Resonance rotation of the plane of polarization in potassium vapor

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Rotation of the plane of polarization of a weak wave in the field of intense monochromatic radiation, as well as rotation of the polarization ellipse, has been investigated theoretically and experimentally. The theoretical calculations are based on the quasiclassical theory without allowance for relaxation of the medium. Rotation of the plane of polarization of a weak wave from a dye laser was observed in potassium vapor near one- and two-photon resonances in the presence of intense circularly polarized radiation from a ruby laser. Self-rotation of the polarization ellipse near one-photon resonances was observed when radiation having a broad frequency spectrum overlapping both *D* lines of the principal doublet traversed the potassium vapor. A qualitative explanation is offered for the dependence of the rotation angle on the pressure and temperature of the potassium vapor.

INTRODUCTION

When an intense laser beam interacts with alkali metal vapors certain nonlinear polarization effects arise, which include splitting and shifting of the magnetic sublevels of the atom in the field of the wave, rotation of the polarization ellipse, and splitting of lines corresponding to multiphoton processes. The effect of the polarization of the transmitted radiation on nonlinear processes in a resonant medium was discussed theoretically in [1]. Later there appeared a number of theoretical papers on polarization effects in systems having different angular momenta [2,3] and on the effect of polarization of the exciting radiation on the self-focusing process [4,5]. Experimental studies of the polarization characteristics of stimulated three-photon scattering, stimulated electronic Raman scattering, and the four-photon parametric process were reported in [6-8]. Observation of the rotation of the polarization ellipse was apparently first reported in [9].

A resonant medium greatly affects the polarization characteristics of the transmitted radiation. This effect is especially clear near possible resonances (one-photon, two-photon, ... resonances) and depends on the nature of the dispersion. Study of resonance polarization effects is therefore of interest in connection with the spectroscopy of one-photon and multiphoton processes [10-12]. Moreover, the rotation of the plane of polarization is strongly frequency dependent and may find interesting applications in the frequency regulation of laser light and may also be useful in the tuning of lasers.

Here we report the results of a detailed theoretical and experimental study of the self-rotation of the polarization ellipse and the rotation of the plane of polarization of a weak wave in the field of intense monochromatic radiation in the presence of one- and two-photon resonances in potassium vapor.

THEORY

Let us consider the propagation of a plane electromagnetic wave in the negative direction of the z axis through a medium consisting of two-level atoms, and let us assume that the electric vector of the wave is given by

$$\mathbf{E}=\mathbf{E}_1(z)\exp\{-i(k_1z+\omega_1t)\}+\text{C.C.},\tag{1}$$

where ω_1 is the carrier frequency, k_1 is the wave vector, and $\mathbf{E}_1$ is a much more slowly varying function of z than is the exponential. As was shown in [1], when the wave traverses such a medium its polarization ellipse rotates, the angle of rotation depending on the angular momenta j_1 and j_2 of the atoms in the ground and excited states, respectively. Below we give the formulas for the angle of rotation for two different media, consisting of atoms having $j_1 = 1/2$, $j_2 = 3/2$, and having $j_1 = j_2 = 1/2$.

For the case $j_1 = 1/2$, $j_2 = 3/2$ we have

$$\alpha=qz[(1+\xi^{(-)}+3\xi^{(+)})^{-1/2}-(1+3\xi^{(-)}+\xi^{(+)})^{-1/2}].\tag{2}$$

Here the $\xi^{(\pm)}$ are intensity parameters defined by the formula

$$\xi^{(\pm)}=\frac{|d_{12}|^2}{6\hbar^2\varepsilon^2}|E_1^{(\pm)}|^2,\tag{3}$$

in which the $E_1^{(\pm)} = E_{1x} \pm iE_{1y}$ are the spherical components of the amplitude (1) of the field, $\epsilon = \omega_0 - \omega_1$ is the difference between the resonance and carrier frequencies, and d_{12} is the reduced matrix element for the transition between the ground and excited states, while the quantity q in Eq. (2) is given by

$$q=\frac{\pi|d_{12}|^2\omega_1}{12c\hbar\varepsilon}N,$$

in which N is the atomic density of the medium. We note that if the incident wave is circularly or linearly polarized its polarization remains unchanged as it propagates through the medium.

