Anisotropy of collisions and the Zeeman structure of nonlinear resonances

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The difference magnetooptical resonances in the absorption of a weak optical field, encountered in the probe-field method and due to the anisotropic relaxation of the gas atoms, is considered. The polarization conditions of formation of such resonances are analyzed. The results of their microscopic calculation are presented. The magnitude of the resonances changes radically with variation of the magnetic field strength and light frequency. These features can be used to experimentally determine the structure of the anisotropic parts of the relaxation matrices of the atomic levels and transitions. The structure depends substantially on the nature of the interaction between the colliding gas particles.

1. INTRODUCTION

Wide use is made in the spectroscopy of gases of the isotropic collision model, which assumes the spherical symmetry of the perturbation of the radiating particles by the buffer particles.[1] The conditions of applicability of this model are not satisfied in many cases of practical interest, e.g., if the mean velocities $\bar{v}$ of the radiating particles exceed the mean velocities of the buffer particles $\bar{v}_b$. In this case the radiating atom, moving with velocity **v**, is "blown" by the counterstream of buffer particles and is thus perturbed not isotropically, but axisymmetrically with symmetry axis aligned with **v**. As a result, the relaxation of the atoms under such conditions becomes anisotropic.

In Refs. 2 and 3 a symmetric approach was developed to the study of the influence of the anisotropy of the collisions on the course of both the linear and the nonlinear optical processes. The symmetry properties of the dielectric susceptibility tensor of the gaseous medium were analyzed, and it was established that in the case of collinear propagation of the light fields the susceptibility of two-photon as well as the one-photon processes is axisymmetric if one allows for the anisotropy of the relaxations. The symmetry properties of the susceptibility under these conditions and in the presence of a magnetic field were also elucidated.

Lowering of the symmetry of the susceptibility tensor due to the anisotropy of the collisions lies at the basis of the difference method for separating the contribution induced solely by the anisotropy of the collisions from those of the other magnetooptical processes. The essence of the method is the following: The polarization of the light fields varies periodically and the difference of the signals of the two-photon process over two half-periods is recorded. The polarizations vary in such a way that the part of the signal of the two-photon process which is typical of isotropic relaxation does not vary. Such a difference signal is due exclusively to the anisotropic relaxation and vanishes in the absence of anisotropy.

In the present paper we analyze the difference magnetooptical absorption signals in one of the main methods of nonlinear laser spectroscopy—the probe-field method (see, e.g., Ref. 4). Difference signals in the probe-field method allow one to obtain more detailed information about the anisotropic relaxation in comparison with fluorescence difference signals described in Refs. 2, 5, and 6, since they are due to the anisotropy of the relaxation of not only the energy states, but also the coherence which exists between them.

In this paper we discuss in detail the polarization conditions of the formation of difference signals. Results of a microscopic calculation are presented, and the behavior of the difference signals is analyzed as a function of the light frequencies and the magnetic field intensity.[7]

2. POLARIZATION CONDITIONS OF THE FORMATION OF DIFFERENCE SIGNALS

The essence of the probe-field method consists in the following. An atom interacts with two light fields. The strong field, resonant with the optical transition m–n, perturbs the structure of the atom, and the weak (probe) field, resonant with the transition m–l (or n–l), probes this perturbation.

An atom of the gas is also perturbed by collisions with other particles. Since the interaction of the atom with the light fields induces a coherence between the combining levels, these collisions perturb this coherence. All of these perturbations manifest themselves in a probe-field absorption spectrum free of the masking effect of Doppler line broadening.

The power P delivered by the probe field per unit volume of the medium can be represented to first order in the intensity of the strong field in the form

$$P=\mathrm{Im}\sum_{ijkl}\chi_{ijkl}E_i^{\mu}E_j^{\mu *}E_kE_l^{*}, \qquad (1)$$

where χ_{ijkl} is the fourth-rank nonlinear susceptibility tensor, and E_i and E_k^{μ} are the Cartesian coordinates of the complex amplitude vectors of the strong and the weak fields, respectively.

To analyze the polarization conditions of the formation of the difference signals, it is convenient to write P not in the Cartesian representation (1), but in the irreducible spherical tensor (IST) representation (see, e.g., Ref. 8). The difference signal ΔP in this representation can also be written as

$$\Delta P\equiv P-\tilde{P}=\mathrm{Im}\sum_{\varkappa,\varkappa_1\varkappa_2 q}\{(-1)^{\varkappa-q}\chi(\varkappa_1,\varkappa_2;\varkappa,q)\times\Delta\Phi(\varkappa_1,\varkappa_2;\varkappa,-q)\}. \qquad (2)$$

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Here P and $\widetilde{P}$ are the values of the power at two successive half-periods of the modulation

$$\Delta\Phi(\varkappa_1,\varkappa_2;\varkappa,q)=\Phi(\varkappa_1,\varkappa_2;\varkappa,q)-\widetilde{\Phi}(\varkappa_1,\varkappa_2;\varkappa,q). \tag{3}$$

$$\Phi(\varkappa_1,\varkappa_2;\varkappa,q)=\sum_{q_1q_2}\langle\varkappa_1q_1\varkappa_2q_2|\varkappa q\rangle I^\mu(\varkappa_1q_1)I(\varkappa_2q_2), \tag{4}$$

$$I(\varkappa q)=\sum_{\sigma\sigma'}(-1)^{1-\sigma'}\langle 1\sigma 1-\sigma'|\varkappa q\rangle E_\sigma E_{\sigma'}^{*},$$

$$I^\mu(\varkappa q)=\sum_{\sigma\sigma'}(-1)^{1-\sigma'}\langle 1\sigma 1-\sigma'|\varkappa q\rangle E_\sigma^\mu E_{\sigma'}^{\mu *}, \tag{5}$$

$I(\varkappa q)$ and $I^\mu(\varkappa q)$ are the polarization IST of the fields, $\Phi(\varkappa_1,\varkappa_2;\varkappa,q)$ is the combined polarization IST of the polarizations of the fields, $\chi(\varkappa_1,\varkappa_2;\varkappa,q)$ is the IST of the susceptibility of the two-photon process of rank $\varkappa$, E_σ and E_σ^μ are the spherical components of the fields:

$$E_0=E_z,\quad E_{\pm1}=\mp(E_x\pm iE_y)/\sqrt{2},$$

and $\langle\varkappa_1q_1\varkappa_2q_2|\varkappa q\rangle$ is a Clebsch–Gordan coefficient.[8] The susceptibilities are determined by the properties of the medium and the type of radiative process, and depend on the frequencies ω and ω^μ, the wave vectors $\mathbf{k}$ and $\mathbf{k}^\mu$ of the fields, and the static magnetic field $\mathbf{H}$.

