Spectral anomalies in the diffusive pulling of particles into a light beam or in the expulsion of particles from the beam

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(Submitted 8 January 1992)
Pis'ma Zh. Eksp. Teor. Fiz. **55**, No. 4, 219–222 (25 February 1992)

The spectral shape of the signal in the diffusive pulling of particles into a light beam (or in the expulsion of particles from the light beam) should be anomalously sensitive to the interparticle interaction potential.

The diffusion coefficients of excited particles (m) may be quite different from those for unexcited particles (n). This circumstance sets the stage for numerous new kinetic effects in a laser beam,[1] in particular, a light-induced diffusive pulling of particles into a light beam or a corresponding expulsion of particles from the beam.[2] This pulling effect has now been detected and studied in detail in experiments with sodium vapor in a variety of buffer gases.[3,4] This effect can be summarized as follows. The intensity of the laser light is spatially nonuniform in the direction transverse with respect to the light wave vector ($\vec{k}$) or in the longitudinal direction, if optical absorption is strong. Since the diffusion coefficient of the particles changes when they are optically excited, absorbing particles will be drawn into the region of elevated light intensity or expelled from this region. The spatial distribution of the light-induced nonequilibrium increment $\delta\rho$ in the density of absorbing particles reproduces the spatial distribution of the light intensity. The theory for the effect in its present form[1,2] states that the spectrum of the pulling signal, $\delta\rho$, will have the same shape as the absorption line.

In the present letter we show that the spectrum of the pulling signal may be qualitatively different from the shape of the absorption line. Specifically, a dip may form near the center of the absorption line on the frequency dependence of $\delta\rho$. Furthermore, $\delta\rho$ becomes a variable-sign function of the light frequency.

The evolution of the gas which absorbs the light $\vec{E}\exp(i\omega t - i\vec{k}\vec{r}) + \text{c.c.}]$ is de-

 0021-3640/92/040214-04$03.00

scribed by the kinetic equations[1]

$$(\Gamma_m + \vec{v}\vec{\nabla})\rho_m(\vec{v}) = S_m(\vec{v}) + \frac{\Gamma_m}{2}\kappa(\vec{v})(\rho(\vec{v}) - 2\rho_m(\vec{v})),$$

$$(\frac{\partial}{\partial t} + \vec{v}\vec{\nabla})\rho(\vec{v}) = S_m(\vec{v}) + S_n(\vec{v}), \quad \rho(\vec{v}) = \rho_m(\vec{v}) + \rho_n(\vec{v}), \tag{1}$$

where $\rho_i(\vec{v})$ is the velocity distribution of the absorbing particles in state i ($= m$ or n), $S_i(\vec{v})$ is the corresponding collision integral, and Γ_m is the relaxation constant of the excited state. In experiments on the pulling effect,[3,4] the transverse dimension of the gas-filled cell is considerably larger than the cross-sectional radius of the light beam. In addition, the homogeneous half-width of the absorption line, $\Gamma(v)$, is large in comparison with the Doppler broadening $k\bar{v}$. It is thus legitimate to ignore the light-induced drift.[5] Here $\bar{v}$ is the mean thermal velocity. By virtue of the relation $\Gamma(v) \gg k\bar{v}$, the Doppler effect can be ignored in the saturation parameter $\kappa(\vec{v})$:

$$\kappa(\vec{v}) \approx \kappa(v) = \frac{4|G|^2\Gamma(v)}{\Gamma_m(\Gamma^2(v) + (\Omega - \Delta(v))^2)}, \tag{2}$$

where $G = Ed_{mn}/2\hbar$ is the Rabi frequency, d_{mn} is the dipole moment of the m–n transition, and $\Omega = \omega - \omega_{mn}$ is the detuning of the light frequency ω from the resonant frequency ω_{mn}. The collisional shift $\Delta(v)$ of the absorption line can be on the same order of magnitude as the collisional broadening $\Gamma(v)$. For example, we would have $\Delta(v)/\Gamma(v) = \tan(\pi/\tau - 1)$ for a power-law potential[1)] $r^{-\tau}$ (Ref. 6).

