Polarization effects in the nonlinear spectroscopy of ground-state atoms having hyperfine sublevels under conditions of optical pumping

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A general approach is suggested to the solution of a broad range of problems in nonlinear-polarization spectroscopy (NPS). The experimental geometry which makes it possible to isolate the individual contributions $C_\varkappa$ of the multipole moments of rank $\varkappa = 0, 1, 2$ induced by a strong light wave is determined. A general expression for $C_\varkappa$ in the case of a rarefied gas of atoms with a hyperfine structure and in the ground state under conditions of optical pumping is proposed. A comparison is made with the experimental data obtained for the D_2-line of ^{87}Rb by isolating the NPS signal generated by the scalar part of the scattering tensor (proportional to C_0). Satisfactory agreement is noted with the calculations carried out according to a theory developed for the amplitudes of nonlinear Doppler-free resonances. Finally, formulas are given for calculating the NPS signal for the most common experimental geometries.

Methods of nonlinear polarization spectroscopy (NPS)[1–3] have made it possible in recent years to initiate an experimental study of nonlinear resonances associated with the existence of hyperfine components of atomic levels. NPS signals in such systems are quite complicated due to the appearance of various types of crossover resonances,[4] optical pumping in the ground state of the atoms,[3,5,6] and effects due to the polarization and the geometry of the interaction of the strong wave and weak waves. Although at present there are a number of experimental papers dedicated to the study of the hyperfine structure by NPS methods, a satisfactory quantitative calculation of the amplitudes of the nonlinear resonances is lacking. Thus, the authors of Ref. 7 make an effort to provide a quantitative analysis numerically. However, it cannot be said to be fully satisfactory since the calculation is made in fact only with allowance for the population redistribution over the hyperfine levels of the ground state, while neglecting polarization effects associated with the presence of induced moments of higher rank (magnetic and quadrupole), whose influence is in fact commensurate with the scalar part. On the other hand, in those few papers[4,8] where the NPS signal is calculated correctly, only one special case of the polarizations of the strong and the weak wave is considered, without a sufficient degree of generality and physical clarity, and the calculations themselves are cumbersome and laborious. Therefore a study of NPS effects based on the most general formulation of the problem for arbitrary elliptical polarizations and arbitrary directions of propagation would be of unquestioned interest, specifically one which would make it possible to determine the various geometries and polarization relations which would be useful in the planning and interpretation of experiments. The present paper is dedicated to a general theoretical analysis and its quantitative experimental confirmation in the case of the D_2-line of ^{87}Rb.

THEORY

1. Geometrical analysis of the NPS signal in media with photoinduced anisotropy

We write the nonlinear polarizability $\mathbf{P}_n(\nu)$ of the medium at the frequency of the probe field ν in two equivalent forms:

$$\mathbf{P}_n(\nu) \propto \sum_{\varkappa=0,1,2} \frac{C_\varkappa}{3^{1/2}} (2\varkappa+1)^{1/2} \{\{\overset{*}{\mathbf{e}}_0 \otimes \mathbf{e}_0\}_\varkappa \otimes \mathbf{e}\}_1 \exp[-i(2\pi\nu t - \mathbf{kr})]$$

$$= \exp[-i(2\pi\nu t - \mathbf{kr})]\Big[\Big(-\frac{C_0}{3}+\frac{C_2}{3}\Big)\mathbf{e} - \frac{C_2+C_1}{2}\mathbf{e}_0(\overset{*}{\mathbf{e}}_0\mathbf{e}) - \frac{C_2-C_1}{2}\overset{*}{\mathbf{e}}_0(\mathbf{e}_0\mathbf{e})\Big]. \quad (1)$$

