Nonlinear spectroscopic phenomena caused by nonlinearity in the collision integral

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The paper analyzes the collision integral (CI) in the quantum transport equation of the Boltzmann type for a gas in an electromagnetic field. The nonlinearity of CI and the polarization of the gas explain the existence of nonlinear interference components in CI, components that are especially important for a one-component gas. In properties and origin these components are similar to the nonlinear interference effects in the two-photon resonance scattering or absorption of light. Specific integral conservation laws following from the generalized optical theorem are established. The collision integral is concretized for a two-level system and in the eikonal approximation. Finally, it is discovered that owing to the nonlinearity of CI a two-level system in a monochromatic field exhibits optical bistability.

1. INTRODUCTION

It is common practice to base the theoretical analysis of kinetic problems of gas spectroscopy on the quantum transport equation of the Boltzmann type.[1–4] The collision integral (CI) in this equation can take into account any nuance in the internal structure of the colliding particles, which is what is needed in spectroscopy since it is interested in transitions between time-independent states of internal motion. From this viewpoint the CI has been studied by many researchers (see, e.g., Refs. 5–15), but some important aspects remain unclear.

As is known, the CI contains the product of matrices of colliding particles and in this sense is a nonlinear operator. This is the nonlinearity mentioned in the heading of the paper. The CI nonlinearity is taken into account in many kinetic problems, say, in transport theory,[16] but spectroscopy is dominated by the idea of a linear or linearized CI: what is studied are either collisions of an absorbing (radiating, scattering) particle with the buffer gas at a given state of the latter, or collisions with excited and unexcited particles of a single species, the unexcited particles being predominant in concentration and acting as the buffer gas.

It is important that the buffer gas or its equivalent is assumed to be at equilibrium. In many cases this is so, but other situations are also of interest. For instance, the buffer gas interacts with an external field that transforms it into a nonequilibrium state. Such a case has been considered by Shalagin and the present author: polarized radiation creates in the buffer gas a preferable direction and ordering, which are transferred to the impurity gas by collisions.[17] A polarized buffer is also realized in inverted light-induced drift.[18]

The nonequilibrium nature of a collision partner becomes important in the case of a one-component gas interacting with high-power radiation, when the number of excited particles is great and a large polarization is induced in the gas. The first to notice this was Kazantsev.[4]

This paper studies the consequences of CI nonlinearity and gas polarization. Section 2 analyzes the general expression for CI that allows for off-diagonal elements of the density matrices of both colliding particles. Section 3 is devoted to the case where the CI is calculated in the eikonal approximation. In Sec. 4 the CI is concretized for the two-level model. Finally, Sec. 5 discusses the problem of saturation in two-level systems.

2. GENERAL RELATIONS

The starting point is the quantum transport equation in the one-particle density matrix in the Wigner representation for translational degrees of freedom:[14]

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\nabla\right)\rho(\mathbf{rv}t) = R + S - i[V\rho(\mathbf{rv}t) - \rho(\mathbf{rv}t)V], \quad (2.1)$$

$$S = -S^{(1)} + S^{(2)}. \quad (2.2)$$

The variables of translational motion are the position coordinator $\mathbf{r}$ and the velocity $\mathbf{v}$ equal (by definition) to the Wigner momentum divided by mass m, and $\rho(\mathbf{rv}t)$ is the density matrix in the internal degrees of freedom in the energy representation. The matrices R, S, and V describe, respectively, radiative (spontaneous) relaxation, collisions, and interactions with the external field (in the quasiclassical approximation in $\mathbf{r}$ and $\mathbf{v}$). The CI is represented in the form of outgoing and incoming terms ($S^{(1)}$ and $S^{(2)}$, respectively).

Let us denote by α and β the sets of quantum numbers of the internal degrees of freedom of the colliding particles. In the case of a buffer gas, whose state is assumed fixed, it is common to write the matrix elements $S^{(1)}(\alpha\alpha'\mathbf{rv})$ and $S^{(2)}(\alpha\alpha'\mathbf{rv})$ as

$$S^{(1)}(\alpha\alpha'\mathbf{rv}) = \nu(\alpha\alpha'|\alpha_1\alpha_2\mathbf{v})\rho(\alpha_1\alpha_2\mathbf{rv}t), \quad (2.3)$$

$$S^{(2)}(\alpha\alpha'\mathbf{rv}) = \int A(\alpha\alpha'\mathbf{v}|\alpha_1\alpha_2\mathbf{v}_1)\rho(\alpha_1\alpha_2\mathbf{rv}_1t)d\mathbf{v}_1\,, \quad (2.4)$$

where summation over repeated arguments is implied. The outgoing rates $\nu(\alpha\alpha'|\alpha_1\alpha_2\mathbf{v})$ and the kernels $A(\alpha\alpha'\mathbf{v}|\alpha_1\alpha_2\mathbf{v}_1)$ are fixed by the scattering amplitudes and the density matrix ρ_b of a collision partner. Since describing collisions with polarized particles or particles of the same species requires isolating ρ and ρ_b explicitly, it is advisable to write $S^{(1)}$ and $S^{(2)}$ in a different manner, namely

$$S^{(1)}(\alpha\alpha'\mathbf{rv}) = \int K^{(1)}(\alpha\alpha'\beta\beta|\alpha_1\alpha_2\beta_1\beta_2\mathbf{v} - \mathbf{v}_b)\rho(\alpha_1\alpha_2\mathbf{rv}t)\rho_b(\beta_1\beta_2\mathbf{rv}_bt)d\mathbf{v}_b, \quad (2.5)$$

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$$S^{(2)}(\alpha\alpha'\mathbf{r}\mathbf{v}) = \int K^{(2)}(\alpha\alpha'\beta\beta\mathbf{v}\mathbf{v}_b|\alpha_1\alpha_2\beta_1\beta_2\mathbf{v}_1\mathbf{v}_{b1})$$

$$\times\rho(\alpha_1\alpha_2\mathbf{r}\mathbf{v}_1t)\rho_b(\beta_1\beta_2\mathbf{r}\mathbf{v}_{b1}t)d\mathbf{v}_1d\mathbf{v}_bd\mathbf{v}_{b1}. \quad (2.6)$$

Here, $\mathbf{v}$, $\mathbf{v}_1$, $\mathbf{v}_b$, and $\mathbf{v}_{b1}$ are the particle velocities after and before a collision. If chemically identical particles collide, $\rho_b(\beta_1\beta_2\mathbf{r}\mathbf{v}_bt)$ differs from $\rho(\alpha_1\alpha_2\mathbf{r}\mathbf{v}t)$ only in the values of the arguments. The kernels $K^{(1)}$ and $K^{(2)}$ are expressed in terms of the matrix elements $f(\alpha\beta\mathbf{u}|\alpha_1\beta_1\mathbf{u}_1)$ of the scattering amplitude:

$$K^{(1)}(\alpha\alpha'\beta\beta|\alpha_1\alpha_2\beta_1\beta_2\mathbf{u})$$

$$= \frac{2\pi\hbar}{i\mu}\{f(\alpha\beta\mathbf{u}|\alpha_1\beta_1\mathbf{u})\delta_{\alpha'\alpha_2}\delta_{\beta\beta_2} - \delta_{\alpha\alpha_1}\delta_{\beta\beta_1}f^*(\alpha'\beta\mathbf{u}|\alpha_2\beta_2\mathbf{u})\},$$

$$\mu = \frac{mm_b}{m+m_b}, \quad E_\alpha + E_\beta = E_{\alpha_1} + E_{\beta_1},$$

$$E_{\alpha'} + E_\beta = E_{\alpha_2} + E_{\beta_2}, \quad \mathbf{u} = \mathbf{v} - \mathbf{v}_b, \quad (2.7)$$

$$K^{(2)}(\alpha\alpha'\beta\beta\mathbf{v}\mathbf{v}_b|\alpha_1\alpha_2\beta_1\beta_2\mathbf{v}_1\mathbf{v}_{b1})$$

$$= 2\delta(u^2 - u_1^2 + \frac{2}{\mu}\Delta E)\delta[\mathbf{v} - \mathbf{v}_1$$

$$- \frac{\mu}{m}(\mathbf{u} - \mathbf{u}_1)]\, f(\alpha\beta\mathbf{u}|\alpha_1\beta_1\mathbf{u}_1)f^*(\alpha'\beta\mathbf{u}|\alpha_2\beta_2\mathbf{u}_1),$$

$$\Delta E = E_\alpha + E_\beta - E_{\alpha_1} - E_{\beta_1} = E_{\alpha'} + E_\beta - E_{\alpha_2} - E_{\beta_2},$$

$$\mathbf{u} = \mathbf{v} - \mathbf{v}_b, \quad \mathbf{u}_1 = \mathbf{v}_1 - \mathbf{v}_{b1}. \quad (2.8)$$

Here E_α and E_β are the energies of the time-independent states α and β; the quantities μ, m, and m_b are the reduced mass and the masses of particles, and $\mathbf{u}$ and $\mathbf{u}_1$ are the relative particle velocities after and before collision.

Equation (2.8) makes it possible to obtain the relation

$$\omega_{\alpha\alpha'} = \omega_{\alpha_1\alpha_2} + \omega_{\beta_1\beta_2}, \quad (2.9)$$

which resembles the one between the photon frequencies and the transition frequencies of the scattering system in Raman scattering of light. An important difference is that in the case at hand all three frequencies are fixed by the internal structure of the colliding particles. If $\omega_{\beta_1\beta_2} = 0$, condition (2.9) is met, for instance, at $E_{\alpha_1} = E_\alpha$ and $E_{\alpha_2} = E_{\alpha'}$, and the CI contains $\rho(\alpha\alpha'\mathbf{v})$ and $\rho(\alpha\alpha'\mathbf{v}_1)$ in $S^{(1)}$ and $S^{(2)}$, respectively (to simplify the notation we drop the arguments $\mathbf{r}$ and t). But if $\omega_{\beta_1\beta_2} \neq 0$, condition (2.9) is met for different particles only if the Bohr frequencies accidentally coincide, which occurs only as an exception. In the absence of such a coincidence the optical coherence of the buffer particles contributes nothing to the CI, which will contain only the diagonal elements of the density matrix ρ_b in the energy.

