

Exact solution of the one- and three-dimensional quantum kinetic equations with velocity-dependent collision rates: Comparative analysis

T. Privalov* and A. Shalagin

Institute of Automation and Electrometry, Universitetskii prospekt 1, 630090 Novosibirsk, Russia

(Received 25 August 1998)

The interaction of a plane monochromatic traveling wave with two-level particles suffering collisions with buffer-gas particles is considered. Collision rates are assumed to be velocity dependent. The collision integral is obtained on the basis of the strong-collision model, generalized to the case of velocity-dependent collision rates (the so-called “kangaroo” model). We obtained the exact analytical solution of the problem for arbitrary intensity of radiation, arbitrary ratio of homogeneous and Doppler widths of the absorption line, and arbitrary mass ratio between absorbing- and buffer-gas particles. The obtained analytical solutions of the quantum kinetic equations allowed us to analyze the spectral shape of the strong-field absorption line as well as the probe-field absorption line (the nonlinear part of the work done by the probe field) and the frequency dependence of the light-induced drift (LID) velocity. A comprehensive comparative analysis for the three- and one-dimensional versions of the model is given. On the basis of this analysis, we reach the conclusion that the one-dimensional quantum kinetic equation has quite a wide range of application. We also reveal the conditions for the strongest manifestation of the velocity dependence of the collision rates, which affects most strongly the anomalous LID. [S1050-2947(99)02904-2]

PACS number(s): 42.50.Ct, 42.62.Fi, 42.50.Vk

I. INTRODUCTION

As is well known, the resonant interaction of laser radiation (plane monochromatic traveling wave) with a gas mixture gives rise to a nonequilibrium velocity distribution of the absorbing-gas particles because of the Doppler effect. Collisions with buffer-gas particles distort this nonequilibrium structure (see [1,2]). The radiation with a specified direction of propagation (say along the z axis) only creates a nonequilibrium distribution along this particular direction, i.e., the direction of the wave vector $\mathbf{k} = k\mathbf{e}_z$. The velocity distributions for the other degrees of freedom are not affected by the radiation itself, but are nonequilibrium due to collisions: The collisions transfer the nonequilibrium distribution to degrees of freedom which are orthogonal to the wave vector \mathbf{k} . The distribution functions of the absorbing-gas particles are governed by the quantum kinetic equation (Boltzmann equation), which is a three-dimensional integro-differential equation in the general case. Therefore we need the solution of the Boltzmann equations for the analysis of the interaction of laser radiation with a gas.

Let us briefly review the most popular models which are used now for the solution of the Boltzmann equations, viz., the models of strong and weak collisions, the Keilson-Storer model for the collision kernel, and the model of Lorentz gas (three- and one-dimensional variants). The kinetic equation may be only analytically solved for arbitrary intensity of resonant radiation in the framework of the strong-collision model [3] (see also [1,2,4]). The model of weak collisions may only be used analytically in the limit of small intensity of the resonant radiation with the help of an iteration method.

The kinetic equation in the framework of the Keilson-Storer kernel may only be solved numerically. All these three models assume the absence of any collisional transfer of the nonequilibrium distribution to the degrees of freedom orthogonal to \mathbf{k} (i.e., \mathbf{v}_\perp). They are also based on velocity-independent collision rates.

It is obvious now that the investigations of nonequilibrium systems inspire new efforts in the modeling of collision integrals, especially in the case when nonequilibrium conditions are created by laser radiation. It is now worthwhile to consider the influence of subtle factors on the collision integral (the behavior of the collision integral kernel). It is clear that the modern problems of nonlinear spectroscopy and gas kinetics definitely demand at least an account of the velocity dependence of the collision rates (see [5–8]). It is precisely this velocity dependence that causes the so-called anomalous light-induced drift (LID) (see [9–12], and references therein). The Lorentz gas model may be used for a qualitative investigation of the role of this velocity dependence, but only in the limit of heavy buffer gas. The Lorentz gas model allows an analytical solution of the kinetic equation, but the conditions for its applicability are seldom realized.

Therefore we evidently need to develop a new model for collision integral with velocity-dependent collision rates and dramatically broader range of application, relative to all the above mentioned models. As far as we need to model the collision integral, we must answer an old but important question. This fundamental question is about validity of the one-dimensional collision integral as a substitution for the three-dimensional one. This question was formulated earlier [1,2], but the validity of the one-dimensional formulation was based only on the qualitative arguments of weak collisional transfer of nonequilibrium distribution on the velocities \mathbf{v}_\perp orthogonal to the wave vector \mathbf{k} .

The one-dimensional kinetic equation is evidently much simpler to solve (both numerically and analytically) than the

*Author to whom correspondence should be addressed. Electronic address: privalov@iae.nsk.su

three-dimensional one. The interpretation of the solutions of the one-dimensional kinetic equation is clearer as well. In addition, the three-dimensional collisional kernel has singularity $A(\mathbf{v}, \mathbf{v}') \propto |\mathbf{v} - \mathbf{v}'|^{-1}$ for $\mathbf{v} = \mathbf{v}'$, and the one-dimensional kernel $A(v_z, v'_z)$ is finite when $v_z = v'_z$.

So it is important to know if one can use the one-dimensional collision integral with a reasonable accuracy. The aim of our paper is to make a comprehensive quantitative comparison of solutions of three- and one-dimensional kinetic equations with velocity-dependent collision rates and to define the area of applicability of the one-dimensional collision integral.

Our paper is organized as follows. We develop a model for the collision integral with velocity-dependent collision rates for the analysis of the kinetic equations in the most general conditions (Sec. II). The velocity-dependent collision rates act as parameters of our model. The kinetic equations are solved analytically (Sec. III) for arbitrary radiation intensity, arbitrary ratio of homogeneous to Doppler widths, and arbitrary mass ratio in the three- and one-dimensional variants of the model (see Secs. III A and III B, respectively). These exact analytical solutions were used for a quantitative comparison of the absorption line shape (Sec. IV), the probe-field absorption line (Sec. V), and the frequency dependence of the LID velocity (Sec. VI) for three- and one-dimensional variants of the model. Our findings are summarized in Sec. VII.

II. FORMULATION OF THE MODEL

The distribution functions $\rho_e(\mathbf{v})$ and $\rho_g(\mathbf{v})$ of two-level particles in the excited, e , and ground, g , states describe the evolution of the absorbing gas in a resonant monochromatic radiation field. They obey the following equations:

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla + \Gamma_e \right) \rho_e(\mathbf{v}) = S_e(\mathbf{v}) + NP(\mathbf{v}),$$

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla \right) \rho_g(\mathbf{v}) = S_g(\mathbf{v}) + \Gamma_e \rho_e(\mathbf{v}) - NP(\mathbf{v}), \quad (1)$$

where $N = \langle \rho_e(\mathbf{v}) + \rho_g(\mathbf{v}) \rangle$ is the total concentration of absorbing particles; $P(\mathbf{v})$ denotes the absorption rate (absorption probability per unit time) of a particle with velocity \mathbf{v} ; Γ_e is the excited state decay rate; and $S_i(\mathbf{v})$ is the integral of collisions between absorbing- and buffer-gas particles only ($i = g, e$). The concentration of the buffer gas is supposed to be much larger than the concentration of the absorbing gas, so we can neglect collisions between the absorbing-gas particles. We denote by angle brackets, $\langle \rangle$, the integration over velocity \mathbf{v} .