Let us write the exact steady-state solution of Eqs. (1) in the limit of a high transport collision rate ($\nu_m \gg \Gamma_m$) and a weak field ($\kappa \ll 1$) for a Lorentz gas: $M \ll M_b$, $\rho \ll \rho_b$, where M and M_b are the masses of the absorbing and buffer particles, and ρ and ρ_b are their densities. Solving Eqs. (1) and (2) as in Ref. 6, we see that the spatial distribution of the density of absorbing particles reproduces the spatial distribution of the light intensity ($\propto |G|^2$) after the saturation parameter in (2):

$$\frac{\delta\rho}{\rho_0} = \frac{\int_0^\infty dv \frac{v^4}{\nu_n(v)} e^{-v^2/\bar{v}^2} \left(\frac{\nu_m(v) - \nu_n(v)}{\nu_m(v)}\right) \frac{\kappa(v)}{2}}{\int_0^\infty dv \frac{v^4}{\nu_n(v)} e^{-v^2/\bar{v}^2}}. \tag{3}$$

The constant ρ_0 represents the density of absorbing particles outside the light beam. It can be seen from (2) and (3) that the frequency dependence of the pulling signal $\delta\rho$ has the same shape as the absorption line $\Gamma/(\Gamma^2 + (\Omega - \Delta)^2)$ if the collisional broadening $\Gamma(v)$ and the collisional shift $\Delta(v)$ are independent of the velocity. In the opposite case, the shape of the pulling signal in (3) may be quite different from Lorentzian, if the relative difference $\alpha(v) = (\nu_m(v) - \nu_n(v))/\nu_m(v)$ between the transport collision rates $\nu_m(v)$ and $\nu_n(v)$ is a variable-sign function of the velocity. That $\alpha(v)$ is of variable sign is demonstrated by two fairly general models which can be solved exactly: The Lennard–Jones model and the Sutherland model.[8] Here we will reproduce the results of a study of (3) for the simpler Sutherland model. In this model the molecules are smooth, hard, elastic spheres of radius a, and are surrounded by weak attractive fields $-\varepsilon(a'/r)^\tau$. For the Sutherland model, the transport rate $\nu_i(v)$, the cross section $\sigma_i(v)$, the collisional broadening, and the shift of the absorption line have the following dependence on the dimensionless velocity $t = v/\bar{v}$:

$$\nu_i(v) = \rho_b v \sigma_i(v), \quad \sigma_i(v) = \sigma_i(1 + \xi_i/t^2), \;\; \Gamma(v) = \Gamma_0 t, \;\; \Delta(v) = 0, \tag{4}$$

where

$$\xi_i = 2\epsilon_i^* \left(\frac{a_i'}{a_i}\right)^{\tau_i} i_1(\tau_i + 1) \ll 1, \quad \sigma_i = \pi a_i^2, \quad \epsilon_i^* = \epsilon_i/k_B T.$$

Values of the function $i_1(\tau)$ are given in a monograph.[8]

The frequency dependence of the spatial distribution of the particles in (3) is described in the Sutherland model, (4), by the function φ:

$$\frac{\delta\rho}{\rho_0} = 2\left(1 - \frac{\sigma_n}{\sigma_m}\right)\frac{\kappa_0 k\bar{v}}{\Lambda\Gamma_0}\varphi, \quad \varphi = \int_0^\infty dt \frac{t^6 e^{-t^2}(t^2 - q)}{(t^2 + \xi_m)(t^2 + \xi_n)(t^2 + (\Omega/\Gamma_0)^2)}. \tag{5}$$

Here $\kappa_0 = 2|G|^2/\Gamma_m k\bar{v}$; $\Lambda = 1 - \xi_n - \xi_n^2 e^{\xi_n} Ei(-\xi_n)$; and $Ei(\xi)$ is the integral ex-