In contrast to what has just been said, condition (2.9) can be met automatically for particles of a single species since here $\rho_b(\beta_1\beta_2\mathbf{v}_b)$ and $\rho(\alpha_1\alpha_2\mathbf{v})$ are elements of a single matrix and the quantities α and β run through the same values. Thus, in collisions of particles of a single species the contribution of the optical coherence of a collision partner is a rule rather than an exception. Hence, in this paper we have focused on just such a case, known in jargon as "proper-pressure broadening."

For purely magnetic coherence we have $\omega_{\alpha\alpha'} = \omega_{\alpha_1\alpha_2} = \omega_{\beta_1\beta_2} = 0$, and condition (2.9) imposes no restrictions. Hence, for instance, the evolutions of magnetic coherence due to collisions with the gas proper or with a buffer gas are perfectly similar.

We use another simplifying assumption, the model of nondegenerate states. In this model, optical coherence manifests itself in collisions quite clearly. Degeneracy and magnetic coherence complicate the analysis considerably, and it has proved expedient to allow for these factors when the problem is stated more specifically (e.g., when the interaction potential, the number of levels, and others have been determined).

Let us start by studying the off-diagonal elements of the outgoing term ($\alpha\neq\alpha'$). Condition (2.9) simplifies because of deltas in Eq. (2.7),

$$\omega_{\alpha\alpha_1} = \omega_{\beta_1\beta}, \quad \omega_{\alpha'\alpha_2} = \omega_{\beta_2\beta}, \quad (2.9a)$$

but the ideas concerning the difference between collisions of particles of the same species and collisions of particles of different species remain valid. It is natural to break down the outgoing term into three parts,

$$S^{(1)}(\alpha\alpha'\mathbf{v}) = S_e^{(1)}(\alpha\alpha'\mathbf{v}) + S_{ex1}^{(1)}(\alpha\alpha'\mathbf{v}) + S_{ex2}^{(1)}(\alpha\alpha'\mathbf{v}), \quad (2.10)$$

$$S_e^{(1)}(\alpha\alpha'\mathbf{v}) = \nu(\alpha\alpha'\mathbf{v})\rho(\alpha\alpha'\mathbf{v}), \quad \nu(\alpha\alpha'\mathbf{v}) = (2\pi\hbar/i\mu)$$

$$\times\sum_\beta\int\{f(\alpha\beta\mathbf{u}|\alpha\beta\mathbf{u}) - f^*(\alpha'\beta\mathbf{u}|\alpha'\beta\mathbf{u})\}\rho(\beta\beta\mathbf{v}_b)d\mathbf{v}_b, \quad (2.11)$$

$$S_{ex1}^{(1)}(\alpha\alpha'\mathbf{v}) = (2\pi\hbar/i\mu)\int\{f(\alpha\alpha'\mathbf{u}|\alpha'\alpha\mathbf{u})\rho(\alpha'\alpha'\mathbf{v})$$

$$- \rho(\alpha\alpha\mathbf{v})f^*(\alpha'\alpha\mathbf{u}|\alpha\alpha'\mathbf{u})\}\rho(\alpha\alpha'\mathbf{v}_b)d\mathbf{v}_b, \quad (2.12)$$

$$S_{ex2}^{(1)}(\alpha\alpha'\mathbf{v}) = (2\pi\hbar/i\mu)\int\{f(\alpha\beta\mathbf{u}|\beta\alpha\mathbf{u})\rho(\beta\alpha'\mathbf{v})\rho(\alpha\beta\mathbf{v}_b)$$

$$- \rho(\alpha\beta\mathbf{v})\rho(\beta\alpha'\mathbf{v}_b)f^*(\alpha'\beta\mathbf{u}|\beta\alpha'\mathbf{u})\}d\mathbf{v}_b. \quad (2.13)$$

The term $S_e^{(1)}$ represents scattering that is elastic for each colliding particle and in what follows is called the elastic part of $S^{(1)}$. The terms $S_{ex1}^{(1)}$ and $S_{ex2}^{(1)}$ represent scattering that is elastic for a quasimolecule consisting of the colliding particles as a whole and not for each particle separately, that is, scattering with excitation exchange, with $S_{ex1}^{(1)}(\alpha\alpha'\mathbf{v})$ incorporating excitation exchange in which only states α and α' participate, and $S_{ex2}^{(1)}(\alpha\alpha'\mathbf{v})$ excitation exchange between states $\alpha\beta$ and $\alpha'\beta$ ($\alpha\neq\alpha'$). The terms $S_{ex1}^{(1)}$ and $S_{ex2}^{(1)}$ will be called the exchange part of $S^{(1)}$.

The elastic part $S_e^{(1)}(\alpha\alpha'\mathbf{v})$ is proportional to the element $\rho(\alpha\alpha'\mathbf{v})$ with the same arguments α, α' and $\mathbf{v}$, and the proportionality factor $\nu(\alpha\alpha'\mathbf{v})$ has the meaning of an outgoing frequency that depends on $\rho(\beta\beta\mathbf{v}_b)$. In other words, $S_e^{(1)}(\alpha\alpha'\mathbf{v})$ is expressed in terms of $\rho(\alpha\alpha'\mathbf{v})$ and the populations $\rho(\beta\beta\mathbf{v}_b)$. In this respect $S_e^{(1)}(\alpha\alpha'\mathbf{v})$ is similar to an off-diagonal outgoing term in a linearized CI. The important difference, however, is that in the case of a buffer gas the outgoing frequency

$$\nu(\alpha\alpha'|\alpha\alpha'\mathbf{v}) = (2\pi\hbar/i\mu)$$

$$\times\sum_\beta\int\{f(\alpha\beta\mathbf{u}|\alpha\beta\mathbf{u}) - f^*(\alpha'\beta\mathbf{u}|\alpha'\beta\mathbf{u})\}\rho_b(\beta\beta\mathbf{v}_b)d\mathbf{v}_b, \quad (2.14)$$

contains not $\rho(\beta\beta\mathbf{v})$ but the elements $\rho_b(\beta\beta\mathbf{v}_b)$ of the density matrix of the buffer gas, which are usually considered given (equilibrium) values.

Let us separate the imaginary and real parts of the forward scattering amplitudes in (2.11). The imaginary parts are expressed, by the optical theorem (see, e.g., Refs. 19 and 20, §12), in terms of the total cross sections of scattering in states α and β:

$$\operatorname{Im} f(\alpha\beta\mathbf{u}|\alpha\beta\mathbf{u}) = \frac{\mu}{2\pi\hbar}\sum_{\alpha_2\beta_2}\int |f(\alpha_2\beta_2\mathbf{u}_2|\alpha\beta\mathbf{u})|^2 \times\delta\left(u_2^2 - u^2 + \frac{2}{\mu}\delta E\right)d\mathbf{u}_2,$$

$$\delta E = E_{\alpha_2} + E_{\beta_2} - E_\alpha - E_\beta, \tag{2.15}$$

and similarly for scattering in states α' and β. The same quantities are present in the outgoing diagonal terms $S_e^{(1)}(\alpha\alpha\mathbf{v})$ and $S_e^{(1)}(\alpha'\alpha'\mathbf{v})$ [see Eq. (2.20) below]. Thus, generally the real part of the outgoing frequency in (2.11) is the half-sum of the diagonal outgoing rates:

$$\operatorname{Re}\nu(\alpha\alpha'\mathrm{v}) = \frac{1}{2}\left[\nu(\alpha\alpha\mathrm{v}) + \nu(\alpha'\alpha'\mathrm{v})\right]. \tag{2.16}$$

This property is also present in the case of a buffer gas.[14]

The difference of the real parts of the scattering amplitudes in (2.11),

$$\operatorname{Re}[f(\alpha\beta\mathbf{u}|\alpha\beta\mathbf{u}) - f(\alpha'\beta\mathbf{u}|\alpha'\beta u)]$$

reflects a possible difference in the scattering in states α and α'. In terms of line-broadening theory, this difference causes the line shift due to the phase modulation of an atomic oscillator.

The term $S_{\mathrm{ex1}}^{(1)}(\alpha\alpha'\mathbf{v})$ incorporates the excitation exchange between levels α and α'. As with the elastic part, this term contains an element $\rho(\alpha\alpha'\mathbf{v}_b)$ with the same numbers α and α' but with the velocity $\mathbf{v}_b$ (rather than $\mathbf{v}$) of the collision partner, with averaging performed over $\mathbf{v}_b$. But the populations $\rho(\alpha\alpha\mathbf{v})$ and $\rho(\alpha'\alpha'\mathbf{v})$ depend on $\mathbf{v}$. In collisions with a buffer gas these properties are inherent in the incoming term, as is especially clear in the model of strong collisions. Here, however, the outgoing term possesses them, and it is the CI nonlinearity that guarantees their existence.