Neglecting the effects of phase memory in the collisions, we have the following well-known expression for $P(\mathbf{v})$:

$$P(\mathbf{v}) = \frac{2|G|^2\Gamma(v)}{\Gamma^2(v) + (\Omega - \mathbf{k}\mathbf{v})^2} \frac{1}{N} [\rho_g(\mathbf{v}) - \rho_e(\mathbf{v})], \quad G = \frac{Ed_{eg}}{2\hbar}, \quad (2)$$

where $\Omega = \omega - \omega_{eg}$ is the detuning of the laser frequency ω from the resonant frequency ω_{eg} of the transition $e \rightarrow g$; E and \mathbf{k} denote the amplitude of the electric field and wave

vector of the radiation, respectively; d_{eg} is the dipole matrix element. The velocity-dependent homogeneous linewidth, $\Gamma(v)$, in expression (2) is the sum of the spontaneous, $\Gamma_e/2$, and collision, $\gamma(v)$, widths:

$$\Gamma_e(v) = \frac{\Gamma_e}{2} + \gamma(v). \quad (3)$$

The collisional shift of the resonant frequency ω_{eg} may be included in Ω .

The general formula for the elastic collision integral reads

$$S_i(\mathbf{v}) = -\nu_i(v)\rho_i(\mathbf{v}) + \int \rho_i(\mathbf{v}')A_i(\mathbf{v}|\mathbf{v}')d\mathbf{v}', \quad i = e, g. \quad (4)$$

Here $A_i(\mathbf{v}|\mathbf{v}')$ is the collision integral kernel.

The collision integral (4) must satisfy particle conservation, $\langle S_i(\mathbf{v}) \rangle = 0$; therefore we have the known relation between the elastic collision frequency $\nu_i(v)$ and the kernel $A_i(\mathbf{v}|\mathbf{v}')$:

$$\nu_i(v) = \int A_i(\mathbf{v}'|\mathbf{v})d\mathbf{v}'. \quad (5)$$

One can say that the model of the collision integral is nothing more than the model of its kernel, $A_i(\mathbf{v}|\mathbf{v}')$.

The starting point of our model is the assumption of factorization of the kernel

$$A_i(\mathbf{v}|\mathbf{v}') = f_i^{(1)}(\mathbf{v})f_i^{(2)}(\mathbf{v}'). \quad (6)$$

According to Eqs. (5) and (6) we have

$$f_i^{(2)}(\mathbf{v}) = \frac{\nu_i(v)}{\langle f_i^{(1)}(\mathbf{v}) \rangle}. \quad (7)$$

The kernels $A_i(\mathbf{v}'|\mathbf{v})$ must satisfy the following condition [1,2]:

$$A_i(\mathbf{v}'|\mathbf{v})W(\mathbf{v}) = A_i(\mathbf{v}|\mathbf{v}')W(\mathbf{v}').$$

This condition together with assumption (6) results in the following relation between $f_i^{(1)}(\mathbf{v})$ and $f_i^{(2)}(\mathbf{v})$:

$$f_i^{(1)}(\mathbf{v}) = \frac{\langle f_i^{(1)}(\mathbf{v}) \rangle f_i^{(2)}(\mathbf{v})W(\mathbf{v})}{\langle f_i^{(2)}(\mathbf{v})W(\mathbf{v}) \rangle}. \quad (8)$$

Taking into account formulas (7) and (8), we obtained the final expression for the collision integral kernel (6):

$$A_i(\mathbf{v}|\mathbf{v}') = \frac{\nu_i(v)W(\mathbf{v})\nu_i(v')}{\langle \nu_i(v)W(\mathbf{v}) \rangle}. \quad (9)$$

The kernel (9) leads to the final expression for the collision integral (4) itself (the so-called ‘‘kangaroo’’ model [13]),

$$S_i(\mathbf{v}) = -\nu_i(v)\rho_i(\mathbf{v}) + \frac{\nu_i(v)W(\mathbf{v})}{\langle \nu_i(v)W(\mathbf{v}) \rangle} \langle \rho_i(\mathbf{v})\nu_i(v) \rangle, \quad i = e, g. \quad (10)$$

The essence of our model (10) is as follows. After each collision the velocity distribution of absorbing particles becomes isotropic, but it differs from the equilibrium Maxwell distribution $W(\mathbf{v})$ due to the velocity dependence of the collision rates, $\nu_i(v)$. In other words, after each collision every direction of velocity has equal probability, but the absolute value of the velocity may be arbitrary, due to the arbitrary velocity dependence of the collision rates. These collisions do not change electronic states of atoms, and are elastic in this sense. Our model (10) coincides with the well-known strong-collision model in the limit of velocity-independent collision rates.

Under the assumption of weak transfer of the nonequilibrium velocity distribution to the orthogonal (to \mathbf{k}) projections, \mathbf{v}_\perp , due to collisions, we may postulate (see also [1,2]) that

$$\rho_i(\mathbf{v}) = W(\mathbf{v}_\perp) \rho_i(v_z), \quad (11)$$

where the z axis is along the wave vector \mathbf{k} . Let us substitute the expression (11) in Eq. (1) and then integrate Eq. (1) over \mathbf{v}_\perp . In view of the collision integral (10), we have the formula for the one-dimensional collision integral

$$\begin{aligned} S_i(v_z) = & -\nu_i(v_z) \rho_i(v_z) \\ & + \frac{\nu_i(v_z) W(v_z)}{\langle \nu_i(v_z) W(v_z) \rangle} \langle \rho_i(v_z) \nu_i(v_z) \rangle, \quad i=e,g \end{aligned} \quad (12)$$

where the angle brackets, denote integration over v_z now; $\nu_i(v_z)$ stand for the one-dimensional collision rates of the states e, g with the following definition:

$$\nu_i(v_z) = \int A_i(v_z' | v_z) dv_z' = \int \nu_i(v) W(\mathbf{v}_\perp) d\mathbf{v}_\perp. \quad (13)$$

The one-dimensional kernel is related to the three-dimensional one by the well-known formula [1,2]

$$A_i(v_z | v_z') = \int A_i(\mathbf{v} | \mathbf{v}') W(\mathbf{v}'_\perp) d\mathbf{v}'_\perp.$$

Therefore the one-dimensional kernel has the evident form:

$$A_i(v_z | v_z') = \frac{\nu_i(v_z) W(v_z) \nu_i(v_z')}{\langle \nu_i(v_z) W(v_z) \rangle}, \quad i=e,g. \quad (14)$$

It is worth mentioning that the model (12) with kernel (14) may be obtained directly from the assumption of the factorization of the one-dimensional kernel, $A_i(v_z | v_z') = f_i^{(1)}(v_z) f_i^{(2)}(v_z')$, similar to expression (10). One needs also to use the conservation of particles and the stability condition for the one-dimensional Maxwell distribution $W(v_z)$.