The susceptibility χ can be represented as consisting of two parts: $\chi=\chi_i+\chi_a$, where χ_i is due to the isotropic part of the relaxation. For a gas in a magnetic field, the tensor χ_i is obviously invariant to rotation about $\mathbf{H}$, i.e., it is an axial tensor. In the IST representation this symmetry is expressed by the fact that in the $\mathbf{H}$-system of coordinates (with z axis aligned with $\mathbf{H}$) the only nonzero components of the tensor $\chi_i(\varkappa_1,\varkappa_2;\varkappa,q)$ are those for which $q=0$. Therefore the conditions under which the part of the difference signal due to χ_i vanishes have the form

$$\Delta P_i=\mathrm{Im}\sum_{\varkappa_1\varkappa_2\varkappa}\{(-1)^\varkappa\chi_i(\varkappa_1,\varkappa_2;\varkappa,0)\Delta\Phi(\varkappa_1,\varkappa_2;\varkappa,0)\}=0. \tag{6}$$

The components of the nonlinear susceptibility $\chi_i(\varkappa_1,\varkappa_2;\varkappa,0)$ depend on the parameters of the medium (pressure, temperature), the external field, and the field frequencies ω and ω^μ. Equation (6) is satisfied independently of the indicated circumstances if the following condition is met for arbitrary $\varkappa_1$, $\varkappa_2$, and $\varkappa$:

$$\Delta\Phi(\varkappa_1,\varkappa_2;\varkappa,0)=0 \tag{7}$$

These conditions lead to a system of equations which connect the polarizations of the fields at different periods of the modulation.

Under conditions of large Doppler broadening, the normal Zeeman effect, the use of the resonance approximation, and a small difference in the magnitudes of the wave vectors $\mathbf{k}$ and $\mathbf{k}^\mu$: $(|\mathbf{k}|-|\mathbf{k}^\mu|)/|\mathbf{k}|\ll1$, the tensor $\chi_i(\varkappa_1,\varkappa_2;\varkappa,0)$ possesses an additional symmetry with respect to interchange of the indices $\varkappa_1$ and $\varkappa_2$ (Ref. 2). In this case, Eq. (2) is satisfied if some linear combination of $\Delta\Phi(\varkappa_1,\varkappa_2;\varkappa,0)$ and $\Delta\Phi(\varkappa_2,\varkappa_1;\varkappa,0)$ vanishes.[2] This case is also analyzed in the present paper. In Ref. 2 it was found that this system of equations has nontrivial solutions only in the case of orthogonality of $\mathbf{H}$ and the wave vectors $\mathbf{k}$ and $\mathbf{k}^\mu$ of the collinearly propagating light fields. The solution of this system is expressed most simply in terms of the polar (θ and θ^μ) and azimuthal (φ and φ^μ) angles of the Stokes vectors

$$\boldsymbol{\xi}=(\xi_1,\xi_2,\xi_3)=(\sin\theta\cos\varphi,\ \sin\theta\sin\varphi,\ \cos\theta),$$

$$\boldsymbol{\xi}^\mu=(\xi_1^\mu,\xi_2^\mu,\xi_3^\mu)=(\sin\theta^\mu\cos\varphi^\mu,\ \sin\theta^\mu\sin\varphi^\mu,\ \cos\theta^\mu)$$

on the Poincaré sphere, which completely describe the polarization states of the light.[9]

The Stokes parameters of each of the fields are determined in their own system of coordinates with the z' axis aligned with the wave vectors of the fields (the $\mathbf{k}$- and $\mathbf{k}^\mu$-systems), whereby the y' axis in these systems is assumed to coincide with the y axis in the $\mathbf{H}$-system. The components of the electric vector $\mathbf{E}$ in the $\mathbf{k}$-system are expressed in terms of the parameters θ and φ in the following way:

$$E_{x'}=E\cos(\theta/2)e^{i\omega t},\quad E_{y'}=E\sin(\theta/2)e^{i(\omega t+\varphi)}. \tag{8}$$

These expressions provide insight into the physical meaning of the azimuthal and polar angles θ and φ of the Stokes vector ξ. The angle φ is the phase difference of the Cartesian components of the electric vector $\mathbf{E}$ of the light wave. The tangent of half the polar angle, $\tan(\theta/2)$, is equal to the ratio of the sides of the rectangle which circumscribes the polarization ellipse and is oriented along the x' and y' axes: $\tan(\theta/2)=b_1/a_1$ (Fig. 1).

Note that the ratio $E_{y'}/E_{x'}$ is the stereographic projection of the Stokes vector $\boldsymbol{\xi}$ from the Poincaré sphere ($|\boldsymbol{\xi}|=1$) onto the equatorial complex plane ($\theta=\pi/2$) from the point $\theta=\pi$:

$$E_{y'}/E_{x'}=\mathrm{tg}(\theta/2)e^{i\varphi}.$$

For reflection-symmetric media there exist three nontrivial solutions corresponding to three types of difference signals, which differ by the way in which the polarizations of the fields change when one goes from one half-period of the modulation to the other. We will dwell on each type of difference signal separately. The first type corresponds to variation of the polarizations of the fields according to the transformations

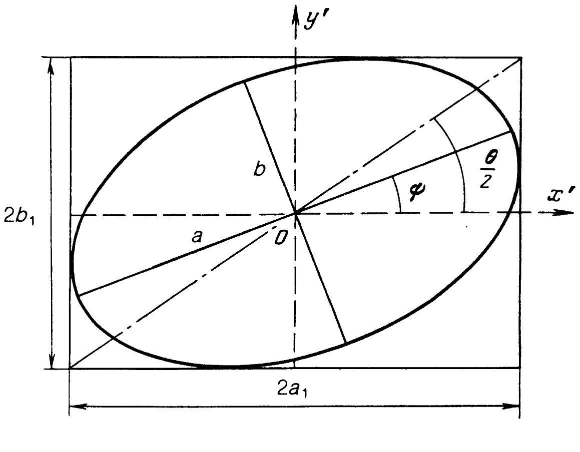

FIG. 1.

$$\tilde{\theta}=\theta^{\mu}, \quad \tilde{\varphi}=\varphi; \quad \tilde{\theta}^{\mu}=\theta, \quad \tilde{\varphi}^{\mu}=\varphi^{\mu}. \tag{9}$$

