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Selective detection of polarisation components of a coherent population trapping signal in hot alkali metal atoms

K.A. Barantsev, E.N. Popov, A.N. Litvinov

Abstract. **A mathematical model of the interaction of bichromatic laser radiation with alkali metal atoms in an optically dense gas cell at above room temperature is constructed. Within the framework of the model, complete hyperfine and Zeeman structures of alkali metal atom levels are considered, which allows the propagation of radiation polarisation along the cell and the effect of a constant magnetic field to be correctly taken into account. It is found that the selective detection of polarisation radiation components carries additional information in comparison with a signal of total intensity.**

Keywords: *coherent population trapping, bichromatic laser radiation, selective detection.*

1. Introduction

It is known that when bichromatic coherent radiation interacts with a three-level quantum system, the latter, under certain conditions, ceases to interact with radiation. In this situation, it is customary to speak of the emergence of destructive quantum interference between the excitation channels, a so-called dark state. Quantum interference manifests itself in an experiment as a narrow peak (several orders of magnitude narrower than the natural linewidth of the excited level) in a transmission spectrum. In other words, resonance laser radiation is not absorbed by the quantum system. The features described above are inherent in the effect of coherent population trapping (CPT) [1–4]. In the case, when an atomic medium is optically dense, or laser radiation is pulsed, one deals with the effect of electromagnetically induced transparency (EIT) [5–6].

The width of the transmission peak (hereinafter, the width of the CPT resonance) in alkali atoms can reach record-high values ranging from hundreds to units of Hertz [7–12]. This feature makes it possible to use the CPT (EIT) resonance in a wide range of applications, including atomic frequency standards [13–18], optical magnetometers [19–23], laser cooling of atoms [24–28], development of devices for recording and processing quantum information [29–32] and inversionless lasing [33–38].

Investigation of the properties of radiation transmitted through a cell with atoms under CPT-resonance conditions has more than 30 years of history. Nevertheless, there are many open issues, as evidenced by recent works. Among these studies, much attention is paid to the investigation of the possibility of narrowing the CPT-resonance line. Xiao et al. [39] succeeded in detecting the narrowing of the CPT-resonance line in a gas cell with a buffer gas under in-band pumping. This is explained by the fact that when the cell is partially illuminated, the atoms do interact with the light all the time, since diffusion takes place in a direction perpendicular to the beam propagation direction. Consequently, there appears an analogue of the Ramsay-type narrowing scheme – diffusion-induced Ramsey narrowing of the CPT resonance. A few years later, a similar effect was observed in buffer-gas-free cells with an antirelaxation wall coating [40–42]. An investigation of the Dicke effect for CPT resonance and the possibility of obtaining a narrow line during its use were also considered in [43, 44]. Kazakov et al. [44] studied cells with an antirelaxation wall coating and found that depending on the width of the laser radiation spectrum, there exist two different narrowing mechanisms: Dicke narrowing and laser-induced narrowing [45].

A number of scientific papers have also been devoted to the study of the polarisation properties of radiation under resonance conditions. Among them, one can especially mention the study of the influence of counterpropagating light wave polarisation on nonlinear EIT resonances and adsorption [46]. The authors proposed a transition from the EIT resonance to the electromagnetically induced adsorption (EIA) resonance by changing the angle between the polarisation planes of waves and the ellipticity parameters. Basalaev et al. [47] studied the effect of stimulated phase modulation by varying the spatial orientation of the polarisation ellipse during the propagation of elliptically polarised light pulses under CPT conditions. It was also shown that depending on the orientation angle of the polarisation ellipse, the propagation velocity of the pulses changes. The presence of polarisation modulation was studied by Yun et al. [48], who showed that such an excitation scheme can also lead to an increase in the CPT-resonance contrast. Lazebnyi et al. [49] examined EIT and EIA effects in the field of elliptically polarised waves. The authors found that the sign of spectral structures, having a width of the order of the natural linewidth, depends on the wave polarisation parameters, while the sign of the super-narrow EIT and EIA resonances does not depend on the polarisation parameters.