Let us make one important remark at the end of this section. The statement that the collisions do not transfer the nonequilibrium distribution to the orthogonal velocities \mathbf{v}_\perp results in the fact that the integral

$$\int A_i(\mathbf{v}' | \mathbf{v}) d\mathbf{v}'$$

does not depend on \mathbf{v}_\perp . This is true only in the case of velocity-independent collision rates. The reverse is not true in general. But it is more or less evident that the transfer of the nonequilibrium distribution to the orthogonal projections of velocity and the velocity dependence of the collision rates are strongly related to each other.

III. EXACT SOLUTION OF THE KINETIC EQUATIONS

A. Three-dimensional collision integral

We are going to solve the three-dimensional kinetic equations (1) with the account of expressions (2) and (10). The distribution functions of absorbing particles, $\rho_e(\mathbf{v})$ and $\rho_g(\mathbf{v})$, depend on the absorption probability $P(\mathbf{v})$ in the following way:

$$\rho_e(\mathbf{v}) = N \left\{ P(\mathbf{v}) \tau_{1e}(v) + W(\mathbf{v}) \tau_{2e}(v) \frac{\langle P(\mathbf{v}) \tau_{2e}(v) \rangle}{\langle W(\mathbf{v}) \tau_{2e}(v) \rangle} \right\}, \quad (15)$$

$$\begin{aligned} \rho_g(\mathbf{v}) = & NW(\mathbf{v}) - N \left\{ P(\mathbf{v}) \tau_{1g}(v) \right. \\ & + W(\mathbf{v}) \tau_{2g}(v) \frac{\langle P(\mathbf{v}) \tau_{2e}(v) \rangle}{\langle W(\mathbf{v}) \tau_{2e}(v) \rangle} \left. \right\} \\ & + \frac{NW(\mathbf{v})}{\langle W(\mathbf{v}) \tau_{2e}(v) \rangle} [\langle P(\mathbf{v}) \tau_{2e}(v) \rangle \langle \tau_{2g}(v) W(\mathbf{v}) \rangle \\ & - \langle P(\mathbf{v}) \tau_{2g}(v) \rangle \langle \tau_{2e}(v) W(\mathbf{v}) \rangle], \end{aligned}$$

where

$$\tau_{1e}(v) = \frac{1}{\Gamma_e + \nu_e(v)}, \quad \tau_{2e}(v) = \frac{1}{\Gamma_e} - \tau_{1e}(v),$$

$$\tau_{1g}(v) = \frac{1}{\Gamma_e + \nu_g(v)} \frac{\nu_e(v)}{\nu_g(v)}, \quad \tau_{2g}(v) = \frac{1}{\Gamma_e} - \tau_{1g}(v). \quad (16)$$

The velocity distribution of excited particles, $\rho_e(\mathbf{v})$, consists of two qualitatively different terms. The first term is proportional to the absorption rate $P(\mathbf{v})$. This is the velocity-selective distribution created by laser radiation and not affected by collisions. The second term in the expression for $\rho_e(\mathbf{v})$ is velocity isotropic. But it deviates from the equilibrium distribution in general. This deviation is only based on the velocity dependence of the collision rates.

The two terms in curly brackets of the expression for $\rho_g(\mathbf{v})$ have the same meaning as the corresponding terms in the expression for $\rho_e(\mathbf{v})$. The third term in $\rho_g(\mathbf{v})$ is the equilibrium Maxwellian distribution. This distribution exists only due to the infinite lifetime of the ground state g ; it is nonzero only if $\nu_g(v) \neq \nu_e(v)$.

The factors (16) are velocity dependent and have a clear physical meaning. The value of $\tau_{1e}(v)$ stands for the time interval between excitation and the first collision with a change in velocity. The sum of $\tau_{2e}(v)$ and $\tau_{1e}(v)$ is the total lifetime $1/\Gamma_e$ of the excited state. Therefore $\tau_{2e}(v)$ is the time complementing $\tau_{1e}(v)$ up to the total lifetime. The

meaning of other quantities, $\tau_{1g}(v)$ and $\tau_{2g}(v)$, is the same as for the factors $\tau_{1e}(v)$ and $\tau_{2e}(v)$.

In the limit of weak radiation intensity one has $\rho_g(\mathbf{v}) \gg \rho_e(\mathbf{v})$ and $\rho_g(\mathbf{v}) \rightarrow NW(\mathbf{v})$, so that $\rho_g(\mathbf{v}) - \rho_e(\mathbf{v}) = NW(\mathbf{v})$. Therefore the absorption rate $P(\mathbf{v})$ is defined beforehand. Consequently in the limit of weak radiation intensity expressions (15) are the final solution of the problem. Of course in the general case (for high radiation intensity, for example) one needs to find the absorption rate $P(\mathbf{v})$ with the help of expressions (16) and (2). The solution of the corresponding integral equation is the following:

$$P(\mathbf{v}) = \frac{f(\mathbf{v})W(\mathbf{v})}{B(\Omega)} [\langle \tau_{2e}(v)W(\mathbf{v}) \rangle + \tau_1(v)\langle f(\mathbf{v})W(\mathbf{v})\tau_{2e}(v) \rangle - \langle f(\mathbf{v})W(\mathbf{v})\tau_1(v)\tau_{2e}(v) \rangle]. \quad (17)$$

The velocity-independent function $B(\Omega)$ in Eq. (17) depends on the detuning Ω as follows:

$$B(\Omega) = \langle \tau_{2e}(v)W(\mathbf{v}) \rangle [1 + \langle \tau_2(v)f(\mathbf{v})W(\mathbf{v}) \rangle + \langle \tau_{2e}(v)f(\mathbf{v})W(\mathbf{v}) \rangle [\langle \tau_1(v)W(\mathbf{v}) \rangle - \tau_1(v) + \langle \tau_2(v)\tau_1(v)f(\mathbf{v})W(\mathbf{v}) \rangle - \tau_1(v) \times \langle \tau_2(v)f(\mathbf{v})W(\mathbf{v}) \rangle], \quad (18)$$

where

$$f(\mathbf{v}) = \frac{1}{\tau_1(v)} \frac{\Gamma^2(v)\kappa(v)}{\Gamma^2(v)[1 + \kappa(v)] + (\Omega - \mathbf{k}\mathbf{v})^2},$$

where $\kappa(v)$ is the so-called saturation parameter

$$\kappa(v) = \frac{2|G|^2}{\Gamma(v)} \tau_1(v) = \kappa_0 \frac{\Gamma_e^2 \tau_1(v)}{\Gamma(v)}, \quad \kappa_0 = \frac{4|G|^2}{\Gamma_e^2}.$$