The difference signal is described by the following components of the tensor $\Delta\Phi(\varkappa_1,\varkappa_2;\varkappa,q)$:

$$\begin{gathered}\Delta\Phi(2,0;2,0)=-2^{-3/2}(\cos\theta-\cos\theta^{\mu})|\mathbf{E}|^2|\mathbf{E}^{\mu}|^2,\\ \Delta\Phi(2,0;2,\pm1)=-6^{-1/2}\Delta\Phi(2,0;2,0),\\ \Delta\Phi(2,2;3,\pm2)=\mp(2/3)^{1/2}\Delta\Phi(2,0;2,0),\\ \Delta\Phi(0,2;2,q)=-\Delta\Phi(2,0;2,q).\end{gathered} \tag{10}$$

The magnitude of the signal, according to Eqs. (10) and (8), depends only on the relative difference of the intensities of the field components aligned with **H** and orthogonal to it. Indeed, from Eqs. (8) we have

$$\begin{aligned}&(|E_{x'}|^2-|E_{y'}|^2)/(|E_{x'}|^2+|E_{y'}|^2)\\ &=\cos^2(\theta/2)-\sin^2(\theta/2)=\cos\theta.\end{aligned} \tag{11}$$

It also follows from Eqs. (2) and (10) that the difference signal changes sign upon interchange of the polarizations of the fields for any counter- or copropagation of the waves. The simplest polarizations that satisfy the given type of difference signal are linear polarizations (Fig. 2). The difference signal is extremal for $\cos\theta - \cos\theta^{\mu} = \pm 2$. This condition corresponds to orthogonal linear polarizations with one of the polarizations aligned with **H**.

The second type of difference a signal corresponds to variation of the polarizations of the fields according to the transformations

$$\tilde{\theta}=\theta, \quad \tilde{\varphi}=\pm\varphi^{\mu}; \quad \tilde{\theta}^{\mu}=\theta^{\mu}, \quad \tilde{\varphi}^{\mu}=\pm\varphi. \tag{12}$$

Here and below, the upper sign corresponds to the case of copropagation of the waves, and the lower signal--to counterpropagation. For this variation of the polarizations of the fields only the following components of the tensor $\Delta\Phi(\varkappa_1,\varkappa_2;\varkappa,q)$ are different from zero:

$$\begin{gathered}\Delta\Phi(1,2;2,0)=\pm2^{-3/2}i\sin\theta\sin\theta^{\mu}\sin(\varphi\mp\varphi^{\mu})|\mathbf{E}|^2|\mathbf{E}^{\mu}|^2,\\ \Delta\Phi(1,2;\varkappa,2\sigma)=2^{-1/2}\sigma^{\varkappa}\langle 1121|\varkappa2\rangle\Delta\Phi(1,2;2,0),\\ \Delta\Phi(2,1;\varkappa,q)=\pm(-1)^{\varkappa}\Delta\Phi(1,2;\varkappa,q), \quad \sigma=\pm1.\end{gathered} \tag{13}$$

To get a clearer picture of the polarization dependence of the difference signals described by these expressions, it is advantageous to express the latter in terms of the more customary polarization parameters of light: the angle of ellipticity ε ($\tan\varepsilon = (b/a)\xi_2/|\xi_2|$, where b and a are the minor and major semiaxes of the polarization ellipse), and the

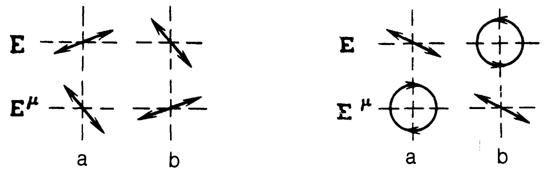

FIG. 2. Two types of variation of the simplest polarizations of the fields in a difference signal induced by the anisotropy of relaxation in the probe-field method for counterpropagating fields a) for $0<t<T/2$ and b) for $T/2<t<T$ (T is the period of variation of the field polarizations).

slope angle ψ of the major axis with respect to the x' axis of the k-system. The relations between the parameters φ, θ and ε, ψ are

$$\cos\theta=\cos(2\psi)\cos(2\varepsilon), \quad \mathrm{tg}\,\varphi=\mathrm{tg}(2\varepsilon)/\sin(2\psi), \tag{14}$$

$$\mathrm{tg}(2\psi)=\mathrm{tg}\,\theta\cos\varphi, \quad \sin(2\varepsilon)=\sin\theta\sin\varphi, \tag{15}$$

with the help of which it is easy to express the polarization dependence (13) of a difference signal of type (12) in terms of the parameters ε and ψ:

$$\begin{aligned}\sin\theta\sin\theta^{\mu}\sin(\varphi\mp\varphi^{\mu})&=\sin(2\varepsilon)\cos(2\varepsilon^{\mu})\sin(2\psi^{\mu})\\ &\mp\sin(2\varepsilon^{\mu})\cos(2\varepsilon)\sin(2\psi).\end{aligned} \tag{16}$$

This expression enables us to determine the transformation group of the polarizations of the fields which doe not change the absolute value of the difference signal. Such transformations can be most diverse. The sole requirement on them is that the absolute value of expression (16) remain constant. From this entire manifold of transformations, let us consider the subgroup of those that do not change the absolute values of any of the factors in expression (16). In this case it is possible to indicate the explicit form of the transformations of the polarization parameters.

First, the symmetry (antisymmetry) of the difference signals with respect to substitution of the polarization of the weak field for the polarization of the strong field and vice versa is immediately obvious for the case of counter- (co-) propagating waves. The second transformation group follows from the fact that the polarization parameters enter into expression (16) as arguments of trigonometric functions, which can take the same value for several different arguments. Thus, the difference signal does not change for either of the substitutions

$$\psi\to\pi/2-\psi+(\pi), \quad \psi^{\mu}\to\pi/2-\psi^{\mu}+(\pi).$$

The additional term (π) is included only to keep the transformed values of ψ and ψ^{μ} within their domain of definition $0\leqslant\psi,\psi^{\mu}<\pi$. Such a transformation corresponds to a reorientation of the polarization ellipse of either of the fields into the orientation symmetric with respect to the axis which makes an angle of 45° with the vector **H**. For the simultaneous substitution

$$\psi\to\pi-\psi-(\pi), \quad \psi^{\mu}\to\pi-\psi^{\mu}-(\pi)$$

the difference signal under consideration changes sign. Physically, this transformation corresponds to a reorientation of both polarization ellipses into orientations symmetric to the initial orientations about the vector **H**. The difference signal also changes sign upon the transformations $\varepsilon\to-\varepsilon$, $\varepsilon^{\mu}\to-\varepsilon^{\mu}$, which correspond to a change in the senses of rotation of the vectors **E** and $\mathbf{E}^{\mu}$ in the polarization plane. Finally, the difference signal is invariant with respect to the simultaneous transformations

$$\psi\to\psi^{\mu}, \quad \psi^{\mu}\to\psi, \quad \varepsilon\to\mp\varepsilon(\pi/4|\varepsilon|-1), \quad \varepsilon^{\mu}\to\mp\varepsilon^{\mu}(\pi/4|\varepsilon^{\mu}|-1).$$

