# Anomalous spectral dependence of the light-induced drift velocity

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(Submitted 28 January 1992)
Zh. Eksp. Teor. Fiz. **102**, 424–440 (August 1992)

Spectral aspects of the light-induced drift (LID) velocity are investigated. A strong dependence of the spectral shape of the LID signal on the interparticle interaction potential is predicted. The Lennard–Jones and Sutherland models are used for the interparticle interaction potential. It is shown that the drift velocity as a function of the radiation frequency can have one, three, or five zeros. The main reason for the appearance of additional zeros in the drift velocity is the dependence on the velocity of the molecules of the relative difference of the collision frequency of the excited and unexcited particles. It is shown that the additional zeros of the drift velocity can arise even in the large collisional broadening limit. In the case of the Sutherland model it is shown that the presence of singular points in the drift velocity makes it possible to extract more detailed information about the interaction potentials from the experimental data.

## 1. INTRODUCTION

Since the prediction of light-induced drift (LID)[1] and its first experimental observation[2] a large number of experimental and theoretical studies have been carried out on this phenomenon. The present state of these investigations is well reflected in Ref. 3. The essence of the effect consists in the appearance of a macroscopic flux of absorbing particles interacting with a traveling light wave and undergoing collisions with the particles of the buffer gas.

Light-induced drift belongs to the category of strongly nonequilibrium effects, and it can be described rigorously only for certain restrictions on the parameters of the system. Thus, at first it was possible to obtain a rigorous expression for the drift velocity only in the limit of large homogeneous half-width $\Gamma$ of the absorption line.[4] In this connection, the importance of having models in the LID problem for which it is possible to obtain exact analytic solutions is obvious. In this sense, the greatest success in the theory of LID has been enjoyed by the strong collision model, which allows an exact solution to be obtained and understand the behavior of the drift velocity for arbitrary values of the field intensity and the parameter $\Gamma/k\bar{v}$ (Ref. 5), where $k$ is the wave number of the radiation $\mathbf{k}$ and $\bar{v}$ is the mean thermal velocity. For certain restrictions on the system parameters it is possible to obtain a rigorous solution in the case of a light buffer gas (the Fokker–Planck limit; Ref. 6), for which, as for the strong collision limit, the transport collision cross sections $\sigma_i^{(1)}(v)$ do not depend on the particle velocity. In the possible case of a heavy buffer gas (a Lorentz gas) it is also possible to obtain a rigorous solution. The LID problem for a Lorentz gas was solved[7-10] for the case of not too great radiation intensity under the assumption that the homogeneous half-width of the absorption line and also the relative difference of the transport cross sections for the excited ($m$) and unexcited ($n$) absorbing particles with the particles of the buffer gas are independent of velocity:

$$\Gamma(v)=\text{const}, \tag{1.1}$$

$$\frac{\sigma_m^{(1)}(v)-\sigma_n^{(1)}(v)}{\sigma_n^{(1)}(v)}=\text{const} \tag{1.2}$$

An analysis of the influence on the spectral dependence of the light-induced drift velocity of the shape of the function $\Gamma(v)$ was carried out in Ref. 9 for a Lorentz gas under assumption (1.2). A quantitative theory of LID, valid for arbitrary masses of the buffer particles, was constructed in Refs. 8 and 10 using the 13-moment Grad method.

Figure 1 shows the characteristic dependence of the light-induced drift velocity $u$ of two-level particles on the detuning $\Omega=\omega-\omega_{mn}$ of the radiation frequency $\omega$ from the optical resonance frequency $\omega_{mn}$.[1] The antisymmetric character of $u(\Omega)$ with only one zero at $\Omega=0$ (Fig. 1) was reproduced in all of the cited papers.

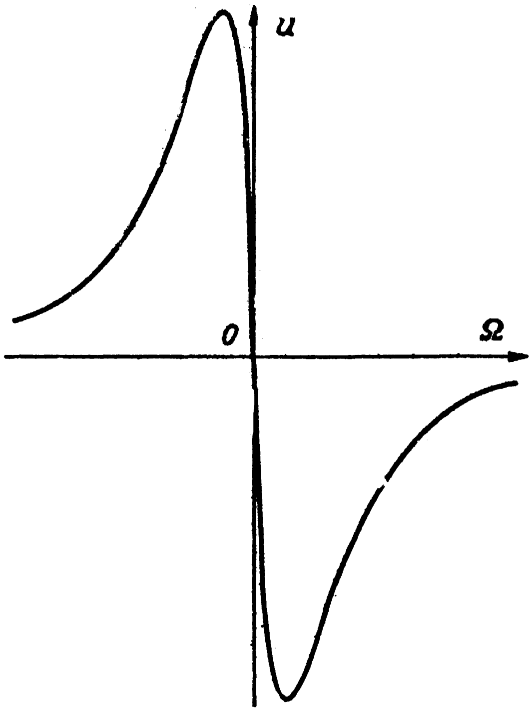


FIG. 1. Dependence of the drift velocity on the detuning $\Omega$ for the case (1.2).

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The question arises, is the spectral dependence of the drift depicted in Fig. 1 universal? The present paper is dedicated to the solution of this fundamental problem. Below we show that besides the spectral dependence with one zero illustrated in Fig. 1, behavior with three and five zeros are also possible. The main reason for these anomalies is the nondefinite character of the velocity dependence of the relative difference of the transport cross sections $\sigma_m^{(1)}(v)$ and $\sigma_n^{(1)}(v)$, i.e., nonfulfillment of condition (1.2). This condition is most strongly violated for the case of heavy buffer particles, i.e., for a Lorentz gas.[8] In this connection, in the analysis of the spectral properties of LID we have assigned particular importance to the Lorentz gas. The drift velocity depends antisymmetrically on the detuning, $u(-\Omega) = -u(\Omega)$; therefore, in what follows we will study $u(\Omega)$ only for positive detuning and speak of the number of zeros of $u(\Omega)$ only in the region $\Omega \geqslant 0$.