We introduced in Eqs. (17) and (18) the following time scales:

$$\tau_1(v) = \tau_{1e}(v) + \tau_{1g}(v) = \frac{1}{\Gamma_e + \nu_e(v)} \left[1 + \frac{\nu_e(v)}{\nu_g(v)} \right],$$

$$\tau_2(v) = \tau_{2e}(v) + \tau_{2g}(v) = \frac{2}{\Gamma_e} - \tau_1(v). \quad (19)$$

These times, $\tau_1(v)$ and $\tau_2(v)$, keep their traditional meaning (see [2–4], for example), with the velocity dependence as the only difference. For example, the factor $\tau_1(v)$ is the effective time interval for an atom being in interaction with the radiation field from the instant of excitation to a collision with velocity change, regardless of the atomic state.

After integration of expression (17) over velocities \mathbf{v} of absorbing particles we have the following formula for the absorption probability per unit time for one atom:

$$P \equiv \langle P(\mathbf{v}) \rangle = \frac{1}{B(\Omega)} [\langle f(\mathbf{v})W(\mathbf{v}) \rangle \langle \tau_{2e}(v)W(\mathbf{v}) \rangle + \langle f(\mathbf{v})W(\mathbf{v})\tau_{2e}(v) \rangle \{ \langle f(\mathbf{v})W(\mathbf{v})\tau_1(v) \rangle - \tau_1(v) \} \times \langle f(\mathbf{v})W(\mathbf{v}) \rangle]. \quad (20)$$

Consider the important case of equal collision rates

$$\nu_g(v) = \nu_e(v). \quad (21)$$

It follows from the condition (21), with the account of Eq. (16), that

$$\tau_{1e} = \tau_{1g} = \tau_1/2, \quad \tau_{2e} = \tau_{2g} = \tau_2/2.$$

The expression for the absorption rate $P(\mathbf{v})$ is

$$P(\mathbf{v}) = \frac{f(\mathbf{v})W(\mathbf{v})}{\tilde{B}(\Omega)} [\langle \tau_2(v)W(\mathbf{v}) \rangle + \tau_1(v)\langle f(\mathbf{v})W(\mathbf{v})\tau_2(v) \rangle - \langle f(\mathbf{v})W(\mathbf{v})\tau_1(v)\tau_2(v) \rangle], \quad (22)$$

and

$$P \equiv \langle P(\mathbf{v}) \rangle = \frac{1}{\tilde{B}(\Omega)} [\langle f(\mathbf{v})W(\mathbf{v}) \rangle \langle \tau_2(v)W(\mathbf{v}) \rangle + \langle f(\mathbf{v})W(\mathbf{v})\tau_2(v) \rangle \{ \langle f(\mathbf{v})W(\mathbf{v})\tau_1(v) \rangle - \tau_1(v)\langle f(\mathbf{v})W(\mathbf{v}) \rangle \}]. \quad (23)$$

The expression for $B(\Omega)$ is now considerably simplified:

$$\tilde{B}(\Omega) = 2B(\Omega) = \langle W(\mathbf{v})\tau_2(v) \rangle - \langle f(\mathbf{v})W(\mathbf{v})\tau_2^2(v) \rangle. \quad (24)$$

The velocity dependence of the collision rates, $\nu_g(v)$ and $\nu_e(v)$, introduces a significant complication to the velocity distribution (15) and to the final expressions for the absorption rate (17), (20), (22), and (23) relative to the results of the model with velocity-independent collision rates. All our results coincide naturally with the previous results for the strong-collision model (see [3,4]) if we neglect the velocity dependence of the collision rates in our formulas.

B. One-dimensional collision integral

One may use the same approach in solving the kinetic equations with the one-dimensional collision integral. Therefore we may use formulas (20), (17), and (15), with the evident permutation $f(\mathbf{v}) \rightarrow f(v_z)$, $W(\mathbf{v}) \rightarrow W(v_z)$, $\nu_j(v) \rightarrow \nu_j(v_z)$.

IV. ABSORPTION LINE SHAPE

We are now ready for a comparison of the absorption line shape in the three- and one-dimensional variants of the model. Our aim is to extract the influence of the velocity dependence of the collision rates on this shape under different conditions. The exact solutions of the preceding section are a nice groundwork for this comparison.

The integration over velocities is supposed to be performed numerically in all expressions. Therefore we need to specify the velocity dependence of the collision rates.

The parameters of our model, $\nu_j(v)$, describe the collisions with loss of the directed velocity. Therefore it is obvious to identify these parameters, $\nu_j(v)$, with the transport collision rates.

The transport collision rates are related to the transport cross section of collisions as follows (see [2]):

$$v_j(v) = \frac{8\mu N_b}{\sqrt{\pi} M \bar{v}_b^5} \int_0^\infty du u^5 \sigma_{tr,j}(u) \exp\left(-\frac{v^2+u^2}{\bar{v}_b^2}\right) \times \left[\frac{q \cosh(q) - \sinh(q)}{q^3} \right], \quad (25)$$

here μ stands for the reduced mass; $q = 2vu/\bar{v}_b^2$. We restrict ourselves to the case of power potentials of the following form: $U(r) \propto r^{-n}$. The transport collision cross section (see [2]) for the classical trajectories reads

$$\sigma_{tr,j}(u) = \sigma_{tr,j}(\bar{v}_b) [u/\bar{v}_b]^{-4/n}, \quad (26)$$

where u is the relative velocity of the colliding particles and \bar{v}_b is the most probable velocity of buffer particles. The buffer-gas particles are supposed to be in equilibrium with the same temperature as the absorbing particles.

Expressions (25) and (26) give the following expression for the three-dimensional collision rates (see also [2]):

$$v_j^{(3)}(v) = v_j(0) \Phi(2/n - 1/2, 5/2; -(u/\bar{v}_b)^2), \quad (27)$$

where

$$v_j(0) = \mu/m N_b \bar{v}_b \sigma_{tr,j}(\bar{v}_b) \frac{4}{3\sqrt{\pi}} \Gamma(3 - 2/n),$$

$\Phi(\alpha, \gamma; y)$ is the confluent hypergeometric function.

The expression for the one-dimensional collision rate is evident from Eqs. (13) and (27):

$$v_j^{(1)}(v_z) = \frac{v_j(0)}{(1+\beta)^\alpha} \frac{\Gamma(\gamma)}{\Gamma(\alpha)} \sum_{k=0}^{\infty} \frac{(-1)^k \Gamma(\alpha+k)}{k! \Gamma(\gamma+k)} \left(\frac{\beta}{1+\beta} \right)^k \times {}_2F_1\left(\alpha+k, \gamma-1+k, \gamma+k; \frac{\beta}{1+\beta}\right) \left(\frac{v_z}{v}\right)^{2k}, \quad (28)$$

the quantity $v_j(0)$ was defined above, and $F(\dots)$ is the hypergeometric function.