It can be concluded that the study of the polarisation properties of radiation under the conditions of CPT (EIT) resonance is an important step towards a deeper understanding of the physical essence of these phenomena. At the same time, in order to construct a theoretical model that takes into account the polarisation properties of radiation, it is necessary

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to consider hyperfine and Zeeman structures of the atom, which leads to a substantial increase in the dimensionality of the system of equations and also to a complication in the interpretation of the results. Nevertheless, allowance for the presence of different polarisation components in describing the interaction of atoms with laser radiation is necessary, since it makes it possible to construct an exact mathematical model characterising the operation of real equipment and quantum electronics devices.

The purpose of this paper is to construct a theoretical model for the interaction of arbitrarily polarised laser radiation with alkali atoms located in an optically dense gas cell with a buffer gas under CPT-resonance conditions. In this case, a special role is assigned to the analysis of the polarisation properties of radiation as it passes through the medium.

2. Statement of the problem and basic equations

Consider the propagation of electromagnetic radiation in a gas cell filled with alkali atoms and a buffer gas. The signal detection scheme is shown in Fig. 1a. The energy spectrum of radiation consists of two broadened lines E_1 and E_2, quasi-resonant to $|g_1\rangle \leftrightarrow |e\rangle$ and $|g_2\rangle \leftrightarrow |e\rangle$ transitions of the alkaline atom (Fig. 1b), where $|e\rangle$ is the excited multiplet of the D_1 or D_2 line. It is assumed that the hyperfine components $|g_1\rangle$ and $|g_2\rangle$ of the lower multiplet are completely resolved by pump radiation, since the width Γ_L of each spectral emission line is less than the hyperfine splitting of the ground state. Moreover, the upper multiplet cannot be resolved at a Doppler width of transitions and a width Γ_L, comparable with the hyperfine splitting of the excited state.

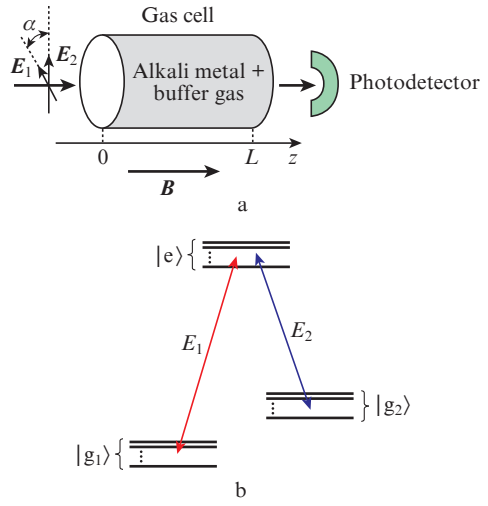

Figure 1. (a) Scheme for the CPT-resonance signal detection from the forward transmitted radiation, consisting of a gas cell irradiated with two-frequency radiation with spectral components E_1 and E_2, and a photodetector, and (b) the energy level diagram of the alkali atom and the transitions excited in it; α is the angle between the polarisation planes. The cell is in a constant magnetic field $\boldsymbol{B}$.

Thus, the vector of the electric field strength of the wave can be written as

$$\boldsymbol{E}(z,t) = \boldsymbol{E}_1^0(z)\exp[-\mathrm{i}(\omega_1 t - k_1 z)] + \boldsymbol{E}_2^0(z)\exp[-\mathrm{i}(\omega_2 t - k_2 z)] + \text{c.c.}, \quad (1)$$

where ω_j are the carrier frequencies; k_j are the wave numbers; z is the axis co-directed with radiation wave vectors and a constant magnetic field; and $j = 1, 2$. The complex amplitudes $\boldsymbol{E}_j^0(z)$ of both frequency components depend on the z coordinate, since the medium is optically dense and radiation absorption and phase incursion of the electromagnetic field take place.

In addition to alkali atoms, a buffer gas is introduced in the cell, which is necessary in order to reduce depolarisation of alkali atoms on the cell walls. At its sufficient concentration (usually 6 to 7 orders of magnitude higher than the concentration of alkali atoms), the mean free path of the alkali atom becomes much smaller than the size of the cell, and the frequency of collisions with the walls sharply decreases. In order to avoid depolarisation of radiation in collisions of alkali atoms with a buffer gas, the latter is selected so that the spin-exchange cross section during the collision is minimal.