The expression inside the braces in (2.12) can be written as (we drop the argument u in the scattering amplitudes)

$$\begin{aligned} &f(\alpha\alpha'|\alpha'\alpha)\rho(\alpha'\alpha'\mathrm{v}) - \rho(\alpha\alpha\mathrm{v})f^*(\alpha'\alpha|\alpha\alpha') \\ &= \frac{1}{2}[f(\alpha\alpha'|\alpha'\alpha) - f^*(\alpha'\alpha|\alpha\alpha')][\rho(\alpha'\alpha'\mathrm{v}) + \rho(\alpha\alpha\mathrm{v})] \\ &+ \frac{1}{2}[f(\alpha\alpha'|\alpha'\alpha) + f^*(\alpha'\alpha|\alpha\alpha')][\rho(\alpha'\alpha'\mathrm{v}) - \rho(\alpha\alpha\mathrm{v})]. \end{aligned} \tag{2.17}$$

Using the generalized optical theorem,[21] we can transform the difference of the amplitudes on the right-hand side as follows:

$$\begin{aligned} &f(\alpha\alpha'\mathbf{u}|\alpha'\alpha\mathbf{u}) - f^*(\alpha'\alpha\mathbf{u}|\alpha\alpha'\mathbf{u}) = \frac{i\mu}{\pi\hbar} \\ &\times\sum_{\alpha_2\beta_2}\int f^*(\alpha_2\beta_2\mathbf{u}_2|\alpha\alpha'\mathbf{u})f(\alpha_2\beta_2\mathbf{u}_2|\alpha'\alpha\mathbf{u}) \\ &\times\delta\left(u_2^2 - u^2 + \frac{2}{\mu}\delta E\right)d\mathbf{u}_2. \end{aligned} \tag{2.18}$$

On the right-hand side cross sections have appeared that describe the interference of the processes

$$\alpha\alpha'\mathbf{u}\rightarrow\alpha_2\beta_2\mathbf{u}_2 \text{ and } \alpha'\alpha\mathbf{u}\rightarrow\alpha_2\beta_2\mathbf{u}_2$$

with the same final state. Equation (2.17) is convenient at high saturation levels, when the difference and sum of the populations of levels α and α' are "good" characteristics.

In contrast to the "two-level" term $S_{\mathrm{ex1}}^{(1)}(\alpha\alpha'\mathbf{v})$, the term $S_{\mathrm{ex2}}^{(1)}(\alpha\alpha'\mathbf{v})$ takes into account excitation exchange in the triplets of levels α, α', and β ($\beta\neq\alpha,\alpha'$). What is important is that $S_{\mathrm{ex2}}^{(1)}$ incorporates the polarizations of the $\beta\alpha'$ and $\alpha\beta$ transitions rather than the level populations. Figure 1 illustrates the processes that determine $S_{\mathrm{ex2}}^{(1)}(\alpha\alpha'\mathbf{v})$. The first term on the right-hand side of (2.13) corresponds to Fig. 1a, in which two level triplets belong to two colliding atoms (velocities $\mathbf{v}$ and $\mathbf{v}_b$). Single arrows designate the polarizations $\rho(\beta\alpha'\mathbf{v})$ and $\rho(\alpha\beta\mathbf{v})$; a double arrow stands for the "generated" polarization of the $\alpha\alpha'$ transition or the polarization for which relaxation is considered. The dashed arrows correspond to collision excitation exchange. Figure 1b illustrates similar processes related to the second term in (2.13). If we do not specify to which of the two atoms one or another polarization belongs, and if we combine Figs. 1a and 1b, and drop the dashed arrows, we arrive at Fig. 1c, which expresses the combination of polarizations of atoms and resembles the diagram for Raman scattering of light. We can say, therefore, that the term $S_{\mathrm{ex2}}^{(1)}(\alpha\alpha'\mathbf{v})$ describes polarization relaxation in an $\alpha\alpha'$ transition of a nonpopulation (interference) origin, that is, relaxation caused by the presence of polarizations in other, adjacent, transitions, whereas $S_e^{(1)}(\alpha\alpha'\mathbf{v})$ and $S_{\mathrm{ex1}}^{(1)}(\alpha\alpha'\mathbf{v})$ contained the off-diagonal elements $\rho(\alpha\alpha'\mathbf{v})$ and $\rho(\alpha\alpha'\mathbf{v}_b)$ for the same transition $\alpha\alpha'$ and population of levels α, α', and β. The exchange and interference effects manifest themselves most vividly in the diagonal elements $S^{(1)}(\alpha\alpha\mathbf{v})$.

Here is the explicit expression for $S^{(1)}(\alpha\alpha\mathbf{v})$:

$$S^{(1)}(\alpha\alpha\mathrm{v}) = S_e^{(1)}(\alpha\alpha\mathrm{v}) + S_{ex}^{(1)}(\alpha\alpha\mathrm{v}), \tag{2.19}$$

$$S_e^{(1)}(\alpha\alpha\mathrm{v}) = \nu(\alpha\alpha\mathrm{v})\rho(\alpha\alpha\mathrm{v}),$$

$$\nu(\alpha\alpha\mathrm{v}) = \frac{4\pi\hbar}{\mu}\sum_{\beta}\int \operatorname{Im}[f(\alpha\beta\mathbf{u}|\alpha\beta\mathbf{u})]\,\rho(\beta\beta\mathbf{v}_b)d\mathbf{v}_b, \tag{2.20}$$

$$S_{ex}^{(1)}(\alpha\alpha\mathrm{v}) = \frac{4\pi\hbar}{\mu}\sum_{\beta\neq\alpha}\int \operatorname{Im}[f(\alpha\beta\mathbf{u}|\beta\alpha\mathbf{u})\rho(\beta\alpha\mathrm{v})\rho(\alpha\beta\mathbf{v}_b)]\,d\mathbf{v}_b. \tag{2.21}$$

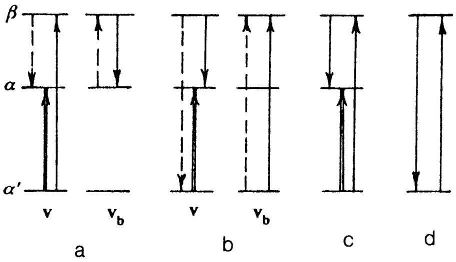

FIG. 1. Diagrams of radiative correlations and collision transitions involving colliding particles.

The elastic part $S_e^{(1)}(\alpha\alpha\mathbf{v})$ contains the amplitudes of scattering which is elastic for each particle, and the populations $\rho(\alpha\alpha\mathbf{v})$ and $\rho(\beta\beta\mathbf{v}_b)$ of levels α and β, that is, this part is quite traditional. In the exchange part $S_{\text{ex}}^{(1)}(\alpha\alpha\mathbf{v})$, however, there are amplitudes of forward scattering with resonant excitation exchange and only the off-diagonal elements of the density matrices, $\rho(\beta\alpha\mathbf{v})$ and $\rho(\alpha\beta\mathbf{v}_b)$ $(\beta\neq\alpha)$. Thus, $S_{\text{ex}}^{(1)}(\alpha\alpha\mathbf{v})$ describes the level-α population relaxation caused by the polarization of $\alpha\beta$ transitions incorporating level α. Such polarization (or interference or coherent) relaxation was first derived by Kazantsev[4] for the case of two levels.

The diagram representing the $S_{\text{ex}}^{(1)}(\alpha\alpha\mathbf{v})$ term is obtained by merging levels α and α' (Fig. 1d). Ultimately, the analogy with Raman scattering transforms into the analogy with resonance fluorescence.

Let us now turn to the incoming term $S^{(2)}(\alpha\alpha'\mathbf{v})$. It contains the fivefold sum over α_1, α_2, β, β_1, and β_2. Condition (2.9) decreases the multiplicity of summation. To clarify this question we write condition (2.9) as

$$E_\alpha + E_{\alpha_2} + E_{\beta_2} = E_{\alpha'} + E_{\alpha_1} + E_{\beta_1}. \tag{2.9b}$$

Let us assume that in the atomic energy spectrum there are no coincidences of the type $E_\alpha = E_{\alpha_1} + E_{\beta_1}$, etc., and that condition (2.9b) is met only if separate terms on the right- and left-hand sides are equal. Then the multiplicity of summation in (2.6) decreases to three.

To simplify the formulas we use angle brackets to denote integration over $\mathbf{v}_1$, $\mathbf{v}_b$, and $\mathbf{v}_{b1}$ with a weight function equal to a double Dirac delta function and, in addition, drop the arguments $\mathbf{u}$ and $\mathbf{u}_1$ in the scattering amplitudes so as not to divert attention from the quantum numbers.

By direct sorting we can easily determine that $S^{(2)}(\alpha\alpha'\mathbf{v})$, $\alpha\neq\alpha'$, consists of four terms,

$$S^{(2)}(\alpha\alpha'\mathbf{v}) = \sum_{l=1}^{4} S_l^{(2)}(\alpha\alpha'\mathbf{v}), \tag{2.22}$$

with each term containing a double sum:

$$S_1^{(2)}(\alpha\alpha'\mathbf{v}) = \sum_{\beta\beta_1} \langle \rho(\alpha\alpha'\mathbf{v}_1)\rho(\beta_1\beta_1\mathbf{v}_{b1}) \times f(\alpha\beta|\alpha\beta_1) f^*(\alpha'\beta|\alpha'\beta_1)\delta(\Delta_{\beta\beta_1})\rangle, \tag{2.23}$$

$$S_2^{(2)}(\alpha\alpha'\mathbf{v}) = \sum_{\beta\alpha_1} \langle \rho(\alpha_1\alpha_1\mathbf{v}_1)\rho(\alpha\alpha'\mathbf{v}_{b1}) \times f(\alpha\beta|\alpha_1\alpha) f^*(\alpha'\beta|\alpha_1\alpha')\delta(\Delta_{\beta\alpha_1})\rangle, \tag{2.24}$$

$$S_3^{(2)}(\alpha\alpha'\mathbf{v}) = \sum_{\beta\alpha_1}{}' \langle \rho(\alpha\alpha_1\mathbf{v}_1)\rho(\alpha_1\alpha'\mathbf{v}_{b1}) \times f(\alpha\beta|\alpha\alpha_1) f^*(\alpha'\beta|\alpha_1\alpha')\delta(\Delta_{\beta\alpha_1})\rangle, \tag{2.25}$$

$$S_4^{(2)}(\alpha\alpha'\mathbf{v}) = \sum_{\beta\alpha_1}{}' \langle \rho(\alpha_1\alpha'\mathbf{v}_1)\rho(\alpha\alpha_1\mathbf{v}_{b1}) \times f(\alpha\beta|\alpha_1\alpha) f^*(\alpha'\beta|\alpha'\alpha_1)\delta(\Delta_{\beta\alpha_1})\rangle, \tag{2.26}$$

$$\Delta_{\beta\beta_1} = u^2 - u_1^2 + \frac{2}{\mu}(E_\beta - E_{\beta_1}),$$

$$\Delta_{\beta\alpha_1} = u^2 - u_1^2 + \frac{2}{\mu}(E_\beta - E_{\alpha_1}). \tag{2.27}$$

The primes on the sums means that in them $\alpha_1\neq\alpha$ and $\alpha_1\neq\alpha'$.