We used the notation m and M_b for the masses of absorbing and buffer particles; N_b is the concentration of the buffer particles; $\Gamma(x)$ is the Γ function; $\alpha = 2/n - 1/2$, $\gamma = 5/2$, and $\beta = M_b/m$.

The collision rates for different states, $v_g(v)$ and $v_e(v)$, are not equal in general. This difference is not so important for the analysis of the absorption line shape, therefore we can neglect it. Consider now the case of state-independent parameter n and $v_g(0) = v_e(0)$. This case corresponds to Eqs. (23) and (24). The role of the velocity dependence of the collision rates is larger for larger values of the ratio $v_j(v)/\Gamma_e$. The obvious limiting case is the infinite value of this ratio: $\zeta = v_g(0)/\Gamma_e \gg 1$.

The velocity dependence of the collision rates is the most important factor in the case $\beta \rightarrow \infty$ and $n \rightarrow \infty$, as one can see from Eqs. (28) and (27). The absorption line shape is shown in Fig. 1(a) for the three- and one-dimensional variants of the model (curves 1 and 2, respectively). We added reference calculations with velocity-independent collision rates, equal

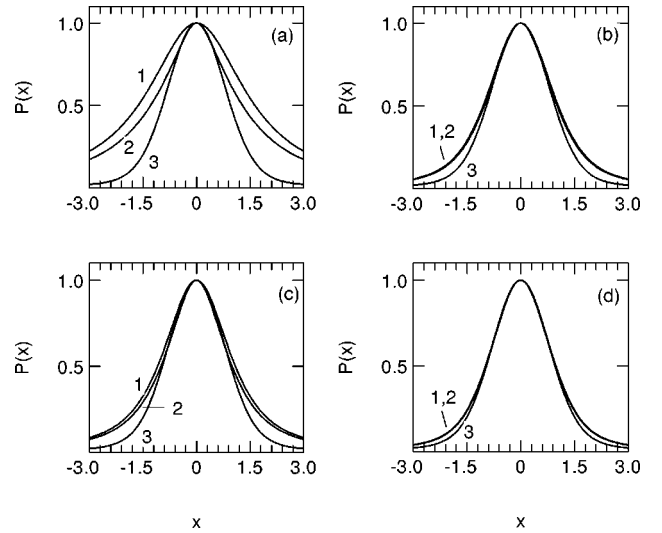


FIG. 1. Absorption rate (in arbitrary units) vs dimensionless detuning, $x = \Omega/(k\bar{v})$. (a) $\beta \rightarrow \infty, n \rightarrow \infty$; (b) $\beta \rightarrow \infty, n = 6$; (c) $\beta = 19, n \rightarrow \infty$; (d) $\beta = 19, n = 6$; Curves 1 and 2 correspond to the one- and three-dimensional approach, curve 3 to the model with velocity-independent collision rates. The values of all other parameters are defined in the text.

to $v^{(3)}(0)$. The following parameters were used: $m = 7$ a.u., $T = 300$ K, $\zeta = 4.0, \kappa_0 = 1.0, \Gamma/k\bar{v} = 0.2$. Calculations with $\beta \rightarrow \infty, n = 6$ and $\beta = 19, n \rightarrow \infty$, are shown in Figs. 1(b) and 1(c), respectively. All other parameters are the same as in Fig. 1(a). The influence of the velocity dependence of the collision rates is small now, and the difference between the one- and three-dimensional models is even much smaller. The calculations for a realistic mixture (the atomic vapors of Li in Xe buffer gas with $\beta = 19$ and $n = 6$) are plotted in Fig. 1(d). The result is that the influence of the velocity dependence of the collision rates is very small. The curves for one- and three-dimensional models are hardly distinguishable.

This result may be formulated in the following way. For realistic parameters of interaction potential and mass ratio the influence of the velocity dependence of the collision rates on the spectral shape is noticeable, but very small. Under the same conditions, the one- and three-dimensional models hardly differ from one another. This is a clear indication that the collisional transfer of the nonequilibrium distribution on \mathbf{v}_\perp is weak and the one-dimensional kinetic equation is valid [see Fig. 1(d)].

Let us focus now on the shape of the nonlinear resonance in the absorption profile of the probe field. This phenomenon is known to be sensitive to the velocity dependence of the collision rates.

V. NONLINEAR PART OF WORK DONE BY A PROBE FIELD

The influence of the velocity dependence of the collision rates on the absorption line of a probe field (namely, the nonlinear part of the work done by the probe field) is now easy to detect in an experiment. Therefore it is important to know exactly the discrepancy between the calculations of the probe-field absorption in the one- and three-dimensional models. We are going to analyze this question in the frame-

work of the ‘‘kangaroo’’ model and calculate the nonlinear part of the work done by the probe field.

We consider the case where the probe field (with wave vector \mathbf{k}_μ) and the strong field (with wave vector \mathbf{k}) are resonant with one and the same transition $g \rightarrow e$ of two-level particles: $\Omega_\mu = \omega_\mu - \omega_{eg}$ and $\Omega = \omega - \omega_{eg}$ are the detunings of the probe- and strong-field frequencies ω_μ and ω from the resonant frequency ω_{eg} , respectively. The \mathbf{k}_μ and \mathbf{k} are assumed to be antiparallel ($\mathbf{k}_\mu = -\mathbf{k}$). The first nonlinear terms in the expression for the work done by the probe field are the solutions of this problem in the limit of large Doppler broadening. This is the case when the nonlinear part of the probe-field absorption line is caused only by active level population change under the interaction with the strong laser field. The corresponding expression for the absorption line of the probe field is the following (see also [1,2]):

$$P_\mu(\Omega_\mu) \propto |G_\mu|^2 \left\langle \left[\rho_g(\mathbf{v}) - \rho_e(\mathbf{v}) \right] \frac{\Gamma(v)}{\Gamma^2(v) + (\Omega_\mu - \mathbf{k}\mathbf{v})^2} \right\rangle. \quad (29)$$