For the case $j_1 = j_2 = 1/2$ we have

$$\alpha=2qz[(1+4\xi^{(+)})^{-1/2}-(1+4\xi^{(-)})^{-1/2}].\tag{4}$$

When $\xi^{(\pm)} \ll 1$, Eqs. (2) and (4) take the form

$$\alpha=qz\xi\eta_2,\quad j_1={}^1/_2,\quad j_2={}^3/_2,\tag{5}$$

and

$$\alpha=4qz\xi\eta_2,\quad j_1=j_2={}^1/_2,\tag{6}$$

where we have written $\xi = \xi^{(+)} + \xi^{(-)}$ and have introduced the Stokes parameter η_2 that specifies the degree of circular polarization.

Now let us assume that in addition to the intense wave (1) there is a weak wave of frequency ω_2 with the electric vector

$$\mathbf{E}=\mathbf{E}_2(z)\exp\{i(k_2z-\omega_2t)\}+\text{C.C.},\ |\mathbf{E}_2|\ll|\mathbf{E}_1|\tag{7}$$

which propagates in the opposite direction (this choice of the propagation direction makes it possible to rule out parametric interaction of the waves).

For the sake of argument let us consider the case in which the strong field is circularly polarized:

$$E_1^{(+)}=0, \quad E_1^{(-)}=E_1, \tag{8}$$

while the weak field is polarized along the x axis as it enters the medium:

$$E_{2y}(0)=0, \quad E_{2x}(0)=E_2. \tag{9}$$

Then simple calculations will give us the angle β through which the plane of polarization of the weak wave is rotated by the action of the strong wave:

$$\beta=rz. \tag{10}$$

The quantity r is given by the formula

$$r=\frac{q\varepsilon}{2}\Big[3\frac{1+\sqrt{1+\xi}}{2\sqrt{1+\xi}}\frac{1}{\varepsilon'-{}^1/_2\varepsilon(1-\sqrt{1+\xi})}+\frac{1+\sqrt{1+3\xi}}{2\sqrt{1+3\xi}}\frac{1}{\varepsilon'-{}^1/_2\varepsilon(1-\sqrt{1+3\xi})}$$
$$-3\frac{(\sqrt{1+3\xi}+1)^2}{4(1+3\xi)}\frac{1}{\varepsilon'-\varepsilon(1-\sqrt{1+3\xi})}-\frac{(\sqrt{1+\xi}+1)^2}{4(1+\xi)}\frac{1}{\varepsilon'-\varepsilon(1-\sqrt{1+\xi})}$$
$$+3\frac{(\sqrt{1+3\xi}-1)^2}{4(1+3\xi)}\frac{1}{\varepsilon'-\varepsilon(1+\sqrt{1+3\xi})}+\frac{(\sqrt{1+\xi}-1)^2}{4(1+\xi)}\frac{1}{\varepsilon'-\varepsilon(1+\sqrt{1+\xi})}\Big] \tag{11}$$

for the case of atoms with angular momenta $j_1 = 1/2$, $j_2 = 3/2$ ($\epsilon' = \omega_0 - \omega_2$ is the detuning of the weak field from the resonance frequency), and by the formula

$$r=2q\varepsilon\Big[\frac{1}{\varepsilon'}-\frac{(1+\sqrt{1+4\xi})^2}{4(1+4\xi)}\frac{1}{\varepsilon'-\varepsilon(1-\sqrt{1+4\xi})} \tag{12}$$
$$+\frac{(1-\sqrt{1+4\xi})^2}{4(1+4\xi)}\frac{1}{\varepsilon'-\varepsilon(1+\sqrt{1+4\xi})}\Big].$$

for the case $j_1 = j_2 = 1/2$. Formulas (11) and (12) simplify considerably for the case $\xi \ll 1$; for that case we have

$$r=\frac{7}{4}q\frac{\varepsilon(\varepsilon+\varepsilon')}{(\varepsilon')^2}\xi, \quad j_1=\frac{1}{2}, \quad j_2=\frac{3}{2}, \tag{13}$$

and

$$r=4q\frac{\varepsilon(\varepsilon+\varepsilon')}{(\varepsilon')^2}\xi, \quad j_1=j_2=\frac{1}{2}. \tag{14}$$