## 2. SOLUTION OF THE KINETIC EQUATIONS FOR A LORENTZ GAS

Let us consider the interaction of a traveling electromagnetic wave $\mathbf{E}\exp(i\omega t - i\mathbf{kr})$ + c.c. with two-level absorbing particles mixed with the buffer gas. The evolution of the absorbing gas is described by the well-known kinetic equations for the density matrix $\rho_{ij}(\mathbf{v})$ (Ref. 3):

$$\begin{aligned}
&\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla+\Gamma_m\right)\rho_m(\mathbf{v})=S_m(\mathbf{v})+\rho p(\mathbf{v}),\\
&\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla\right)\rho(\mathbf{v})=S_m(\mathbf{v})+S_n(\mathbf{v}),\\
&\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla+\frac{\Gamma_m}{2}-i(\Omega-\mathbf{kv})\right)\rho_{mn}(\mathbf{v})\\
&\qquad=S_{mn}(\mathbf{v})+iG(\rho_n(\mathbf{v})-\rho_m(\mathbf{v})),
\end{aligned}\tag{2.1}$$

where

$$p(\mathbf{v})=-\frac{2}{\rho}\,\mathrm{Re}(iG^*\rho_{mn}(\mathbf{v})),\qquad G=\frac{Ed_{mn}}{2\hbar},\tag{2.2}$$

and $\rho(\mathbf{v}) = \rho_m(\mathbf{v}) + \rho_n(\mathbf{v})$ is the velocity distribution of the absorbing particles as a whole, being a sum of the velocity distributions of the excited $\rho_m(\mathbf{v}) \equiv \rho_{mm}(\mathbf{v})$ and unexcited $\rho_n(\mathbf{v}) = \rho_{nn}(\mathbf{v})$ particles; $\rho_i(\mathbf{v})$ is normalized to the density $\rho_i$, $d_{mn}$ is the matrix element of the dipole moment for the $m$–$n$ transition, and $\Gamma_m$ is the radiative decay rate of the $m$th level. For a Lorentz gas ($M \ll M_b$ and $\rho \ll \rho_b$, where $M$ and $M_b$ are the masses of the absorbing and buffer particles, and $\rho$ and $\rho_b$ are their densities) the collision integrals have the form[11]

$$S_i(\mathbf{v})=-\frac{1}{v^2}\frac{\partial}{\partial v}(v^2 s_i(\mathbf{v}))+\rho_b v\int d\mathbf{n}'\,\sigma_i(v,\theta)\,(\rho_i(\mathbf{v})-\rho_i(\mathbf{v}')).\tag{2.3}$$

Here

$$s_i(\mathbf{v})=-\frac{M}{M_b}\nu_i(v)\,v\left(1+\bar{v}^2\frac{\partial}{\partial v^2}\right)\rho_i(\mathbf{v})$$

is the flux of particles of type $i = m, n$ in $v$ space: $\bar{v} = (2k_B T/M)^{1/2}$,

$$\begin{aligned}
&\sigma_i^{(1)}(v)=2\pi\int_0^\pi d\theta\,\sin\theta\,(1-\cos\theta)\,\sigma_i(v,\theta),\\
&\nu_i(v)=\rho_b v\sigma_i^{(1)}(v),\quad \cos\theta=\mathbf{v}'\mathbf{v}/v^2,\quad \mathbf{n}'=\mathbf{v}'/v,
\end{aligned}\tag{2.4}$$

$\sigma_i(v,\theta)$ is the elastic scattering ($v = v'$) cross section of the absorbing particles in the state $i = m,n$ for scattering by a buffer particle; $\mathbf{v}$ and $\mathbf{v}'$ are the velocities of the absorbing particle before and after the collision. The differential and integral terms on the right-hand side of Eq. (2.3) describe respectively the variation of the magnitude and direction of the velocity of the light absorbing particles during their collisions with the heavy buffer particles. In what follows we will neglect phase memory effects, the effect of which on light-induced drift was investigated in Ref. 12; i.e., we will use the following simplification for the non-diagonal collision integral:

$$S_{mn}(\mathbf{v})\approx-(\gamma(v)+i\Delta(v))\rho_{mn}(\mathbf{v}),\tag{2.5}$$

where $\gamma(v)$ and $\Delta(v)$ are the collisional broadening and collisional shift of the absorption line.

In the present work we will restrict ourselves to the investigation of the spectral peculiarities of the drift velocity. Therefore, we will solve the kinetic equations (2.1) for the stationary and spatially homogeneous case. Noting the relation $\Gamma(v) = \Gamma_m/2 + \gamma(v)$ between collisional $\gamma(v)$ and homogeneous $\Gamma(v)$ broadening of the absorption line, we rewrite Eqs. (2.1) in the form

$$\begin{aligned}
&\Gamma_m\rho_m(\mathbf{v})=S_m(\mathbf{v})+\rho p(\mathbf{v}),\\
&S_m(\mathbf{v})+S_n(\mathbf{v})=0,\\
&p(\mathbf{v})=\frac{\Gamma_m}{2\rho}\varkappa(\mathbf{v})\,(\rho(\mathbf{v})-2\rho_m(\mathbf{v})),\\
&\varkappa(\mathbf{v})=\frac{4|G|^2}{\Gamma_m((\Omega-\Delta(v)-\mathbf{kv})^2+\Gamma^2(v))}.
\end{aligned}\tag{2.6}$$

From these equations it is clear that the distribution functions $\rho_i(\mathbf{v})$ depend only on $v$ and the angle $\zeta$ between $\mathbf{v}$ and the wave vector $\mathbf{k}$. This allows us to search for the solution of the kinetic equations (2.6) in the form of an expansion in Legendre polynomials $P_l(\cos\zeta)$:

$$\begin{aligned}
&\rho_i(\mathbf{v})=\sum_{l=0}^{\infty}(2l+1)\rho_i^{(l)}(v)P_l(\cos\zeta),\\
&\rho_i^{(l)}(v)=\frac{1}{2}\int_0^\pi\rho_i(\mathbf{v})P_l(\cos\zeta)\sin\zeta\,d\zeta.
\end{aligned}\tag{2.7}$$

The formal reason for the solvability of the kinetic equations for a Lorentz gas is the following property of the collision integrals:[8]

$$\begin{aligned}
S_i^{(l)}(v)&=\frac{1}{2}\int_0^\pi S_i(\mathbf{v})P_l(\cos\zeta)\sin\zeta\,d\zeta\\
&=-\frac{1}{v^2}\frac{\partial}{\partial v}(v^2 s_i^{(l)}(v))-\nu_i^{(l)}(v)\rho_i^{(l)}(v),
\end{aligned}\tag{2.8}$$

where

$$\begin{aligned}
&s_i^{(l)}(v)=-\frac{M}{M_b}\nu_i(v)\,v\left(1+\bar{v}^2\frac{\partial}{\partial v^2}\right)\rho_i^{(l)}(v),\\
&\sigma_i^{(l)}(v)=2\pi\int_0^\pi\sigma_i(v,\theta)\,(1-P_l(\cos\theta))\sin\theta\,d\theta,
\end{aligned}\tag{2.9}$$

$$\nu_i^{(l)}(v)=\rho_b v \sigma_i^{(l)}(v).$$

Substituting expansion (2.7) in Eq. (2.6) and using Eq. (2.8), we obtain the following equations for the moments $\rho_i^{(l)}(v)$ of the distribution functions $\rho_i(\mathbf{v})$:

$$(\Gamma_m+\nu_m^{(l)}(v))\rho_m^{(l)}(v)$$
$$=\frac{\Gamma_m}{2}\sum_{l',l_1=0}^{\infty}\frac{(2l_1+1)(2l'+1)}{(2l+1)}$$
$$\times(C_{l_10l'0}^{l0})^2\varkappa^{(l_1)}(v)(\rho^{(l')}(v)-2\rho_m^{(l')}(v)),$$
$$S_m^{(l)}(v)+S_n^{(l)}(v)=0. \qquad (2.10)$$