Using Eqs. (29) and (15) one gets the following expression for the probe-field absorption line:

$$\begin{aligned} P_\mu(\Omega_\mu) \propto & |G_\mu|^2 \langle W(\mathbf{v}) f(\mathbf{v}, \Omega_\mu) \rangle \\ & - 2|G_0|^2 |G_\mu|^2 \left\langle \langle W(\mathbf{v}) f(\mathbf{v}, \Omega_\mu) f(\mathbf{v}, \Omega) \tau_1(v) \rangle \right. \\ & + \left\langle W(\mathbf{v}) f(\mathbf{v}, \Omega_\mu) \left[\langle \tau_2(v) f(\mathbf{v}, \Omega) W(\mathbf{v}) \rangle \right. \right. \\ & \left. \left. - \frac{\langle \tau_{2e}(v) f(\mathbf{v}, \Omega) W(\mathbf{v}) \rangle}{\langle \tau_{2e}(v) W(\mathbf{v}) \rangle} \right] \right\rangle \\ & \left. \times \left[\tau_1(v) - \langle \tau_1(v) W(\mathbf{v}) \rangle \right] \right\rangle. \quad (30) \end{aligned}$$

In first order approximation

$$f(\mathbf{v}, \Omega) = \frac{\Gamma(v)}{\Gamma^2(v) + (\Omega - \mathbf{k}\mathbf{v})^2}.$$

Consider expression (30) for $P(\Omega_\mu)$ as a function of Ω_μ . The first term in the curly brackets of expression (30) describes the nonlinear resonance, the second term stands for the homogeneous saturation band.

It is not possible to analytically evaluate the integrals in expression (30) in the three-dimensional model. The simplest expression for the probe-field absorption line in this three-dimensional case is the following:

$$\begin{aligned} P_\mu^{(3)}(x_\mu) = & \frac{\sqrt{\pi} |G_\mu|^2}{k\bar{v}} \left\{ e^{-x_\mu^2} \left[1 - \frac{2\sqrt{\pi} |G_0|^2}{k\bar{v}} \right. \right. \\ & \times \int_{|x|}^{\infty} e^{-t^2} \frac{t}{2} \left(\tau_2^{(3)}(t) - \tau_{2e}^{(3)}(t) \right. \\ & \left. \left. \times \frac{\tau_1^{(3)}(t) - \langle \tau_1^{(3)}(t) W(\mathbf{v}) \rangle}{\langle \tau_{2e}^{(3)}(t) W(\mathbf{v}) \rangle} \right) dt \right] \\ & \left. - \frac{2|G_0|^2}{k\bar{v}} \int_{|x_\mu|}^{\infty} e^{-t^2} \frac{t}{2} \frac{y(t) \tau_1^{(3)}(t)}{y^2(t) + (x + x_\mu)^2} dt \right\}. \quad (31) \end{aligned}$$

The superscript ‘‘(3)’’ denotes the three-dimensional time factors (17) and (19); $x_\mu = \Omega_\mu / (k\bar{v})$; $t = v/\bar{v}$; all other notations are defined above.

The one-dimensional version of Eq. (30) may be obtained by the trivial permutation $f(\mathbf{v}, \Omega) \rightarrow f(v_z, \Omega)$, $W(\mathbf{v}) \rightarrow W(v_z)$, $v_j(v) \rightarrow v_j(v_z)$ (see also Secs. IV and III B). It is remarkable that in the one-dimensional case it is possible to evaluate the integrals analytically. We have the following rather simple expression in the one-dimensional model [compare with Eq. (30)] without integrals:

$$\begin{aligned} P_\mu^{(1)}(x_\mu) = & \frac{\sqrt{\pi} |G_\mu|^2}{k\bar{v}} e^{-x_\mu^2} \left\{ 1 - \frac{2\sqrt{\pi} |G_0|^2}{k\bar{v}} \right. \\ & \times \left[e^{-x^2} \left(\tau_2^{(1)}(x) - \tau_{2e}^{(1)}(x) \right. \right. \\ & \left. \left. \times \frac{\tau_1^{(1)}(x_\mu) - \langle \tau_1^{(1)}(v_z) W(v_z) \rangle}{\langle \tau_{2e}^{(1)}(v_z) W(v_z) \rangle} \right) \right. \\ & \left. \left. - \frac{1}{\sqrt{\pi}} \frac{y(x) \tau_1^{(1)}(x)}{y^2(x) + (x + x_\mu)^2} \right] \right\}. \quad (32) \end{aligned}$$

A comparison between Eqs. (32) and (31) clearly reveals the advantage of the one-dimensional version of the model. Expressions (32) and (31) do coincide in the case of velocity-independent collision rates.

We again need to use the velocity-dependent collision rates (27) and (28) for the numerical comparison of expressions (32) and (31) and the analysis of the behavior of the nonlinear resonance.

The results of the calculations are plotted in Fig. 2 in the limiting case of the strongest velocity dependence of the collision rates $\beta \rightarrow \infty$ and $n \rightarrow \infty$; solid lines correspond to three-dimensional and dotted lines correspond to one-dimensional models.

The homogeneous saturation band appears to be only weakly sensitive to the model of the collision integral (one or three dimensional). The nonlinear resonance may be considered to be quite sensitive to the type of model. The deviation may be up to 50% of the absolute value. The more realistic parameters, $n=6$ and $\beta=19$, are used for the calculations presented in Fig. 3. This case reveals a small discrepancy

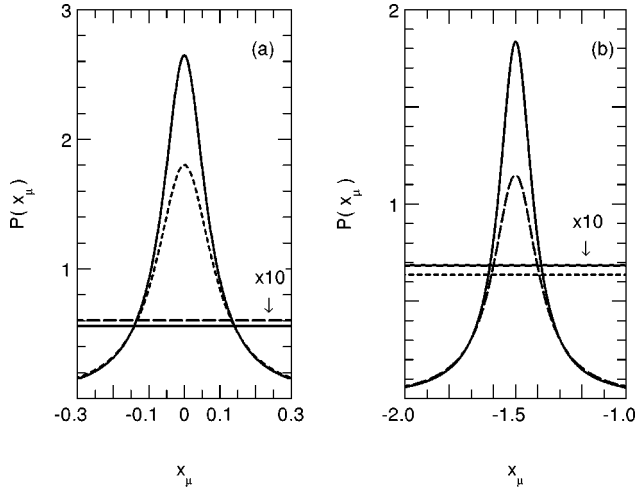


FIG. 2. The nonlinear part of the probe-field absorption rate (in arbitrary units) vs dimensionless detuning, $x_\mu = \Omega_\mu / (k\bar{v})$, of the probe field. $\beta \rightarrow \infty, n \rightarrow \infty, \zeta = 1.0, \kappa_0 = 1.0$; (a) $x = 0$; (b) $x = 1.5$. $x = \Omega / (k\bar{v})$ is the dimensionless detuning of the strong field. The amplitude of the homogeneous saturation band is increased by a factor of 10. The values of all other parameters are defined in the text.

between the three- and one-dimensional models (less than 10% of the absolute value). This discrepancy tends to decrease with decreasing parameter β .

So we see that the nonlinear resonance in the probe-field absorption line is much more sensitive to the velocity dependence of the collision rates than the absorption line shape of only one laser field. Consequently, the nonlinear resonance is much more sensitive to the type of model (three- or one-dimensional collision integral). Nevertheless, one can use the one-dimensional collision integral with good accuracy even in such problems.

