Eigenvalues of collision operators: Properties and methods of computation

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The linear Boltzmann equation for active atoms submerged in the much denser perturber gas contains a collision rate and a kernel. These two quantities are combined into a single entity—the collision operator. The collision operator possesses several interesting properties, the most important being that it is Hermitian. The eigenvalues are negative with the exception of one eigenvalue, which is zero and corresponds to the Maxwellian (steady-state) velocity distribution. A set of functions, closely related to the eigenfunctions of the quantum-mechanical harmonic oscillator, is postulated to approximate the true eigenfunctions. This assumption was a basis of the method of modeling various physical phenomena occurring in the gaseous mixtures, subjected to a radiation field. The eigenvalues of the collision operator were treated as free parameters. In this paper we establish a direct relationship between the eigenvalues and the collision integrals, or transport coefficients, known from the kinetic theory of gases. The generating function approach is employed to derive expressions yielding the eigenvalues. The obtained results form a bridge between kinetic theory, atomic physics, and quantum optics. [S1050-2947(97)00711-7]

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I. INTRODUCTION

A. General discussion

Light-induced kinetic effects (LIKE) in gases continue to be of interest [1–4], with light-induced drift (LID) being the most well-known one [5]. Such effects occur due to the light-induced modifications of the velocity distributions of atoms or molecules interacting collisionally between themselves and with incoming light. LID is possible when the active particles immersed in the (usually much denser) buffer gas are excited in a velocity-selective manner. This induces oppositely directed fluxes of ground- and excited-state atoms. When the atoms in either of the states sustain different diffusive friction, the two fluxes do not cancel and the macroscopic drift is observed. Some other LIKE phenomena are described, for example, in [6,7].

There were several different approaches to theoretical description of LIKE in gases (see, for example, Refs. [2–7], and references given therein). One of the possible methods of modeling and/or describing LIKE was introduced in [8], and then further elaborated on in [9] and [10]. The proposed method concerns the physical situation in which the active atoms, i.e., those coupled to the incoming radiation field, also interact collisionally with the perturbers that constitute the thermal bath. The evolution of the velocity distributions of the active atoms, in either the excited or ground state, is then described by the Boltzmann equation.

The Boltzmann equation in either its classical or quantum-mechanical form is extremely complicated. Its solution is indeed a formidable task. Therefore, some approximate methods are needed in practical calculations. A considerable simplification is achieved when the perturber gas is much denser than active-atom vapor. Since the collisions between perturber particles occur much more frequently than any other ones, it is reasonable to assume that the perturbers reach the equlibrium state rapidly, and their velocity distribution may be taken as the Maxwellian

$$f_P(\mathbf{v}_P,t)=W_P(\mathbf{v}_P)=\left(\frac{1}{\pi u_P^2}\right)^{3/2}\exp\left(-\frac{\mathbf{v}_P^2}{u_P^2}\right), \qquad (1)$$

with $u_P^2=2k_BT/m_P$ being the square of the most probable velocity of perturbers at temperature T. Moreover, in the case of $N_A \ll N_P$ (N_A and N_P being the densities of active atoms and perturbers, respectively), the Boltzmann equation for the velocity distribution of active atoms can be put into linear form [8–14]. The linear approximation to the Boltzmann equation should be distinguished from the linearized one. The latter is obtained when one seeks small deviations of the atomic velocity distribution from the Maxwellian one. The Boltzmann equation is then expanded in terms of the corrections to the Maxwellian and only the terms linear in the sought corrections are retained. Moreover, the linearized Boltzmann equation possesses five collisional invariants (particle number, three components of velocity, and the magnitude of velocity), while the linear one has only one invariant—the particle number. This is due to the fact that the much denser perturber gas serves as the reservoir of momentum and energy (see, e.g., [12]).

In this work we will deal with the linear form of the Boltzmann equation, which is obtained due to the assumption that much denser perturbers are in the equilibrium state described by the Maxwellian (1). Such an approach allows one to describe the influence of collisions between active atoms and perturbers on the velocity distributions of the former by collision kernels and rates.

The theoretical method discussed in detail in [9] is based on the presented assumptions. Its main idea can be summarized as follows. The collision kernel and rate are combined into one entity called the collision operator. The collision operator has several interesting and useful properties, the most important being that it is Hermitian in the suitably cho-

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sen space of velocity distributions. Hence, such a collision operator possesses a set of orthonormal eigenfunctions and real eigenvalues. The solution of the corresponding eigenproblem is not simpler than the solution of the initial Boltzmann equation. However, a useful modeling method can be easily devised. We adopt, as an assumption, a certain set of functions to be the eigenfunctions. Such a choice seems to be reasonable and well justified (see [9]). Then all physically important quantities can be expressed in terms of the chosen eigenfunctions and by the eigenvalues. The eigenvalues are, however, unspecified and enter the theory as free parameters. They can be adjusted, for example, by fitting the experimental data to the theoretical predictions. Moreover, assuming that the adopted eigenfunctions are good approximations to the true ones, we avoid the introduction (or derivation) of any particular collision kernel. Such a modeling method is presented in [9]. Since this method does not give any prescription to determine the eigenvalues, we address this point in the present work. Thus, the aim of this paper is to find an explicit connection between the eigenvalues of the collision operator and the collision integrals known from the kinetic theory of gases. The latter integrals are, in turn, closely related to transport coefficients. Thereby, the eigenvalues are not free parameters anymore, but are given a direct physical meaning. Such a procedure allows us to close the method that will thus contain only the quantities with direct experimental relevance.

The outline of the paper is as follows. In the next two subsections we summarize the main concepts, which constitute the basic theoretical framework. In the second section we recall the definition of the collision operator and we review its most important properties. We also explicitly give and discuss the adopted eigenfunctions, which are closely related to the eigenfunctions of the quantum-mechanical harmonic oscillator. Section II also includes some remarks on the eigenvalues of the collision operator and on the integral methods of their calculation. In Sec. III we present a generating function that allows us to derive the expressions connecting the eigenvalues with collision integrals and transport coefficients known from the kinetic theory of gases. The last section contains some additional remarks and comments on the obtained results and their potential applications. The technicalities of our approach are presented in two appendixes.

B. Collision kernels and rates

In our approach we assume that the density of active atoms is much smaller than that of perturbers. Therefore, we may linearize the Boltzmann equation. This is a standard procedure thoroughly discussed in the literature (e.g., [11]). We, therefore, only state the final results. When $N_A \ll N_P$, the time evolution of velocity distribution $f_A(\mathbf{v})$ of active atoms can be, within a very good approximation, attributed solely to the collisions with perturbers and the corresponding linear Boltzmann equation [11–13] can be written as

$$\left.\frac{d}{dt} f_A(\mathbf{v})\right|_{\text{coll}} = -\gamma(\mathbf{v}) f_A(\mathbf{v}) + \int d\mathbf{v}' \mathcal{K}(\mathbf{v} \leftarrow \mathbf{v}') f_A(\mathbf{v}'). \tag{2}$$

The time and position dependencies of the distribution function f_A are suppressed, since they are of little relevance in the present context.

The collision kernel $\mathcal{K}(\mathbf{v} \leftarrow \mathbf{v}')$ and the corresponding rate $\gamma(\mathbf{v})$ appearing in Eq. (2) are easily derived by linearization of the Boltzmann equation. The collision kernel is of the form

$$\mathcal{K}(\mathbf{v} \leftarrow \mathbf{v}') = N_P \left(\frac{m_A}{\mu}\right)^3 \int d\mathbf{v}_r \int d\mathbf{v}_r' \, \frac{d\sigma(\chi, v_r)}{d\Omega} W_P(\mathbf{v}' - \mathbf{v}_r') \times \frac{\delta(v_r - v_r')}{v_r} \delta^3\left(\mathbf{v}_r - \mathbf{v}_r' - \frac{m_A}{\mu}(\mathbf{v} - \mathbf{v}')\right), \tag{3}$$

which accounts for the momentum and energy conservation. The collisional rate $\gamma(\mathbf{v})$ is given as

$$\gamma(\mathbf{v}) = N_P \int d\mathbf{v}_P \int d\Omega(\chi) v_r W_P(\mathbf{v}_P) \frac{d\sigma(\chi, v_r)}{d\Omega}, \tag{4}$$

where $\mathbf{v}$ (or $\mathbf{v}'$) are the velocities of an active atom, and $\mathbf{v}_P$ (or $\mathbf{v}_P'$) of the perturber after (or before) collision. $\mathbf{v}_r = \mathbf{v} - \mathbf{v}_P$ (or $\mathbf{v}_r' = \mathbf{v}' - \mathbf{v}_P'$) are the corresponding relative velocities. m_A, m_P, and μ denote the masses of the active atom, perturber, and the reduced mass, respectively. Expressions (3) and (4) contain also the differential cross section for the active-atom-perturber scattering in the center-of-mass frame, with χ being the usual scattering angle.