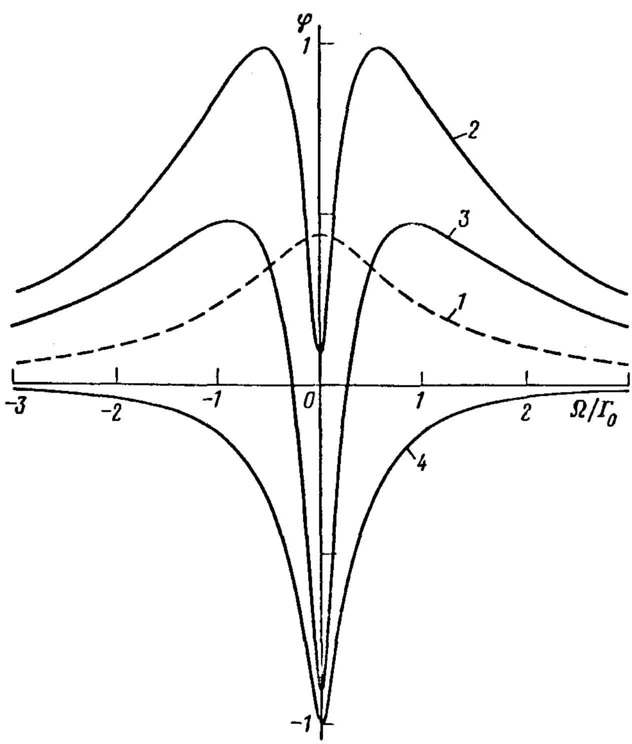

FIG. 1. The pulling signal in (5) versus the detuning Ω for various values of the parameter q, which characterizes the interaction potential ($\xi_m = 0.0002$, $\xi_n = 0.0001$). 1—φ, $q = 0$; 2—$\varphi \times 7$, $q = 1/2$; 3—$\varphi \times 6$, $q = 0.7$; 4—φ, $q = 1.7$.

ponential function. The integrand in (5), which is proportional to the relative difference between the transport collision rates, $\alpha(v)$, is a variable-sign function if

$$q \equiv \frac{\xi_m \sigma_m/\sigma_n - \xi_n}{1-\sigma_m/\sigma_n} > 0,$$

i.e., if the attractive and repulsive parts of the Sutherland potential change "in opposite directions" upon excitation. As long as the condition $q \leqslant 0$ holds (line 1 in Fig. 1), the spectrum of the pulling signal will differ only slightly from a Lorentzian shape. When q goes positive, the pulling signal in (5) begins to acquire a dip near the center of the absorption line (line 2 in Fig. 1). If q is increased further, the dip becomes deeper, and in the region $1/2 < q < 3/2$ the pulling signal becomes a variable-sign function of Ω (line 3 in Fig. 1). Finally, at $q > 3/2$, the function φ shrinks and becomes negative everywhere (line 4 in Fig. 1). The limiting values (1/2 and 3/2) of the parameter q are reached in the case $\xi_m = \xi_n = 0$.

Figure 1 demonstrates the strong dependence of the shape of the spectrum of the pulling signal on the parameter q, which corresponds to the relation between the attractive and repulsive parts of the interparticle interaction potential. The parameter q has a T^{-1} dependence on the gas temperature. The change in the shape of the frequency dependence of the pulling signal as the temperature changes means that q can be determined experimentally, so information on the interaction potential for excited particles can be obtained in more detail. One can show that spectral anomalies similar to this pulling effect occur in other, related effects,[1] in particular, light-induced drift.[5]

The results which we are reporting here are not peculiar to a Lorentzian gas ($M_b/M \gg 1$). Corresponding spectral features are found at $M_b/M \lesssim 1$. The main reason for the spectral features in the pulling signal is the velocity dependence of the collision rate, $\nu_i(v)$. It was shown in Ref. 7 that $\nu_i(v)$ remains a function of v in the case $M_b/M \lesssim 1$ and that this dependence disappears, strictly speaking, only in the case of heavy absorbing particles: $M_b/M \ll 1$.

I wish to thank A. I. Parkhomenko, S. G. Rautian, and A. M. Shalagin for valuable comments.

[1)] This circumstance was pointed out to the author by S. G. Rautian.

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Translated by D. Parsons