Here $\{\ldots \otimes \ldots\}$ represents a tensor product of rank $\varkappa$ (Ref. 9); $\mathbf{e}_0$ and $\mathbf{e}$ are the unit complex vectors of the strong and weak fields, respectively, which in general have arbitrary elliptical polarizations. The coefficients $C_\varkappa$ describe the nonlinear interaction of the probe wave

$$\mathbf{E} = E(r)\mathbf{e}\exp[-i(2\pi\nu t - \mathbf{kr})] + \text{c.c.} \quad (2)$$

and the pump wave

$$\mathbf{E}_0 = E_0(r)\mathbf{e}_0\exp[-i(2\pi\nu_0 t - \mathbf{k}_0\mathbf{r})] + \text{c.c.} \quad (3)$$

and contain all the spectral information. Their explicit form is determined by the model of the medium, the angular momenta of the resonance levels, collision processes (in the gas), etc. However, all geometric and polarizational aspects of the interaction can be studied independently of the explicit form of $C_\varkappa$.

As to their physical content, the quantities $C_\varkappa$ determine, respectively, the scalar (C_0), the antisymmetric (magnetic dipole) (C_1), and the symmetric (quadrupole) (C_2) scattering.

The experiments usually yield some linear combination of the constants C_0, C_1, and C_2. Since the spectral properties of these coefficients are in general diverse, it is more convenient for the sake of physical visualization to isolate in the polarization signal each individual term proportional to the corresponding multipole moment of rank $\varkappa = 0$, 1, or 2 induced by the strong field. Toward this end, starting with Eq. (1), we obtain expressions for the intensities $I^{\parallel} \propto |E|^2$ and $I^{\perp} \propto |E^{\perp}|^2$ by isolating the components of the probe field at the exit from the medium—the component whose direction coincides with the direction of the polarization vector $\mathbf{e}$ at the entrance to the medium ($I^{\parallel}$) and the component whose direction coincides with the direction of the polarization vector $\mathbf{e}_{\perp}$ orthogonal to it ($I^{\perp}$):

$$I^{\parallel}(L)=I(0)\left\{1-2\alpha L\,\mathrm{Re}\left[D(\nu)-\frac{C_0}{3}+\frac{C_2}{3}-\frac{C_2+C_1}{2}\,|(\overset{*}{\mathbf{e}}_0\mathbf{e})|^2-\frac{C_2-C_1}{2}\,|(\mathbf{e}_0\mathbf{e})|^2\right]\right\},\qquad(4)$$

$$I^{\perp}(L)=I(0)\,(\alpha L)^2\times\left|\frac{C_2+C_1}{2}\,(\overset{*}{\mathbf{e}}{}^{\perp}\mathbf{e}_0)\,(\overset{*}{\mathbf{e}}_0\mathbf{e})+\frac{C_2-C_1}{2}\,(\overset{*}{\mathbf{e}}{}^{\perp}\overset{*}{\mathbf{e}}_0)\,(\mathbf{e}_0\mathbf{e})\right|^2.\qquad(5)$$

$\alpha L<1.$

Here α is the linear coefficient of absorption by the weak field, L is the interaction length of the waves, $I(0)$ is the intensity of the probe wave at the entrance to the medium, and $D(\nu)$ is the Doppler contour of linear absorption [see Eq. (12)]. The scalar products of the unit vectors contain all of the dependence on the relative polarizations and on the geometry of the interaction. We note that in general $\mathbf{e}_0$, $\mathbf{e}$, and $\mathbf{e}^{\perp}$ have elliptical polarization.

From Eqs. (1), (4), and (5) it is possible to find the experimental geometries (polarization of the waves and their directions of propagation) for which the terms in C_0, C_1, and C_2 separate out in the NPS signal. We carry out such a separation.

a) The scalar scattering signal.

Scalar scattering C_0 is not present in the signal of the orthogonal component (5) since it does not lead to a change in the polarization of the probe wave. The very existence of scalar scattering is associated with the formation of a population difference in the atomic levels which are resonant with the field, and also with the redistribution of the population among the hyperfine levels of the ground state under optical pumping. C_0 usually does not separate out in pure form. Finding the conditions under which scalar scattering separates out would correspond to the determination of the limits of applicability of the scalar model of the interaction of light with the medium, when it is possible to ignore the degeneracy of the atomic levels and polarization effects.