Here $C_{l_10l'0}^{l0}$ are the Clebsch–Gordan coefficients; the moments $\varkappa^{(l)}(v)$ are expressed exactly in terms of the saturation parameter $\varkappa(\mathbf{v})$ as are the moments $\rho_i^{(l)}(v)$ in terms of $\rho_i(\mathbf{v})$ [Eqs. (2.7)]. In a Lorentz gas the magnitude of the velocity of the absorbing particles hardly varies during the collisions since the masses satisfy $M\ll M_b$. This allows us to neglect diffusion in velocity space for $l\geqslant 1$, i..e, the differential term in Eq. (2.8). An exception is the case $l=0$ in the second of Eqs. (2.10), for which other relaxation processes are absent ($\nu_i^{(0)}(v)=0$).

Let us solve Eqs. (2.10) for fields of moderate intensity

$$\frac{\Gamma_m}{\Gamma_m+\nu_m}\varkappa(\mathbf{v})\ll 1. \qquad (2.11)$$

This condition allows us to replace $\rho_i^{(l')}(v)$ in the right-hand side of Eqs. (2.10) by $\rho_i^{(0)}(v)\delta_{l',0}$ and obtain the following expressions for the moments $\rho_i^{(l)}(v)$:

$$\rho^{(l)}(v)=\left(\frac{\nu_n^{(l)}(v)-\nu_m^{(l)}(v)}{\nu_n^{(l)}(v)}\right)\rho_m^{(l)}(v), \quad l\geqslant 1, \qquad (2.12)$$

where

$$\rho_m^{(0)}(v)=\frac{\varkappa^{(0)}(v)}{2(1+\varkappa^{(0)}(v))}\rho^{(0)}(v),$$

$$\rho_m^{(l)}(v)=\frac{\Gamma_m\varkappa^{(l)}(v)}{2(\Gamma_m+\nu_m^{(l)}(v))(1+\varkappa^{(l)}(v))}\rho^{(0)}(v). \qquad (2.13)$$

The distribution $\rho^{(0)}(v)$ is the solution of the second of Eqs. (2.10) for $l=0$:

$$\frac{1}{v^2}\frac{\partial}{\partial v}(v^2 s^{(0)}(v))=0, \qquad s^{(0)}(v)=s_m^{(0)}(v)+s_n^{(0)}(v).$$

Hence it follows that $s^{(0)}(v)=\mathrm{const}/v^2$. However, the total flux of the absorbing particles $s(\mathbf{v})$ in velocity space should be finite for all values of $v$; therefore $s^{(0)}(v)=0$, whence

$$\left(1+\bar{v}^2\frac{\partial}{\partial v^2}\right)\rho^{(0)}(v)+\left(\frac{\nu_m(v)}{\nu_n(v)}-1\right)\left(1+\bar{v}^2\frac{\partial}{\partial v^2}\right)\rho_m^{(0)}(v)=0.$$

Substituting $\rho_m^{(0)}(v)$ from Eq. (2.13) in this equation, we find that the isotropic part $\rho^{(0)}(v)$ of the distribution function of the absorbing particles has the form

$$\rho^{(0)}(v)=\rho W(v)\frac{\varepsilon(v)}{\bar{\varepsilon}},$$

$$\varepsilon(v)=\frac{1+\varkappa^{(0)}(v)}{1+(1+\alpha(v)/2)\varkappa^{(0)}(v)} \qquad (2.14)$$
$$\times\exp\left\{\frac{1}{2}\int_0^\infty\frac{\varkappa^{(0)}(v_1)\dfrac{\partial\alpha(v_1)}{\partial v_1}dv_1}{1+(1+\alpha(v_1)/2)\varkappa^{(0)}(v_1)}\right\},$$

$$\bar{\varepsilon}=\int dv\,\varepsilon(v)W(v), \qquad \alpha(v)=\frac{\nu_m(v)-\nu_n(v)}{\nu_n(v)}$$

and is different from the Maxwellian distribution $W(v)=(\pi\bar{v}^2)^{-3/2}\exp(-v^2/\bar{v}^2)$ if $\nu_m(v)\neq\nu_n(v)$. The dependence of the collision frequencies on the velocity influences the shape of the absorption line, which is described by the probability of absorption per unit time

$$p=\int d\mathbf{v}p(\mathbf{v})=4\pi\int_0^\infty dv\,v^2p^{(0)}(v)$$
$$=\frac{2\pi}{\rho}\Gamma_m\int_0^\infty dv\,v^2\rho^{(0)}(v)\left(\frac{\varkappa^{(0)}(v)}{1+\varkappa^{(0)}(v)}\right). \qquad (2.15)$$

## 3. THE DRIFT VELOCITY

The drift velocity is expressed in terms of the first moment of the distribution function:

$$\mathbf{u}=\frac{1}{\rho}\int\mathbf{v}\rho(\mathbf{v})d\mathbf{v}=4\pi\frac{\mathbf{k}}{k}\int_0^\infty dv\,v^3\rho^{(1)}(v). \qquad (3.1)$$

The solution (2.12)–(2.14) of the kinetic equations (2.10), taking into account the explicit expressions for the first two moments of the saturation parameter (2.6)

$$\varkappa^{(0)}(v)=\varkappa_0 t^{-1}\psi(t), \quad \varkappa^{(1)}(v)=\varkappa_0 t^{-2}f(t)$$

allows one to obtain the following expression for the light-induced drift velocity:

$$\mathbf{u}=\frac{\mathbf{k}}{k}\frac{2}{\pi^{1/2}}\varkappa_0\Gamma_m\bar{v}\int_0^\infty dt\,t\left(\frac{\nu_n(v)-\nu_m(v)}{\nu_n(v)}\right)\frac{f(t)e^{-t^2}}{(\Gamma_m+\nu_m(v))}\eta(v), \qquad (3.2)$$

where we have introduced the dimensionless velocity $t=v/\bar{v}$, detuning $x=(\Omega-\Delta(v))/k\bar{v}$, and homogeneous half-width $y=\Gamma(v)/k\bar{v}$, and also made use of the following notation:

$$\psi(t)=\mathrm{arctg}\left(\frac{t-x}{y}\right)+\mathrm{arctg}\left(\frac{t+x}{y}\right),$$
$$f(t)=x\psi(t)+\frac{y}{2}\ln\left(\frac{y^2+(t-x)^2}{y^2+(t+x)^2}\right), \qquad (3.3)$$
$$\varkappa_0=\frac{2|G|^2}{\Gamma_m k\bar{v}}, \qquad \eta(v)=\frac{\rho^{(0)}(v)}{\rho(1+\varkappa^{(0)}(v))W(v)}.$$

The functions $f(t)$ and $\psi(t)$ are positive for $t\geqslant 0$.