The state of alkali atoms is described by the density matrix $\hat{\sigma}(v,z,t)$, where v is the projection of the atomic velocity on the z axis. In the one-dimensional approximation, the quantum kinetic equation for the density matrix has the form [50]

$$\left(\frac{\partial}{\partial t} + v\frac{\partial}{\partial z}\right)\hat{\sigma}(v,z,t) = -\frac{\mathrm{i}}{\hbar}[\hat{H}_0 + \hat{V}, \hat{\sigma}(v,z,t)] + R[\hat{\sigma}(v,z,t)] + S[\hat{\sigma}(v,z,t)], \quad (2)$$

where $\hat{H}_0$ is the Hamiltonian of the alkali atom in the absence of an electromagnetic field; the operator R describes spontaneous relaxation; and S is the collision integral. In the calculations of this paper, S is used in the strong collision model, which is applicable in the case of approximate equality of the masses of the active and buffer atoms [51–53]. The Hamiltonian of the interaction of an atom with an electromagnetic field in the dipole approximation can be expressed as

$$\hat{V} = -\hat{\boldsymbol{d}}\boldsymbol{E} = -\sum_{e,g_1}(\boldsymbol{d}_{eg_1}\cdot\boldsymbol{e}_1)E_1^0|e\rangle\langle g_1|\exp[\mathrm{i}(k_1 z - \omega_1 t)] - \sum_{e,g_2}(\boldsymbol{d}_{eg_2}\cdot\boldsymbol{e}_2)E_2^0|e\rangle\langle g_2|\exp[\mathrm{i}(k_2 z - \omega_2 t)] + \text{h.c.}, \quad (3)$$

where $\hat{\boldsymbol{d}}$ is the dipole moment operator; and $\boldsymbol{e}_j = \boldsymbol{E}_j^0/E_j^0$ are unit polarisation vectors of the jth radiation component. We expand these vectors in cyclic vectors:

$$\boldsymbol{d}_{ab} = d_{ab}^{+}\boldsymbol{e}_{+} + d_{ab}^{0}\boldsymbol{e}_0 + d_{ab}^{-}\boldsymbol{e}_{-}, \qquad \boldsymbol{e}_j = p_j^{+}\boldsymbol{e}^{+} + p_j^{-}\boldsymbol{e}^{-},\ j = 1, 2, \quad (4)$$

where the unit vectors with subscripts and superscripts denote co- and contravariant cyclic unit vectors. Since the z axis is directed along the radiation wave vectors, there is no π component in the expansion of the polarisation unit vectors. The polarisation coefficients have a normalisation:

$$p_j^{+*}p_j^{+} + p_j^{-*}p_j^{-} = 1. \quad (5)$$

We denote the scalar products from expression (3) as

$$\hbar D_{eg_j} = (\boldsymbol{d}_{eg_j}\cdot\boldsymbol{e}_j) = d_{eg_j}^{+}p_j^{+} + d_{eg_j}^{-}p_j^{-}. \quad (6)$$

In the off-diagonal elements of the density matrix, we select a slow envelope:

$$\sigma_{g_j e}(v,z,t) = \rho_{g_j e}(v,z,t)\exp[i(\omega_j t - k_j z)],\ j = 1, 2,$$
$$\sigma_{g_2 g_1}(v,z,t) = \rho_{g_2 g_1}(v,z,t)\exp[i(\omega_2 - \omega_1)t - i(k_2 - k_1)z], \quad (7)$$
$$\sigma_{nn}(v,z,t) = \rho_{nn}(v,z,t).$$