The physical meaning of the collision kernel and rate is evident from the kinetic equation (2) (see, e.g., [12]). First of all, we note that the number of particles must be conserved during the collisions. This requirement implies that

$$\gamma(\mathbf{v}) = \int d\mathbf{v}_1 \mathcal{K}(\mathbf{v}_1 \leftarrow \mathbf{v}). \tag{5}$$

The integral term in Eq. (2) is the gain one and it gives the number of particles that change velocity from $\mathbf{v}'$ before, to $\mathbf{v}$ after the collision. Hence, the collision kernel $\mathcal{K}(\mathbf{v} \leftarrow \mathbf{v}')$ is a measure of transition probability between $\mathbf{v}'$ and $\mathbf{v}$ velocity groups. The rate $\gamma(\mathbf{v})$ is the loss term in Eq. (2) and it gives the number of particles escaping from velocity interval $(\mathbf{v}, \mathbf{v} + d\mathbf{v})$ to any other one. Let us note that $\gamma(\mathbf{v})$ can be also viewed as the collision frequency, and its inverse $\tau(\mathbf{v}) = 1/\gamma(\mathbf{v})$ can be interpreted as the average time between collisions. Hence, the names collision rate and frequency can be used interchangeably. The given probabilistic interpretation of the kernel and frequency is fully consistent with requirement (5).

Since the kernel gives the transition probability between various velocity groups, it satisfies the detailed balance condition in equilibrium [12]:

$$\mathcal{K}(\mathbf{v} \leftarrow \mathbf{v}') W_A(\mathbf{v}') = \mathcal{K}(\mathbf{v}' \leftarrow \mathbf{v}) W_A(\mathbf{v}), \tag{6}$$

where $W_A(\mathbf{v})$ is the Maxwellian distribution for active atoms

$$W_A(\mathbf{v}) = \left(\frac{1}{\pi u_0^2}\right)^{3/2} \exp\left(-\frac{\mathbf{v}^2}{u_0^2}\right), \tag{7}$$

with $u_0^2=2k_BT/m_A$ being the square of the most probable velocity. Integrating requirement (6) over $d\mathbf{v}'$ and using Eq. (5) we obtain

$$0=-\gamma(\mathbf{v})W_A(\mathbf{v})+\int d\mathbf{v}'\mathcal{K}(\mathbf{v}\leftarrow\mathbf{v}')W_A(\mathbf{v}'). \tag{8}$$

So, the Maxwellian $W_A(\mathbf{v})$ must be the stationary solution to the kinetic equation. It is equivalent to say that $W_A(\mathbf{v})$ is the eigenfunction of the right-hand side of Eq. (2) with the eigenvalue zero. This result is also evident from the notion of the equilibrium distribution (which is known to be Maxwellian) as the stationary solution to the kinetic equation. With the aid of the particle number conservation requirement it can be shown [12] that $W_A(\mathbf{v})$ is a unique steady-state solution. Hence the discussed zero eigenvalue is nondegenerate.

Furthermore, it can be shown [12] that the collision rate $\gamma(\mathbf{v})$ and collision kernel $\mathcal{K}(\mathbf{v}\leftarrow\mathbf{v}')$ satisfy the relation

$$\int d\mathbf{v}\int d\mathbf{v}'\frac{f_A(\mathbf{v})}{W_A(\mathbf{v})}\mathcal{K}(\mathbf{v}\leftarrow\mathbf{v}')f_A(\mathbf{v}') \leqslant\int d\mathbf{v}\frac{f_A(\mathbf{v})}{W_A(\mathbf{v})}\gamma(\mathbf{v})f_A(\mathbf{v}). \tag{9}$$

The above given relations summarize the most important properties of the collision kernel and rate. Although these properties are simple and physically understandable, they do not facilitate the computation of the kernel and rate. The task of finding the explicit form of these quantities from the interatomic potential (via the corresponding center-of-mass cross sections) is not really simpler than the solution to the initial Boltzmann equation. Moreover, we have presented here the classical approach. The fully quantum-mechanical treatment (see, for example, [13,15,16]) via the scattering amplitudes is still more complex.

Hence, in practical investigations one usually adopts an analytical model kernel, while the rate follows by Eq. (5). The simplest model, sometimes called the strong collision one, is based on the assumption that even a single collision fully thermalizes the velocity distribution of active atoms. In such a case, Eq. (2) reduces to

$$\left.\frac{d}{dt}f_A(\mathbf{v})\right|_{\text{coll}}=-\gamma_{sc}f_A(\mathbf{v})+\gamma_{sc}W_A(\mathbf{v})\int d\mathbf{v}'f_A(\mathbf{v}'), \tag{10}$$

where γ_{sc} is a constant.

Another frequently used model is the Keilson-Storer kernel

$$\mathcal{K}_{\text{KS}}(\mathbf{v}\leftarrow\mathbf{v}')=\frac{\gamma_{\text{KS}}}{(\sigma\sqrt{\pi})^3}\exp\left[-\frac{(\mathbf{v}-\alpha_{\text{KS}}\mathbf{v}')^2}{\sigma^2}\right], \tag{11}$$

where γ_{KS} is a constant, while $\sigma^2=(1-\alpha_{\text{KS}}^2)u_0^2$, and parameter $\alpha_{\text{KS}}\in(0,1)$. It is straightforward to check that both given models satisfy all the necessary requirements. The strong-collision and Keilson-Storer models seem to be the most frequently used ones. It is, however, possible to construct other models. For example, a kernel for hard spheres or the difference kernel are also sometimes employed.

When a model analytical kernel is employed in practical computations there immediately arises the question of whether the adopted model can be found from a physically justified cross section. That is, whether a given model can be derived from relation (3), which relates the kernel and the interatomic potential (via a corresponding cross section). The answer to such a question is usually either negative or very difficult to give. Berman *et al.* [17] addressed this question by relating the suitable moments of the collision kernel to the collisional integrals $\Omega^{(l,s)}$ known from kinetic theory of gases that are closely connected with transport coefficients (see, e.g., [11]). These authors analyzed the Keilson-Storer kernel, the one corresponding to hard spheres and the difference kernel. They have found that the Keilson-Storer kernel gives the results closest to the expectations following from the kinetic theory of gases. Thus, they have concluded that the Keilson-Storer kernel may be treated as the model yielding the results more reliable physically than the other models. This is so, regardless of the fact that the KS model is not derivable from interatomic potentials.

The interesting feature of the Keilson-Storer kernel is that it possesses a set of eigenvalues and eigenfunctions [18]. These eigenfunctions coincide with the ones of the kernel found from Eq. (3) for Maxwell molecules, i.e., for the potential $V(r)\propto r^{-4}$. These eigenfunctions play an important role in the present considerations, so they will be discussed in more detail below.

C. Collision integrals

It is our aim to express the eigenvalues of the collision operators via the collision integrals frequently used in the classical kinetic theory of gases. It might be therefore useful to recall some basic definitions.

Collision integrals $\Omega^{(l,s)}$ appear in the kinetic theory of gases [11] when calculating the transport coefficients and are defined as follows:

$$\Omega^{(l,s)}=\frac{u_R}{2\sqrt{\pi}}\int_0^\infty dx\ \exp(-x^2)x^{2s+3}Q^{(l)}(xu_R), \tag{12}$$

where $u_R^2=2k_BT/\mu=u_P^2+u_0^2$ is the square of the most probable speed of the relative motion of A and P particles. The quantity $Q^{(l)}$ appearing in Eq. (12) is defined as the integral

$$Q^{(l)}(v_r)=2\pi\int_0^\infty bdb\{1-\cos^l[\chi(v_r,b)]\}, \tag{13}$$

with b being the collision parameter. The scattering angle χ depends on b and on the relative velocity v_r of the colliding species. The integral $Q^{(l)}$ can be easily related to the center-of-mass cross section. The relation equivalent to (13) can be thus written as

$$Q^{(l)}(v_r)=\int d\Omega(\chi)\left(\frac{d\sigma(\chi,v_r)}{d\Omega(\chi)}\right)[1-\cos^l(\chi)]. \tag{14}$$

The cross section $d\sigma/d\Omega$ is positive due to its probabilistic interpretation and the factor $(1-\cos^l\chi)$ is also positive, except perhaps for some isolated points where it is zero. Hence,

integrals Q^l are non-negative and this implies that collision integrals $\Omega^{(l,s)}$ defined in Eq. (12) are positive for all combinations of superscripts.

For the sake of further reference, it is useful to review some additional facts apart from the above given definitions. The collision integral $\Omega^{(l,s)}$ can be written equivalently as (see [17])

$$\Omega^{(l,s)}=\frac{1}{8}\int d^3v\, W_R(\mathbf{v})v\left(\frac{v}{u_R}\right)^{2s}Q^{(l)}(v) \tag{15}$$

with

$$W_R(\mathbf{v})=\left(\frac{1}{\pi u_R^2}\right)^{3/2}\exp\left(-\frac{\mathbf{v}^2}{u_R^2}\right), \tag{16}$$

which clearly corresponds to the distribution of the velocity of relative motion of the collision partners.

The total scattering cross section can be expressed by any of the following relations:

$$\sigma_T=2\pi\int_0^\infty b\,db, \tag{17a}$$

$$=\int d\Omega(\chi)\left(\frac{d\sigma(\chi,v_r)}{d\Omega(\chi)}\right), \tag{17b}$$

$$=Q^{(\infty)}(v_r), \tag{17c}$$

where the last equality follows from Eq. (14).

Presented relations can be manipulated in many ways suitable for particular applications. We do not pursue this subject since it is not needed in the present context. All other necessary information can be found, for example, in Ref. [11] (see also [17]).