The terms in the double sums with equal summation indices ($\beta_1=\beta$ or $\alpha_1=\beta$) correspond to elastic scattering and resonant exchange, and in such terms the delta function contains only $u^2-u_1^2$. The other terms are related to inelastic scattering and quasiresonant excitation exchange.

The sum $S_1^{(2)}$ describes the elastic scattering of a particle whose velocity is $\mathbf{v}_1$ and the elastic or quasielastic scattering of the particle's collision partner (velocity $\mathbf{v}_{b1}$). This explains the important fact that $S_1^{(2)}(\alpha\alpha'\mathbf{v})$ contains $\rho(\alpha\alpha'\mathbf{v}_1)$. The sum $S_2^{(2)}$ incorporates excitation exchange. A common factor in all the terms in this sum is $\rho(\alpha\alpha'\mathbf{v}_{b1})$. The sums $S_3^{(2)}$ and $S_4^{(2)}$ result from the interference of scattering with excitation exchange and the elastic scattering of the $\mathbf{v}_1$-particle. As with $S_{\text{ex}2}^{(1)}$ in the outgoing term, the sums $S_3^{(2)}(\alpha\alpha'\mathbf{v})$ and $S_4^{(2)}(\alpha\alpha'\mathbf{v})$ are "generated" by polarizations in the $\alpha\alpha_1$ and $\alpha_1\alpha'$ transitions adjacent to the $\alpha\alpha'$ transition rather than by populations. Hence, Fig. 1c can serve as an illustration to processes contributing to $S_3^{(2)}(\alpha\alpha'\mathbf{v})$ and $S_4^{(2)}(\alpha\alpha'\mathbf{v})$. The only difference is that for the incoming term single arrows indicate the polarizations of atoms prior to collision (velocities $\mathbf{v}_1$ and $\mathbf{v}_{b1}$) while for the outgoing term the velocities are $\mathbf{v}$ and $\mathbf{v}_b$ (forward scattering).

In the case of the diagonal element $S^{(2)}(\alpha\alpha\mathbf{v})$ condition (2.9) simplifies,

$$E_{\alpha_2} + E_{\beta_2} = E_{\alpha_1} + E_{\beta_1} \tag{2.9c}$$

and the incoming term breaks down into two triple sums,

$$S_1^{(2)}(\alpha\alpha\mathbf{v}) = \sum_{\alpha_1\beta\beta_1} \langle \rho(\alpha_1\alpha_1\mathbf{v}_1)\rho(\beta_1\beta_1\mathbf{v}_{b1}) |f(\alpha\beta|\alpha_1\beta_1)|^2 \delta(\Delta)\rangle, \tag{2.28}$$

$$S_2^{(2)}(\alpha\alpha\mathbf{v}) = \sum_{\substack{\alpha_1\beta\beta_1\\ \beta_1\neq\alpha_1}} \langle \rho(\alpha_1\beta_1\mathbf{v}_1)\rho(\beta_1\alpha_1\mathbf{v}_{b1}) \times f(\alpha\beta|\alpha_1\beta_1) f^*(\alpha\beta|\beta_1\alpha_1)\delta(\Delta)\rangle, \tag{2.29}$$

$$\Delta = u^2 - u_1^2 + \frac{2}{\mu}(E_\alpha + E_\beta - E_{\alpha_1} - E_{\beta_1}). \tag{2.30}$$

The part $S_1^{(2)}(\alpha\alpha\mathbf{v})$ of the diagonal incoming term incorporates the diagonal elements of the density matrix and the squares of the absolute values of the $\alpha_1\beta_1\mathbf{u}_1\to\alpha\beta\mathbf{u}$ scattering amplitudes. In other words, $S_1^{(2)}(\alpha\alpha\mathbf{v})$ describes "ordinary" collision transitions from all states onto level α of particles with a velocity $\mathbf{v}$. The second part, $S_2^{(2)}(\alpha\alpha\mathbf{v})$, describes the collision "arrival" onto level α due to polarization of $\alpha_1\beta_1$ transitions of the colliding atoms and is a corollary of the interference of two scattering processes, $\alpha_1\beta_1\mathbf{u}_1\to\alpha\beta\mathbf{u}$ and $\beta_1\alpha_1\mathbf{u}_1\to\alpha\beta\mathbf{u}$, with a common final state, $\alpha\beta\mathbf{u}$. As before, the interference properties of the incoming part of the process are closely related to resonant and quasiresonant excitation exchange. Note that $S_2^{(2)}$ contains the product of off-diagonal elements of the density matrix on one and the same transition of the colliding particles.

Up to this point we have emphasized the equal status of the density matrices, which represent the colliding particles in the CI and enter into as factors. Now let us examine the differences stemming from their dependence on the velocities $\mathbf{v}$, $\mathbf{v}_b$, $\mathbf{v}_{b1}$ and $\mathbf{v}_1$. For the outgoing part of the process it is important that there is integration over $\mathbf{v}_b$, which influences only the factor $\rho(\beta_1\beta_2\mathbf{v}_b)$. The forward scattering amplitudes, which are fairly smooth functions of the relative velocity $\mathbf{u}=\mathbf{v}-\mathbf{v}_b$, act in such averaging as weight factors. In the incoming term integration is carried out over the velocity arguments $\mathbf{v}_1$ and $\mathbf{v}_{b1}$ of both factors, which depend on $\mathbf{v}_1$ and $\mathbf{v}_{b1}$ quite differently. The law of momentum conservation implies

$$\mathbf{v}_1-\mathbf{v}=\frac{1}{2}(\mathbf{u}_1-\mathbf{u}),\qquad \mathbf{v}_{b1}-\mathbf{v}=-\frac{1}{2}(\mathbf{u}_1+\mathbf{u}) \tag{2.31}$$

(like particles, $\mu=m/2$). For the most important elastic and resonant processes,

$$(\mathbf{u}_1-\mathbf{u})(\mathbf{u}_1+\mathbf{u})=u_1^2-u^2=0,$$

and, hence, $\mathbf{v}_1-\mathbf{v}$ is orthogonal to $\mathbf{v}_{b1}-\mathbf{v}$. The second difference is important for small-angle scattering, where $|\mathbf{u}-\mathbf{u}_1|$ is small but $|\mathbf{u}+\mathbf{u}_1|\approx 2|\mathbf{u}|$ is not. This fact manifests itself most vividly in the "relaxation-constants model," in which no variation in velocity is taken into account and integration over $\mathbf{v}_1$ and $\mathbf{v}_{b1}$ is not present while integration over $\mathbf{v}_b$ remains and singles out $\rho(\beta_1\beta_2\mathbf{v}_b)$ to the same extent as it does in the outgoing term (see Sec. 3).

It is expedient to bear in mind that in collisions of like particles we have

$$f(\alpha\beta\mathbf{u}\,|\,\alpha_1\beta_1\mathbf{u}_1)=f(\beta\alpha-\mathbf{u}\,|\,\beta_1\alpha_1-\mathbf{u}_1), \tag{2.32}$$

which explicitly reflects the fact that the colliding particles are equivalent.

There is also an additional conservation law characteristic of interference phenomena of any kind. It is known that

$$\sum_\alpha\int d\mathbf{v}S(\alpha\alpha\mathbf{v})=0, \tag{2.33}$$

which means that the number of particles in collision processes is conserved. The optical theorem makes it possible to obtain a more explicit conservation law,

$$\sum_\alpha\int d\mathbf{v}S_e^{(1)}(\alpha\alpha\mathbf{v})=\sum_\alpha\int d\mathbf{v}S_1^{(2)}(\alpha\alpha\mathbf{v}). \tag{2.34}$$

Combining (2.33) and (2.34) yields

$$\sum_\alpha\int d\mathbf{v}S_{ex}^{(1)}(\alpha\alpha\mathbf{v})=\sum_\alpha\int d\mathbf{v}S_2^{(2)}(\alpha\alpha\mathbf{v}), \tag{2.35}$$

whose validity can also be proved directly by employing the generalized optical theorem and the symmetry relations (2.32).

The reader will recall that $S_e^{(1)}$ and $S_1^{(2)}$ contain products of the populations of the colliding particles, while $S_{\text{ex}}^{(1)}$ and $S_2^{(2)}$ contain products of the off-diagonal elements of the density matrices. Allowing for a certain leeway, we can say that the number of particles is conserved both in "collisions of populations (of particles)" and in "collisions of dipoles" [Eqs. (2.34) and (2.35)]. In the absence of external fields there are no induced dipoles and only Eq. (2.34) is important. An external field changes the populations (the saturation effect) and induces polarization, but Eq. (2.34) retains its appearance, while for polarization there is its own conservation law (2.35).