For this it is necessary, as follows from Eq. (1), that the following conditions be fulfilled when the expressions for C_2 and C_1 vanish:

$$\begin{gathered}{}^1/_2(\overset{*}{\mathbf{e}}_\mu\mathbf{e}_0)(\overset{*}{\mathbf{e}}_0\mathbf{e})+{}^1/_2(\overset{*}{\mathbf{e}}_\mu\overset{*}{\mathbf{e}}_0)(\mathbf{e}_0\mathbf{e})=(\overset{*}{\mathbf{e}}_\mu\mathbf{e})/3,\\(\overset{*}{\mathbf{e}}_\mu\mathbf{e}_0)(\overset{*}{\mathbf{e}}_0\mathbf{e})-(\overset{*}{\mathbf{e}}_\mu\overset{*}{\mathbf{e}}_0)(\mathbf{e}_0\mathbf{e})=0,\end{gathered}\qquad(6)$$

where $\mathbf{e}_\mu$ is the probe-field component which separates out from the NPS exit signal.

As an example let us consider the case of oppositely directed, linearly polarized waves with angle θ between $\mathbf{e}$ and $\mathbf{e}_0$. Then, in order that conditions (6) be satisfied, it is necessary to isolate (with a polarizer) $\mathbf{e}_\mu$ at the exit from the medium the probe-field component oriented at the angle Ω to $\mathbf{e}$:

$$\mathrm{tg}\,\Omega=\frac{3\cos^2\theta-1}{3\cos\theta\sin\theta}.\qquad(7)$$

In particular, if one isolates in the signal only the projection $\mathbf{e}_\mu=\mathbf{e}$, i.e., the same as at the entrance to the medium ($\Omega=0$), then $\theta=54.7°$ ($\cos^2\theta=1/3$). If condition (7) is fulfilled, the exit signal has the form

$$I^{\parallel}(L)=I(0)\left\{1-2(\alpha L)\,\mathrm{Re}\left[D(\nu)-\frac{C_0}{3}\right]\right\}\cos^2\Omega.\qquad(8)$$

Precisely such an expression is used in the resonance-medium model which does not take account of the degeneracy of the levels or polarization effects.[10]

b) The antisymmetric scattering signal.

To isolate the signal proportional to $|C_1|^2$, it is necessary, as follows from Eq. (5), to satisfy the condition

$$(\overset{*}{\mathbf{e}}{}^{\perp}\mathbf{e}_0)(\overset{*}{\mathbf{e}}_0\mathbf{e})+(\overset{*}{\mathbf{e}}{}^{\perp}\overset{*}{\mathbf{e}}_0)(\mathbf{e}_0\mathbf{e})=0.\qquad(9)$$

In this case the exit signal contains a component $\mathbf{e}^{\perp}$ orthogonal to the initial signal $\mathbf{e}$. Condition (9) is fulfilled, in particular, when the strong wave is circularly polarized, while the counterpropagating probe wave can have arbitrary ellipticity, including even linear polarization. Just this case was considered in Refs. 4 and 8.

c) The symmetric scattering signal.

To isolate in the orthogonal component the signal due only to the quadrupole moment induced by the strong field, it is necessary that the magnetic-dipole component in Eq. (5) vanish:

$$(\overset{*}{\mathbf{e}}{}^{\perp}\mathbf{e}_0)(\overset{*}{\mathbf{e}}_0\mathbf{e})-(\overset{*}{\mathbf{e}}{}^{\perp}\overset{*}{\mathbf{e}}_0)(\mathbf{e}_0\mathbf{e})=0,\qquad(10)$$

which is equivalent to the fulfillment of the condition

$$\cos\varphi\sin2\varepsilon_0\cos2\varepsilon=0,\quad-\pi/4\leqslant\varepsilon_0,\ \varepsilon\leqslant\pi/4,\qquad(11)$$

where φ is the angle between the wave vectors $\mathbf{k}_0$ and $\mathbf{k}$, and ε_0 and ε are the ellipticities of the strong and the weak wave, respectively. Then, setting $\varepsilon=\varepsilon_0=0$ (both waves linearly polarized) and $\varphi=0$ ($\mathbf{k}\|\mathbf{k}_0$), we obtain from Eq. (5) that $I^{\perp}\sim|C_2|^2\sin^2 2\theta$. It is clear that in this situation the angle $\theta=45°$ ($\sin2\theta=1$) is the most preferable.