II. THE COLLISION OPERATOR AND ITS PROPERTIES

A. General theory

The concept of the collision operator was introduced in previous papers [8,9]. Collision operators stem directly from the linear Boltzmann equation and are defined by the right-hand side of Eq. (2). Thus, if $f(\mathbf{v},t)$ is an arbitrary velocity distribution, the collision operator $\hat{\mathcal{C}}$ on $f(\mathbf{v},t)$ is defined as

$$(\hat{\mathcal{C}}f\,)(\mathbf{v},t)=-\gamma(\mathbf{v})f(\mathbf{v},t)+\int d\mathbf{v}'\mathcal{K}(\mathbf{v}\leftarrow\mathbf{v}')f(\mathbf{v}',t), \tag{18}$$

hence it combines the concepts of the kernel and rate into a single entity. The corresponding equation of motion,

$$\frac{\partial}{\partial t}f(\mathbf{v},t)=(\hat{\mathcal{C}}f\,)(\mathbf{v},t), \tag{19}$$

together with the given initial distribution $f(\mathbf{v},t_0)$, is the kinetic equation that gives the time evolution of the velocity distribution $f(\mathbf{v},t)$ of active atoms due to the influence of the velocity-changing collisions with perturber particles. It is worth stressing that the introduced collision operator is, by definition, neither time nor position dependent.

The properties of collision operators were thoroughly discussed in [9]. For the sake of completeness, it seems, however, useful to review briefly the main concepts.

It is convenient to introduce formal notation, much resembling the formalism of standard quantum mechanics. We introduce a vector space $\mathcal{F}$ of velocity distributions

$$\mathcal{F}=\left\{f(\mathbf{v}):\int\frac{d\mathbf{v}}{W(\mathbf{v})}|f(\mathbf{v})|^2<\infty\right\}. \tag{20}$$

For $f,g\in\mathcal{F}$, we define the scalar product

$$\langle f|g\rangle=\int\frac{d\mathbf{v}}{W(\mathbf{v})}f^*(\mathbf{v})g(\mathbf{v}), \tag{21}$$

which may be rewritten as

$$\langle f|g\rangle=\int d\mathbf{v}\langle f|\mathbf{v}\rangle\langle\mathbf{v}|g\rangle. \tag{22}$$

Comparing the above relations we obtain

$$\langle\mathbf{v}|f\rangle=\frac{f(\mathbf{v})}{\sqrt{W(\mathbf{v})}}. \tag{23}$$

In this manner we represent a velocity distribution by a vector in the Hilbert space.

The analogy of the introduced formalism and the quantum-mechanical one allows us to prove easily some important and useful properties of the collision operator. As a consequence of the detailed balance condition (6) for the collision operator we find that for any two distribution functions $f,g\in\mathcal{F}$,

$$\langle f|\hat{\mathcal{C}}|g\rangle=\langle g|\hat{\mathcal{C}}|f\rangle, \tag{24}$$

which means that the collision operator is Hermitian in space $\mathcal{F}$.

Moreover, the rate $\gamma(\mathbf{v})$ and kernel $\mathcal{K}(\mathbf{v}\leftarrow\mathbf{v}')$ satisfy relation (9), which yields [9] the inequality

$$\langle f|\hat{\mathcal{C}}|f\rangle\leqslant 0, \tag{25}$$

which holds for arbitrary velocity distribution $f(\mathbf{v})$. This relation may be called the non-positivity property of the collision operator.

Since we know that the collision operator $\hat{\mathcal{C}}$ is Hermitian in the space $\mathcal{F}$, we can state that there exist left and right eigenvectors of $\hat{\mathcal{C}}$ that are equal and that the eigenvalues of the collision operator are real. Denoting the eigenstates by $|\varphi_\alpha\rangle$ and the real eigenvalues by λ_α we can write

$$\hat{\mathcal{C}}|\varphi_\alpha\rangle=\lambda_\alpha|\varphi_\alpha\rangle$$

and

$$\langle\varphi_\alpha|\hat{\mathcal{C}}=\langle\varphi_\alpha|\lambda_\alpha. \tag{26}$$

Due to the properties of Hermitian operators, eigenfunctions $|\varphi_\alpha\rangle$ form a complete set of orthonormal vectors

$$\sum_\alpha|\varphi_\alpha\rangle\langle\varphi_\alpha|=1$$

and

$$\langle\varphi_\alpha|\varphi_\beta\rangle=\delta_{\alpha\beta}. \tag{27}$$

The general property (25) of the collision operator implies that the eigenvalues are nonpositive

$$\lambda_\alpha = \langle \varphi_\alpha | \hat{\mathcal{C}} | \varphi_\alpha \rangle \leqslant 0. \tag{28}$$

As discussed after Eq. (8) the Maxwellian $W(\mathbf{v})$ is a unique stationary solution to the kinetic equation and it corresponds to the nondegenerate zero eigenvalue. Hence, there must exist such an index α, henceforward denoted by 0, that

$$\lambda_0 = 0$$

and

$$\varphi_0(\mathbf{v}) = W(\mathbf{v}). \tag{29}$$

Since it is a unique eigenvalue, we deduce that for all other indices eigenvalues are strictly negative:

$$\lambda_\alpha < 0 \quad \text{for all} \quad \alpha \neq 0. \tag{30}$$

Expanding the solution of the kinetic equation (19) we see that all initially present deviations from Maxwellian (i.e., equilibrium) distributions behave as $\exp[\lambda_\alpha(t-t_0)]$. Hence, relation (30) ensures that the Maxwellian is the stationary solution corresponding to thermal equilibrium, as it should be.

Having reviewed the basic theoretical methods, let us note that within the presented formalism the kinetic equation (19) can be written as

$$\frac{\partial}{\partial t} f(\mathbf{v},t) = \int d\mathbf{v}' \mathcal{C}(\mathbf{v} \leftarrow \mathbf{v}') f(\mathbf{v},t), \tag{31}$$

with the kernel expanded in terms of the eigenfunctions as

$$\mathcal{C}(\mathbf{v} \leftarrow \mathbf{v}') = \sum_\alpha \lambda_\alpha \frac{\varphi_\alpha(\mathbf{v}) \varphi_\alpha(\mathbf{v}')}{W(\mathbf{v}')}. \tag{32}$$

This relation is the basis of the modeling method introduced and thoroughly discussed in [9]. Namely, we choose a certain set of functions $\{\varphi_\alpha(\mathbf{v})\}$ to be the eigenfunctions of the collision operator. Then, any collision operator can be constructed according to Eq. (32), provided the eigenvalues are known. Thus, we can model a whole class of collision operators by a suitable choice of eigenvalues. In Ref. [9] we treated the eigenvalues as free parameters, and we have also shown how to reconstruct the Keilson-Storer and strong-collision models by the proper choice of the eigenvalues. In this paper we intend to show that the eigenvalues are not free parameters, but can be computed via the quantities that possess a well-defined physical meaning. As already stated in the Introduction, such an approach allows us to circumvent all the questions that immediately arise when one adopts any specific collision kernel, either a model one (as the Keilson-Storer one), or any other one derivable from real interatomic potentials.

B. Eigenfunctions and eigenvalues

Finding exact eigenfunctions and eigenvalues for the collision operator $\hat{\mathcal{C}}$ is as difficult as finding a collision kernel and rate from the interatomic potential. Therefore, we propose a different approach introduced in [8] and [9] and briefly discussed above. Namely, we adopt as the eigenfunctions of the collision operator the following set of orthonormal functions:

$$\varphi_\alpha(\mathbf{v}) = W_A(\mathbf{v}) \phi_\alpha(\mathbf{v}), \tag{33}$$

where $\phi_\alpha(\mathbf{v})$ might be taken in the Cartesian coordinates as

$$\phi_\alpha(\mathbf{v}) = \phi_{n_1 n_2 n_3}(\mathbf{v}) = \prod_{i=1}^{3} \frac{1}{\sqrt{2^{n_i} n_i!}} H_{n_i}\left(\frac{v_i}{u_0}\right), \tag{34}$$

where v_i are Cartesian components of the velocity and $H_n(\cdot)$ are the Hermite polynomials (all special functions in our work are taken according to Ref. [19]). Equivalently, we may choose spherical coordinates and then the eigenfunctions $\phi_\alpha(\mathbf{v})$ become

$$\phi_\alpha(\mathbf{v}) = \phi_{nlm}(\mathbf{v}) = \sqrt{\frac{2\pi^{3/2} n!}{\Gamma(n+l+3/2)}} \left(\frac{v}{u_0}\right)^l \times L_n^{(l+1/2)}\left(\frac{v^2}{u_0^2}\right) Y_{lm}(\theta_{\hat{\mathbf{v}}}, \varphi_{\hat{\mathbf{v}}}), \tag{35}$$

with $L_n^{(l+1/2)}$ being the associated Laguerre polynomials and Y_{lm} the spherical harmonics of the angles determined by the spatial orientation of the vector $\mathbf{v}$. The two sets of eigenfunctions for either of the coordinate systems are distinguished by different subscripts and the context should make it clear which eigenfunctions are considered.

It is certainly worth noting that the eigenfunctions modified by an additional factor, that is, the functions

$$\psi_\alpha(\mathbf{v}) = \sqrt{W_A(\mathbf{v})} \phi_\alpha(\mathbf{v}) = \frac{\varphi_\alpha(\mathbf{v})}{\sqrt{W_A(\mathbf{v})}}, \tag{36}$$

are the usual eigenfunctions of the standard quantum-mechanical harmonic oscillator [20] of mass m_0 and frequency ω_0, such that the factor $\hbar/m_0\omega_0$ is replaced by u_0^2.