Such a rotation of the plane of polarization of one wave under the influence of another as was considered above also takes place in a medium consisting of three-level atoms. Let us consider the passage through a three-level medium of the wave

$$\mathbf{e}_0=\mathbf{e}_1\exp\{i(k_1z-\Omega_1t)\}+\text{к.с.}+\mathbf{e}_2\exp\{-i(k_2z+\Omega_2t)\}+\text{C.C.}, \tag{15}$$

Here the wave of frequency Ω_1 propagates in the positive z direction and we assume it to be in resonance with the atomic transition of frequency ω_{01} between the ground state 1 and the excited state 2, while the wave of frequency Ω_2 propagates in the negative z direction, and we assume it to be in resonance with the transition of frequency ω_{02} between the excited states 2 and 3.

If we assume that $|\mathbf{e}_1| \ll |\mathbf{e}_2|$ and limit ourselves to the linear approximation in the weak field we can obtain the following expression for the angle γ through which the plane of polarization of the weak wave is rotated (it being assumed, as in the preceding case, that the weak wave is linearly polarized, and the strong wave, circularly polarized):

$$\gamma=\delta z. \tag{16}$$

For the case in which the medium consists of atoms with the angular momenta $j_1 = 1/2$, $j_2 = 3/2$, $j_3 = 1/2$, the quantity δ is given by

$$\delta=q'\varepsilon_1\Big[\frac{9\xi'}{\sqrt{1+3\xi'}(1+\sqrt{1+3\xi'})}\frac{1}{\varepsilon_1+\varepsilon_2(1+\sqrt{1+3\xi'})/2}$$
$$+\frac{\xi'}{\sqrt{1+\xi'}(1+\sqrt{1+\xi'})}\frac{1}{\varepsilon_1+\varepsilon_2(1+\sqrt{1+\xi'})/2}-$$
$$-\frac{9\xi'}{\sqrt{1+3\xi'}(1-\sqrt{1+3\xi'})}\frac{1}{\varepsilon_1+\varepsilon_2(1-\sqrt{1+3\xi'})/2}-$$
$$-\frac{\xi'}{\sqrt{1+\xi'}(1-\sqrt{1+\xi'})}\frac{1}{\varepsilon_1-\varepsilon_2(1-\sqrt{1+\xi'})/2}-8\frac{1}{\varepsilon_1}\Big], \tag{17}$$

where

$$\xi'=\frac{|d_{23}|^2|\mathbf{e}_2|^2}{6\hbar^2\varepsilon_2^2}, \quad q'=\frac{\pi\Omega_1|d_{12}|^2}{48c\hbar\varepsilon_1}N,$$

$\epsilon_1 = \omega_{01} - \Omega_1$ is the detuning of the weak field, $\epsilon_2 = \omega_{02} - \Omega_2$ is the detuning of the strong field, and d_{12} and d_{23} are the reduced matrix elements for the transitions 1—2 and 2—3, respectively. The first two terms in Eq. (17) are due to two-photon resonances shifted in the field of the strong wave, the next two terms to shifted one-photon resonances, and the last term to an unshifted one-photon resonance.

For the case $j_1 = j_2 = j_3 = 1/2$, the quantity δ is given by

$$\delta=4q'\varepsilon_1\Big[\frac{4\xi'}{2\sqrt{1+4\xi'}(1+\sqrt{1+4\xi'})}\frac{1}{\varepsilon_1+\varepsilon_2(1+\sqrt{1+4\xi'})/2}$$
$$-\frac{4\xi'}{2\sqrt{1+4\xi'}(1-\sqrt{1+4\xi'})}\frac{1}{\varepsilon_1+\varepsilon_2(1-\sqrt{1+4\xi'})/2}-\frac{1}{\varepsilon_1}\Big], \tag{18}$$

in which the first term is due to a two-photon resonance and the last two terms to one-photon resonances.

When $\xi' \ll 1$, we have

$$\delta=5q'\varepsilon_1\xi'\frac{\varepsilon_2^2}{\varepsilon_1^2(\varepsilon_1+\varepsilon_2)}, \quad j_1=\frac{1}{2}, \quad j_2=\frac{3}{2}, \quad j_3=\frac{1}{2}, \tag{19}$$

$$\delta=8q'\varepsilon_1\xi'\frac{\varepsilon_2^2}{\varepsilon_1^2(\varepsilon_1+\varepsilon_2)}, \quad j_1=j_2=j_3=\frac{1}{2}. \tag{20}$$

EXPERIMENTAL SETUP

The apparatus diagrammed in Fig. 2 was used for an experimental study of the effects mentioned above in potassium vapor (the level scheme is given in Fig. 1). Part of the radiation from the ruby laser 1 (working in the Q-switched mode, power density ~60 MW/cm^2) was focused by the cylindrical lens 13 onto the cell 3 containing a methanol solution of the dye 1,1′ diethyl quino + 4,4′ carbocyanin iodite. The cell was mounted in a resonator with plane mirrors.