2. An expression for the nonlinear polarizability of atomic gases under conditions of optical pumping, taking hyperfine splitting into account

As was already noted, the coefficients $C_\varkappa$ are determined by the medium. In the case of an isotropic gas of atoms in a ground state which is degenerate in the projections of the angular momenta and in which there is hyperfine splitting, flow-through effects associated with the finite time τ of interaction of the atoms with the field in bounded light beams[11] play an important role. A correct account of these effects leads to the construction of a perturbation theory in the parameter $\gamma G\tau<1$, where $\gamma\tau\gg1$ (γ is the rate of radiative relaxation of the excited levels, and $G=|E_0d/h\gamma|^2$ is the saturation parameter).

Using a technique described in Refs. 11 and 12, it is possible to obtain (in the absence of a magnetic field) the following expression for the coefficients $C_\varkappa$ when the frequencies of the strong and the weak fields coincide and their directions of propagation are antiparallel ($\mathbf{k}=-\mathbf{k}_0$):

$$C_\varkappa = 2\gamma G\tau \sum_{m,n,m_1,n_1} (-1)^{F_m - F_{m_1}} \frac{N^{(n_1)}}{2F_{n_1}+1} \times |d_{mn}|^2 |d_{m_1n_1}|^2 \begin{Bmatrix} 1 & 1 & \varkappa \\ F_n & F_n & F_m \end{Bmatrix}$$

$$\times \left[\delta_{nn_1} \begin{Bmatrix} 1 & 1 & \varkappa \\ F_n & F_n & F_{m_1} \end{Bmatrix} + (-1)^{F_{m_1}+F_{n_1}} (2J_1+1) \times |d_{m_1n}|^2 \begin{Bmatrix} F_{m_1} & F_{m_1} & \varkappa \\ F_n & F_n & 1 \end{Bmatrix} \times \begin{Bmatrix} 1 & 1 & \varkappa \\ F_{m_1} & F_{m_1} & F_n \end{Bmatrix} \right] \exp\{-\pi^2[(\nu_{mn}-\nu_{m_1n_1})/kv_0]^2\}$$

$$\times \left[1 - i\frac{2\pi}{\gamma}(2\nu - |\nu_{mn} - \nu_{m_1n_1}) \right]^{-1}, \tag{12}$$

$$D(\nu) = \sum_{m,n} \frac{N^{(n)}}{3(2F_n+1)} |d_{mn}|^2 \left\{ \exp\left\{ -\left[\frac{2\pi(\nu-\nu_{mn})}{kv_0} \right]^2 \right\} + i\Phi\left[\frac{2\pi(\nu-\nu_{mn})}{kv_0} \right] \right\},$$

$$\Phi(z) = \frac{2}{\pi^{1/2}} \exp(-z^2) \int_0^z \exp(p^2)\, dp.$$

Here $\begin{Bmatrix} a & b & c \\ d & e & f \end{Bmatrix}$ is the 6*j*-symbol.[9] The indices n and n_1 number the hyperfine levels of the ground state, m and m_1—the excited state, d_{mn} is the normalized reduced matrix element, whose square is equal to

$$|d_{mn}|^2 = (2F_m+1)(2F_n+1) \begin{Bmatrix} J_1 & I_R & F_m \\ F_n & 1 & J_0 \end{Bmatrix}^2,$$

where J_1 and J_0 are the total angular momenta of the electron shell in the excited and the ground states, respectively, $F_{(m,n)}$ is the total angular momentum of the hyperfine level, and I_n is the angular momentum of the nucleus. Further, ν_{mn} is the frequency of the transition between the m and n levels, $2kv_0$ is the Doppler linewidth (it is assumed that $2kv_0 \gg \gamma$), and $N^{(n)}$ is the equilibrium population of the nth ground state;