We write the system of equations for the elements of the density matrix in the rotating-wave approximation:

$$\dot\rho_{g_j g_j}(v) = i\sum_e [E_j^{0*} D_{g_j e}\rho_{e g_j}(v) - E_j^0 D_{e g_j}\rho_{g_j e}(v)] + \frac{\gamma}{N_g}\sum_e \rho_{ee}(v) + S\{\hat\rho(v)\}_{g_j g_j}, \quad (8)$$

$$\dot\rho_{ee}(v) = i\sum_{g_1} [E_1^0 D_{e g_1}\rho_{g_1 e}(v) - E_1^{0*} D_{g_1 e}\rho_{e g_1}(v)] + i\sum_{g_2} [E_2^0 D_{e g_2}\rho_{g_2 e}(v) - E_2^{0*} D_{g_2 e}\rho_{e g_2}(v)] - \gamma\rho_{ee}(v) + S\{\hat\rho(v)\}_{ee}, \quad (9)$$

$$\dot\rho_{g_j g_j'}(v) = i\sum_e [E_j^{0*} D_{g_j e}\rho_{e g_j'}(v) - E_j^0 D_{e g_j'}\rho_{g_j e}(v)] + i\omega_{g_j g_j'}\rho_{g_j g_j'} + S\{\hat\rho(v)\}_{g_j g_j'}, \quad (10)$$

$$\dot\rho_{g_1 g_2}(v) = i\sum_e [E_1^{0*} D_{g_1 e}\rho_{e g_2}(v) - E_2^0 D_{e g_2}\rho_{g_1 e}(v)] + i(\omega_{g_1 g_2} + \omega_2 - \omega_1 + qv)\rho_{g_1 g_2}(v) + S\{\hat\rho(v)\}_{g_1 g_2}, \quad (11)$$

$$\dot\rho_{e g_j}(v) = i\sum_{g_1'} E_1^0 D_{e g_1'}\rho_{g_1' g_j}(v) + i\sum_{g_2'} E_2^0 D_{e g_2'}\rho_{g_2' g_j}(v) - iE_j^0 D_{e g_j}\rho_{ee}(v) - \frac{\gamma}{2}\rho_{e g_j}(v) + i(\omega_j - \omega_{e g_j} - k_j v)\rho_{e g_j}(v) + S\{\hat\rho(v)\}_{e g_j}. \quad (12)$$

For brevity, the arguments z and t in the density matrix are omitted. For a fixed value of j, the subscript g_j runs through all levels of the first ($j = 1$) or second ($j = 2$) lower multiplet, the subscript g runs through all lower levels and the subscript e runs through all upper levels; ω_{ab} are the frequencies of the transitions between the levels $|a\rangle$ and $|b\rangle$; N_g is the number of all lower levels; γ is the rate of spontaneous decay of the excited state of an isolated atom; and $q = k_1 - k_2$ is the difference wave number. The dot denotes the total derivative: $\dot\rho_{ab} = (\partial/\partial t + v\partial/\partial z)\rho_{ab}$.

In the class of problems of interest to us, in particular in the field of optical pumping of quantum rotation sensors, quantum frequency standards and quantum magnetometry, use is made of weak electromagnetic fields that slightly modify the equilibrium gas-kinetic distribution function of atoms. All collision processes are much faster than the processes associated with the field excitation. This means that the Rabi frequency Ω of such a field is much lower than the collision frequency ν of the alkali atom. At a buffer gas concentration $n_{buf} \sim 10^{17}$ cm^{-3} and a temperature of 60 °C, the collision frequency is $\nu \sim 10^8$ s^{-1}. The radiation intensities used in frequency standards and magnetometers are equal to 0.001–1 mW cm^{-2}, which corresponds to the frequencies $\Omega \approx 10^5$–10^7 s^{-1}. This allows us to assume that the population of the excited level is much less than the population of the ground level: $\rho_{ee} \ll \rho_{gg}$. Therefore, in equation (12) for optical coherences one can neglect the term containing ρ_{ee} (adiabatic approximation). This also allows coherences between excited levels to be neglected: $\rho_{ee'} \approx 0$.