In contrast with previous papers, here the relative difference of the transport collision frequencies in the expression for the drift velocity (3.2) is inside the integral. This result was first obtained in Ref. 12. Equation (3.2) shows that the dependence of the factor $\alpha(v)=[\nu_m(v)-\nu_n(v)]/\nu_n(v)$ on the velocity $v$ can qualitatively alter the character of the spectral dependence of the drift velocity $u$ (even to the appearance of the additional zeros) if the sign of $\alpha(v)$ varies as $v$ varies. Below we will convince ourselves that such a property of $\alpha(v)$ is not exotic.

If the ends of the gas cell are closed, then the drift of the absorbing particles along the $z$ axis ($k_z = k$) leads to their accumulation at one of the ends of the cell. It can be shown that the corresponding distribution of the absorbing particle concentration for an optically thin medium has exponential form:

$$\rho(z)=\rho(0)\exp\left(\frac{u}{D}z\right).$$

Thanks to the difference of the transport cross sections of the excited and unexcited particles, the diffusion coefficient of the absorbing particles

$$D=\frac{4\pi}{3}\int_0^\infty dv\frac{v^4}{\nu_n(v)}\frac{\varepsilon(v)}{\bar{\varepsilon}}W(v)$$

depends on the radiation parameters. It is not difficult to convince oneself that formula (3.2) coincides with the corresponding expression for the drift velocity in Ref. 8 if the hypothesis of similarity of cross sections (1.2) is fulfilled.

## 4. THE LENNARD–JONES AND SUTHERLAND MODELS

There are two quite general interaction potentials which allow one to obtain simple analytical expressions for the scattering cross sections. We refer to the Lennard–Jones and Sutherland interaction potentials or models.[13] We will study the spectral properties of the light-induced drift velocity in the case of these models, directing our main attention to the more realistic Sutherland model.

1. The Lennard–Jones model. This model is based on the interaction potential

$$V(r)=\varepsilon\left(\left(\frac{a}{r}\right)^\tau-\left(\frac{a'}{r}\right)^2\right),\quad \tau>2, \tag{4.1}$$

in which the attractive part of the potentials is assumed to be small in comparison with its repulsive part ($a' \ll a$). In the classical approximation the transport cross section for the Lennard–Jones model has the form ($i = m,n$)

$$\sigma_i^{(1)}(v)=\sigma_i t^{-4/\tau_i}(1+\xi_i t^{4/\tau_i-2}),\quad t=\frac{v}{\bar{v}}, \tag{4.2}$$

where

$$\sigma_i=2\pi a_i^2\left(\frac{\tau_i\varepsilon_i^*}{2}\right)^{2/\tau_i}A_1(\tau_i+1),$$

$$\xi_i=2\tau_i^{-2/\tau_i}\left(\frac{a_i'}{a_i}\right)^2\frac{B_1(\tau_i+1)}{A_1(\tau_i+1)}\left(\frac{\varepsilon_i^*}{2}\right)^{1-2/\tau_i}\ll 1,$$

$$\varepsilon_i^*=\frac{\varepsilon_i}{k_B T}.$$

The values of the functions $A_1(\tau)$ an $B_1(\tau)$ are given in Ref. 13. The change in the transport collision frequencies during excitation

$$\frac{1}{\nu_m(v)}-\frac{1}{\nu_n(v)}=\frac{t[t^{2-4/\tau_n}-(\sigma_m/\sigma_n)t^{2-4/\tau_m}+\xi_n-\xi_m\sigma_m/\sigma_n]}{\nu_m(t^{2-4/\tau_m}+\xi_m)(t^{2-4/\tau_n}+\xi_n)},$$
$$\nu_i=\rho_b\bar{v}\sigma_i \tag{4.3}$$

is controlled by the four parameters $\tau_i$, $\varepsilon_i^*$, $a_i$, and $a_i'$ of the potential (4.1). If $\tau_m = \tau_n = \tau$, then the buffer particle, passing at a distance $r$ from the absorbing particle, shifts the frequency of the absorbing particle by an amount proportional to $r^{-\tau}$. According to Weisskopf–Lindholm theory such instantaneous shifts of the resonance frequency lead to collisional broadening $\gamma(v)$ and a shift $\Delta(v)$ of the absorption line[14,15]

$$\gamma(v)=\Gamma_0 t^{(\tau-3)/(\tau-1)},\quad \frac{\Delta(v)}{\gamma(v)}=\mathrm{tg}\left(\frac{\pi}{\tau-1}\right). \tag{4.4}$$

The latter formula is valid for $\tau > 3$. The dependence of the collisional shift $\Delta(v)$ of the absorption line on the velocity must be taken into account in LID theory since $\Delta(v)$ can be of the same order of magnitude as the collisional broadening $\gamma(v)$;[1] for example, for the ratio $\Delta(v)/\gamma(v)$ we obtain

$$\Delta(v)/\gamma(v)=\begin{cases}1{,}7, & \tau=4,\\ 1, & \tau=5,\\ 0{,}7, & \tau=6.\end{cases}$$

Note that the velocity dependence of the absorption line shift $\Delta(v)$ leads to a violation of the antisymmetric dependence of the LID velocity on $\Omega$.[12,16]

2. The Sutherland model. In this model the molecules are smooth, rigid, elastic spheres of radius $a$, surrounded by weak attractive fields ($a' \ll a$):

$$-\varepsilon\left(\frac{a'}{r}\right)^\tau.$$

Formally, the Sutherland model can be considered as a particular case of the Lennard–Jones model for $\tau_m = \tau_n \to \infty$. In the Sutherland model the transport cross section, the collisional broadening, and the absorption line shift in the classical approximation depend on the dimensionless velocity $t = v/\bar{v}$ in the following way:

$$\sigma_i^{(1)}(v)=\sigma_i(1+\xi_i t^{-2}),\quad \gamma(v)=\Gamma_0 t,\quad \Delta(v)=0, \tag{4.5}$$

where

$$\xi_i=2\varepsilon_i^*\left(\frac{a_i'}{a_i}\right)^{\tau_i}i_1(\tau_i+1)\ll 1,\quad \sigma_i=\pi a_i^2,\quad i=m,n. \tag{4.6}$$

The values of the integrals $i_1(\tau)$ are tabulated in Ref. 13.