$$\sum_n N^{(n)} = 1,$$

in the case $h\nu_{nn_1} \ll k_B T$ we have

$$N^{(n)} = (2F_n+1) \Big/ \sum_{n'} (2F_{n'}+1).$$

Considering the frequency dependence in formula (12), it is easy to identify the various types of nonlinear processes. Thus, the terms with $m = m_1$ and $n = n_1$ correspond to direct resonances. For $m = m_1$ and $n \neq n_1$ we have the crossover Λ-resonances (Fig. 1a), and for $m \neq m_1$ and $n = n_1$ we have the *V*-resonances (Fig. 1b). The case $m \neq m_1$ and $n \neq n_1$ corresponds to the *X*-resonances (Fig. 1c). It is quite obvious that the terms obtained by simultaneously transposing the index pairs (m,n) and (m_1, n_1) correspond to the same resonance process. It should also be noted that expression (12) was obtained in the limit of a sufficiently rarefied gas, when it is possible to neglect depolarizing collisions.

It was noted above that the majority of experimental studies contain all of the coefficients $C_\varkappa$ in the NPS signal, which complicates the analysis of the results and sometimes leads to misunderstandings. As an example let us consider the standard setup of the experiment[4]: the strong field is circularly polarized, while the weak, linearly polarized wave propagates in the opposite direction. The cell containing the atomic gas is placed between crossed polaroids the angle between which can be arbitrary. Then the expression for the signal in such an experimental scheme has the form

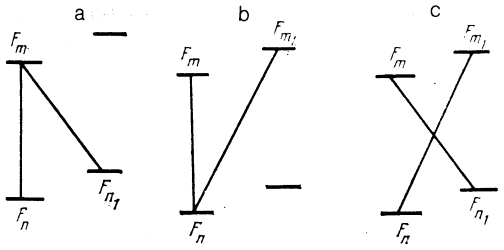

FIG. 1. Possible optical transitions between atomic levels: a) Λ-transitions, b) *V*-transitions, and c) *X*-transitions.

$$I^\Omega(L) = I(0) \left| \cos\Omega - \alpha L \left\{ [D(\nu) - {}^1\!/_3 C_0 - {}^1\!/_6 C_2] \times \cos\Omega \pm \frac{i}{2} C_1 \sin\Omega \right\} \right|^2,$$

$$-\frac{\pi}{2} \leqslant \Omega \leqslant \frac{\pi}{2}. \tag{13}$$

The $\pm$ sign depends on the sign of the spirality of the strong wave. This, however, is not fundamental since one can always create an equivalent situation for the two types of circularly polarized waves by changing the sign of Ω. For $\Omega = 90°$ there remains only $|C_1|^2$. However, for $\Omega \neq 90°$ the signal is substantially deformed as a consequence of the interference of Re $C_\varkappa$ and Im $C_\varkappa$ in Eq. (13) since C_1 enters into the expression with the imaginary unit i. Such a deformation is clearly evident in Ref. 13. In the previously mentioned references 4 and 8 only a numerical calculation of $|C_1|^2$ for the D_1-line of some atoms was in fact presented, along with a qualitative interpretation.

We also present an expression for the NPS signal in the case when the linear polarization vectors of the counter propagating waves have the same direction:

$$I^{\parallel}(L) = I(0)\{1 - 2\alpha L\, \mathrm{Re}[D(\nu) - {}^1\!/_3 C_0 - {}^2\!/_3 C_2]\}. \tag{14}$$

Hence it follows that there are present in the NPS signal both a scalar and a quadrupole part, and this again demonstrates the necessity of a careful theoretical analysis in the interpretation of the results of experiments with polarized laser radiation. It is clear thus that it is wrong to compare the results of the experiments in Refs. 14–16 with the calculations in Ref. 7, where in essence only C_0 is calculated.