A larger subgroup of explicit transformations of the polarizations of the fields follows from an analysis of the left side of expression (16). The difference signal is invariant to the following transformations of the parameters θ and φ:

$$1)\ \theta\to\pi-\theta, \quad 2)\ \theta^{\mu}\to\pi-\theta^{\mu}, \quad 3)\ \theta\rightleftharpoons\theta^{\mu},$$

$$4)\ \varphi\to\mp\varphi^{\mu}, \quad \varphi^{\mu}\to\mp\varphi, \quad 5)\ \varphi\to\varphi+\alpha, \ \varphi^{\mu}\to\varphi^{\mu}\pm\alpha$$

(α is arbitrary). Change of its sign occurs in the case of the transformations

$$1)\ \varphi\to\varphi+\pi,\ 2)\ \varphi^\mu\to\varphi^\mu+\pi,\ 3)\ \varphi\to2\pi-\varphi,\ \varphi^\mu\to2\pi-\varphi^\mu.$$

The signal reaches its extreme value for polarizations of the fields which satisfy the conditions ($\sigma = \pm 1$)

$$\theta_e=\theta_e^\mu=\pi/2, \tag{17}$$

$$\varphi_e\mp\varphi_e^\mu=\pi/2+\pi\sigma. \tag{18}$$

Conditions (17) are equivalent, as follows from relations (14), to the following conditions:

$$\cos(2\psi_e)=\cos(2\psi_e^\mu)=0, \tag{19}$$

which corresponds to an orientation of the polarization ellipses of the fields at an angle of 45° with the vector **H** (i.e., $\psi_e,\psi_e^\mu = \pi/4, 3\pi/4$). The parameters ε and ε^μ for the extremal difference signal are connected by the following relation, which follows from relations (16), (18), and (19):

$$|\varepsilon|+|\varepsilon^\mu|=\pi/4. \tag{20}$$

If conditions (19) are satisfied, the principal axes of the ellipses of the fields can be oriented either identically or at an angle of 45° with respect to each other. In the first case the ellipticity parameters, in addition to being related by (20), are also related by the inequality

$$\varepsilon\varepsilon^\mu\leqslant0, \tag{21}$$

which points out the different helicity of the fields. In this case for $\psi = \pi/4$ and $\varepsilon > 0$ expression (16) is positive for copropagating waves and negative for counterpropagating waves. In the case of orthogonal relative orientation of the ellipses, in inverse (21) is true. Therefore the extremal difference signals that differ only in the orientation of one of the polarization ellipses (while preserving its shape) differ also in the sign of one of the ellipticity parameters ε and ε^μ. The sign of the difference signal with changed orientation of one of the polarization ellipses is the opposite of the sign of the initial difference signal if the ellipticity parameter of the reoriented ellipse also reverses sign. In the opposite case the sign of the difference signal does not change. In the case of simultaneous change of the signs of ε and ε^μ the extremal difference signal changes sign in accordance with the general invariance property for the given type of difference signal indicated above.

On the basis of what has been said, one can verify that there exist only three different polarization states for the extremal difference signals which have fixed values of $|\varepsilon|$ and $|\varepsilon^\mu|$ related by (20).

The simplest polarizations that satisfy the extremum conditions for the given type of difference signal are circular polarization and linear polarization oriented at an angle of 45° with respect to **H** (Fig. 2).

In the third type of difference signal the law of variation of the initial light wave polarizations is obtained by successive application of the polarization transformations corresponding to the difference signals of the first two types.

3. RESULTS OF MICROSCOPIC CALCULATION OF THE DIFFERENCE SIGNALS

An explicit calculation of the absorption difference signals in the probe-field method was carried out according to the standard scheme.[4] In this method the density-matrix operator $\hat\rho$ is represented in the form $\hat\rho = \hat\rho^0 + \hat\rho^\mu$, where $\hat\rho^\mu$ is a small quantity which is due to the interaction with the probe field. Within the framework of the model of the relaxation constants, to first order in the amplitude of the probe field E^μ the operators $\hat\rho^0$ and $\hat\rho^\mu$ obey the equations[4]

$$\frac{\partial\hat\rho^0}{\partial t}+\hat\Gamma\hat\rho^0+\frac{i}{\hbar}g[\hat\mu\mathbf{H},\hat\rho^0]+\frac{i}{\hbar}[\hat V,\hat\rho^0]=\hat Q, \tag{22}$$

$$\frac{\partial\hat\rho^\mu}{\partial t}+\hat\Gamma\hat\rho^\mu+\frac{i}{\hbar}g[\hat\mu\mathbf{H},\hat\rho^\mu]+\frac{i}{\hbar}[\hat V,\hat\rho^\mu]=-\frac{i}{\hbar}[\hat V^\mu,\hat\rho^0], \tag{23}$$

where $\hat\Gamma$ is the relaxation operator which accounts for the anisotropy of the collisions, $\hat{\boldsymbol\mu}$ is the magnetic dipole moment operator, g is the Landé factor, $\hat V$ and $\hat V^\mu$ are the interaction Hamiltonians of the strong and weak fields **E** and $\mathbf{E}^\mu$ with the atom in the dipole approximation: $\hat V = -\mathbf{E}\cdot\hat{\mathbf{d}}$, $\hat V^\mu = -\mathbf{E}^\mu\cdot\hat{\mathbf{d}}$, where $\hat{\mathbf{d}}$ is the dipole-moment operator, and $\hat Q$ describes the excitation of the atoms in the absence of the light fields. In what follows, the anisotropic part of the relaxation operator $\hat\Gamma$ is assumed to be small in comparison with the isotropic part (which in all known cases is fulfilled—see Ref. 10 and the literature cited). This circumstance allows us to search for the solution of system (22), (23) by the method of iterations in the anisotropic part of the relaxation matrix.