Optical excitation of the absorbing particle causes its transport collision frequency to change in the following way:

$$\frac{1}{\nu_m(v)}-\frac{1}{\nu_n(v)}=\frac{(1-\sigma_m/\sigma_n)t(t^2-q)}{\nu_m(t^2+\xi_m)(t^2+\xi_n)}. \tag{4.7}$$

Here

$$q=\frac{\xi_m\sigma_m/\sigma_n-\xi_n}{1-\sigma_m/\sigma_n},\quad \nu_i=\rho_b\bar{v}\sigma_i. \tag{4.8}$$

Equations (4.3) and (4.7) directly demonstrate the possibility of change of sign of $\alpha(v) = (\nu_m(v) - \nu_n(v))/\nu_n(v)$ with variation of $v$. For example, if $q > 0$ in the Sutherland model (4.3), then the sign of $\alpha(v)$ changes at $t = q^{1/2}$. As can be seen from Eq. (4.8), the sign of $\alpha(v)$ changes if the attractive and repulsive parts of the Sutherland potential change in different directions; i.e., $q > 0$ if $\xi_m/\xi_n < \sigma_n/\sigma_m < 1$ or $\xi_n/\xi_m < \sigma_m/\sigma_n < 1$. In the Sutherland model $\alpha(v)$ does not change its sign if the attractive part of the potential is absent ($\xi_i = 0$).

## 5. DEPENDENCE OF THE DRIFT VELOCITY ON THE RADIATION FREQUENCY

We proceed now to a direct study of the spectral properties of the LID velocity. Toward this end, we restrict ourselves to the Sutherland model, but in addition we will take as given the following conditions, which are frequently realized in experiments:

$$\varkappa_0 \ll 1, \quad \Gamma_m \ll \nu_m, \quad \Gamma(v) \approx \gamma(v). \tag{5.1}$$

These nonessential restrictions allow us to substantially simplify the expression for the drift velocity (3.2), (4.7)

$$u = cu_0, \quad c = \frac{2}{\pi^{1/2}} \bar{v} \varkappa_0 \frac{\Gamma_m}{\nu_m}\left(1 - \frac{\sigma_m}{\sigma_n}\right),$$
$$u_0 = \int_0^\infty dt\, Q(t) f(t), \quad Q(t) = \frac{(t^2 - q) t^2 \exp(-t^2)}{(t^2 + \xi_m)(t^2 + \xi_n)} \tag{5.2}$$

and for the probability of absorption per unit time (2.15)

$$p = \frac{2}{\pi^{1/2}} \varkappa_0 \Gamma_m \int_0^\infty dt\, t \psi(t) \exp(-t^2). \tag{5.3}$$

Many experimental papers on LID[3,17] address the spectral properties not only of the drift velocity (5.2), but also of the function $\tilde{\varphi}$ associated with it[8]

$$u = \bar{v} \cdot \frac{(\tilde{\nu}_n - \tilde{\nu}_m)}{(\Gamma_m + \tilde{\nu}_m)\tilde{\nu}_n} p \tilde{\varphi}, \tag{5.4}$$

where

$$\tilde{\nu}_i = \frac{3\pi^{1/2}}{8}\left[\int_0^\infty dt \frac{t^4}{\nu_i(v)} \exp(-t^2)\right]^{-1} \tag{5.5}$$

is the transport collision frequency in the absence of radiation, related to the diffusion coefficient $D_i$ by the formula $D_i = \bar{v}^2/2\tilde{\nu}_i$. For the Sutherland model (4.5) the transport collision frequency is calculated exactly:

$$\tilde{\nu}_i = \frac{3\pi^{1/2}}{4} \nu_i (1 - \xi_i - \xi_i^2 \exp(\xi_i \mathrm{Ei}(-\xi_i))^{-1}.$$

Here

$$\mathrm{Ei}(-\xi) = -\int_\xi^\infty \frac{e^{-t}}{t} dt$$

is the exponential integral function, which for small $\xi$ behaves like $\ln \gamma\xi$, where $\gamma \approx 1.781$ is Euler's constant.

If conditions (1.1) and (1.2) are satisfied, then for $\Gamma/k\bar{v} \gtrsim 0.1$ the function $\tilde{\varphi}$ is essentially identical to the function $\varphi$ (Ref. 18):

$$\varphi = \frac{\mathrm{Re}(zw(z))}{\mathrm{Re}(w(z))}, \quad z = \frac{\Omega + i\Gamma}{k\bar{v}},$$
$$w(z) = \exp(-z^2)\left[1 + \frac{2i}{\pi^{1/2}} \int_0^z \exp(\zeta^2) d\zeta\right]. \tag{5.6}$$

A representation of the drift velocity similar to expression (5.4) was first suggested in Ref. 18 with $\tilde{\varphi} = \varphi$.

The drift velocity (5.2) is equal to the integral of the product of the function $f(t)$ [Eq. (3.3)] and the function $Q(t)$ [Eq. (5.2)], which is proportional to the relative difference of the transport collision frequencies (4.6). Such a representation for the drift velocity makes it possible to understand in an easily visualized way the reason for the appearance of additional zeros of the drift velocity and the function $\tilde{\varphi}$ besides the one at $\Omega = 0$. Of special interest for us is the region $q > 0$ since for negative values of $q$ the function $Q(t)$ is positive everywhere and, consequently, the drift velocity has no additional zeros.

Let us consider the case of large Doppler broadening

$$\Gamma \ll k\bar{v} \tag{5.7}$$

and

$$q > 0. \tag{5.8}$$

Figure 2 shows how the functions $Q(t)$ [Eq. (5.2)] and $f(t)$ [Eq. (3.3)] depend on the dimensionless velocity $t = v/\bar{v}$ typical of the region (5.7) and (5.8).