The connection between the adopted eigenfunctions $\varphi_\alpha(\mathbf{v})$ of collision operator and oscillator eigenfunctions allows one to see that all the requirements imposed on $\varphi_\alpha(\mathbf{v})$ are indeed satisfied. Moreover, the adopted eigenfunctions of the collision operator are the eigenfunctions of the Keilson-Storer collision kernel, as demonstrated by Snider [18]. The Cartesian eigenfunctions correspond to the eigenvalues $\lambda_{n_1,n_2,n_3} = \gamma_{\mathrm{KS}}(1 - \alpha_{\mathrm{KS}}^{n_1+n_2+n_3})$, while the spherical ones to $\lambda_{nl} = \gamma_{\mathrm{KS}}(1 - \alpha_{\mathrm{KS}}^{2n+l})$.

We should, however, stress the fact that adopting the given functions, in either of the coordinate systems, as the eigenfunctions of the collision operator is a *postulate*. This is a postulate because our choice is in fact arbitrary, although it is well justified. We assume that the chosen eigenfunctions are good approximations to the true eigenfunctions of any physically reasonable collision operator. The fact that the selected eigenfunctions $\varphi_\alpha(\mathbf{v})$ are the ones corresponding to the Keilson-Storer model is one of the arguments supporting our choice. In our earlier paper [9] we have given a detailed discussion and justification that such an approach is reasonable and that the functions $\varphi_\alpha(\mathbf{v})$ can be considered as good approximations to the true eigenfunctions. On the other hand, in [9] we have assumed that the eigenvalues are unknown, and thereby treated as free parameters of the theory. In particular, the eigenvalues can be different from the ones corresponding to the Keilson-Storer kernel. In this manner, using expansion (32) and allowing the eigenvalues to be free parameters we are able to model a whole class of collision

operators. These assumptions allowed us to propose a method for modeling various physical phenomena in gaseous mixtures subjected to electromagnetic irradiation. The examples of applications of our modeling approach together with full discussion and justification of the assumed modeling method are given in [9] (see also [21]). In this sense the eigenfunctions (33) are postulated to be, albeit approximate, eigenfunctions of any collision operator and our current aim is to present a method of computation of the eigenvalues.

We can use either Cartesian form (34) or the spherical one (35) of the eigenfunctions of the collision operator. The latter form of the eigenfunctions is sometimes more convenient and it is usually used in the literature (see, for example, [17,18]). However, in practical calculations the choice between the forms (34) and (35) is, in fact, a matter of convenience. In the papers devoted to modeling the physical phenomena eigenfunctions $\varphi_\alpha(\mathbf{v})$ as given in Eq. (34) were mainly used. One of the advantages of the eigenfunctions taken in the form (34) is that they are factorized in a way that facilitates calculations of various integrals. Moreover, the case with axial symmetry (the symmetry axis being determined by the incident radiation) is essentially a one-dimensional one. Then, the eigenfunctions (34) are especially convenient since all the physically interesting quantities are expressed by integrals over a single component of velocity.

It should be noted that the choice of either of the possible forms of the adopted eigenfunctions determines the numbering of the eigenvalues. The situation is then similar to the case of the harmonic oscillator. When necessary, we shall distinguish two different cases by suitable superscripts.

The eigenvalues of the collision operator obviously follow from the relation $\lambda_\alpha=\langle\varphi_\alpha|\hat{\mathcal{C}}|\varphi_\alpha\rangle$. Substituting the collision operator according to its definition (18) into scalar product (21) and using the property (5) of the collision rate we arrive at the expression

$$\lambda_\alpha=\int d\mathbf{v}\int d\mathbf{v}'\,W_A(\mathbf{v}')\mathcal{K}(\mathbf{v}\leftarrow\mathbf{v}')\times[\phi_\alpha^*(\mathbf{v})\phi_\alpha(\mathbf{v}')-\phi_\alpha^*(\mathbf{v}')\phi_\alpha(\mathbf{v}')],\tag{37}$$

and either Cartesian eigenfunctions (34) or spherical ones (35) can be used.

It is perhaps worth noting that eigenvalues (or rather energies) of the harmonic oscillator are strongly degenerate. In the spherical case the energy corresponds to the principal quantum number $N^{(s)}=2n+l$, while in the Cartesian case $N^{(c)}=n_1+n_2+n_3$. The energy levels are $g(N)=\frac{1}{2}(N+1)(N+2)$ times degenerate (with N being the principal quantum number for either of the case). However, in this work we consider the collision operator which differs considerably from the oscillator Hamiltonian. Therefore, it is reasonable to expect that the degeneracy specific to the harmonic oscillator will be, at least partially, lifted.

C. Eigenvalues for spherical case

It is a straightforward matter to substitute the eigenfunctions (35) into relation (37). We also note that due to symmetry considerations the eigenvalues should be independent of the magnetic quantum number m. Hence, the eigenvalues are labeled only by quantum numbers n and l. Therefore, both sides of the obtained relation can be summed over quantum number m and the addition theorem for spherical harmonics can be used. The resulting integral is then transformed according to the method outlined in Appendix A. According to the relation (A4) we obtain

$$\lambda_{nl}=N_P\frac{2^{n+l}n!}{(2n+2l+1)!!}\int d\mathbf{v}W_R(\mathbf{v})|\mathbf{v}|\int d\Omega(\chi)\frac{d\sigma(\chi,v)}{d\Omega}\int d\mathbf{z}W_T(\mathbf{z})\times\left\{\left(\frac{vv'}{u_0^2}\right)^l L_n^{(l+1/2)}\left(\frac{v^2}{u_0^2}\right)L_n^{(l+1/2)}\left(\frac{v'^2}{u_0^2}\right)P_l\left(\frac{\mathbf{v}\cdot\mathbf{v}'}{vv'}\right)-\left(\frac{v'^2}{u_0^2}\right)^l\left[L_n^{(l+1/2)}\left(\frac{v'^2}{u_0^2}\right)\right]^2\right\},\tag{38}$$

where P_l denote the Legendre polynomials of the cosine of an angle between vectors $\mathbf{v}$ and $\mathbf{v}'$.

In this manner we can relatively easily express the eigenvalues of the collision operator via the corresponding collision integrals (12) or (15). The simplest way to find explicit expressions for the eigenvalues (at least some initial ones) is to use the method outlined in the Appendix. Firstly, we take the corresponding Laguerre and Legendre polynomials to construct the second line of Eq. (38). Then we perform the substitutions that follow from Eq. (A6). Integration over auxiliary variable $\mathbf{z}$ poses no problems. The remaining integrals over $\mathbf{v}_r'$ and $d\Omega(\chi)$ can be easily done when one recalls the basic geometry of the scattering process, which allows us to write

$$\mathbf{v}_r'\cdot(v_r'\mathbf{n}_0-\mathbf{v}_r')=-2v_r'^2\sin^2(\chi/2)=-v_r'^2(1-\cos\chi).\tag{39}$$

Due to the appearance of the cosine of the scattering angle χ we recognize the integrals $Q^{(l)}(v_R)$ defined in Eq. (14) and then collision integrals $\Omega^{(l,s)}$ as in Eq. (15). A few first eigenvalues are obtained in that manner and they read as follows:

$$\lambda_{01}=-\frac{16}{3}N_P\left(\frac{\mu}{m_A}\right)\Omega^{(1,1)},\tag{40a}$$

$$\lambda_{02}=-\frac{32}{3}N_P\left(\frac{\mu^2}{m_Am_P}\right)\left[\Omega^{(1,1)}+\frac{3}{10}\left(\frac{m_P}{m_A}\right)\Omega^{(2,2)}\right],\tag{40b}$$

$$\lambda_{10}=-\frac{32}{3}N_P\left(\frac{\mu^2}{m_Am_P}\right)\Omega^{(1,1)},\tag{40c}$$

Calculation of the eigenvalues corresponding to higher numbers n,l can be done similarly, though it is more and more involved. The obtained eigenvalues for the spherical case are degenerate with respect to the quantum number m. Other degeneracies, typical to the harmonic oscillator are, as expected, lifted.

D. Eigenvalues for Cartesian case

In this case the eigenvalues are labeled by three integers. The calculation goes exactly along the same lines as in the spherical case, hence we state only the results. We have obtained

$$\lambda_{001}=\lambda_{010}=\lambda_{100}=-\frac{16}{3}N_P\left(\frac{\mu}{m_A}\right)\Omega^{(1,1)}, \tag{41a}$$

$$\lambda_{011}=\lambda_{101}=\lambda_{011}=-\frac{32}{3}N_P\left(\frac{\mu^2}{m_A m_P}\right)\times\left[\Omega^{(1,1)}+\frac{3}{10}\left(\frac{m_P}{m_A}\right)\Omega^{(2,2)}\right], \tag{41b}$$

$$\lambda_{002}=\lambda_{020}=\lambda_{200}=-\frac{32}{3}N_P\left(\frac{\mu^2}{m_A m_P}\right)\times\left[\Omega^{(1,1)}+\frac{1}{5}\left(\frac{m_P}{m_A}\right)\Omega^{(2,2)}\right]. \tag{41c}$$

We see that the eigenvalues that differ by the permutation of the indices are degenerate. We shall later show that this is a general property of the Cartesian eigenvalues obtained from the eigenfunctions constructed with Hermite polynomials as in Eq. (34).