It is important to note that under real conditions the experiments are carried out, as a rule, in the presence of a weak constant magnetic field **H** ($\Omega_n \ll \gamma$, kv_0, where $\Omega_n = g_n H$ is the Larmor frequency in the ground state), which leads to a reorientation of the multipole moments of rank $\varkappa \geqslant 1$ (Refs. 11 and 17). As a result, the above formulas and techniques for isolating $C_\varkappa$ become invalid in the general case. However, the effect of the weak magnetic field can be easily excluded by choosing its direction in the following way: if the strong field $\mathbf{E}_0$ is linearly polarized, then $\mathbf{H}$ should be aligned with $\mathbf{E}_0$; if, however, $\mathbf{E}_0$ is circularly polarized, then $\mathbf{H}$ should be aligned with $\mathbf{k}_0$.

EXPERIMENT

As an illustration, we present the results of an experiment in which we observed an NPS signal proportional to the multipole moment of rank $\varkappa = 0$, i.e., scalar scattering.

Experiments of nonlinear, Doppler-free spectroscopy of the D_2-line of rubidium (Fig. 2) were carried out using injection lasers with external cavities, which have recently found widespread use and have become, along with dye lasers, a basic instrument in high-resolution spectroscopy.[18,19] Figure 3 shows a diagram of the experimental setup. The laser diode was mounted flush against a copper block whose temperature could be varied within 10°C of room temperature. The external dispersive cavity consisted of a matching, high-aperture microobjective, a one-way dielectric piezoceramic-based mirror, and a holographic diffraction grating. All of the component parts of the injection laser with the external resonator (ILER) were set up on a massive glass-ceramic slab. Laser diodes were used which had opaque faces as well as a partially translucent face (up to 2–6%) turned towards the external cavity. This translucency leads to a broadening of the range of working wavelengths and the possibility of continuous tuning in the interval between the modes of the laser diode, but also to a decrease of the power of the output radiation from the output opposite the external cavity.

The spectrum of the laser radiation was monitored with a confocal interferometer (region of free dispersion 320 MHz, sharpness 35) and a monochromator. Rough tuning of the lasing wavelength was realized by rotating the diffraction grating. Continuous scanning of the radiation frequency within the limits of the intermode internal $\Delta\nu_{\text{ILER}} \simeq c/2l \sim 0.05$ GHz (l is the length of the external resonator) was carried out by displacing the piezoceramic-based mirror. In order to increase the stability of the ILER frequency, the radiation frequency was stabilized at the resonances of the tunable interferometer (region of free dispersion 600 MHz, sharpness 30) by the method of extremum adaptive control.

In order to observe the nonlinear resonances we used an optical setup with counter propagating waves—a probe wave and a pump wave. The absorption due to the saturated rubidium vapor in a glass cell of length 3 cm did not exceed 30% at room temperature. The cell was placed in a magnetic field $H \approx 0.6$ Oe. The pump and the probe waves were linearly polarized. The electric field vector of the pump radiation was aligned with $\mathbf{H}$ with an error less than 10%. The electric field vector of the probe radiation was tilted by an angle $\theta = 54.7°$ ($\cos\theta = 1/3$) with respect to E_0. Such a geometry, as follows from Eqs. (6)–(8), makes it possible to exclude the influence of the orientation and the alignment of

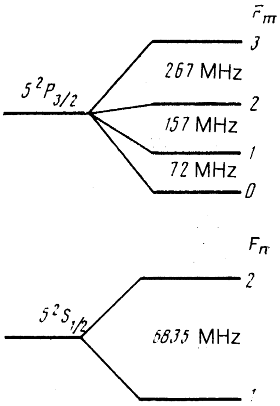

FIG. 2. Diagram of the energy levels of the ^{87}Rb atom for the D_2-line ($\lambda = 780$ nm). The radiative width of the transitions $\gamma/2\pi = 6$ MHz, the Doppler broadening $kv_0/\pi = 612$ MHz at room temperature (300 K).