For moderate detunings ($0 \leqslant x \lesssim 1$) the function $f(t)$ can be approximated in the limit (5.7) by the step function $\Theta(t)$ (curve *1*, Fig. 2):

$$f(t) \approx \pi x \Theta(t - x). \tag{5.9}$$

The function $Q(t)$ [Eq. (5.2)] changes sign at the point $t = q^{1/2}$. In the limit $x = \Omega/k\bar{v} \to 0$ the dimensionless drift

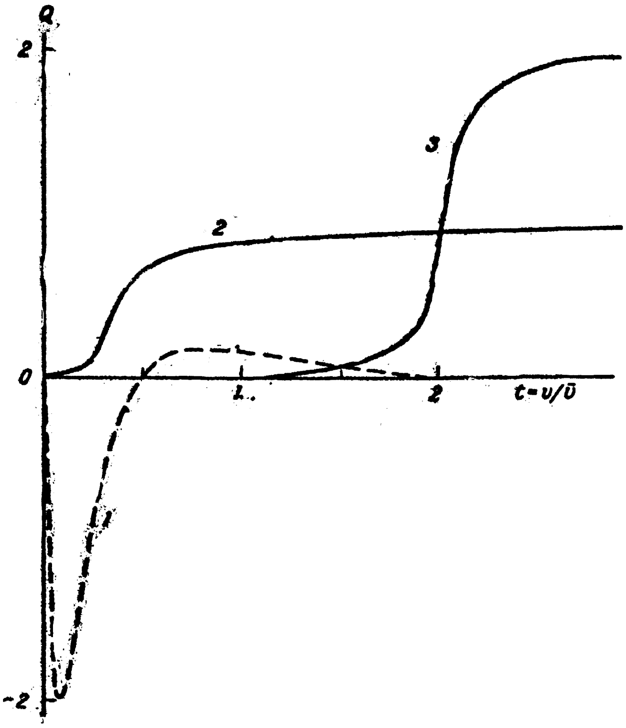


FIG. 2. Dependence of the function $Q(t)$ (5.2), proportional to the relative change in the transport cross sections upon excitation, and the "field" function $f(t)$ (3.3) on the dimensionless velocity $t = v/\bar{v}$; $\Gamma(v) = \Gamma_0$, $\Gamma_0/k\bar{v} = 0.1$: *1*) $Q(t) \times \frac{1}{2}$, *2*) $f(t)$ for $x \equiv \Omega/k\bar{v} = 0.3$, *3*) $f(t) \times \frac{1}{3}$ for $x \equiv \Omega/k\bar{v} = 2$.

velocity $u_0$ [Eq. (5.2)] is proportional to the difference of the areas under the positive part, $Q_+(t)$, and the negative part, $Q_-(t)$, of the function $Q(t)$ (Fig. 2). If the area under $Q_-(t)$ is larger than the area under $Q_+(t)$, then for small values of $x$ the dimensionless drift velocity we have $u_0<0$. At the center of the absorption line ($x=0$) the velocity satisfies $u_0=0$ since, according to relation (5.9), $f(t)\sim x$. With increasing $x$ the step function $\Theta(t-x)$ decreases the contribution of the negative part of $Q(t)$ to the integral $u_0$ (5.2). At some detuning $x_0$ the drift velocity vanishes and for $x>x_0$ its sign is reversed.

Approximation (5.9) and condition $\xi_i \ll 1$ [relation (4.6)] allow us to obtain the following expressions for the dimensionless drift velocity (5.2):

$$u_0 \approx \pi^{1/2} x\beta(x)(a^2(x)-q), \tag{5.10}$$

where

$$a^2(x)=\frac{1-\Phi(x)}{2\beta(x)}>0, \qquad \beta(x)=\frac{\exp(-x^2)}{\pi^{1/2}x}-(1-\Phi(x))>0,$$

$$\Phi(x)=\frac{2}{\pi^{1/2}}\int_0^x dt\exp(-t^2).$$

The detuning $\Omega_0$ at which $u_0=0$ holds is the solution of the equation ($x_0=\Omega_0/k\bar{v}$)

$$a^2(x_0)=q. \tag{5.11}$$

The dependence of $x_0$ on the parameter $q$ (4.8) is depicted in Fig. 3. The drift velocity $u_0$ is negative for $x<x_0$, vanishes at $x=x_0$, and is positive for $x>x_0$. Note that the shape of the function $\Gamma(v)$ of the condition for the appearance of the first additional zero $x_0$ (5.11) is independent of the drift velocity.

With further increase of the dimensionless detuning $x=\Omega/k\bar{v}\gtrsim 1$ a second additional zero can appear. Looking at Fig. 2, we note that if the "step" function $f(t)$ departs from the region of values of $x$ for which the function $Q(t)$ is different from zero (curve *2* in Fig. 2), then the function $Q(t)$ will be multiplied not by the step part (5.9) of the function $f(t)$ (3.3), but by its tail. In this case the areas under the positive and negative parts of $Q(t)$ (Fig. 2) will be subtracted from one another with different weights than in the case $0<x\lesssim 1$. For a quantitative illustration of what we have just said, consider the case $x\gg 1$. As a result of the factor $\exp(-t^2)$ the main contribution to the integral (5.2) comes from the values of $t\lesssim 1$. Therefore, in this region the tail of the function $f(t)$ depends on $t$ like $f(t)\approx\frac{4}{3}y(t/x)^3$. Recall that $y=\Gamma(v)/k\bar{v}$ depends on $t/v/\bar{v}$.

Thus, the asymptotic limit of the drift velocity (5.2) at large detunings ($\Omega/k\bar{v}\gg 1$) has the form

$$u_0\approx\frac{\pi^{1/2}(k\bar{v})^2\Gamma_0}{3\Omega^3}\left(\frac{3}{2}-q\right), \tag{5.12}$$

for $\Gamma(v)=\Gamma_0(t)$ (the Sutherland model), and

$$u_0\approx\frac{2(k\bar{v})^2\Gamma_0}{3\Omega^3}(1-q), \tag{5.13}$$

for $\Gamma(v)=\Gamma_0=\text{const}$. The obtained asymptotic dependences show that the condition for the appearance of a second additional zero of the drift velocity ($q>3/2$ if $\Gamma(v)=\Gamma_0 t$, and $q>1$ if $\Gamma(v)=\Gamma_0$), in contrast with Eq. (5.11), depends on the shape of the function $\Gamma(v)$.

Figures 4 and 5 show the dependences on the detuning $\Omega$ of the drift velocity $u$ [Eq. (5.2)] and the functions $\tilde{\varphi}$ [Eq. (5.4)] and $\varphi$ [Eq. (5.6)]. Since $u(-\Omega)=-u(\Omega)$, Figs. 4 and 5 demonstrate the presence in the drift velocity $u$ and the function $\tilde{\varphi}$ of three and five zeros.

Is the appearance of additional zeros in the drift velocity possible for large homogeneous broadening

$$\Gamma\gg k\bar{v}? \tag{5.14}$$

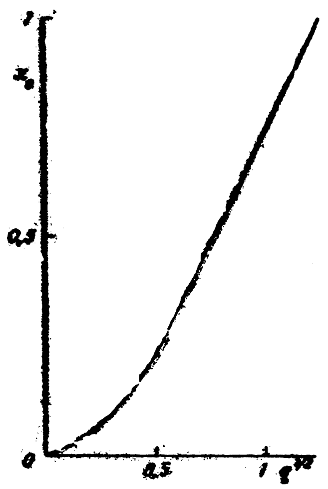


FIG. 3. Dependence of the first additional zero $x_0\equiv\Omega_0/k\bar{v}$ (5.11) of the drift velocity on the parameter $q$ (4.8), which characterizes the change in the potential.