Comparing Eqs. (40) and (41) one easily sees that the eigenvalues for spherical and Cartesian case are related,

$$\lambda^{(c)}_{001}=\lambda^{(s)}_{01}, \tag{42a}$$

$$\lambda^{(c)}_{011}=\lambda^{(s)}_{02}, \tag{42b}$$

$$\lambda^{(c)}_{002}=\tfrac{1}{3}\lambda^{(s)}_{10}+\tfrac{2}{3}\lambda^{(s)}_{02}, \tag{42c}$$

where the superscripts denote either the Cartesian or spherical case. Certainly the Cartesian eigenvalues are degenerate in the sense indicated by Eqs. (41). Relations (42) follow from the presented results, but can be also derived. The eigenfunctions of the harmonic oscillator in spherical or Cartesian coordinates are connected by unitary transformation [20]. Since eigenfunctions of the collision operator are proportional to the oscillator eigenfunctions, the mentioned transformation can be employed yielding relations (42) between the eigenvalues corresponding to two coordinate systems.

III. GENERATING FUNCTION FOR THE EIGENVALUES

A. Generating function for eigenfunctions

As stated in the Introduction our aim is to find expressions connecting the eigenvalues of the collision operator with the collision integrals $\Omega^{(l,s)}$ known from the kinetic theory of gases. We will do so by the generating function method. To achieve this end we introduce the following function:

$$G(\mathbf{x},\mathbf{a})=\exp(-\mathbf{x}^2+2\mathbf{a}\cdot\mathbf{x}), \tag{43}$$

where $\mathbf{x}$ and $\mathbf{a}$ are two independent vectors. This function is a product of three one-dimensional generating functions of Hermite polynomials. Hence, we have

$$G(\mathbf{x},\mathbf{a})=\prod_{i=1}^{3}\sum_{n_i=0}^{\infty}\frac{x_i^{n_i}}{n_i!}H_{n_i}(a_i), \tag{44}$$

where x_i and a_i denote Cartesian components of the vectors entering the definition of function G. The function G can also be expanded into power series (Kumar's generating function [22]) of Laguerre polynomials and spherical harmonics as follows:

$$G(\mathbf{x},\mathbf{a})=2\pi^{3/2}\sum_{n=0}^{\infty}\sum_{l=0}^{\infty}\sum_{m=-l}^{l}\frac{(-1)^n}{\Gamma(n+l+3/2)}x^{2n+l}a^l\times L_n^{(l+1/2)}(a^2)Y^*_{lm}(\hat{\mathbf{x}})Y_{lm}(\hat{\mathbf{a}}), \tag{45}$$

where a and x denote the lengths, while $\hat{\mathbf{x}}$ and $\hat{\mathbf{a}}$ specify the angular orientation of the corresponding vectors.

Thus, one clearly sees that the function G is closely related with the eigenfunctions of collision operator (33) given either in Cartesian frame by Eq. (34) or in spherical coordinates (35). Indeed, by substitution $\mathbf{a}=\mathbf{v}/u_0$ and some simple transformations we obtain

$$W_A(\mathbf{v})G(\mathbf{x},\mathbf{v}/u_0)=\sum_{n_1=0}^{\infty}\sum_{n_2=0}^{\infty}\sum_{n_3=0}^{\infty}x_1^{n_1}x_2^{n_2}x_3^{n_3}\sqrt{\frac{2^{n_1}2^{n_2}2^{n_3}}{n_1!n_2!n_3!}}\times\varphi^{(c)}_{n_1n_2n_3}(\mathbf{v}). \tag{46}$$

Similarly, function G may be expanded in eigenfunctions expressed in spherical coordinates. In this case we have

$$W_A(\mathbf{v})G(\mathbf{x},\mathbf{v}/u_0)=\sqrt{2\pi^{3/2}}\sum_{n=0}^{\infty}\sum_{l=0}^{\infty}\sum_{m=-l}^{l}x^{2n+l}\times\frac{(-1)^n}{\sqrt{n!\Gamma(n+l+3/2)}}Y^*_{lm}(\hat{\mathbf{x}})\varphi^{(s)}_{nlm}(\mathbf{v}). \tag{47}$$

The two last expressions may be considered as generating functions for the eigenfunctions of the collision operator in either of the coordinate frames.

B. Generating function for eigenvalues

The next step consists in finding the generating function for the eigenvalues. Hence, we define the following matrix element:

$$J(\mathbf{x},\mathbf{y})=\langle G(\mathbf{x},\mathbf{v}/u_0)W_A|\hat{\mathcal{C}}|G(\mathbf{y},\mathbf{v}/u_0)W_A\rangle, \tag{48}$$

where $\mathbf{x}$ and $\mathbf{y}$ are two independent vectors playing the role of parameters. Using relations (22) and (23) this matrix element may be transformed into an integral:

$$J(\mathbf{x},\mathbf{y})=\int d\mathbf{v}G^*(\mathbf{x},\mathbf{v}/u_0)[\hat{\mathcal{C}}G(\mathbf{y},\mathbf{v}/u_0)W_A(\mathbf{v})]. \quad (49)$$

Integral $J(\mathbf{x},\mathbf{y})$ may be further expressed in terms of expansions, either Eq. (46) or Eq. (47). The collision operator acting on its eigenfunctions produces corresponding eigenvalues. Then, due to orthonormality of the eigenfunctions, we obtain in the Cartesian case

$$J(\mathbf{x},\mathbf{y})=\sum_{n_1,n_2,n_3=0}^{\infty}\lambda^{(c)}_{n_1n_2n_3}\frac{2^{n_1}2^{n_2}2^{n_3}}{n_1!n_2!n_3!}\times(x_1y_1)^{n_1}(x_2y_2)^{n_2}(x_3y_3)^{n_3}. \quad (50)$$

The obtained expression is symmetrical with respect to interchange of the cordinate axes. This clearly suggests that we may expect the Cartesian eigenvalues $\lambda^{(c)}_{n_1n_2n_3}$ to be degenerate with respect to permutation of the indices. Hence, the conclusion drawn after Eqs. (41) seems, at least partially, to be confirmed.

An equivalent expansion is obtained for eigenfunctions expressed in spherical coordinates, namely,

$$J(\mathbf{x},\mathbf{y})=\sum_{n,l=0}^{\infty}\lambda^{(s)}_{nl}\frac{2^{n+l}(2l+1)}{n!(2n+2l+1)!!}(xy)^{2n+l}P_l\left(\frac{\mathbf{x}\cdot\mathbf{y}}{xy}\right), \quad (51)$$

where we have used the addition theorem for spherical harmonics, in a manner similar to that leading to Eq. (38). For the spherical case it is difficult to draw conclusions about degeneracies. It rather should be expected that all eigenvalues $\lambda^{(s)}_{nl}$ are different (of course, apart from trivial degeneracy with respect to the magnetic quantum number m).

The function $J(\mathbf{x},\mathbf{y})$ may be thus regarded as the generating function for eigenvalues of collision operators. Knowing the explicit form of this integral we can then find eigenvalues either for the Cartesian or for the spherical case by simple differentiation of both sides of Eq. (50) or Eq. (51). Explicit computation of $J(\mathbf{x},\mathbf{y})$ must be done by substituting the functions G given in Eq. (43) into the integral (49), using the form of the collision operator as specified by Eq. (18) and by performing all necessary integrations.

Proceeding along the outlined lines and employing the neccessary properties of the collision kernel we easily arrive at the expression for integral $J(\mathbf{x},\mathbf{y})$:

$$J(\mathbf{x},\mathbf{y})=\exp(-x^2-y^2)\int d\mathbf{v}\int d\mathbf{v}'W_A(\mathbf{v}')\mathcal{K}(\mathbf{v}\leftarrow\mathbf{v}')\times\exp\left[\frac{2}{u_0}(\mathbf{x}+\mathbf{y})\cdot\mathbf{v}'\right]\left\{\exp\left[\frac{2}{u_0}\mathbf{x}\cdot(\mathbf{v}-\mathbf{v}')\right]-1\right\}. \quad (52)$$

Explicit evaluation of this integral can be done by employing the calculational methods presented in the Appendix.

The integrand in Eq. (52) is a function of two velocities as the function integrated in Appendix A. Hence performing the substitutions indicated by Eq. (A4), we recast Eq. (52) into a similar form and we easily perform the integration over the auxiliary variable $\mathbf{z}$, obtaining

$$J(\mathbf{x},\mathbf{y})=N_PN(\mathbf{x},\mathbf{y})\int d\mathbf{v}W_R(\mathbf{v})|\mathbf{v}|\int d\Omega(\chi)\frac{d\sigma(\chi,v)}{d\Omega}\times\exp[a(\mathbf{x}+\mathbf{y})\cdot\mathbf{v}]\{\exp[a\mathbf{x}\cdot(\mathbf{n}_0v-\mathbf{v})]-1\}, \quad (53)$$

where we have dropped unnecessary subscripts for the integration variable and we have denoted

$$a=\frac{2\mu}{u_0m_A} \quad (54)$$

and

$$N(\mathbf{x},\mathbf{y})=\exp\left[-x^2-y^2+\frac{u_T^2}{u_0^2}(\mathbf{x}+\mathbf{y})^2\right]. \quad (55)$$

Technical details of the calculation are presented in Appendix B. Here, we only state the final result, which reads

$$J(\mathbf{x},\mathbf{y})=N(\mathbf{x},\mathbf{y})\sum_{m=0}^{\infty}\sum_{n=1}^{\infty}A_{nm}(\alpha)x^ny^m, \quad (56)$$

where the coefficients $A_{nm}(\alpha)$ are given in Eq. (B14), and α is the angle between vectors $\mathbf{x}$ and $\mathbf{y}$, i.e., $\cos\alpha=\mathbf{x}\cdot\mathbf{y}/(xy)$. We again see that the obtained expression is symmetric under the interchange of three Cartesian axes. Therefore, we finally conclude that eigenvalues $\lambda^{(c)}_{n_1n_2n_3}$ are unchanged (degenerate) when the indices are permuted.