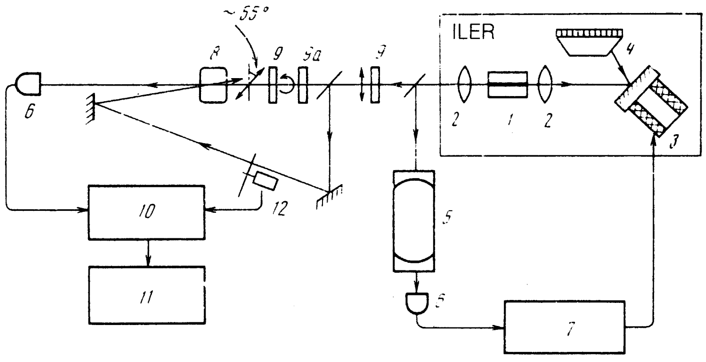

FIG. 3. Diagram of the experimental setup: 1) injection laser, 2) matching and exit microobjectives with numerical apertures 0.65 and 0.3, respectively, 3) piezoceramic-based mirror, 4) holographic selector, 5) scanning confocal interferometer, 6) photodiodes, 7) electronic frequency self-tuning system, 8) rubidium vapor cell, 9) polarizer, 9a) quarter-wave plate, 10) synchronous detector, 11) recorder, and 12) beam chopper.

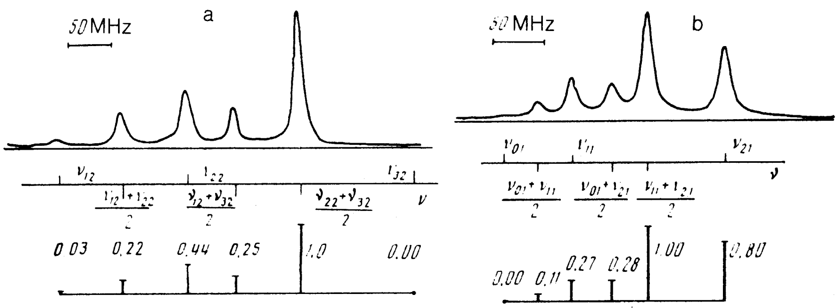

FIG. 4. Nonlinear Doppler-free resonances at the transitions in the D_2-line of ^{87}Rb at $\theta = 54.7°$. The experimental dependences are shown in the upper part of the figures, and in the lower part the relative amplitudes of the resonances, calculated according to formula (12) for $\varkappa = 0$. The frequencies $\nu_{F_m F_n}$ correspond to the transitions $5^2S_{1/2}(F_n)$–$5^2P_{3/2}(F_m)$: a) the transitions with $F_n = 2$, b) the transitions with $F_n = 1$.

the atoms of the amplitudes of the nonlinear resonances. Modulation of the intensity of the pump beam with subsequent synchronous detection was used to record the Doppler-free resonances with zero background. For the saturation parameter, allowing for optical pumping, $\gamma G\tau < 1$, the amplitudes of the nonlinear resonances were proportional to Re C_0. The intensity of the pump wave was ~ 0.1 mW/cm^2, and of the probe wave, $\sim 5\ \mu$W/cm^2.

DISCUSSION OF RESULTS

Figures 4a and b show the nonlinear resonances of the D_2-line of ^{87}Rb, recorded against a zero background, which are proportional to C_0. In the lower part of the figures are shown the relative amplitudes of these resonances calculated according to the above theory. Satisfactory agreement to within 10% is observed between experiment and theory. It is interesting to note the absence of direct nonlinear resonances at the transitions $0 \rightarrow 1 (\nu_{01})$ and $3 \rightarrow 2 (\nu_{32})$. In the approximation $\gamma G\tau < 1$, $\gamma\tau \gg 1$ the upper level plays the role of a virtual level, and the nonlinear resonances in C_0 describe a redistribution of the population only between the hyperfine levels of the ground state. But since the above transitions are optically forbidden, it is obvious that such a redistribution is absent. In the case of other experimental geometries the presence of resonances at the forbidden transitions is due to multipole moments of rank $\varkappa \geqslant 1$ (see, e.g., Ref. 14).