Within the framework of the probe-field method, a few concrete schemes of its realization are possible which differ in the number and arrangement of the combining energy levels, and also in the geometry of the propagating waves. Such an abundance of schemes allows us to use the probe-field method to obtain a vast amount of information about the anisotropic part of the relaxation matrices of both the energy levels and the coherences between them. A detailed microscopic calculation and analysis of the difference levels in all of the variants of the probe-field method was carried out in Ref. 11. The calculation was carried out within the framework of the semiclassical resonance approximation, assuming conditions of the normal Zeeman effect and isotropic excitation of the levels, and implementing a model of the relaxation constants which allows for the anisotropy of the relaxation.

Here we present results of a calculation of the difference signals for a three-level system with arrangement of the combining levels of Λ-type (a Raman-scattering-type process) in which the lower levels, n and l, have total angular momentum $J_n = J_l = 0$, and the upper level m, common to both transitions m–n and m–l, has total angular momentum $J_m = 1$.

For counterpropagating waves in this case the difference signal of type (9) has the form

$$\Delta P=P_0\,\mathrm{Im}\{i\langle(1-\Gamma_{m0}/\Gamma_{m2})(N_m-N_n)[\tilde\Gamma_{ml}(112;v_\perp)$$
$$-\tilde\Gamma_{mn}(112;v_\perp)]\gamma^{-1}F_1(\tilde\Omega_+,\tilde\Delta)$$
$$-N_m\tilde\Gamma_{mm}(022;v_\perp)\Gamma_{m2}^{-1}F_2(\tilde\Omega_+,\tilde\Delta)\rangle_{v_\perp}\}, \tag{24}$$

$$P_0\equiv\frac{\pi}{8}\left(\frac{\pi}{6}\right)^{1/2}\frac{\hbar\omega^\mu}{k\bar v}\frac{|G|^2|G^\mu|^2}{\gamma\Gamma_{m0}}(\cos\theta^\mu-\cos\theta), \tag{25}$$

$$F_1(\tilde\Omega_+,\tilde\Delta)=\sum_{\sigma=\pm1} L(\tilde\Omega_++\sigma\tilde\Delta)\,[L(\tilde\Omega_++\sigma\tilde\Delta)-L(\tilde\Omega_+-\sigma\tilde\Delta)\,], \tag{26}$$

$$F_2(\tilde\Omega_+,\tilde\Delta)=\sum_{\sigma=\pm1} L(\tilde\Omega_++\sigma\tilde\Delta)\,[1-L(2\sigma\Delta/\Gamma_{m2})\,], \tag{27}$$

$$L(x)=1/(1-ix), \tag{28}$$

$$\tilde\Omega_\pm=(\Omega_\mu\pm\Omega)/\gamma,\quad \tilde\Delta=\Delta/\gamma,\quad \gamma=\Gamma_{mn}+\Gamma_{ml}, \tag{29}$$

$$\begin{aligned}\Gamma_{ij}(\varkappa\varkappa_1L;v_\perp)&=[(2\varkappa+1)(2\varkappa_1+1)]^{1/2}\\ &\times\left\{\Gamma_{ij}(\varkappa\varkappa_1L;v)P_2\left(\frac{\Omega}{kv}\right)\exp\left[-\left(\frac{\Omega}{k\bar v}\right)^2\right]\right.\\ &\left.+\,\Gamma_{ij}(\varkappa\varkappa_1L;v^\mu)P_2\left(\frac{\Omega^\mu}{k^\mu v}\right)\exp\left[-\left(\frac{\Omega^\mu}{k^\mu\bar v}\right)^2\right]\right\},\end{aligned} \tag{30}$$

$$v=[v_\perp{}^2+(\Omega/k)^2]^{1/2},\quad v^\mu=[v_\perp{}^2+(\Omega^\mu/k)^2]^{1/2}, \tag{31}$$

$$G=Ed_{mn}/2\hbar,\quad G^\mu=E^\mu d_{ml}/2\hbar, \tag{32}$$

$$\Omega=\omega-\omega_{mn},\quad \Omega^\mu=\omega^\mu-\omega_{mn}. \tag{33}$$

Here ω_{ij}, d_{ij}, and Γ_{ij} are respectively the frequency, the reduced matrix element of the dipole moment, and the isotropic relaxation constant of the i–j transition; N_i, Γ_{i0}, and Γ_{i2} are the population and the isotropic relaxation constants of the population and the alignment[1] of level i; $\Delta\equiv g\mu_0H/\hbar$, where g and μ_0 are the Landé factor and the Bohr magneton: $\Gamma_{ij}(\varkappa\varkappa_1L;v)$ are the anisotropic relaxation constants of the levels ($i=j$) and the transitions ($i\neq j$) [Ref. 10]; $P_2(x)$ is a Legendre polynomial; and $\bar v$ and $v_\perp$ are the mean thermal velocity and the modulus of the transverse component (with respect to $\mathbf{k}$) of the velocity of the atom. The angular brackets denote averaging over the Maxwellian distribution $W(v_\perp)$ of the transverse velocities $v_\perp$:

$$\langle f(v_\perp)\rangle_{v_\perp}=\int_0^\infty W(v_\perp)f(v_\perp)v_\perp\,dv_\perp.$$

Graphs of the functions $\mathrm{Re}[F_1(\tilde\Omega_+,\tilde\Delta)]$ and $\mathrm{Re}[F_2(\tilde\Omega_+,\tilde\Delta)]$ are shown in Figs. 3 and 4. In agreement with phenomenological predictions,[2] these functions are even with respect to inversion of $\mathbf{H}$ and vanish when $\mathbf{H}=0$. The area under the graphs of $\mathrm{Re}[F_1(\tilde\Omega_+,\tilde\Delta)]$ (see Fig. 3a) plotted vs $\tilde\Omega_+$ is equal to zero, i.e., the difference signal integrated over ω^μ is expressed only in terms of $\int F_2(\tilde\Omega_+,\tilde\Delta)d\omega^\mu$.