Suppose that for some reason inelastic processes and nonresonant excitation exchange can be ignored. Then collisions do not change the number of particles on a level:

$$\int S_e^{(1)}(\alpha\alpha\mathbf{v})d\mathbf{v}=\int S_1^{(2)}(\alpha\alpha\mathbf{v})d\mathbf{v}, \tag{2.36}$$

$$\int S_{ex}^{(1)}(\alpha\alpha\mathbf{v})d\mathbf{v}=\int S_2^{(2)}(\alpha\alpha\mathbf{v})d\mathbf{v}. \tag{2.37}$$

On the whole the situation is almost trivial: by its very definition elastic scattering does not change the number of particles on a level, and in resonant excitation exchange the departing particle is replaced on the same level by its collision partner. What is worth noting is also that the population and interference parts in the integrals of $S(\alpha\alpha\mathbf{v})$ with respect to $\mathbf{v}$ vanish separately.

An analysis of the matrix structure of the CI leads to the following conclusion. For collisions with a buffer gas the following mnemonic rule is valid: "like is excited by like." According to this rule, a population CI is formed only by the populations $\rho(\alpha\alpha\mathbf{v})$ and $\rho(\alpha_1\alpha_1\mathbf{v}_1)$, and the CI for the polarization of the $\alpha\alpha'$ transitions is formed only by the polarizations $\rho(\alpha\alpha'\mathbf{v})$ and $\rho(\alpha\alpha'\mathbf{v}_1)$. In both cases the buffer gas contributes only its populations to the CI. On the other hand, in the collision of like particles polarized by an external field the above rule does not hold, and relaxation or excitation of particles occurs because of the presence of off-diagonal elements of the density matrices. For the diagonal CI to contain such an interference term, polarization must be induced at least on one transition, and for off-diagonal CI polarization must be induced on two transitions adjacent to the $\alpha\alpha'$ transition.

3. THE EIKONAL APPROXIMATION

Collision-integral theory often uses scattering amplitude calculated in the eikonal approximation, according to which (see, e.g., Refs. 12, 14, 15, and 19)

$$f(\alpha\beta\mathbf{u}\,|\,\alpha_1\beta_1\mathbf{u}_1)=\frac{\mu u}{2\pi\hbar i}\int F(\alpha\beta\,|\,\alpha_1\beta_1\mathbf{u}_1\mathbf{R})\exp\left[\frac{i\mu(\mathbf{u}_1-\mathbf{u})\mathbf{R}}{\hbar}\right]d\mathbf{R}, \tag{3.1}$$

$$F(\alpha\beta\,|\,\alpha_1\beta_1\mathbf{u}_1\mathbf{R})=S(\alpha\beta\,|\,\alpha_1\beta_1\mathbf{u}_1\mathbf{R})-\delta_{\alpha\alpha_1}\delta_{\beta\beta_1}, \tag{3.2}$$

where integration over the impact parameter $\mathbf{R}$ is performed in a plane perpendicular to $\mathbf{u}_1$ and separated from the interaction region by a distance $(\mathbf{r}\mathbf{u}_1)/u_1$ such that

$$\lambda\!\!\!^{-}\ll\rho_W\ll\mathbf{r}\hat{\mathbf{u}}_1\ll\rho_W^2/\lambda\!\!\!^{-},\qquad \hat{\mathbf{u}}_1=\mathbf{u}_1/u_1, \tag{3.3}$$

where ρ_W and $\lambda\!\!\!^{-}$ are the radius of the interaction region (the Weisskopf radius) and the de Broglie wavelength. We restrict our discussion to elastic and resonant processes. The functions $S(\alpha\beta\,|\,\alpha_1\beta_1\mathbf{u}_1\mathbf{R})$ are solutions of the system of equations (the approximation of straight trajectories)

$$\hat{\mathbf{u}}_1\nabla S(\alpha\beta\,|\,\alpha_1\beta_1\mathbf{u}_1\mathbf{r})=-\frac{i}{\hbar}\sum_{\alpha_2\beta_2}W(\alpha\beta\,|\,\alpha_2\beta_2\mathbf{r})S(\alpha_2\beta_2\,|\,\alpha_1\beta_1\mathbf{u}_1\mathbf{r}) \tag{3.4}$$

in the plane just mentioned, and satisfy the boundary conditions

$$S(\alpha\beta|\alpha_1\beta_1\mathbf{u}_1\mathbf{r}) = \delta_{\alpha\alpha_1}\delta_{\beta\beta_1}, \qquad \mathbf{r}\hat{\mathbf{u}}_1 \to -\infty \tag{3.5}$$

$[W(\alpha\beta|\alpha_1\beta_1\mathbf{r})$ is the matrix element of the interaction Hamiltonian of the colliding particles]. In approximation (3.1) we get

$$\begin{gathered} K^{(1)}(\alpha\alpha'\beta\beta|\alpha_1\alpha_2\beta_1\beta_2\mathbf{u}) \\ = -\int\{F(\alpha\beta|\alpha_1\beta_1\mathbf{u}\mathbf{R})\delta_{\alpha'\alpha_2}\delta_{\beta\beta_2} + \delta_{\alpha\alpha_1}\delta_{\beta\beta_1}F^*(\alpha'\beta|\alpha_2\beta_2\mathbf{u}\mathbf{R})\}d\mathbf{R}, \end{gathered} \tag{3.6}$$

$$\begin{gathered} K^{(2)}(\alpha\alpha'\beta\beta\mathbf{v}\mathbf{v}_b|\alpha_1\alpha_2\beta_1\beta_2\mathbf{v}_1\mathbf{v}_{b1}) \\ = \frac{1}{2}\left(\frac{\mu u}{\pi\hbar}\right)^2 \delta\left[\mathbf{v} - \mathbf{v}_1 - \frac{\mu}{m}(\mathbf{u} - \mathbf{u}_1)\right]\delta(u^2 - u_1^2) \\ \times\int F(\alpha\beta|\alpha_1\beta_1\mathbf{u}_1\mathbf{R}_1)F^*(\alpha'\beta|\alpha_2\beta_2\mathbf{u}_1\mathbf{R}_2) \\ \times\exp\left[\frac{i\mu(\mathbf{u}_1 - \mathbf{u})(\mathbf{R}_1 - \mathbf{R}_2)}{\hbar}\right]d\mathbf{R}_1 d\mathbf{R}_2. \end{gathered} \tag{3.7}$$

Introducing the scattering matrix $S(\alpha\beta|\alpha_1\beta_1\mathbf{u}_1\mathbf{R})$ by (3.2) makes it possible to write the CI in the form[12]

$$\begin{aligned} S(\alpha\alpha'\mathbf{v}) = 2\int & d\mathbf{v}_1 d\mathbf{v}_b d\mathbf{v}_{b1} d\mathbf{R}_1 d\mathbf{R}_2 u\delta(u^2 \\ & - u_1^2)\delta\left[\mathbf{v} - \mathbf{v}_1 - \frac{\mu}{m}(\mathbf{u} - \mathbf{u}_1)\right] \\ & \times\{S(\alpha\beta|\alpha_1\beta_1\mathbf{u}_1\mathbf{R}_1)S^*(\alpha'\beta|\alpha_2\beta_2\mathbf{u}_1\mathbf{R}_2) \\ & - \delta_{\alpha\alpha_1}\delta_{\alpha'\alpha_2}\delta_{\beta\beta_1}\delta_{\beta\beta_2}\}\exp[i\mu(\mathbf{u}_1 - \mathbf{u})(\mathbf{R}_1 \\ & - \mathbf{R}_2)/\hbar]\,\rho(\alpha_1\alpha_2\mathbf{v}_1)\rho(\beta_1\beta_2\mathbf{v}_{b1}). \end{aligned} \tag{3.8}$$

In its dependence on the scattering matrix Eq. (3.8) resembles the expression for the scattering amplitude [compare the expression in braces in (3.8) with Eqs. (3.1) and (3.2)] and, hence, agree with intuitive ideas. Sometimes the terms with the product SS^* and with the delta symbols are called the incoming and outgoing terms, respectively (see, e.g., Ref. 4). Note, however, that the integrals of SS^* and of the delta terms are infinite and have no meaning of their own. Only their difference has a definite physical meaning. The reason for this is that the S matrix describes the entire scattered wave while F describes the difference of the scattered and incident waves.

Equation (3.8) for the CI is less convenient than (3.6) and (3.7) also because it combines the terms $S^{(1)}$ and $S^{(2)}$, which have much different analytical structures, namely, different multiplicities of the integrals and different velocity arguments of the density matrix elements. This fact becomes unimportant if for some reason the changes in velocity in collisions are ignored. In a linear CI this case is known as the relaxation-constants model. The law of momentum conservation implies that

$$\mathbf{v}_1 = \mathbf{v} - \mu(\mathbf{u} - \mathbf{u}_1)/m, \qquad \mathbf{v}_{b1} = \mathbf{v}_b + \mu(\mathbf{u} - \mathbf{u}_1)/m_b.$$

If the terms $\mu(\mathbf{u}-\mathbf{u}_1)/m$ and $\mu(\mathbf{u}-\mathbf{u}_1)/m_b$ can be dropped, we obtain

$$\begin{gathered} S(\alpha\alpha'\mathbf{v}) = \int d\mathbf{v}_b d\mathbf{R} u\rho(\alpha_1\alpha_2\mathbf{v})\rho(\beta_1\beta_2\mathbf{v}_b) \\ \times\{F(\alpha\beta|\alpha_1\beta_1\mathbf{u}\mathbf{R})\delta_{\alpha'\alpha_2}\delta_{\beta\beta_2} + \delta_{\alpha\alpha_1}\delta_{\beta\beta_1}F^*(\alpha'\beta|\alpha_2\beta_2\mathbf{u}\mathbf{R}) \\ + F(\alpha\beta|\alpha_1\beta_1\mathbf{u}\mathbf{R})F^*(\alpha'\beta|\alpha_2\beta_2\mathbf{u}\mathbf{R})\} \\ = \int d\mathbf{v}_b d\mathbf{R} u\rho(\alpha_1\alpha_2\mathbf{v})\rho(\beta_1\beta_2\mathbf{v}_b) \\ \times\{S(\alpha\beta|\alpha_1\beta_1\mathbf{u}\mathbf{R})S^*(\alpha'\beta|\alpha_2\beta_2\mathbf{u}\mathbf{R}) - \delta_{\alpha\alpha_1}\delta_{\alpha'\alpha_2}\delta_{\beta\beta_1}\delta_{\beta\beta_2}\}, \end{gathered} \tag{3.9}$$

with $S^{(1)}$ and $S^{(2)}$ corresponding to the terms that are linear and quadratic in F. Since approximation (3.9) does not allow for velocity variation, the fact that the CI splits into $S^{(1)}$ and $S^{(2)}$ becomes unimportant and the merging of the two terms seems natural. Also, using a scattering matrix to describe the CI yields a more compact notation.