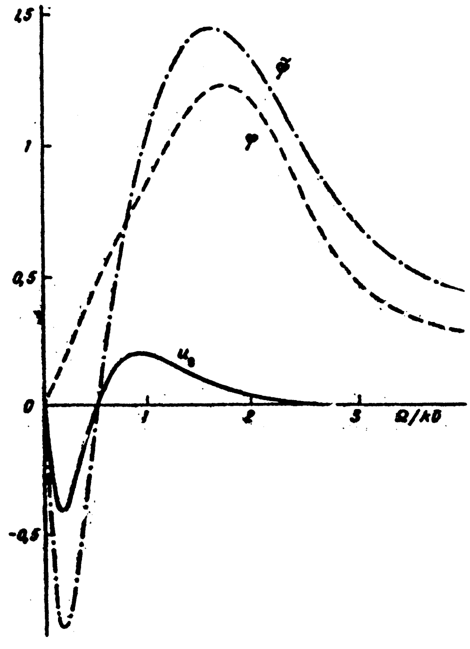


FIG. 4. Spectral dependence of the LID velocity (5.2), the functions $\tilde{\varphi}$ (5.4) and $\varphi$ (5.6) for low pressure of the buffer gas: $\Gamma_0/k\bar{v}=0.1$, $q=0.6317$, $\xi_m=0.02$, $\xi_n=0.04$.

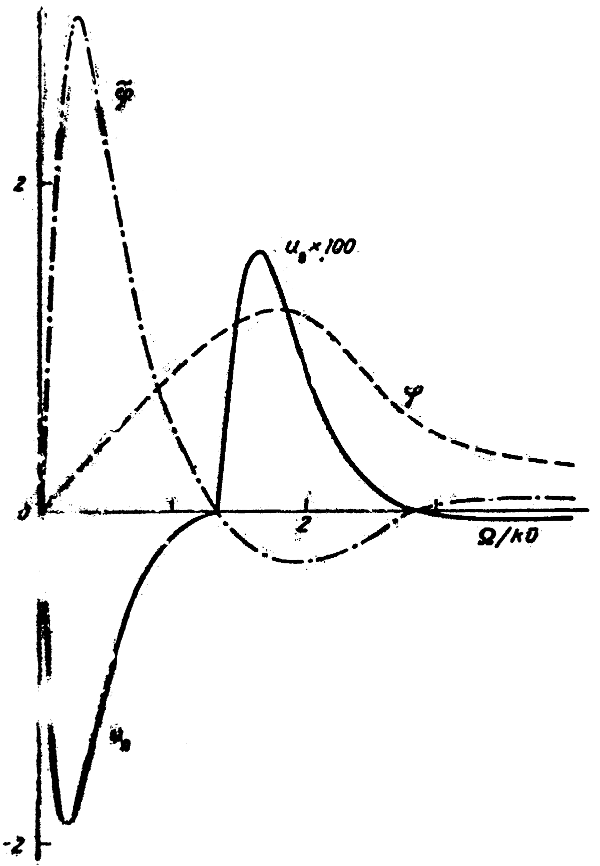


FIG. 5. Spectral dependence of the LID velocity (5.2), the functions $\tilde{\varphi}$ (5.4) and $\varphi$ (5.6) for low pressure of the buffer gas: $\Gamma_0/k\bar{v} = 0.1$, $q = 1.965$, $\xi_m = 0.02$, $\xi_n = 0.04$.

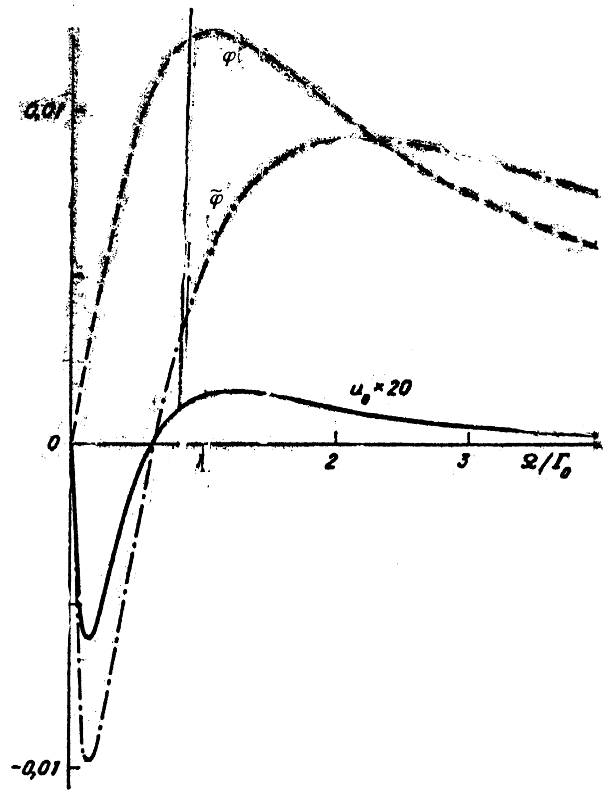


FIG. 6. Spectral dependence of the LID velocity (5.2), the functions $\tilde{\varphi}$ (5.4) and $\varphi$ (5.6) for high pressure of the buffer gas: $\Gamma_0/k\bar{v} = 10$, $q = 0.6317$, $\xi_m = 0.02$, $\xi_n = 0.04$.

At first glance, the answer to this question would appear to be negative. To a significant extent, the results of Ref. 8, obtained under assumptions (1.1) and (1.2), support this conviction. However, as we will now see, when conditions (1.1) and (1.2) are simultaneously violated the drift can disappear not only at $\Omega = 0$, but elsewhere as well. Taking into account that in the homogeneous-broadening limit of the absorption line (5.14) the function $f(t)$ given by Eq. (3.3) is equal to $\frac{4}{3}yxt^3/(y^2 + x^2)^2$, we note that the dimensionless drift velocity (5.2) at small detunings for $\Gamma(v) = \Gamma_0 t$ behaves like

$$u_0 \approx \frac{4\pi (k\bar{v})^2}{3\Gamma_0^3 \varepsilon} \left( \frac{\zeta}{2\pi^{1/2}} - q \right) \Omega, \quad |\Omega| \ll \Gamma_0. \qquad (5.15)$$

For large detunings ($|\Omega| \gg k\bar{v}$, $\Gamma_0$) the expression for the drift velocity coincides with expression (5.12) if $\Gamma(v) = \Gamma_0 t$ and the expression (5.13) if $\Gamma(v) = \Gamma_0$. Here $\zeta = (\xi_m + \xi_n)^{1/2}$.