As discussed in Appendix B, the obtained generating function $J(\mathbf{x},\mathbf{y})$ is a complicated combination of the collision integrals $\Omega^{(l,s)}$. Thus the main aim of our work may be considered as achieved. This is so because substituting expression (56) as the right-hand side of relation (50) we can find the sought eigenvalues. Then, by standard generating function techiques, taking the necessary derivatives and setting the parameters to zero we can obtain the corresponding Cartesian eigenvalues $\lambda^{(c)}_{n_1n_2n_3}$ of the collision operator. A similar procedure can be applied to Eqs. (51) and (56) to obtain the spherical eigenvalues $\lambda^{(s)}_{nl}$. The described approach allows easy recovery of the eigenvalues calculated above by direct integration.

The generating function procedure in either of the considered cases poses no conceptual difficulties. It is possible to derive general expressions for the eigenvalues, but the results of differentiation are lengthy and not very illuminating. This is due to the complicated form of the coefficients $A_{nm}(\alpha)$ where the angle α enters at several places and the derivatives obtained via the Leibniz formula are quite involved. Therefore, instead of discussing the general case, we will focus our attention on a more specific situation, which, however, covers a wide range of realistic experimental situations.

C. The case with axial symmetry

The formalism described so far concerns the simple gas mixture and is, therefore, closely related to the kinetic theory of gases. It is interesting in itself, but we would like to consider another physically important case, which seems to be more important in quantum optics. When the sample contain-

ing a mixture is irradiated by electromagnetic radiation, the system has axial symmetry with the axis specified by the direction of the incoming field. Such a case is of special interest since then one may observe various light-induced kinetic phenomena.

As is well known, the velocity distributions are modified only along the symmetry axis. The reason for this is as follows. Let us assume that the incoming radiation propagates along the z axis and has the wave vector $\mathbf{k}=(0,0,k_z)$. Interaction of radiation with active atoms is described by optical Bloch equations, which contain Doppler shift $\mathbf{k}\cdot\mathbf{v}=k_z v_z$. Thereby radiation may modify the velocity distributions only with respect to the v_z component. On the other hand, collisions with perturbers drive the distributions towards equilibrium, i.e., towards Maxwellian. Due to the radiation influence the steady-state velocity distributions usually differ from Maxwellian and can be written as $f_A(\mathbf{v})=W_A(v_x)W_A(v_y)f_A(v_z)$. Therefore, expanding $f_A(\mathbf{v})$ in the eigenfunctions of the collision operator we see that the Cartesian case is the most suitable. This is so because the eigenfunctions (34) are correspondingly factorized. Thus, all physically interesting quantities can be expanded into the series of Cartesian eigenfunctions φ_{00n}, that is, those with two first indices being zeros. Hence, the relavant distributions are essentially one dimensional. Physical quantities expressible via expectation values, or, in general, via integrals over the distributions would then depend on the corresponding eigenvalues λ_{00n}. Practical examples of the considered case together with the velocity distributions and other quantities of interest are presented in [9].

The discussed eigenvalues λ_{00n} are most easily obtained when we take $\mathbf{x}=(0,0,x)$ and $\mathbf{y}=(0,0,y)$. In such a case the sum in Eq. (50) reduces to a single one and upon differentiation we simply get

$$[y^k]\,\frac{1}{2^k}\,\frac{\partial^k}{\partial x^k}J(x,y)\bigg|_{x=0}=\lambda_{00k}^{(c)}, \tag{57}$$

where the notation $[z^k]f(z)$ denotes the coefficient of z^k in the Taylor expansion of the function $f(z)$ (this notation follows that introduced by Graham *et al.* [23]).

To use relation (57) effectively we need the explicit expression for the generating function $J(x,y)$ as given by Eq. (56). Due to our assumption about parameter vectors $\mathbf{x}$ and $\mathbf{y}$ we have an angle $\alpha=0$. Therefore the coefficients A_{nm} defined in Eq. (B14) are effectively simpler and read

$$A_{nm}(0)=-2\pi N_P\frac{a^{n+m}}{m!}\int_0^\infty dv\,W_R(\mathbf{v})|\mathbf{v}|^{3+n+m}\int_{-1}^{1}d(\cos\theta)\cos^{n+m}\theta\left[\frac{Q^{(n)}(v)}{n!}-\sum_{k=1}^{[n/2]}\frac{B_{nk}(v)\tan^{2k}\theta}{2^{2k}(k!)^2(n-2k)!}\right]. \tag{58}$$

Differentiating Eq. (56) we make use of the Leibniz formula; we express the factor $N(x,y)$ as the generating function of Hermite polynomials. Taking the derivative at $x=0$, we obtain

$$\lambda_{00n}^{(c)}=[y^k]\,\frac{k!}{2^k}\exp[-y^2\kappa^2]\sum_{p=0}^{k-1}\frac{\kappa^p}{p!}H_p(\beta y)\sum_{m=0}^{\infty}A_{k-p,m}y^m, \tag{59}$$

where H_p are Hermite polynomials and $\kappa^2=\mu/m_A$, $\beta=u_T^2/(u_0^2\kappa)$. Evaluation of the eigenvalue $\lambda_k^{(c)}$ reduces then to finding the coefficient accompanying the parameter y in the kth power. Finding several first eigenvalues is a simple matter. We get

$$\lambda_{001}^{(c)}=\frac{1}{2}A_{11}(0), \tag{60a}$$

$$\lambda_{002}^{(c)}=-\frac{\kappa^2}{2}A_{20}(0)+\left(\frac{1}{2}A_{22}(0)+\frac{u_T^2}{u_0^2}A_{11}(0)\right), \tag{60b}$$

$$\lambda_{003}^{(c)}=\frac{3}{4}\left[A_{33}(0)-\frac{\mu}{m_A}[A_{13}(0)+A_{31}(0)]+2\frac{\mu}{m_P}A_{22}(0)+\left(2\frac{\mu^2}{m_P^2}+\frac{\mu^2}{m_A^2}\right)A_{11}(0)\right]. \tag{60c}$$

When coefficients $A_{nm}(0)$ are substituted according to Eqs. (B16), then two first eigenvalues reproduce relations (41a) and (41c). While the third one yields

$$\lambda_{003}^{(c)}=-8N_P\frac{\mu^3}{m_A^2m_P}\left[\left(\frac{m_P}{m_A}+2\frac{m_A}{m_P}\right)\Omega^{(1,1)}+\frac{4}{5}\Omega^{(2,2)}-\frac{4}{5}\left(\frac{m_P}{m_A}\right)\Omega^{(1,2)}+\frac{4}{35}\left(\frac{m_P}{m_A}\right)\left(\Omega^{(1,3)}+\frac{2}{3}\Omega^{(3,3)}\right)\right]. \tag{61}$$

Calculation of further eigenvalues $\lambda_{00n}^{(c)}$ requires only coefficients $A_{nm}(0)$, which can be easily found from Eq. (58). Then, the corresponding eigenvalues are simply read from Eq. (59). The case with axial symmetry, important from the quantum-optical point of view, is thus simpler than the general one, considered in the previous section. The given results provide, it seems, a useful tool to describe collisional effects and light-induced kinetic effects in gases.

IV. DISCUSSION AND REMARKS

We have presented a general method allowing one to express the eigenvalues of the collision operator by the collision integrals. Equation (50) or (51) with left-hand sides given by Eq. (56) can, by suitable differentiation, yield the eigenvalues of collision operators either in Cartesian or in spherical coordinate systems. The eigenvalues with arbitrary indices can thus be obtained, although the procedure is

somewhat tedious albeit quite straightforward and much simpler than direct integration. The general case is of interest within kinetic theory of gases. In such a case the Chapman-Enskog approach gives a method to find succesive approximations to the various physically significant quantities. Within the first order (for the case when $N_A \ll N_P$, i.e., when P gas is much denser, as assumed in this paper) this method gives, for example, the following results.

(1) The diffusion coefficient for molecules of active-atom gas in the perturber gas is given as

$$D_{AP}=\frac{3k_BT}{16N_P\mu\Omega^{(1,1)}}. \tag{62}$$

(2) Viscosity of the considered gaseous mixture is

$$\eta=\frac{5k_BT}{8\Omega^{(2,2)}}. \tag{63}$$

(3) The heat conductivity is expressed as

$$\lambda^{(Q)}=\frac{75k_B^2T}{32m_A\Omega^{(2,2)}}. \tag{64}$$

It is thus evident that only several eigenvalues of the collision operator suffice to express the above given transport coefficients, which are most frequently used in kinetic theory. Transport coefficients (62)–(64) can easily be expressed by the eigenvalues given in Eqs. (41). Conversely, eigenvalues can be rewritten in terms of combinations of transport coefficients. This fact puts the eigenvalues on firm physical ground. They are no longer free parameters, but are fully expressed by physically understandable transport coefficients.