The optical theorem for the F and S matrices has the form

$$SS^+ = 1, \qquad F + F^+ = -FF^+. \tag{3.10}$$

The fact that the velocity does not change in collisions leads to additional conservation laws that follow from (3.10):

$$\sum_\alpha S(\alpha\alpha\mathbf{v}) = 0, \qquad \sum_\alpha S_1(\alpha\alpha\mathbf{v}) = 0, \qquad \sum_\alpha S_2(\alpha\alpha\mathbf{v}) = 0, \tag{3.11}$$

where $S_1(\alpha\alpha\mathbf{v})$ and $S_2(\alpha\alpha\mathbf{v})$ are the population and interference parts of the CI similar to (2.28) and (2.29). Thus, as before, there are independent conservation laws for the population and interference parts of the CI.

4. THE COLLISION INTEGRAL IN THE TWO-LEVEL MODEL

The two-level model is often used in many fields of physics, for instance, in spectroscoy and quantum electronics. In this section the general formulas of Sec. 2 are concretized for a system of two levels m and n, where it is assumed that n is the ground level and m the excited.

The two-level model adopts a simplified system of notation, making the problem of labeling the internal states more manageable:

$$\begin{gathered} \rho_\alpha(\mathbf{v}) = \rho(\alpha\alpha\mathbf{v}), \qquad \rho(\mathbf{v}) = \rho(mn\mathbf{v}), \\ f(mn\mathbf{u}|mn\mathbf{u}_1) = p_{mn}(\mathbf{u}|\mathbf{u}_1), \qquad f(mn\mathbf{u}|nm\mathbf{u}_1) = q_{mn}(\mathbf{u}|\mathbf{u}_1), \\ f(\alpha\alpha\mathbf{u}|\alpha\alpha\mathbf{u}_1) = r_\alpha(\mathbf{u}|\mathbf{u}_1), \qquad \alpha = m, n. \end{gathered}$$

More than that, without risk of misunderstanding, the velocity arguments can be dropped from the scattering amplitudes: $r_\alpha(\mathbf{u}|\mathbf{u}_1) = r_\alpha$, etc. Combining (2.19)–(2.21), (2.28), and (2.29), we get

$$S_e^{(1)}(mm\mathbf{v}) = (4\pi\hbar/\mu)\rho_m(\mathbf{v})\int \mathrm{Im}[r_m\rho_m(\mathbf{v}_b) + p_{mn}\rho_n(\mathbf{v}_b)]d\mathbf{v}_b, \tag{4.1}$$

$$S_{ex}^{(1)}(mm\mathbf{v}) = (4\pi\hbar/\mu)\mathrm{Im}\{\rho^*(\mathbf{v})\int q_{mn}\rho(\mathbf{v}_b)d\mathbf{v}_b\}, \tag{4.2}$$

$$\begin{gathered} S_1^{(2)}(mm\mathbf{v}) = \langle\{\rho_m(\mathbf{v}_1)[|r_m|^2\rho_m(\mathbf{v}_{b1}) + |p_{mn}|^2\rho_n(\mathbf{v}_{b1})] \\ + \rho_n(\mathbf{v}_1)|q_{mn}|^2\rho_m(\mathbf{v}_{b1})\}\delta(u^2 - u_1^2)\rangle, \end{gathered} \tag{4.3}$$

$$S_e^{(2)}(mm\mathbf{v}) = 2\mathrm{Re}\langle\rho(\mathbf{v}_1)\rho^*(\mathbf{v}_{b1})p_{mn}q_{mn}^*\delta(u^2 - u_1^2)\rangle. \tag{4.4}$$

In (4.3) and (4.4) we have retained only the terms that describe elastic scattering and resonant exchange, since in the two-level scheme and the optical spectral range, the range of interest to us, inelastic processes contribute little. The collision integral $S(nn\mathbf{v})$ for the ground level n is obtained from (4.1)–(4.4) by replacing m with n and $\rho(\mathbf{v})$ and $\rho^*(\mathbf{v})$. In the "elastic" approximation adopted here collisions do not change the number of particles on each level, which is true separately for the population and interference terms in the CI,

$$\int S_e^{(1)}(mm\mathbf{v})d\mathbf{v} = \int S_1^{(2)}(mm\mathbf{v})d\mathbf{v}, \qquad \int S_{ex}^{(1)}(mm\mathbf{v})d\mathbf{v} = \int S_2^{(2)}(mm\mathbf{v})d\mathbf{v} \tag{4.5}$$

and similary for $S(nn\mathbf{v})$.

Let us write the expression for the CI in the approximation where no changes of velocity in collisions are taken into account. From (3.9) we find that

$$S(mm\mathbf{v}) = \rho_m(\mathbf{v})\langle\langle \rho_n(\mathbf{v}_b)[|P_{mn}|^2 - 1]\rangle\rangle + \rho_n(\mathbf{v})\langle\langle \rho_m(\mathbf{v}_b)|Q_{mn}|^2\rangle\rangle + 2\mathrm{Re}\{\rho(\mathbf{v})\langle\langle \rho^*(\mathbf{v}_b)P_{mn}Q^*_{mn}\rangle\rangle\}, \tag{4.6}$$

where the following notation has been introduced:

$$P_{mn} = S(mn|mn\mathbf{u}\mathbf{R}), \qquad Q_{mn} = S(mn|nm\mathbf{u}\mathbf{R}),$$
$$R_\alpha = S(\alpha\alpha|\alpha\alpha\mathbf{u}\mathbf{R}), \qquad \langle\langle A\rangle\rangle = \int Au\,d\mathbf{v}_b d\mathbf{R}. \tag{4.7}$$

According to the conservation laws (3.11), the following equalities are true in the (4.6) approximation:

$$S_e^{(1)}(mm\mathbf{v}) + S_e^{(1)}(nn\mathbf{v}) = S_1^{(2)}(mm\mathbf{v}) + S_1^{(2)}(nn\mathbf{v}), \tag{4.8}$$
$$S_{ex}^{(1)}(mm\mathbf{v}) + S_{ex}^{(1)}(nn\mathbf{v}) = S_2^{(2)}(mm\mathbf{v}) + S_2^{(2)}(nn\mathbf{v}), \tag{4.9}$$

that is, the sum of collision integrals, $S(mm\mathbf{v}) + S(nn\mathbf{v}) = 0$, vanishes, and so do separately the population and interference terms in this sum.

According to (2.10)–(2.12), (2.23), and (2.24), for the off-diagonal CI in the two-level system we obtain

$$S^{(1)}(mn\mathbf{v}) = (2\pi\hbar/i\mu)\int\{\rho(\mathbf{v})[(p_{mn} - r_n^*)\rho_n(\mathbf{v}_b) - (p^*_{nm} - r_m)\rho_m(\mathbf{v}_b)] + [\rho_n(\mathbf{v})q_{mn} - \rho_m(\mathbf{v})q^*_{nm}]\,\rho(\mathbf{v}_b)\}d\mathbf{v}_b, \tag{4.10}$$

$$S^{(2)}(mn\mathbf{v}) = \langle\langle\{\rho(\mathbf{v}_1)[p_{mn}r_n^*\rho_n(\mathbf{v}_{b1}) + p^*_{nm}r_m\rho_m(\mathbf{v}_{b1})] + [\rho_n(\mathbf{v}_1)q_{mn}r_n^* + \rho_m(\mathbf{v}_1)q^*_{nm}r_m]\,\rho(\mathbf{v}_b)\}\delta(u^2 - u_1^2)\rangle\rangle. \tag{4.11}$$

In the model equivalent to the relaxation-constants model we get

$$S(mn\mathbf{v}) = \rho(\mathbf{v})\langle\langle(R_mP^*_{nm} - 1)\rho_m(\mathbf{v}_b) + (P_{mn}R_n^* - 1)\rho_n(\mathbf{v}_b)\rangle\rangle + \rho_m(\mathbf{v})\langle\langle R_mQ^*_{nm}\rho(\mathbf{v}_b)\rangle\rangle + \rho_n(\mathbf{v})\langle\langle Q_{mn}R_n^*\rho(\mathbf{v}_b)\rangle\rangle. \tag{4.12}$$

The two-level approximation is often used for a resonant transition. In this case the dipole-dipole interaction plays the main role in collisions of neutral particles, and for such an interaction the scattering amplitudes $f(\alpha\beta|\alpha_1\beta_1)$ with equal values of the quantum numbers α and β and of α_1 and β_1 vanish.[4] Hence,

$$f(\alpha\alpha|\alpha\alpha) = f(\alpha\alpha|\alpha\alpha') = f(\alpha\alpha|\alpha'\alpha') = f(\alpha\alpha'|\alpha\alpha) = 0. \tag{4.13}$$

It was the dipole-dipole interaction that was considered by Kazantsev[4] (without allowing for degeneracy and with degeneracy, level moments 0 and 1, and without allowing for velocity variations).