With this setup the dye laser generated radiation at a power level of about 1 MW. The spectrum of this radiation overlapped both the D lines of the principal doublet of potassium, and the spectral power was about the same at both doublet lines. The discreteness of the structure of the spectrum was eliminated to the necessary extent by appropriate choice of the concentration of the solution (the absorption coefficient at ~7600 Å was 10 cm^{-1}). The linearly polarized radiation from the dye laser passed through the 35-cm cell 4 containing potassium vapor and through the polarizer 11, which enabled the type ISP-51

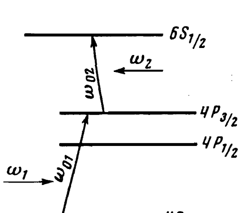

FIG. 1. Relevant portion of the potassium level scheme.

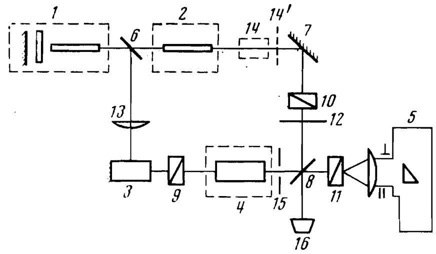

FIG. 2. Experimental setup: 1—ruby laser used as light source, 2—ruby laser used as light amplifier, 3—dye laser, 4—cell containing potassium vapor, 5—spectrograph, 6–8—mirrors, 9–11—polarizers, 12—quarter-wave plate, 13—cylindrical lens, 14—cell containing nitrobenzol, 14′—FS-6 filter, 15—diaphragm, 16—type IKT-1M energy meter.

FIG. 3. Spectra of the X-polarized (a) and Y-polarized (b) radiation leaving the potassium-vapor cell in the absence of radiation from the ruby laser; potassium-vapor temperature: I—180°C, II—230°C. An iron comparison spectrum can also be seen.

spectrograph (reciprocal linear dispersion 21 Å/mm in the 7600 Å region) to record two mutually perpendicular polarizations, one of which was chosen to be the initial polarization. The setup also made it possible to investigate the polarization characteristics of the "weak" radiation in the presence of an intense circularly polarized monochromatic wave propagating through the cell containing the potassium vapor in the direction opposite to the direction of the weak radiation.

Part of the radiation from the ruby laser passed through the semitransparent mirror 6, was amplified, and was directed by the mirrors 7 ($R \approx 99\%$) and 8 ($R \approx 25\%$) through the stop 15 and into the cell 4. A quarter-wave plate 12 was used to obtain circularly polarized radiation. The energy of this radiation was monitored during the experiment with the type IKT-1M energy meter 16.

Sometimes radiation from stimulated Raman scattering in cell 14 containing nitrobenzol was directed through the cell containing the potassium vapor. In this case the light from the ruby laser was removed by the FS-6 filter 14′.

EXPERIMENTAL RESULTS

We made an experimental study of the rotation of the plane of polarization when a linearly polarized test signal from the dye laser passed through potassium vapor, the test signal having a broad frequency spectrum and being in resonance with the $4S_{1/2} \to 4P_{1/2}$ and $4S_{1/2} \to 4P_{3/2}$ transitions. For convenience we shall take the z axis in the direction of propagation of the test signal and the x axis in the direction of its polarization. According to Eqs. (5) and (6), even a slight degree of elliptical polarization can lead to rotation of the plane of polarization, the angle of rotation depending linearly on the parameter ξ and the atomic density N of the medium, and also depending on the frequency detuning ϵ from resonance, being proportional to ϵ^{-3}. Under conditions in which the temperature of the potassium vapor reached 200°C we observed not only the x polarization, but also a y component of the polarization near the principal doublet absorption lines D_1 and D_2 (Fig. 3, I). When the temperature was raised to 230°C, rotation of the plane of polarization was observed only near the frequency of the potassium D_1 line (Fig. 3, II). On further increasing the temperature, the line near the D_1 line also dropped out.