Expressing some of the first eigenvalues by the collision integrals or transport coefficients we can construct an approximate (because of finite number of eigenvalues used) collision operator by its integral kernel (32). Thus, the collision operator is then written in terms of the quantities that are either directly measurable or can be expressed via such quantities. In this way we are able to construct the collision operator without invoking any particular analytical model of the collision kernel and rate. Therefore, we avoid all the difficult questions arising when adopting any specific model. For example, the interatomic potential determines the differential cross section that appears in the collision integrals and hence in the transport coefficients. The former ones, in turn, determine the eigenvalues, thereby the right potential is automatically accounted for in our (although approximate) collision operator constructed according to Eq. (32). Circumventing the problems connected with any particular model of the collision kernel, we provide a method to construct a whole class of collision operators.

Our work focuses on the theoretical approach towards determination of the collision kernels. On the other hand, some experimental measurements were also done to determine the collision operators (or kernels) (see [24,25], and the references given therein). The basic idea of these experiments is to excite a resonance transition in a velocity selective manner, and then to probe it by a second (usually weak) laser field by excitation to a higher third level. The spectra of fluorescence from this third level provide then information about velocity redistribution in the first excited level. Haverkort, Woerdman, and Berman [24] applied such a method to obtain the best fit to the Keilson-Storer kernels. Gibble and Gallagher obtained the necessary spectra and then, by means of an appropriate deconvolution procedure, found the corresponding (one-dimensional) collision kernels. It is very interesting to relate the experimental fluorescence spectra directly to collision operators. An attempt in this direction, however, with only one exciting laser field, was presented in [21]. The main difficulty in such an approach is that ground-state and excited-state atoms interact with perturbers in a different way, we simply have two different interaction potentials. This fact entails the necessity to introduce two different collision operators and thereby two different sets of eigenvalues. Due to this, as shown previously [7], the radiative and collisional processes are strongly interwoven—radiation modifies transport properties. Hence, it may be expected that a general and possibly rigorous treatment of the fluorescence spectra corresponding to the experimental cases will be quite complex and probably will require a suitable approximation scheme. Nevertheless, it might be expected that, at least for the weak probe case, it will be possible to express the spectra via the eigenvalues of the collision operators. Then, comparison of experimental spectra to the theoretical ones should directly yield the eigenvalues and hence the (approximate) collision operators according to Eq. (32).

The obtained relationship between the eigenvalues of the collision operators and collision integrals $\Omega^{(l,s)}$ or transport coefficients clarifies the previously employed modeling method. We have shown that only several eigenvalues of the collision operator usually suffice to describe physical phenomena occurring in the gaseous mixtures [9,21]. Hence, it would be of interest to reexamine the results of modeling in the view of the present ones. It would be interesting to use explicit expressions for the eigenvalues in the equations for the quantities modeled by eigenvalues—previously taken as free parameters. This may provide some new information on the light-induced kinetic effects in gases. Moreover, it will be easier to compare the theory with potential experiments, since the physically significant quantities will be expressed by other ones, which are measurable and have well-known physical meaning. This may be an interesting subject for further investigations.

These conclusions shed additional light on the modeling method developed and used elsewhere [9]. The power of this method is obviously not affected by the present results. Just the contrary, the modeling procedure is now fully justified physically, since due to the present results, it does not include free (unknown) parameters, but the quantities expressible by other ones, which have a clear physical meaning.

Moreover, as we have discussed, Cartesian eigenvalues $\lambda_{00n}^{(c)}$ (which correspond to the case with axial symmetry) play a special role in a variety of spectroscopical or quantum-optical problems. The above given comments obviously apply also to this—more specialized—case. It seems that the obtained results fully justify the use of $\lambda_{00n}^{(c)}$ instead of spherical eigenvalues $\lambda_{nl}^{(s)}$ in the description of experiments where the radiation beam specifies an axis of symmetry.

The transport coefficients given, as examples above, are found within the Chapman-Enskog method in the first-order approximation. It is, however, possible to find the corresponding equations in higher orders. The resulting expressions are quite complex, but nevertheless still include the collision integrals $\Omega^{(l,s)}$, although not in simple linear combinations. The connection between the eigenvalues and the collision integrals might be also useful in better understanding of the Boltzmann equation. Following this train of thought one may, perhaps, gain some additional data for more dense gases, and study their properties by relatively simple theoretical methods provided by the formalism of collision operators and their eigenvalues. The discussed connection between higher-order Chapmann-Enskog results and collision operators may lead to interesting physical results. It may also facilitate the investigations of the phenomena occurring in the radiation field.

The Chapmann-Enskog method may be equally well applied to single species gases as to mixtures. The collision operator considered in this paper concerns the mixture with one component much denser than the other one. Study of the derived relationships might also prove useful in a suitable adaptation of our method to single-component gases. This is, however, the subject for future studies. Thus, the modeling method used in previous papers can, perhaps, be suitably modified and then applied for single-component gases.

The collision integrals $\Omega^{(l,s)}$ or transport coefficients are well known from the literature (see [11]). They follow when one knows the differential cross sections. Hence, adopting either analytic or numerical interatomic potentials, one can relatively easily compute the cross sections, and then the collision integrals and eigenvalues of collision operators. This procedure will yield eigenvalues directly via the potentials. Moreover, collision integrals are explicitly temperature dependent. Therefore, our modelling method allows investigations of the temperature dependencies involved in LIKE in gases. This fact may prove useful in the view of the recent communications [1,2] on the anomalous light-induced drift effect.

The approach via eigenfunctions and eigenvalues of the collision operator seems to be quite elastic and applicable to the description of a large collection of experimental situations. Results of this paper together with previously published modeling method [9] seem to have considerable potential and a consistent, well justified physical basis.

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APPENDIX A: AN AUXILIARY INTEGRAL

In this Appendix we present an important calculational method. Let us consider the quantity of the type

$$\mathcal{F}=\int d\mathbf{v}\int d\mathbf{v}'\,W_A(\mathbf{v}')\mathcal{K}(\mathbf{v}\leftarrow\mathbf{v}')F(\mathbf{v},\mathbf{v}'). \tag{A1}$$

This quantity is clearly of the type of integral necessary to find the eigenvalues according to Eq. (37). In order to find this integral we need the collision kernel. We take it in the form given by Eq. (3). Substituting the collision kernel into the integral (A1) we obtain a four-fold three-dimensional integral. We proceed to simplify it as follows. Firstly we consider the product W_AW_P of two Maxwellian distributions, which appear under the integrals. By simple manipulation this product can be transformed into

$$W_A(\mathbf{v}')W_P(\mathbf{v}'-\mathbf{x})=W_R(\mathbf{x})W_T\left(\mathbf{v}'-\frac{\mu}{m_A}\mathbf{x}\right), \tag{A2}$$

and where $\mathbf{x}$ denotes arbitrary vector. The distribution W_R is given in Eq. (16) and W_T is defined as follows:

$$W_T(\mathbf{v})=\left(\frac{1}{\pi u_T^2}\right)^{3/2}\exp\left(-\frac{\mathbf{v}^2}{u_T^2}\right), \tag{A3}$$

with $u_T^2=2k_BT/(m_A+m_P)$. Hence, W_T describes the velocity distribution of the motion of the collision partners as a single whole.

Secondly, we simplify and compute some of the obtained integrals. Instead of the integration variable $\mathbf{v}'$ we introduce $\mathbf{z}=\mathbf{v}'-(\mu/m_a)\mathbf{v}_r'$. Transforming the argument of the three-dimensional delta function we are able to perform an integration over velocity $\mathbf{v}$. Thus we obtain

$$\mathcal{F}=N_P\int d\mathbf{v}_r\int d\mathbf{v}_r'\int d\mathbf{z}\,\frac{d\sigma(\chi,v_r)}{d\Omega}W_R(\mathbf{v}_r')W_T(\mathbf{z})\times\frac{\delta(v_r-v_r')}{v_r}F\left(\mathbf{z}+\frac{\mu}{m_A}\mathbf{v}_r,\mathbf{z}+\frac{\mu}{m_A}\mathbf{v}_r'\right). \tag{A4}$$

In order to perform the integration over the velocity, say dv_r, some basic geometric considerations are helpful. Vectors $\mathbf{v}_r$ and $\mathbf{v}_r'$ are relative velocities before and after the collision. Due to energy conservation their lengths are equal. The unit vector

$$\mathbf{n}_0=(\sin\chi\cos\varepsilon,\sin\chi\sin\varepsilon,\cos\chi) \tag{A5}$$

specifies the orientation of velocity $\mathbf{v}_r=v_r\mathbf{n}_0$ with respect to the vector $\mathbf{v}_r'$. Hence integration over $d\mathbf{v}_r$ is equivalent to integration over scattering angles $d\Omega(\chi)$ and over the modulus dv_r. The last one is easily done due to the presence of the delta function in Eq. (A4). Thus, we obtain a much simpler expression, namely,

$$\mathcal{F}=N_P\int d\mathbf{v}_r'W_R(\mathbf{v}_r')|\mathbf{v}_r'|\int d\Omega(\chi)\,\frac{d\sigma(\chi,v_r')}{d\Omega}\times\int d\mathbf{z}W_T(\mathbf{z})F\left(\mathbf{z}+\frac{\mu}{m_A}v_r'\mathbf{n}_0,\mathbf{z}+\frac{\mu}{m_A}\mathbf{v}_r'\right), \tag{A6}$$

which clearly indicates the variable substitutions necessary to compute the integral explicitly. Moreover, it allows one to use the cross section, which is usually easier to find than the collision kernel. Hence, we conclude that the obtained relation expresses the sought integral (A1) in a manner greatly simplifying the calculations.