The product of scattering amplitudes in the incoming term [Eqs. (2.23)–(2.26)] contains in the left-hand arguments the same numbers β, while the numbers α and α' are different. Hence, for a two-level system, in one of the amplitudes the left arguments must be the same and for the dipole-dipole interaction this amplitude vanishes. Thus, for this interaction the incoming term of the off-diagonal CI is exactly zero and $S(mn\mathbf{v})$ is specified by the following expression:

$$S(mn\mathbf{v}) = (2\pi\hbar/i\mu)\int\{\rho(\mathbf{v})[p_{mn}\rho_n(\mathbf{v}_b) - p^*_{nm}\rho_m(\mathbf{v}_b)] + [\rho_n(\mathbf{v})q_{mn} - \rho_m(\mathbf{v})q^*_{nm}]\,\rho(\mathbf{v}_b)\}d\mathbf{v}_b. \tag{4.14}$$

If the population of the upper level is moderate and the terms with $\rho_m(\mathbf{v})$ and $\rho_m(\mathbf{v}_b)$ can be dropped,

$$S(mn\mathbf{v}) = \rho(\mathbf{v})(2\pi\hbar/i\mu)\int p_{mn}\rho_n(\mathbf{v}_b)d\mathbf{v}_b + \rho_n(\mathbf{v})(2\pi\hbar/i\mu)\int q_{mn}\rho(\mathbf{v}_b)d\mathbf{v}_b. \tag{4.15}$$

The role of the nonlinearity in the CI is clearly seen in Eq. (4.15). With $\rho_n(\mathbf{v}_b)$ corresponding to a Maxwellian velocity distribution, Eq. (4.15) for atoms in the ground state resembles the linear CI in the strong-collision model. The second term on the right-hand side of (4.15), however, is due to the nonlinearity of the CI and originates in the outgoing term in the CI, while in the canonical linear CI the corresponding term is the incoming one.

Adopting the relaxation-constants model and Eqs. (4.6) and (4.12), we can obtain the Kazantsev collision integral[4] in the nondegenerate state approximation). According to Ref. 4,

$$P_{mn} = P_{nm} = P^*_{mn}, \qquad Q_{mn} = Q_{nm} = -Q^*_{mn}, \tag{4.16}$$

that is, the elastic-scattering amplitudes are real and the resonant-exchange amplitudes are imaginary. Thus,

$$S(mm\mathbf{v}) = \langle\langle \rho_m(\mathbf{v})\rho_n(\mathbf{v}_b)P_{mn}(2 + P_{mn}) + \rho_n(\mathbf{v})\rho_m(\mathbf{v}_b)|Q_{mn}|^2 + 2\mathrm{Re}\{\rho(\mathbf{v})\rho^*(\mathbf{v}_b)Q_{mn}(1 + P_{mn})\}\rangle\rangle, \tag{4.17}$$

$$S(mn\mathbf{v}) = \rho(\mathbf{v})\langle\langle[\rho_n(\mathbf{v}_b) + \rho_m(\mathbf{v}_b)]P_{mn}\rangle\rangle + [\rho_n(\mathbf{v}) - \rho_m(\mathbf{v})]\langle\langle \rho(\mathbf{v}_b)Q_{mn}\rangle\rangle. \tag{4.18}$$

Note that in the last formula the real factor of $\rho(\mathbf{v})$ contains the sum of populations, which does not change in the two-level approximation, while the imaginary coefficient in the second term on the right-hand side of (4.18), the term with averaged product of $\rho(\mathbf{v}_b)$, contains the difference of populations, which changes under radiation. These facts prove to be important for nonlinear optical phenomena.

5. THE SATURATION EFFECT IN A TWO-LEVEL SYSTEM. OPTICAL BISTABILITY

Let us consider the simple problem of absorption of monochromatic radiation by a two-level system. We ignore particle motion and Doppler broadening. In such a model, equivalent to the Karplus–Schwinger model,[22] Eq. (2.1) and formulas of Sec. 4 yield the following system of equations:

$$\dot{\rho}_m = -\gamma_m \rho_m + 2\mathrm{Re}(iG\rho), \tag{5.1}$$

$$\dot{\rho} = -[\Gamma - i\gamma_2(\rho_n - \rho_m) + i\Omega]\,\rho - iG^*(\rho_n - \rho_m), \tag{5.2}$$

$$\rho_m + \rho_n = N, \tag{5.3}$$

$$\Gamma = \gamma_m/2 + \gamma_1 N, \qquad G = d_{mn}E/2\hbar, \qquad \Omega = \omega - \omega_{mn}. \tag{5.4}$$

Here ω and E are the field's frequency and strength, ω_{mn} and d_{mn} the Bohr frequency and the matrix element for the mn transition, N the total number of particles, γ_m the spontaneous decay rate of level m. Inelastic processes are caused only by spontaneous emission and are described by the terms $-\gamma_m\rho_m$ and $-\frac{1}{2}\gamma_m\rho$. Collisional nonlinear effects are represented by the terms $\gamma_1 N$ and $\gamma_2(\rho_n - \rho_m)$ in Eqs. (5.2) and (5.4). The collision-broadened linewidth depends on the total concentration N. On the other hand, the line shift is proportional to the difference $\rho_n - \rho_m = N - 2\rho_m$, which depends on the field's intensity. This fact leads to interesting consequences.

The steady-state regime ($\dot{\rho}_m = 0$ and $\dot{\rho} = 0$) is described by the following formulas:

$$\rho = -i\frac{G^*}{\Gamma}Ny, \qquad y = \frac{1-2x}{1+i(\varepsilon+\eta x)} = \frac{1-i(\varepsilon+\eta x)}{1+\varkappa+(\varepsilon+\eta x)^2}, \tag{5.5}$$

$$\frac{\rho_m}{N} = x = \frac{\varkappa/2}{1+\varkappa+(\varepsilon+\eta x)^2}, \tag{5.6}$$

$$\varkappa = 4|G|^2/\gamma_m\Gamma, \qquad \varepsilon = (\Omega - \gamma_2 N)/\Gamma, \qquad \eta = 2\gamma_2 N/\Gamma. \tag{5.7}$$

The solutions of Eqs. (5.5) and (5.6) coincide, to within notation, with the respective results of Karplus and Schwinger,[22] except for the term ηx.

Equation (5.6) must be considered a cubic equation with respect to x, and we are, naturally, interested in its real roots. For small and large values of $\varkappa$ there is only one physical root and, obviously,

$$x = \varkappa/2(1+\varepsilon^2), \qquad \varkappa \ll 1+\varepsilon^2, \tag{5.8}$$

$$x = 1/2, \qquad \varkappa \gg 1+\varepsilon^2. \tag{5.9}$$

Equation (5.8) gives the linear root in low fields, and (5.9) the complete equalizing of the populations of levels m and n in high, saturating fields. In the intermediate range, $\varkappa \sim 1$, there can be three real roots.

The values of the roots of Eq. (5.6) depend on three parameters: $\varkappa$ is proportional to the radiation intensity, ε is specified by the difference between the field frequency and the transition frequency, and η is determined by the interrelationship between the line shift and linewidth. The value of η is determined by the object of investigation, while both $\varkappa$ and ε are in the hands of the experimenter and can be varied in a continuous manner. Hence, it is expedient to examine the roots of Eq. (5.6) as functions of $\varkappa$ and ε for fixed values of η.

Figure 2 depicts x plotted against $\varkappa$ for a number of values of η and ε. The reader can see that at $\eta = 1$ and $\varepsilon = \pm 1$ the $x(\varkappa)$ curves have the usual shape of curves with saturation and do not differ qualitatively from the case where $\eta = 0$ and $\varepsilon = 0$ (curve *1*). As η grows ($\eta = 5$, 8, and 10), the appearance of the diagrams changes and, as the case where $\eta = 10$ and $\varepsilon = -4$ shows, Eqs. (5.6) can have three different positive roots, that is, optical bistability sets in. The physical reason for bistability is obvious. As the field's intensity grows, so does the upper-level population. This changes the line shift (the ηx term), which may balance the initial frequency detuning ε. If the line shift is much greater than the linewidth ($\eta \gg 1$), even a small increase in x may put the system in a resonance state and dramatically increase the absorption coefficient, and this generates other steady states. This interpretation agrees with the fact that the points where the "anomalous" diagrams touch curve *1* ($\eta = 0$ and $\varepsilon = 0$) correspond to the resonance condition $\varepsilon + \eta x = 0$.

By Gibb's classification, the above case of bistability belongs to optical bistability with increasing absorption. The main feature of such bistability is that it emerges as a result of an increase in absorption due to an increase in intensity,[23] which corresponds to what has just been said. Our case is special in that optical bistability emerges, so to say, on a purely atomic level, while situations described earlier required additional external devices.[23]

Let us examine the conditions in which bistability is realized. Note, first, that the problem must have two parameters rather than three, since any cubic equation can be reduced to a two-parameter form. It is convenient to introduce the following quantities:

$$z = \frac{\eta x}{\sqrt{1+\varkappa}}, \qquad \delta = \frac{\varepsilon}{3\sqrt{1+\varkappa}}, \qquad g = \frac{\eta\varkappa}{4(1+\varkappa)^{3/2}}. \tag{5.10}$$

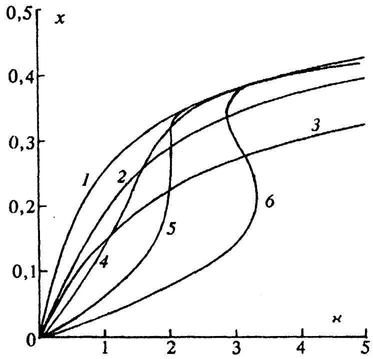

FIG. 2. The relative population of the excited level as a function of the field's intensity. Curve *1*, $\eta = 0$ and $\varepsilon = 0$; curve *2*, $\eta = 1$ and $\varepsilon = -1$; curve *3*, $\eta = 1$ and $\varepsilon = 1$; curve *4*, $\eta = 5$ and $\varepsilon = -2$; curve *5*, $\eta = 8$ and $\varepsilon = -3$; curve *6*, $\eta = 10$ and $\varepsilon = -4$.