VI. LIGHT-INDUCED DRIFT

The aim of this section is to evaluate the light-induced drift velocity for the three- and one-dimensional versions of

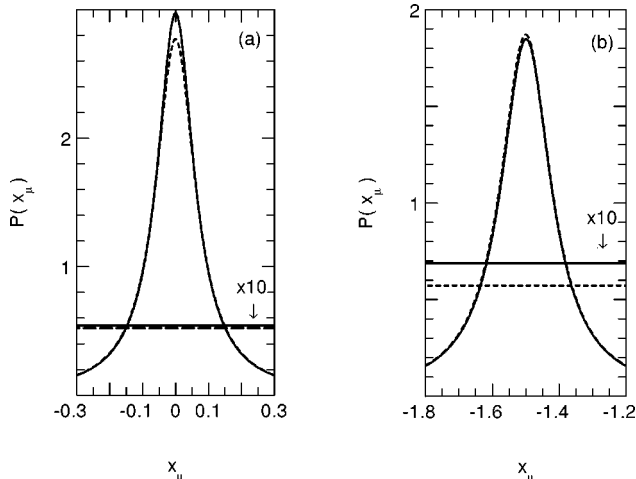


FIG. 3. The same as in Fig. 2, but $\beta = 19, n = 6$. All other parameters are the same. Arbitrary units are used, the notation is the same as in Fig. 2.

our model. Let us briefly recall the essence of the LID effect [14,15,2] (see also reviews [16,17]). LID arises when atoms or molecules, mixed with some buffer gas, are excited by radiation. The bases of the phenomena are the velocity-selective interaction of the laser radiation with the absorbing particles (due to the Doppler effect) and the change of transport collision properties of the absorbing particles because of the excitation (state-dependent friction by the buffer gas).

The LID velocity is given by the expression

$$\mathbf{u} = \frac{1}{N} \langle \mathbf{v} \{ \rho_g(\mathbf{v}) + \rho_e(\mathbf{v}) \} \rangle. \quad (33)$$

The distribution functions (15) and expression (33) give the following formula for the drift velocity:

$$\mathbf{u} = \int \frac{\nu_g(v) - \nu_e(v)}{\nu_g(v) [\Gamma_e + \nu_e(v)]} \mathbf{v} P(\mathbf{v}) d\mathbf{v}, \quad (34)$$

the absorption rate, $P(\mathbf{v})$, Eq. (17), was discussed in Sec. IV. The one-dimensional variant of Eq. (34) is quite obvious:

$$u = \int \frac{\nu_g(v_z) - \nu_e(v_z)}{\nu_g(v_z) [\Gamma_e + \nu_e(v_z)]} v_z P(v_z) dv_z. \quad (35)$$

Expressions (34) and (35) coincide in the case of velocity-independent collision rates, as one may expect. These formulas show that the difference of the collision rates of different states is absolutely essential for the existence of LID. Therefore we consider now the values of the parameters $\nu_j(0)$ and n in the transport collision rates as state dependent with the evident notation: $\nu_e(0)$ and $\nu_g(0), n_e$ and n_g . Now it is also convenient to introduce the parameter $\Delta = \nu_e(0) / \nu_g(0)$.

A. Traditional LID effect

Let us consider the frequency dependence of the LID velocity (34) and (35) in the case of normal LID, when the values of the transport collision rates of two different states are significantly different. The relative difference of the transport collision rates in Eqs. (34) and (35) is very weak, with small influence on the frequency dependence of corresponding integrals. Therefore the frequency dependence of the drift velocity is described by a dispersionlike curve (the first frequency derivative of the absorption line shape). It is obvious to expect the maximum difference between the three- and one-dimensional models in the case of the strongest velocity dependence of the collision rates, when $n_g, n_e \rightarrow \infty$. We simulated the frequency dependence of LID velocity (34) and (35) for parameters $n_g = n_e \rightarrow \infty, \beta = 19, \Delta = 1.2, \zeta = 4.0, \kappa_0 = 1.0$. The results are shown in Fig. 4(a) (in arbitrary units). The deviation of the ‘‘three-dimensional’’ formula (34) from that of the ‘‘one-dimensional’’ formula (35) (curves 1 and 2, respectively) is much smaller than the difference of these results from the result obtained in the model with velocity-independent collision rates. The influ-

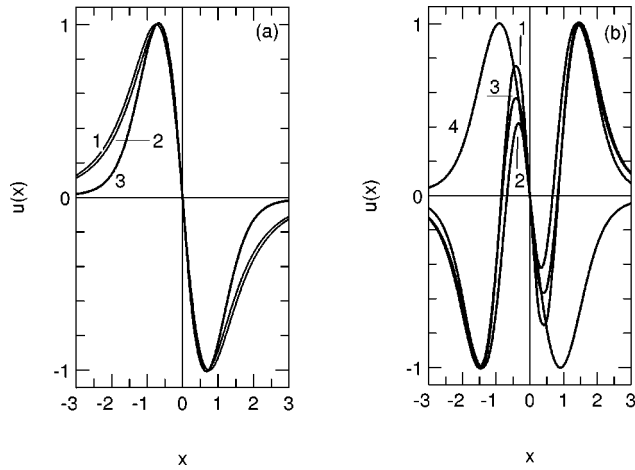


FIG. 4. (a) The frequency dependence of the drift velocity of “normal” LID (in arbitrary units) vs dimensionless detuning, $x = \Omega/(k\bar{v})$. Curves 1 and 2 correspond to the one- and three-dimensional approaches; curve 3 is the model with velocity-independent collision rates. The values of all parameters are defined in the text. (b) The frequency dependence of the drift velocity of anomalous LID (in arbitrary units) vs dimensionless detuning, $x = \Omega/(k\bar{v})$. Curves 1 and 2 correspond to the one- and three-dimensional approach, curve 3 to the calculation taking into account velocity dependence of the collision rates only in the factor $\Delta\nu$, curve 4 to the model with velocity-independent collision rates. The values of all parameters are defined in the text.

ence of the velocity dependence of the collision rates on the LID velocity is small, as one can expect under the above mentioned conditions. The difference between the curves 1 and 2 decreases with the decrease of the parameters n_g and n_e . This difference is hardly noticeable on the plot for the values $\beta < 19$ and $n_{g,e} < 10$.

B. The anomalous LID

The so-called anomalous LID (see [9–12], and references therein) is an exceptional effect, because it is completely caused by the velocity dependence of the collision rates. Indeed, the transport collision rates may have close values but different velocity dependence. The difference in transport collision rate, $\Delta\nu$, in Eqs. (34) and (35) may change sign as a function of velocity near the most probable velocity \bar{v} . Therefore particles with positive and negative values of $\Delta\nu$ make comparable contributions to Eq. (34) or Eq. (35). This is the cause of the dramatic deviation of the frequency dependence of LID velocity, $u(x)$, from the dispersionlike curve: The dispersionlike curve has only one zero for $x=0$ [see Fig. 4(a)], whereas the anomalous LID velocity $u(x)$ has additional zeros as a function of the detuning x .

