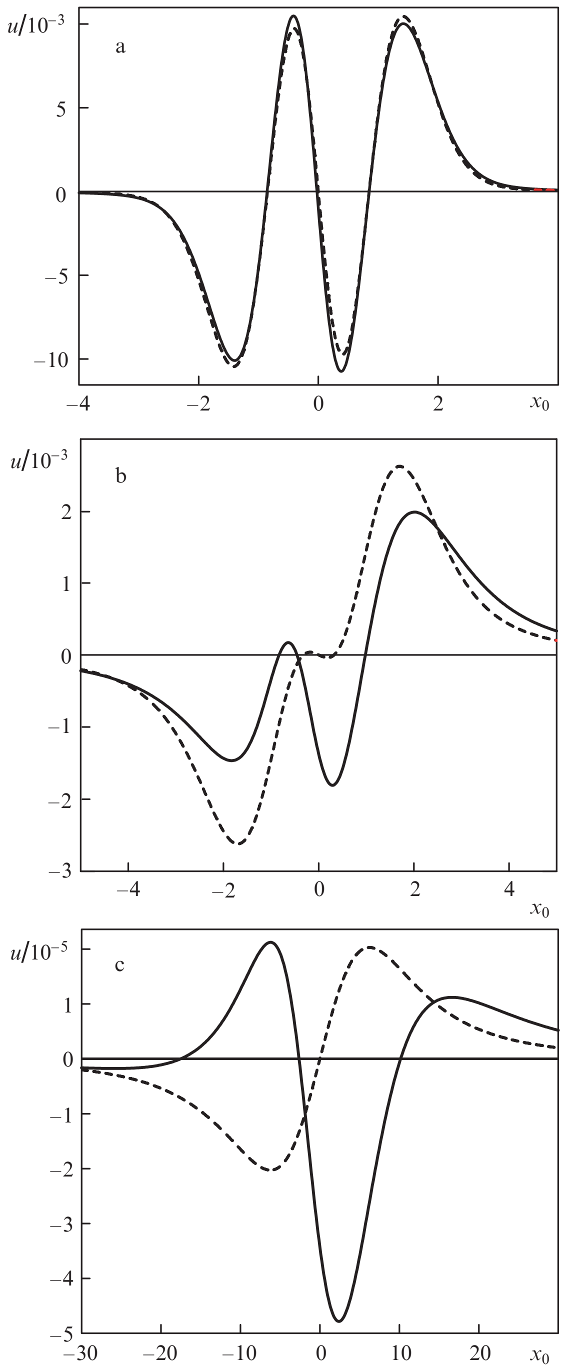

Figure 2. Dimensionless drift velocity u vs. dimensionless detuning of the radiation frequency x_0 at the ratios of the collision and Doppler widths of the absorption line $\gamma_0/(kv_T) = 0.1$ (a), 1 (b) and 10 (c), $t_0 = 1.27$ (a, b) and 1.43 (c), $\beta = 3$, $p = 6$, $\Delta_0 > 0$, $(\Gamma_m/2)/(kv_T) = 0.01$. Dashed curves are given for $\gamma(v) = \gamma_0 = \mathrm{const}$, $\Delta(v) = \Delta_0 = \mathrm{const}$, solid curves are calculated taking into account the effect of the dependences $\gamma(v)$ and $\Delta(v)$ on the LID line shape.

LID can occur only when the difference of transport collision frequencies $\Delta\nu(v)$ at the combining (affected by radiation) levels changes its sign near the average thermal velocity v_T of resonance particles (this is the criterion of possible manifestation of anomalous LID).

Sign-alternating behaviour of $\Delta\nu(v)$ is not at all exotic. In the case of the molecules such behaviour of $\Delta\nu(v)$ may be due to inelastic collisional transitions between rotational levels,