Formulas (5.12) and (5.15) imply that the drift velocity has an additional zero besides $\Omega = 0$ if

$$\frac{\zeta}{2\pi^{1/2}} < q < \frac{3}{2}. \qquad (5.16)$$

The results of numerical analysis of formulas (5.2) and (5.4) in the limit (5.14) confirm this (see Fig. 6).

As can be seen from Fig. 6, the function $\hat{\varphi}$ (5.4), even in the limit (5.14),

$$\tilde{\varphi} = \frac{\pi^{1/2}}{4} \Omega k\bar{v}$$

$$\times \frac{\int_0^\infty dt\, t^4 \exp(-t^2) [\nu_m^{-1}(v) - \nu_n^{-1}(v)] \frac{\Gamma(v)}{\Gamma^2(v) + \Omega^2}}{\int_0^\infty dt\, t^4 \exp(-t^2) [\nu_m^{-1}(v) - \nu_n^{-1}(v)] \int_0^\infty dt t^2 \exp(-t^2) \frac{\Gamma(v)}{\Gamma^2(v) + \Omega^2}}$$

can differ greatly from $\varphi$ given by (5.6). These two functions coincide exactly with each other ($\tilde{\varphi} = \varphi = k\bar{v}\Omega/(\Gamma^2 + \Omega^2)$) only when conditions (1.1) and (1.2) are simultaneously satisfied. For large detunings ($\Omega \gg \Gamma$) the formula $\tilde{\varphi} = (k\bar{v}/\Omega)\Lambda$, where $\Lambda = 1$ if both conditions are satisfied, is valid. If either of these two conditions is violated, $\Lambda$ depends on the parameters of the interaction potential between the particles. Attention was first directed to this fact in Ref. 9 for the case (1.2).

## 6. TEMPERATURE DEPENDENCE OF THE CONDITIONS FOR APPEARANCE OF ADDITIONAL ZEROS OF THE DRIFT VELOCITY

The condition for the appearance of additional zeros of the drift velocity depends on the parameter $q$ given by Eqs. (4.6), (4.8),

$$q \propto \frac{1}{T},$$

and $\zeta$ (5.16),

$$\zeta = (\xi_m + \xi_n)^{1/2} \propto T^{-1/2}.$$

The temperature dependence of these two parameters can be conveniently expressed in terms of their values at some fixed temperature $T_0$, e.g., room temperature:

$$q=q(T_0)\frac{T_0}{T},\quad \zeta=\zeta(T_0)\left(\frac{T_0}{T}\right)^{1/2}. \tag{6.1}$$

Let us consider first the case of large Doppler broadening $\Gamma \ll k\bar{v}$. The first additional zero appears at the detuning determined by Eq. (5.11) (Fig. 3), which, taking Eqs. (6.1) into account, can be rewritten in the form

$$a^2(x_0)=q(T_0)\frac{T_0}{T}. \tag{6.2}$$

From the definition of the function $a(x)$ of Eq. (5.10) it is not hard to see that $a^2(x_0)\approx x_0\pi^{1/2}/2$ holds for $x_0 \ll 1$ $(q \ll 1)$ and $a(x_0)\approx x_0$ for $x_0 \gg 1$ $(q \gg 1)$. Thus, from Eq. (6.2) we find that for small values of the parameter $q$ (6.1) the position of the first zero depends on the temperature as

$$x_0 \approx \frac{2}{\pi^{1/2}} q(T_0)\frac{T_0}{T}. \tag{6.3}$$

As the parameter $q$ the temperature dependence of $x_0$ becomes weaker:

$$x_0 \approx \left[q(T_0)\frac{T_0}{T}\right]^{1/2},\quad q \gg 1. \tag{6.4}$$

The second additional zero (5.12) arises if $q > 3/2$. Taking the temperature dependence of the parameter $q$ (6.1) into account, we note that the second additional zero appears at temperatures

$$\frac{T}{T_0} < \frac{2}{3} q(T_0). \tag{6.5}$$

In the opposite case of large collisional broadening $(\Gamma \gg k\bar{v})$ the drift velocity has one additional zero if condition (5.6) is satisfied, i.e., when the temperature is in the interval

$$\frac{2}{3} q(T_0) < \frac{T}{T_0} < \left[\frac{2\pi^{1/2} q(T_0)}{\zeta(T_0)}\right]^{1/2}.$$

The temperature dependence of the spectral shape of the LID signal can be used to measure the parameter $q$ (4.8), which characterizes the change in the interaction potential associated with optical excitation.

## 7. CONCLUSION

The appearance of additional zeros in the drift velocity makes it possible to extract more detailed information from the experimental data on the interaction potentials between the particles, specifically by measuring the change in the attractive and repulsive parts of the potential upon optical excitation. For example, for the Sutherland potential measuring the position of the first additional zero $x_0$ gives the value of the parameter $q$ of Eq. (5.11) (Fig. 3). The behavior of the drift velocity (5.2) at large detunings (5.12) allows one to measure $1-\sigma_m/\sigma_n$. Next, with the help of the values of $q$ and $1-\sigma_m/\sigma_n$ thus found one can determine the change in the attractive part of the potential $\xi_m - \xi_n = (\sigma_n/\sigma_m - 1)(q+\xi_n)$. Here the parameters of the interaction potential for the unexcited particles are assumed to be known. Naturally, in the actual extraction of the parameters of the interaction potential from LID experiments, as in classical gas dynamics, it is necessary to choose an interaction potential model.

Note that the results of the present work are not specific to the Lorentz gas $(M_b/M \gg 1)$; i.e., analogous spectral properties should also hold for $M_b/M \lesssim 1$. Indeed, the main reason for the spectral properties of the drift velocity obtained here is the dependence of the collision frequencies $\nu_i(v)$ on the velocity. In Ref. 8 it was shown that the dependence of $\nu_i(v)$ on $v$ is the preserved even for $M_b/M \lesssim 1$ and disappears completely only for heavy absorbing particles: $M_b/M \ll 1$. With the goal of carrying out a qualitative study of the effect of the mass ratio $M_b/M$ on the spectral anomalies of LID we have, following Ref. 8, used the 13-moment Grad method. However, a comparison of the Grad expansion with the exact solution for a Lorentz gas (3.2), (5.2) shows that the convergence of the Grad method worsens drastically just in that region of the potential parameters where the spectral anomalies of the LID effect are observed.

It is not difficult to convince oneself that analogous LID spectral anomalies should also be manifested in the light-induced heat flux and pressure tensor, in the effect of diffusive traction of particles into the light beam (or ejection), and in other LID-related effects.[3]

We would like to thank C. G. Rautian and A. M. Shalagin for fruitful discussions and valuable remarks.

[1)] S. G. Rautian brought to our attention the necessity of taking account of the collision shift $\Delta(v)$ of the absorption line in LID theory and formula (4.4) for $\Delta(v)$.


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Translated by P. F. Schippnick