APPENDIX B: EXPLICIT EVALUATION OF GENERATING FUNCTION

Evaluation of the integration over the scattering angles χ and ε in Eq. (53) requires some geometrical considerations. First of all, we note that vectors **x** and **y** are arbitrary—they are just parameters. Therefore we can take a vector, say **x**, as determining the z axis. Then vector **y** is directed so that it forms an angle α with **x**. Hence, we can take

$$\mathbf{y}=y(\sin\alpha,0,\cos\alpha), \tag{B1}$$

while the arbitrarily directed velocity vector **v** is then

$$\mathbf{v}=v(\sin\theta\cos\varphi,\sin\theta\ \sin\varphi,\cos\theta), \tag{B2}$$

where θ and φ are the spherical angles taken with respect to the vector **x**, which specifies the z axis. The unit vector $\mathbf{n}_0$ is defined in Eq. (A5) with respect to velocity **v**. Thus it has to be rotated to the new coordinate system. We obtain

$$\mathbf{n}_o=\begin{pmatrix}\cos\theta\cos\varphi\sin\chi\cos\varepsilon-\sin\varphi\sin\chi\sin\varepsilon+\sin\theta\cos\varphi\cos\chi\\ \cos\theta\sin\varphi\sin\chi\cos\varepsilon+\cos\varphi\sin\chi\sin\varepsilon+\sin\theta\sin\varphi\cos\chi\\ -\sin\theta\sin\chi\cos\varepsilon+\cos\theta\cos\chi\end{pmatrix}. \tag{B3}$$

We then rewrite integral (53) as

$$J(\mathbf{x},\mathbf{y})=-N_PN(\mathbf{x},\mathbf{y})\int d\mathbf{v}W_R(\mathbf{v})|\mathbf{v}|e^{a\mathbf{y}\cdot\mathbf{v}}\times[e^{a\mathbf{x}\cdot\mathbf{v}}\sigma_T(v)-H(v)], \tag{B4}$$

with σ_T defined in Eq. (17) and

$$H(v)=\int d\Omega(\chi)\frac{d\sigma(\chi,v)}{d\Omega}e^{av\mathbf{x}\cdot\mathbf{n}_0}. \tag{B5}$$

Using the above given vectors we write explicitly an equivalent form of integral $H(v)$. Changing the integration variables in a typical way we obtain

$$H(v)=\int_0^\infty bdbe^{axv\ \cos\theta\cos\chi}\int_0^{2\pi}d\varepsilon e^{-axv\ \sin\theta\sin\chi\cos\varepsilon}. \tag{B6}$$

The integral over the angle ε can be expressed via the modified Bessel function I_0. Returning to standard notation we thus have

$$H(v)=\int d\Omega(\chi)\frac{d\sigma(\chi,v)}{d\Omega}e^{a\mathbf{x}\cdot\mathbf{v}\ \cos\chi}I_0(avx\sin\theta\sin\chi). \tag{B7}$$

Next we expand the exponential and the Bessel function into power series. Separating the zeroth term of the Bessel function expansion, we find that it reproduces the term containing σ_T in Eq. (B4), which, therefore, cancels out. Thus, we have

$$J(\mathbf{x},\mathbf{y})=-N_PN(\mathbf{x},\mathbf{y})\int d\mathbf{v}W_R(\mathbf{v})|\mathbf{v}|e^{a\mathbf{y}\cdot\mathbf{v}}H_1(v), \tag{B8}$$

where function $H_1(v)$ has the expansion

$$H_1(v)=\sum_{n=1}^{\infty}\frac{a^n}{n!}(\mathbf{x}\cdot\mathbf{v})^nQ^{(n)}(v)-\sum_{n=0}^{\infty}\sum_{k=1}^{\infty}\frac{(axv)^{n+2k}}{2^{2k}n!(k!)^2}\times\cos^n\theta\sin^{2k}\theta\int d\Omega(\chi)\frac{d\sigma(\chi,v)}{d\Omega}\times\cos^n\chi\ \sin^{2k}\chi. \tag{B9}$$

Changing the order of summation in the second sum we can rearrange the terms in $H_1(v)$ so that we obtain just one power series with respect to parameter x obtaining the expansion for the generating function in the form

$$J(\mathbf{x},\mathbf{y})=-N_PN(\mathbf{x},\mathbf{y})\int d\mathbf{v}W_R(\mathbf{v})|\mathbf{v}|e^{a\mathbf{y}\cdot\mathbf{v}}\times\sum_{n=1}^{\infty}(axv\cos\theta)^n\left[\frac{Q^{(n)}(v)}{n!}-\sum_{k=1}^{[n/2]}\frac{B_{nk}(v)\tan^{2k}\theta}{2^{2k}(k!)^2(n-2k)!}\right], \tag{B10}$$

where $[n/2]$ denotes the entier function, while coefficients B_{nk} are defined as

$$B_{nk}(v)=\int d\Omega(\chi)\frac{d\sigma(\chi,v)}{d\Omega}\cos^n\chi\tan^{2k}\chi. \tag{B11}$$

Let us note that the subscripts in quantity B_{nk} must satisfy the requirements $n\geqslant 2$, $k\geqslant 1$, and $k\leqslant[n/2]$. It seems to be evident that coefficients B_{nk} by simple trigonometric manipulations can be expressed by the combinations of the integrals $Q^{(l)}(v)$, which are defined in Eq. (14). For the sake of completeness we give several first coefficients B_{nk} explicitly:

$$B_{21}(v)=Q^{(2)}(v), \tag{B12a}$$

$$B_{31}(v)=Q^{(3)}(v)-Q^{(1)}(v), \tag{B12b}$$

$$B_{41}(v)=Q^{(4)}(v)-Q^{(2)}(v), \tag{B12c}$$

$$B_{42}(v)=2Q^{(2)}(v)-Q^{(4)}(v). \tag{B12d}$$

The next step of evaluation of the integral (B10) consists in expressing the scalar product $\mathbf{y}\cdot\mathbf{v}$ via the corresponding angles and in performing the integration over the angle φ, which determines the orientation of the variable $\mathbf{v}$. This angle appears in the exponential still remaining in the integrand of Eq. (B8). The obtained integral is again expressible via the modified Bessel function I_0. We, however, expand the resulting functions, and, after some straightforward manipulations we obtain an expansion:

$$J(\mathbf{x},\mathbf{y})=N(\mathbf{x},\mathbf{y})\sum_{m=0}^{\infty}\sum_{n=1}^{\infty}A_{nm}(\alpha)x^n y^m, \tag{B13}$$

where $A_{nm}(\alpha)$ denotes a rather complicated coefficient

$$A_{nm}(\alpha)=-2\pi N_P a^{n+m}\cos^m\alpha\int_0^\infty dv\,W_R(\mathbf{v})|\mathbf{v}|^{3+n+m}\int_{-1}^{1}d(\cos\theta)\cos^{n+m}\theta$$
$$\times\left[\frac{Q^{(n)}(v)}{n!}-\sum_{k=1}^{[n/2]}\frac{B_{nk}(v)\tan^{2k}\theta}{2^{2k}(k!)^2(n-2k)!}\right]\sum_{p=0}^{[m/2]}\frac{(\tan\theta\,\tan\alpha)^{2p}}{2^{2p}(p!)^2(m-2p)!}. \tag{B14}$$

It is perhaps worth noting that these coefficients have the property

$$A_{nm}(\alpha)=0 \quad \text{when } n+m \text{ is odd.} \tag{B15}$$

This is so, because for $n+m$ being odd the integrand is an odd function in the variable $\cos\theta$ and the integral vanishes. Moreover, we note that the factor in square brackets in Eq. (B14) is a combination of integrals $Q^{(l)}$. Hence, the integral over $d\mathbf{v}$ will produce a combination of the collision integrals $\Omega^{(l,s)}$. The remaining integral over $\cos\theta$ gives some numerical factors. Therefore, we may say that the generating function $J(\mathbf{x},\mathbf{y})$ is expressed as a combination of collision integrals.

In the applications discussed in the main text a special role is played by coefficients $A_{nm}(\alpha=0)$. It is straightforward to find them, and several first ones read as follows:

$$A_{11}(0)=-\frac{32}{3}N_P\left(\frac{\mu}{m_A}\right)\Omega^{(1,1)}, \tag{B16a}$$

$$A_{13}(0)=-\frac{64}{15}N_P\left(\frac{\mu^2}{m_A^2}\right)\Omega^{(1,2)}, \tag{B16b}$$

$$A_{20}(0)=0, \tag{B16c}$$

$$A_{22}(0)=-\frac{64}{15}N_P\left(\frac{\mu^2}{m_A^2}\right)\Omega^{(2,2)}, \tag{B16d}$$

$$A_{31}(0)=A_{13}(0), \tag{B16e}$$

$$A_{33}(0)=-\frac{128}{105}N_P\left(\frac{\mu^3}{m_A^3}\right)\left[\Omega^{(1,3)}+\frac{2}{3}\Omega^{(3,3)}\right]. \tag{B16f}$$

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