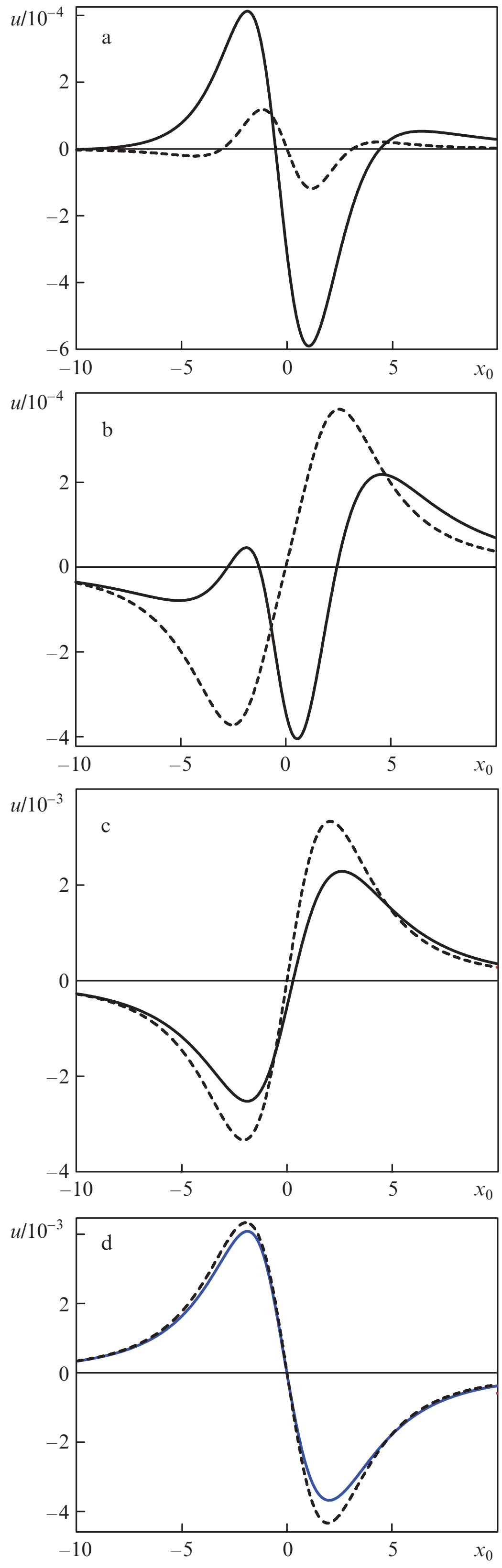

Figure 3. Sensitivity of the LID line shape to the value of the dimensionless velocity t_0, at which the difference between the transport collision frequencies [or the factor $\tau(t)$] is zero: $t_0 = 1.5$ (a), 1.35 (b), 0.3 (c) and 3 (d), $\gamma_0/(kv_T) = 3$, $\beta = 3$, $p = 6$, $\Delta_0 > 0$, $(\Gamma_m/2)/(kv_T) = 0.01$. Dashed curves are given for $\gamma(v) = \gamma_0 = \mathrm{const}$, $\Delta(v) = \Delta_0 = \mathrm{const}$, solid curves are calculated taking into account the effect of the dependences $\gamma(v)$ and $\Delta(v)$ on the LID line shape.

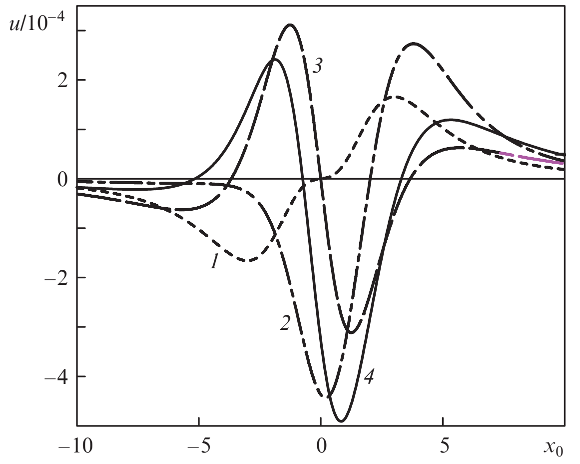

Figure 4. Dimensionless drift velocity u vs. dimensionless detuning of the radiation frequency x_0 at $\gamma_0/(kv_T) = 3$, $\beta = 3$, $t_0 = 1.43$, $p = 6$, $\Delta_0 > 0$, $(\Gamma_m/2)/(kv_T) = 0.01$. Curve (*1*) is given for $\gamma(v) = \gamma_0 = \text{const}$, $\Delta(v) = \Delta_0 = \text{const}$, curve (*2*) – for $\gamma(v) = \gamma_0 = \text{const}$, $\Delta(v) \neq \text{const}$, curve (*3*) – for $\gamma(v) \neq \text{const}$, $\Delta(v) = \Delta_0 = \text{const}$, curve (*4*) – for $\gamma(v) \neq \text{const}$, $\Delta(v) \neq \text{const}$.

and, therefore, the manifestation of anomalous LID of the molecules may have some regular pattern. In [23] it is shown that for any linear molecules with a small rotational constant one can always observe anomalous LID with an appropriate choice of the experimental conditions [temperature, rotational quantum number, transition type (P or R)].

In the case of atoms the sign-alternating behaviour of $\Delta\nu(v)$, necessary to observe anomalous LID, can be caused only by certain peculiarities in the behaviour of the interaction potentials of atoms in the combining states with buffer particles. Therefore, to calculate anomalous LID of atoms it is needed to know the interaction potentials for each system of colliding particles (resonance atom–buffer particle). For atoms anomalous LID was theoretically predicted and calculated in the Li–Ne and Rb–Kr systems [21, 22] for the resonant excitation of Li and Rb atoms. To observe anomalous LID in these systems requires very high temperatures ($T \sim 1000$ K), and, therefore, targeted experiments on observation of anomalous LID in them have not been performed.

For some of the atoms in a binary buffer gas mixture, the sign-alternating behaviour of $\Delta\nu(v)$ can be controlled by changing the concentration of one of the components of the buffer mixture. This is how the authors of the experiment [8] observed anomalous LID of potassium atoms in a buffer medium consisting of a mixture of neon and some other inert gas. In a theoretical paper [9] it was shown that for lithium atoms anomalous LID can be observed virtually at any temperature, depending on the concentration of neon atoms in mixtures of Ne–Ar, Ne–Kr and Ne–Xe.

Since the transport collision frequencies $\nu_i(v)$ are entirely determined by the interaction potentials of resonance and buffer particles, the line shape of anomalous LID is very sensitive to differences in the interaction potentials of resonant atoms in the ground and excited states with buffer particles. This allows for high-precision experimental testing of LID of interatomic interaction potentials used to calculate the spectral shape of the anomalous LID signal, and, therefore, the possibility of a relatively simple experimental testing of the accuracy of various theoretical methods for calculating the interaction potentials.

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