Then instead of Eq. (5.6) we have

$$z = \frac{2g}{1 + (z + 3\delta)^2}, \tag{5.6a}$$

where there are only two (albeit combined) parameters δ and g.

As is known, the three roots of a cubic equation are real if the equations's discriminant is negative, which in relation to Eq. (5.6a) means that

$$[g + \delta(1 + \delta^2)]^2 < (\delta^2 - 1/3)^3. \tag{5.11}$$

In Fig. 3 the region in the δ, g plane where condition (5.11) is met is hatched. The initial point of the region,

$$\delta_0 = -1/\sqrt{3}, \quad g_0 = 4\sqrt{3}/9 \tag{5.12}$$

at $\varkappa = 2$ corresponds to $\eta = 8$ and $\varepsilon = -3$ (curve *5* in Fig. 2). Here are simple approximate equations for the lower and upper boundaries of the bistability region:

$$g_1 = -3\delta/2, \quad g_2 = -\delta(2\delta^2 + 1/2), \tag{5.11a}$$

which follow from Eq. (5.11) and provide good accuracy if the distance from point (δ_0, g_0) is not too great.

Many types of interaction that result in broadening and shift of spectral lines lead to $\eta \approx 2$, while values of η greater than eight are unknown, to my knowledge. Nevertheless, large values of η do not contradict the general laws of physics, and objects with such properties may exist. At any rate they can be considered at least as models.

Of great interest is population x [or $\operatorname{Re} y(\varepsilon)$] as a function of ε at fixed values of $\varkappa$ and η, that is, the absorption profile. Figure 4 depicts the ε-dependence of $2x/\varkappa$ for some values of $\varkappa$ and η. At $\eta = 1$ there is a slight asymmetry, which increases with η. Curve *4* at $\varepsilon = -3$ corresponds to point (δ_0, g_0) of Fig. 3. Figure 5 shows that the x vs ε dependence is not unique. The manifestation of bistability in the real part of polarizability [or $\operatorname{Im} y(\varepsilon)$], which fixes the refractive index, is nontrivial. Figure 5 shows that an increase in η violates the antisymmetry of the dispersion curve, which begins to represent a multivalued function.

As is known, bistability is accompanied by hysteresis phenomena in both active optical systems[24] and passive optical systems.[23] In our case hysteresis phenomena manifest themselves in variation of the field's frequency and intensity.

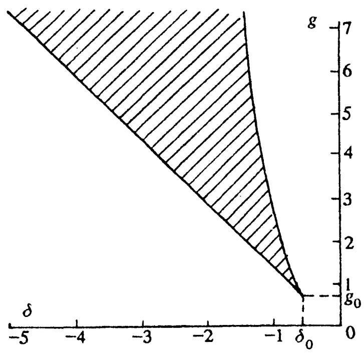

FIG. 3. The region of optical bistability (hatched region).

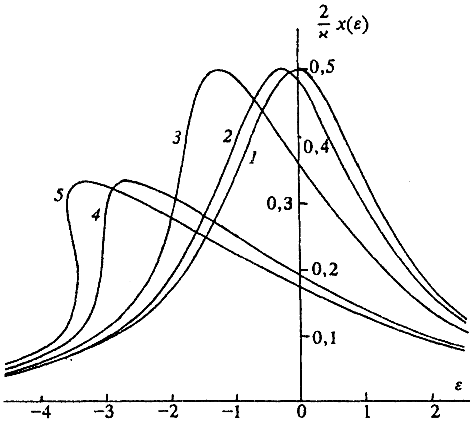

FIG. 4. Absorption profiles: curve *1*, $\eta = 0$ and $\varkappa = 1$; curve *2*, $\eta = 1$ and $\varkappa = 1$; curve *3*, $\eta = 5$ and $\varkappa = 1$; curve *4*, $\eta = 8$ and $\varkappa = 2$; curve *5*, $\eta = 10$ and $\varkappa = 2$.

In addition to absorption and dispersion, mentioned above, bistability manifests itself in all quantities related to the populations ρ_m and ρ_n and the polarization induced in the *mn* transition. For instance, this is true of absorption and scattering of a test field in resonance with the adjacent *ml* and *nj* transitions, of other multiphoton processes, and of diffusion of excited atoms in the presence of the radiation field.

An equation similar to (5.6) has long been known in nonlinear oscillation theory: it describes forced oscillations of a nonlinear oscillator driven by a harmonic force. The equation contains a similar term reponsible for the variation of the natural frequency due to the nonlinearity of the oscillations [see, e.g., Ref. 25, Sec. 14, and Ref. 26, §29, where a point similar to (δ_0, g_0) is defined]. This analogy produces a richer picture of the known correspondence between a classical oscillator and a transition from one quantum state to another caused by an electromagnetic field.

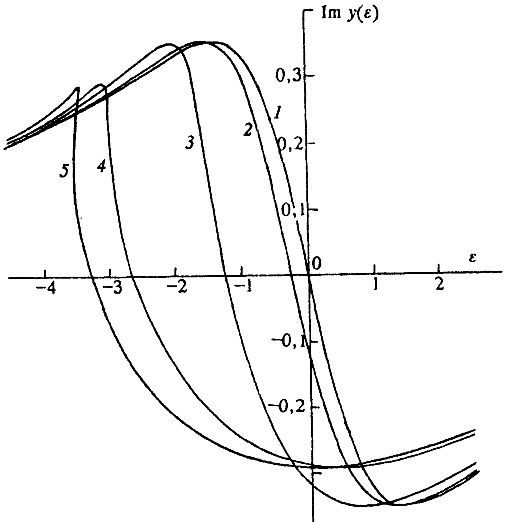

FIG. 5. Dispersion curves: curve *1*, $\eta = 0$ and $\varkappa = 1$; curve *2*, $\eta = 1$ and $\varkappa = 1$; curve *3*, $\eta = 5$ and $\varkappa = 1$; curve *4*, $\eta = 8$ and $\varkappa = 2$; curve *5*, $\eta = 10$ and $\varkappa = 2$.

6. CONCLUSION

The above analysis has revealed the nontrivial role of the nonlinearity in the CI and the strong polarization of a gas in spectroscopic phenomena. The principal new feature of the collision integral is believed to be the existence of interference terms in the integral. The diagonal element $S(\alpha\alpha\mathbf{v})$ of the "ordinary" CI is expressed, for a low polarization of the gas or absence of such polarization, in terms of the level populations $\rho(\alpha_1\alpha_1\mathbf{v})$ and $\rho(\beta\beta\mathbf{v}_b)$ of the colliding particles, while the off-diagonal element $S(\alpha\alpha'\mathbf{v})$ for the $\alpha\alpha'$ transition is expressed in terms of the combinations $\rho(\alpha\alpha'\mathbf{v})\rho(\beta\beta\mathbf{v}_b)$ and $\rho(\alpha\alpha'\mathbf{v}_1)\rho(\beta\beta\mathbf{v}_{b1})$ of the populations and polarization of the same transition. Of course, a strong external field influences the level populations (a saturation effect) and, hence, the CI, but this in itself does not alter the general structure of the CI. The polarization induced by the external field generates unusual components in the CI, components that do not incorporate populations explicitly and contain products of only off-diagonal elements, $\rho(\alpha\beta\mathbf{v})\rho(\beta\alpha'\mathbf{v}_b)$. Terms of this kind appear because of interference or correlation of steady states (a process caused by the external field) and, therefore, it is natural to call them interference terms. There are also exchange terms since they contribute to the CI only through collision exchange processes. The special nature of the interference terms is stressed by the fact that the integral relations associated with the conservation of the number of particles in collisions are formulated independently for the population terms and for the interference terms in the CI [see Eqs. (2.34)–(2.37)].

Equation (2.9) and Fig. 1c specify the analogy between the interference effects in the CI and in radiative processes. This analogy is deeply rooted in the general physical laws. In weak fields, as is known, the probabilities of radiative processes obey Einstein relations, according to which such probabilities are proportional to the populations of the initial levels in the process. Such is the case when different steady states evolve independently. A strong resonant field causes transitions to emerge between levels, thus establishing a correlation between the levels. The evolution of these levels can no longer be considered independent, and the probability of a radiative process is determined not only by the initial-level population, even though the field has altered the population, but also by the off-diagonal elements for adjacent transitions. The respective phenomena, known in spectroscopy as nonlinear interference effects (NIE), change the spectral density of the transition probability, in a different manner for direct and reverse transitions. However, spectrum-integrated probabilities depend only on populations, and the integral contribution of NIE is nil.[14,15]

The origin of the interference terms in the CI is the same: the interdependence of the evolution of different steady states mixed by a resonant field. Equation (2.9) and the integral conservation laws (2.34)–(2.37) serve as a direct quantitative corollary of the discussed analogy.

For a polarized gas, the nonlinear collision integral incorporating the collisions of like particles proves to be extremely complex even in the model of nondegenerate states. A well-developed theory concerning particle interaction is of a phenomenological nature, and calculations involving a concrete type of interaction are, undoubtedly, of interest. All this complexity is the reverse side of the richness and diversity of the phenomena potentially contained in the CI. One of these, the nonlinear line shift in saturation, has been discussed in Sec. 5. Systems with three or more levels will exhibit other phenomena, which hopefully will be investigated in future studies. Note the possibility of experimentally including or excluding transitions by varying the spectral composition of the resonance radiation.

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