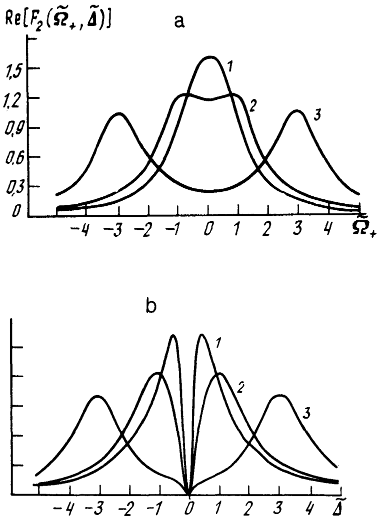

FIG. 4. Dependence of the real part of the nonlinear difference signal (24) due to the anisotropy of the relaxation of the degenerate level m on a) the total frequency detuning of the fields $\tilde\Omega_+$ for various values of the Zeeman level splitting $\tilde\Delta$: 1) $\tilde\Delta=0.5$, 2) $\tilde\Delta=1$, 3) $\tilde\Delta=3$ and b) the magnitude of the Zeeman level splitting $\tilde\Delta$ for various values of the total frequency detuning of the fields $\tilde\Omega_+$: 1) $\tilde\Omega_+=0$, 2) $\tilde\Omega_+=1$, 3) $\tilde\Omega_+=3$. In the construction of the graphs the relaxation constant of the common level m was taken to be equal to the half-sum of the relaxation constants of the Raman transitions: $\Gamma_{m2}=(\Gamma_{mn}+\Gamma_{ml})/2$.

This statement is valid in general.[11] Its physical meaning is that the spectrally integrated signal does not depend on any of the peculiarities of the perturbation of the radiation process, which in the case under consideration are described in Eq. (24) by the term proportional to $F_1(\tilde\Omega_+,\tilde\Delta)$.

The behavior of the function $\mathrm{Re}[F_1(\tilde\Omega_+,\tilde\Delta)]$ changes radically upon variation of any of its parameters. The zeros of the function $\mathrm{Re}[F_1(\tilde\Omega_+,\tilde\Delta)]$ are located at the points $\tilde\Omega_+^{(0)}=\pm1\pm(2+\tilde\Delta^2)^{1/2}$ (Fig. 3a) and $\tilde\Delta^{(0)}=0$, $\pm(\tilde\Omega_+^2\pm2\tilde\Omega_+-1)^{1/2}$ (Fig. 3b), where it is to be noted that the zeros $\tilde\Delta^{(0)}$ do not exist for all values of $\tilde\Omega_+$. Thus, if $|\tilde\Omega_+|\leqslant\sqrt2-1$, then $\mathrm{Re}[F_1(\tilde\Omega_+,\tilde\Delta)]$ vanishes only at $\tilde\Delta^{(0)}=0$ (curve 1 in Fig. 3b). For $\sqrt2-1<|\tilde\Omega_+|\leqslant\sqrt2+1$ two additional zeros appear: $\tilde\Delta^{(0)}=\pm(\tilde\Omega_+^{(2)}+2\tilde\Omega_+-1)^{1/2}$ (curve 2 in Fig. 3b).

The extrema of the function $\mathrm{Re}[F_1(\tilde\Omega_+,\tilde\Delta)]$ are located at the points $\tilde\Omega_+^{(e)}=0$, $\pm(\tilde\Delta^2+1)^{1/2}$ (the main positive extremum), $\pm\{\tilde\Delta^2+1)^{1/2}[(\tilde\Delta^2+1)^{1/2}+2]\}^{1/2}$, and

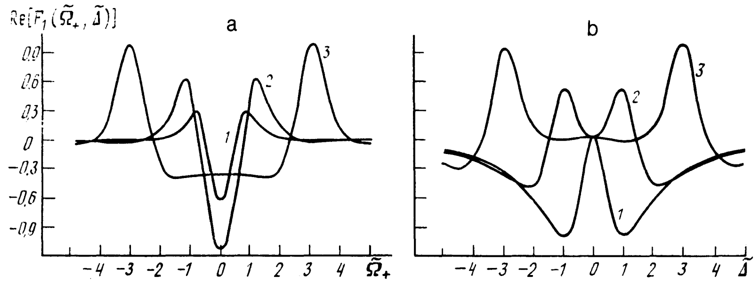

FIG. 3. Dependence of the real part of the nonlinear difference signal (24) due to the anisotropy of the relaxation of the optical coherence on a) the total frequency detuning of the fields $\tilde\Omega_+$ for various values of the Zeeman level splitting $\tilde\Delta$: 1) $\tilde\Delta=0.5$, 2) $\tilde\Delta=1$, 3) $\tilde\Delta=3$ and b) the Zeeman level splitting $\tilde\Delta$ for various values of the total frequency detuning of the fields $\tilde\Omega_+$: 1) $\tilde\Omega_+=0$, 2) $\tilde\Omega_+=1$, 3) $\tilde\Omega_+=3$.

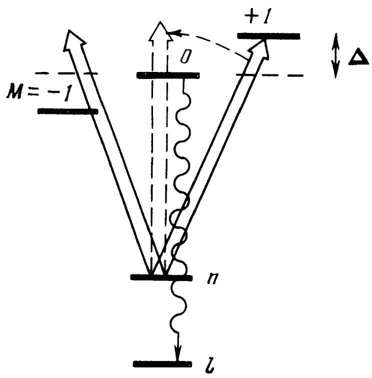

FIG. 5.

$\pm\{(\tilde\Delta^2+1)^{1/2}[(\tilde\Delta^2+1)^{1/2}-2]\}^{1/2}$, where the last two extrema arise only for $\tilde\Delta > \sqrt{3}$ (curve 3 in Fig. 3a). The magnitude of the extrema of $\mathrm{Re}[F_1(\tilde\Omega_+, \tilde\Delta)]$ grows monotonically with increasing H. At $\tilde\Delta = 1$ the negative extremum ($\tilde\Omega_+^{(e)} = 0$) reaches its maximal value, equal to 1. With further increase of $\tilde\Delta$ its magnitude falls off, tending toward zero at large $\tilde\Delta$. The positive extrema grow monotonically with increasing $\tilde\Delta$ toward a value of 1. In the limit $\tilde\Delta \gg 1$ the function $\mathrm{Re}[F_1(\tilde\Omega_+, \tilde\Delta)]$ can be represented as a sum of two symmetrically shaped resonances with unit amplitude:

$$\mathrm{Re}[F_1(\tilde\Omega_+, \tilde\Delta)] \approx \mathrm{Re}\left\{\left[\frac{1}{1-i(\tilde\Omega_+-\tilde\Delta)}\right]^2 + \left[\frac{1}{1-i(\tilde\Omega_++\tilde\Delta)}\right]^2\right\}. \tag{34}$$