The insignificant discrepancy between the results of experiment and calculation can be explained, for example, by the presence of a residual angle between the direction of the magnetic field and the electric vector of the strong field, which leads to the appearance in the NPS signal of a contribution due to the reoriented quadrupole moment (C_2).[11] Another possibility is the influence of nonlinear effects of higher orders in $\gamma G\tau$.

In conclusion we note that in this paper we have indicated a general approach to the solution of a broad range of problems of nonlinear polarization spectroscopy for atoms with arbitrary hyperfine structure and for arbitrary polarizations of the strong and the weak waves. Explicit expressions for $C_\varkappa$ (12) have been obtained here for the first time. Using tunable injection lasers, we have recorded Doppler-free nonlinear resonances at the D_2-line of rubidium corresponding to the scalar part of the scattering tensor C_0. The results are in agreement with the theory. It is obvious that further detailed experimental and theoretical study of nonlinear resonances due to multipole moments of rank $\varkappa \geqslant 1 (C_1, C_2)$ is necessary.

[1)] P. N. Lebedev Physical Institute, Academy of Sciences of the USSR

[1]C. Wieman and T. W. Hansch, Phys. Rev. Lett. **36**, 1170 (1976).

[2]A. Corney, B. P. Kibble, and G. W. Series, Proc. Roy. Soc. **A293**, 70 (1966).

[3]S. G. Rautian, G. I. Smirnov, and A. M. Shalagin, *Nonlinear Resonances in the Spectra of Atoms and Molecules* [in Russian], Nauka, Novosibirsk (1979).

[4]S. Nakayama, G. W. Series, and W. Gawlik, Opt. Comm. **34**, 382 (1980).

[5]H. Rinneberg, T. Hühle, E. Methies, and A. Timmerman, Z. Phys. **A295**, 17 (1980).

[6]W. Happer, Rev. Mod. Phys. **44**, 169 (1972).

[7]S. Nakayama, Jpn. J. Appl. Phys. **23**, 879 (1984).

[8]S. Nakayama, Opt. Comm. **50**, 19 (1984).

[9]D. A. Varshalovich, A. N. Moskalev, and V. K. Khersonskii, *The Quantum Theory of Angular Momenta* [in Russian], Nauka, Leningrad (1975).

[10]V. S. Letokhov and V. P. Chebotaev, *The Principles of Nonlinear Laser Spectroscopy* [in Russian], Nauka, Moscow (1975).

[11]M. V. Krashenninikov, V. S. Smirnov, M. B. Sultanov *et al.*, Zh. Eksp. Teor. Fiz. **94**, No. 1, 203 (1988) [Sov. Phys. JETP **67**, 113 (1988)].

[12]M. V. Krashenninikov, V. S. Smirnov, M. B. Sultanov *et al.*, *ibid.* No. 11, 24 (1988) [**67**, 2190 (1988)].

[13]W. Gawlik, Acta Physica Polonica **A66**, 401 (1984).

[14]T. Yabuzaki, T. Ibaraki, H. Hori *et al.*, Jpn. J. Appl. Phys. **20**, L451 (1981).

[15]H. Hori, Y. Kitayama, M. Kitano *et al.*, IEEE J. Quantum Electron. **QE-19**, 169 (1983).

[16]T. Tsuchida, M. Ohtsu, T. Tako *et al.*, Jpn. J. Appl. Phys. **21**, L561 (1982).

[17]S. Slijkhuis, G. Nienhuis, and R. Morgenstern, Phys. Rev. **A33**, 3977 (1986).

[18]A. M. Akul'shin, V. Yu. Bazhenov, V. L. Velichanskiĭ *et al.*, Kvant. Elektron. **13**, 1391 (1986) [Sov. J. Quant. Electron. **16**, 912 (1986)].

[19]A. M. Akul'shin, V. L. Velichanskiĭ, A. S. Zibrov *et al.*, *ibid.* **15**, 1945 (1988) [**18**, 1214 (1988)].

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