The observed temperature dependence of the rotation of the plane of polarization is due to the change in the width of the absorption line. Under our experimental conditions, the spectral power density was such that rotation of the plane of polarization could take place only for frequencies falling within the half-width of the absorption lines. At low temperatures (2 to 20°C) the line shape is due to Doppler broadening and is therefore independent of the atomic density. This accounts for the threshold for rotation of the plane of polarization at 180°C. Allowing for the difference between the oscillator strengths of the $4S_{1/2} \to 4P_{1/2}$ and $4S_{1/2} \to 4P_{3/2}$ transitions, we conclude from Eqs. (5) and (6) that the angles of rotation near the potassium D_1 and D_2 lines are equal. At high temperatures the line shape is due to collision (pressure) broadening, which depends linearly on the atomic density of the medium and the oscillator strength [13].

The half-width of the D_2 absorption line is 1.4 times greater than that of the D_1 line; as the atomic density of the medium increases, therefore, the rotation of the plane of polarization near the D_2 line is the first to disappear, the rotation near the D_1 line disappearing later.

It must be borne in mind that Eqs. (2) and (4) were derived for a monochromatic wave. Hence we can give only a rough description of the observed rotation for the case of a broad spectrum.

In addition to the effect discussed above, we investigated the rotation of the plane of polarization of a weak linearly polarized wave in the field of a powerful circularly polarized wave from a ruby laser in resonance with the 2—3 transition, the detuning ϵ_2 of the wave from the resonance frequency being $\sim -7\ \mathrm{cm}^{-1}$. In accordance with the theoretical equations (17) and (19), rotation of the plane of polarization should be observed at frequencies close to the one-photon resonance ($\epsilon_1 \sim 0$) and at frequencies near the two-photon resonance ($\epsilon_1 + \epsilon_2 \sim 0$) when the potassium vapor pressure is low.

At a temperature of 125°C (this corresponds to the potassium vapor density $N = 3 \times 10^{12}$ atoms/cm^3), where there is no possibility of self-rotation, a narrow line of y polarization is seen at frequencies close to the $4S_{1/2} \to 4P_{3/2}$ transition (Fig. 4, I). The width of this line does not exceed the instrumental width of the spectrograph. A slight increase in temperature results in the appearance of the two-photon resonance line, in addition to the one-photon one (Fig. 4, II). As the temperature is further increased, the range of frequencies at which rotation of the plane of polarization takes place expands sharply and in some cases reaches a width of $\sim 300\ \mathrm{cm}^{-1}$ (Fig. 4, III). This makes it possible to investigate the nonlinear susceptibility of the medium at frequencies far from resonance.

A similar rotation of the plane of polarization of a test signal was also observed when the first Stokes com-

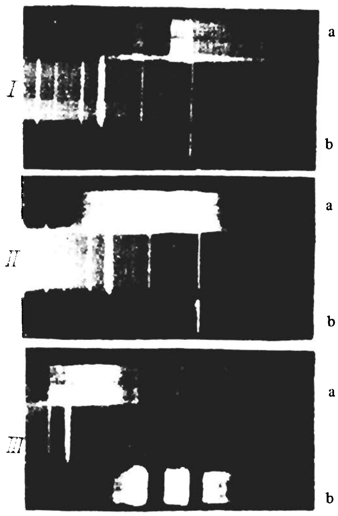

FIG. 4. Spectra of the X-polarized (a) and Y-polarized (b) radiation leaving the potassium-vapor cell after traversing it in the presence of radiation from the ruby laser; potassium-vapor temperature: I–125°C, II–135°C, III–270°C.

ponent of the radiation obtained by stimulated Raman scattering of the ruby laser light in nitrobenzol was used as the powerful wave.

From the results presented above we may conclude that rotation of the plane of polarization in the case of two-photon resonance is extremely sensitive to the frequency. If the frequencies of the waves propagating in opposite directions are equal, the Doppler broadening of the absorption lines is excluded from the two-photon process. This makes it possible to use more refined experimental techniques to investigate the structure of atomic levels via measurements of two-photon rotation of the plane of polarization.

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