The physical interpretation of these resonances is as follows. Let the polarization of the strong field in one of the two polarization states of the difference signal be orthogonal, and that of the weak field parallel, to **H** (for such polarizations the difference signal is maximal). The atoms whose velocities satisfy the conditions $-\mathbf{k}^\mu\cdot\mathbf{v} = \Omega^\mu$ and $-\mathbf{k}\cdot\mathbf{v} = \Omega \pm \Delta$ interact resonantly with the strong and weak fields. In the absence of collisions, nonlinear resonances are absent in the action of the probe field, since the weak and strong fields interact with various sublevels. Isotropic collisions lead to a redistribution of the population of the Zeeman sublevels [the factor $1-\Gamma_{m0}/\Gamma_{m2}$ in Eq. (24)]. As a consequence of the anisotropic collisions, the optical polarization in the transition with $\Delta M \pm 1$ is transferred to the transition with $\Delta M = 0$ (Fig. 5), which is impossible in principle for isotropic relaxation (see, e.g., Ref. 4). The result is a nonlinear resonance described by the function $F_1(\tilde\Omega_+, \tilde\Delta)$.

The component of the difference signal proportional to $F_2(\tilde\Omega_+, \tilde\Delta)$ is due to the hidden[1)] alignment of the level m, where the first and second terms in the square brackets of the expression for $F_2(\tilde\Omega_+, \tilde\Delta)$ [Eq. (27)] are respectively due to longitudinal and transverse alignment (in the **H**-system). The additional decrease of the second term with growth of the magnetic field in comparison with the first is due to the rotation of the transverse alignment in the magnetic field, which, in turn, lowers the efficiency of coherent interaction with the light. Therefore at large values of **H** (or Ω_+) the function $F_2(\tilde\Omega_+, \tilde\Delta)$ is the sum of two independent Lorentzians of unit amplitude, centered at the points $\tilde\Omega_+$ ($\tilde\Delta$) (see Fig. 4).

For a resonant dipole–dipole interaction between colliding particles,[13] at $(\Omega\pm\Delta)/k\bar v \ll 1$ and $\cos\theta = -\cos\theta^\mu = 1$ the difference signal (24) is roughly 6% of the usual nonlinear magnetooptical resonance.

The difference signal of the second type (12) is expressed in this particular case ($J_m = 1, J_n = J_l = 0$) in the form

$$\Delta P = P_1\,\mathrm{Im}\langle (N_m-N_n)[\Gamma_{ml}(112;\,v_\perp)-\Gamma_{mn}(112;\,v_\perp)] \times \gamma^{-1}F_3(\tilde\Omega_+,\,\tilde\Delta)+N_m\Gamma_{mm}(022;\,v_\perp)\Gamma_{m2}^{-1}F_4(\tilde\Omega_+,\,\tilde\Delta)\rangle_{v_\perp}, \tag{35}$$

$$P_1 \equiv \frac{\pi}{4}\left(\frac{3\pi}{2}\right)^{1/2}\frac{\hbar\omega^\mu}{k^\mu\bar v}\,\frac{|G|^2|G^\mu|^2}{\gamma\Gamma_{m2}}\sin\theta\sin\theta^\mu\sin(\varphi+\varphi^\mu), \tag{36}$$

$$F_3(\tilde\Omega_+,\tilde\Delta) \equiv \sum_{\sigma=\pm1} L(\tilde\Omega_++\sigma\tilde\Delta)[L(\tilde\Omega_++\sigma\tilde\Delta)-L(\tilde\Omega_+-\sigma\tilde\Delta)] \times\left[\frac{\Gamma_{m2}}{\Gamma_{m1}}L\left(\frac{\sigma\Delta}{\Gamma_{m1}}\right)-L\left(\frac{\sigma\Delta}{\Gamma_{m2}}\right)\right], \tag{37}$$

$$F_4(\tilde\Omega_+,\tilde\Delta) \equiv \sum_{\sigma=\pm1} L(\tilde\Omega_++\sigma\tilde\Delta)\left[1-L\left(\frac{2\sigma\Delta}{\Gamma_{m2}}\right)\right] \times\left[\frac{\Gamma_{m2}}{\Gamma_{m1}}L\left(\frac{\sigma\Delta}{\Gamma_{m1}}\right)-L\left(\frac{\sigma\Delta}{\Gamma_{m2}}\right)\right]. \tag{38}$$

Graphs of the functions $\mathrm{Im}[F_3(\tilde\Omega_+, \tilde\Delta)]$ and $\mathrm{Im}[F_4(\tilde\Omega_+, \tilde\Delta)]$ are shown in Figs. 6 and 7. The difference signal is even with respect to inversion of **H**, in agreement with phenomenological predictions.[2] However, in contrast to difference signals of the previous type, it is asymmetric with respect to $\tilde\Omega_+$ (see Figs. 6a and 7a). This property is valid also in general. Specifically, it turns out that for difference signals of type (12) the asymmetric part of the difference signal is proportional to the real parts of the anisotropic relaxation constants, and the symmetric part—to the imagi-

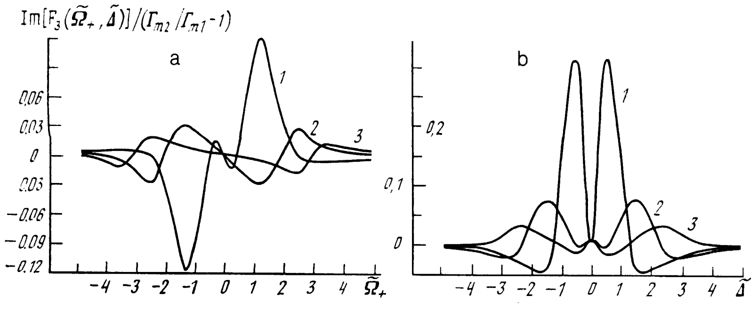

FIG. 6. Dependence of the imaginary part of the nonlinear difference signal (35) due to the anisotropy of the relaxation of the optical coherence on a) the total frequency detuning of the fields $\tilde\Omega_+$ for various values of the Zeeman level splitting $\tilde\Delta$: 1) $\tilde\Delta = 1$, 2) $\tilde\Delta = 2$, 3) $\tilde\Delta = 3$ and b) the Zeeman level splitting $\tilde\Delta$ for various values of the total frequency detuning of the fields $\tilde\Omega_+$: 1) $\tilde\Omega_+ = 1$, 2) $\tilde\Omega_+ = 2$, 3) $\tilde\Omega_+ = 3$. In addition to the conditions indicated in Fig. 4a, the ratio of the relaxation constants of the alignment Γ_{m2} and the orientation Γ_{m1} is equal to $\Gamma_{m2}/\Gamma_{m1} = 1.03$.