The anomalous LID velocity is caused by the factor $\Delta\nu$. Therefore account of the velocity dependence of the collision rates in this factor only for the calculation of expressions (34) and (35) must correctly describe the anomalous LID with good accuracy. It is remarkable that expressions (34) and (35) coincide due to the definition (13). Taking the velocity dependence in all factors into account may only produce small corrections to this approximate shape of the spectral dependence of LID velocity. The importance of this correction increases with increasing velocity dependence of

the collision rates and the role of collisions. Our numerical calculations [see Fig. 4(b)] prove this conclusion. We used the following values of parameters: $\beta = 19, \zeta = 4.0, \kappa_0 = 150, n_e = 6, n_g = 7$. Curve 3 in Fig. 4(b) corresponds to the calculation with account of velocity dependence of the collision rates only in the factor $\Delta\nu$ in Eq. (34) or Eq. (35). Curves 2 and 1 correspond to the calculations according to Eqs. (34) and (35) with complete account of velocity dependence of the collision rates. The results of the calculation in the frame of velocity-independent collision rates are plotted as curve 4. The comparison of curves 1, 2, 3 with curve 4 (see Fig. 4) clearly reveals the manifestation of anomalous LID. We may also conclude that the account of velocity dependence of the collision rates only in the factor $\Delta\nu$ describes the anomalous LID with good accuracy. The correction caused by the type of model (three or one dimensional) is small (less than 10% for $\beta < 6$).

So, there is a range of parameters where the one-dimensional collision integral is valid even for the description of anomalous LID. Moreover, it is possible to correctly describe the anomalous LID without taking into account the velocity dependence of the collision rates in the absorption rate $P(\mathbf{v})$, but taking this dependence into account in the difference of the collision rates $\Delta\nu$ only (see also [17]).

VII. CONCLUSION

The model of collision integral (10) is a generalization of the strong-collision model in the case of velocity-dependent collision rates. The analytical solution of the three-dimensional quantum kinetic equation is possible for an arbitrary system of levels of absorbing particles and arbitrary parameters in the problem. We have found this solution for two-level particles. The exact analytical solution enables us to analyze the absorption probability of monochromatic radiation, the nonlinear part of the work done by the weak probe field, and the light-induced drift velocity.

One of the advantages of the model is that it allows us to analyze in detail the validity of the one-dimensional kinetic equation. The comparison of the three- and one-dimensional versions of the present model, with the help of calculations of the absorption rate, probe-field absorption, and LID velocity revealed a remarkable “hierarchy.” The derivation caused by the velocity dependence of the collision rates is much larger than the difference between the three- and one-dimensional solutions.

Our approach is not supposed to be universal and completely general. However, the “kangaroo” model opens the possibility of quantitatively justifying the idea of small collisional transfer of the nonequilibrium distribution on the orthogonal degrees of freedom and proves the validity of the one-dimensional kinetic equation.

ACKNOWLEDGMENTS

We would like to thank Professor S. Rautian, Professor F. Gel'mukhanov, and Dr. L. Il'ichov for fruitful discussions. The work was supported by the Russian Foundation for Fundamental Research (Grant No. 98-02-17924) and the program “Laser Physics” (Grant No. 7.41).

- [1] S.G. Rautian, G.I. Smirnov, and A.M. Shalagin, *Nonlinear Resonances in the Spectra of Atoms and Molecules (Nelineinie rezonansi v spektrakh atomov i molekul)* (Nauka, Novosibirsk, 1979).
- [2] S.G. Rautian and A.M. Shalagin, *Kinetic Problems of Non-Linear Spectroscopy* (North-Holland, Amsterdam, 1991).
- [3] A.P. Kolchenko and S.G. Rautian, Zh. Éksp. Teor. Fiz. **54**, 959 (1968) [Sov. Phys. JETP **27**, 511 (1968)].
- [4] V.R. Mironenko and A.M. Shalagin, Izv. Akad. Nauk SSSR, Ser. Fiz. **45**, 995 (1981) [Bull. Acad. Sci. USSR, Phys. Ser. **45**, 87 (1981)].
- [5] R. Berman *et al.*, in *Spectral Line Shapes*, Volume 9, at the 13th ICSLS, Firenze, Italy, edited by Marco Zoppi and Lorenzo Ulivi, AIP Conf. Proc. No. 386 (AIP, New York, 1996), p. 331.
- [6] R. Berman, P.M. Sinclair, J.R. Drummond, and A.D. May, in *14th International Conference on Spectral Line Shapes (ICSLS XIV)*, State College, Pennsylvania, June 22–26, 1998 (book of abstracts) (Pennsylvania State University, 1998), p. 105.
- [7] R. Ciurulo and J. Szudy, in *14th International Conference on Spectral Line Shapes (ICSLS XIV)*, State College, Pennsylvania, June 22–26, 1998 (book of abstracts) (Pennsylvania State University, 1998), p. 75.
- [8] A. Bielski *et al.*, in *14th International Conference on Spectral Line Shapes (ICSLS XIV)*, State College, Pennsylvania, June 22–26, 1998 (book of abstracts) (Pennsylvania State University, 1998), p. 94.
- [9] G.J. van der Meer, J. Smeets, S.P. Pod'yachev, and L.J.F. Hermans, Phys. Rev. A **45**, R1303 (1992).
- [10] I. Kuščer, L.J.F. Hermans, P.L. Chapovsky, J.J.M. Beenakker, and G.J. van der Meer, J. Phys. B **26**, 2837 (1993).
- [11] P.L. Chapovsky, G.J. van der Meer, J. Smeets, and L.J.F. Hermans, Phys. Rev. A **45**, 8011 (1992).
- [12] F.Kh. Gel'mukhanov, A.I. Parkhomenko, T.I. Privalov, and A.M. Shalagin, J. Phys. B **30**, 1819 (1997).
- [13] A. Brissaud and U. Frisch, J. Math. Phys. **15**, 5 (1974); **15**, 524 (1974).
- [14] F.Kh. Gel'mukhanov and A.M. Shalagin, Pis'ma Zh. Éksp. Teor. Fiz. **29**, 773 (1979) [JETP Lett. **29**, 711 (1979)].
- [15] F.Kh. Gel'mukhanov and A.M. Shalagin, Zh. Éksp. Teor. Fiz. **78**, 1672 (1980) [Sov. Phys. JETP **51**, 839 (1980)].
- [16] E.R. Eliel, Adv. At., Mol., Opt. Phys. **30**, 199 (1993).
- [17] L.J.F. Hermans, Int. Rev. Phys. Chem. **11**, 289 (1992).