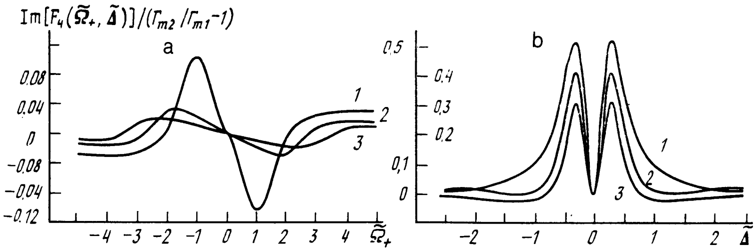

FIG. 7. Dependence of the imaginary part of the nonlinear difference signal (35) due to the anisotropy of the relaxation of the degenerate level m on a) the total frequency detuning of the fields $\widetilde{\Omega}_+$ for various values of the Zeeman level splitting $\widetilde{\Delta}$: 1) $\widetilde{\Delta}=1$, 2) $\widetilde{\Delta}=2$, 3) $\widetilde{\Delta}=3$ and b) the magnitude of the Zeeman level splitting $\widetilde{\Delta}$ for various values of the total frequency detuning of the fields $\widetilde{\Omega}_+$: 1) $\widetilde{\Omega}_+=1$, 2) $\widetilde{\Omega}_+=2$, 3) $\widetilde{\Omega}_+=3$. The conditions are the same as in Fig. 6.

nary parts. For difference signals of the other kind (9) the situation is the reverse.[11]

The area under the plot of F_3 vs Ω_+ is equal to zero, and the difference signal integrated over ω^μ is expressed only in terms of $\int f_4 d\omega^\mu$. As in the case of the difference signal (24), this is a general property and has the same physical meaning.

The functions F_3 and F_4 differ from the functions F_1 and F_2 only by the factor $(\Gamma_{m2}/\Gamma_{m1})L(\sigma\Delta/\Gamma_{m1}) - L(\sigma\Delta/\Gamma_{m2})$. This characteristic factor reflects a physical property of the given type of difference signal. It is due to the difference between the interaction of an atom in the magnetic field with circularly polarized light and with linearly polarized light. Let us clarify this circumstance by an example. In one of the two polarization states let the strong field be circularly polarized and the weak field be linearly polarized at an angle of 45° with respect to **H**. In the other polarization state let the situation be the reverse. For such polarizations the difference signal is maximal. In these polarization states the light interacts with all the Zeeman sublevels, and the resonance conditions and the intensity of the interaction with each of the sublevels are identical for these two polarizations. The difference consists only in the phase relations between the circular components of the fields. The latter circumstance explains the falloff of the difference signals of the given type at large $\widetilde{\Delta}(\widetilde{\Omega}_+)$. Indeed, for large magnetic fields (frequency detunings) the atom interacts resonantly with only one of the circular components. The phase relations between the different components in this case are not important. Therefore, the interaction of the atom with the indicated circular and linear polarizations is identical, and as a consequence of this the difference signal vanishes.

In conclusion we note that the results which we have presented of the calculation of the difference signals in the absorption of the probe field for the case in which the common level m for the transitions m–n and m–l is located above the others (Raman-scattering-type process) are easily extended to the case of two-photon absorption or two-photon fluorescence (in which the level m is located between the levels n and l) for copropagating waves. For this it is only necessary to make the substitution $\Omega_+ \to -\Omega_-$.

4. CONCLUSION

The symmetry properties of the nonlinear susceptibility tensor χ of a gas is closely connected with the anisotropy of the relaxation of its constituent molecules. For isotropic collisions of the molecules the tensor χ is invariant to rotations (isotropic). For anisotropic collisions it is axial with symmetry axis along the direction of the collinearly propagating light waves.

This difference in the symmetry of χ lies at the basis of the formation of those difference magnetooptic signals of two-photon processes which are proportional to the anisotropy of the relaxation. The difference signal is the difference of the intensities of the two-photon process which arises as a result of replacing the initial polarizations of the light fields (which can be arbitrary) by others which are related to the initial polarizations by some definite law.

Nontrivial difference signals, proportional to the anisotropy of the relaxation, exist only when the magnetic field is perpendicular to the wave-propagation direction (deviation from this configuration produces a contribution proportional to the isotropic relaxation and to the square of the deviation angle). For a reflection-symmetric medium there are only three such types of difference signals, differing by their law of variation of the field polarizations. The symmetry groups of each type of difference signal with respect to the transformations of the initial polarizations of the fields have been found. The class of light-field polarizations for which the difference signal attains its extremum has been found.

The absorption difference signals in the probe-field method are even with respect to inversion of the magnetic field. The magnitude of the magnetooptical difference signal undergoes a radical change upon variation of the magnetic field and of the frequency detuning of the wave fields from the resonant frequencies of the transitions in the atom (Figs. 3, 4, 6, and 7). For a reflection-symmetric medium the even (with respect to sign reversal of the detunings) and odd parts of the difference signal of one type are respectively proportional to the real and imaginary parts of the anisotropic relaxation matrices. For the other type of difference signal the situation is the reverse.

The different types of difference signals are described by a diverse set of elements of the anisotropic part of the relaxation matrices both of the combining levels and the transitions. Only the terms of the difference signal which correspond to the anisotropic reorientation of the levels contribute to the intensity integrated over the spectrum.

Using the different variants of the difference signals of both types (counter- and copropagating waves, signals integrated over ω^μ), it is possible to obtain information on the various elements of the anisotropic part of the relaxation of the levels and the transitions which describe the difference signal. Knowledge of the relaxation matrix can be used to elucidate the nature of the interaction between the colliding particles. The magnitude of the signal for the 1–0 transitions and the resonant dipole–dipole interaction between the particles for $(\Omega-\Delta)/k\bar{v}$, $\Gamma/k\bar{v} \ll 1$ is around 6% of the ordinary nonlinear signal.

[1)] The term "hidden" alignment, first introduced in Ref. 12, denotes the presence of alignment of several subensembles of atoms with given velocity, but its absence for the gas as a whole.

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