The propagation of the field in a cell is described by the wave equation for complex amplitudes [54], which in the one-dimensional approximation has the form

$$\left(\frac{\partial}{\partial z} + \frac{1}{c}\frac{\partial}{\partial t}\right)\boldsymbol{E}_j^0(z,t) = 4\pi i k_j \boldsymbol{P}_j^0(z,t),\ j = 1, 2, \quad (13)$$

where the slow amplitude of the gas polarisation can be calculated as a quantum-mechanical mean value:

$$\boldsymbol{P}^0(z,t) = n_a \mathrm{Sp}(\hat\rho(z,t)\hat{\boldsymbol{d}}) = \boldsymbol{e}_+ n_a \mathrm{Sp}(\hat\rho(z,t)\hat d^+) + \boldsymbol{e}_0 n_a \mathrm{Sp}(\hat\rho(z,t)\hat d^0) + \boldsymbol{e}_- n_a \mathrm{Sp}(\hat\rho(z,t)\hat d^-), \quad (14)$$

where n_a is the concentration of active atoms; and $\hat\rho(z,t) = \int\hat\rho(v,z,t)\mathrm{d}v$. The complex amplitude of each of the two frequency components ($j = 1, 2$) can be decomposed in a cyclic basis into the left and right circular components:

$$\boldsymbol{E}_j^0(z) = E_j^+(z)\boldsymbol{e}^+ + E_j^-(z)\boldsymbol{e}^-,\ j = 1, 2,$$
$$E_j^\pm(z) = p_j^\pm(z)E_j^0(z). \quad (15)$$

Then, equation (13) eventually transforms into four equations for each frequency and circular component. In the stationary case, they can be written in the form:

$$\frac{\partial E_j^\pm(z)}{\partial z} = 4\pi i k_j n_a \sum_{e,g_j}\rho_{e g_j}(z)\, d_{g_j e}^\pm,\ j = 1, 2. \quad (16)$$

Thus, solving the system of equations (8)–(12) for the density matrix of atoms in the stationary case together with field transfer equations (16), one can find the distributions of the field and density matrix over the cell volume.

3. Discussion of the results

Usually, in the schemes of optical detection of the CPT resonance, for example, in work [16], the total intensity of light is measured at the output from the cell. However, the isolation of individual polarisation components, both linear and cyclic, can provide additional information. Let us follow the propagation of the polarisation components of two-frequency laser radiation in the case of tuning to the D_1 line and under the action on an optically dense gas cell with ^{87}Rb atoms. Transitions excited in the ^{87}Rb atom are shown in Fig. 2.

At the cell input, both field components are linearly polarised, the polarisation vectors being parallel to the x axis (the lin || lin configuration [14]):

$$\boldsymbol{e}_{1,2}\big|_{z=0} = \boldsymbol{e}_x = \frac{1}{\sqrt2}(\boldsymbol{e}^- - \boldsymbol{e}^+), \quad (17)$$

where $\boldsymbol{e}_x$ is the unit vector in the direction of the x axis; and $\boldsymbol{e}^\pm$ are the cyclic unit vectors [55]. A constant external magnetic field $\boldsymbol{B}$ is applied to the cell, which is necessary to remove the degeneracy of the energies of the Zeeman sublevels and to resolve the CPT resonances on the $1 \to 7$ and $3 \to 5$ transitions.

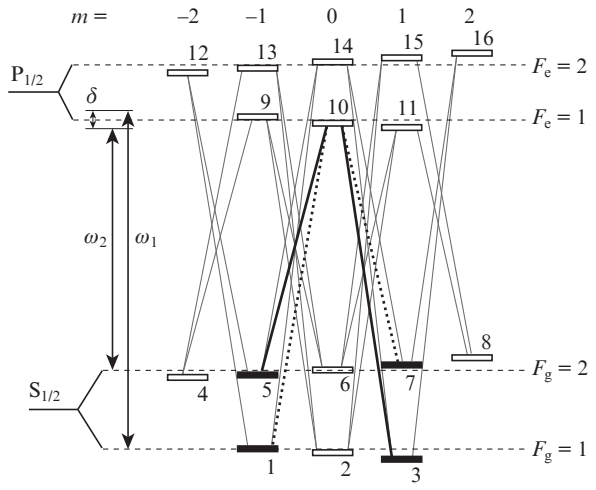

Figure 2. Energy level diagram of the D_1 line of the ^{87}Rb atom and transitions (thin gray lines) excited by a laser field with carrier frequencies ω_1 and ω_2 and two-photon detuning δ. Skew thick lines indicate two Λ-schemes (solid and dashed lines), leading to the appearance of magnetically dependent dark resonances during lin||lin excitation when tuning to a level with a total moment $F_e = 1$.

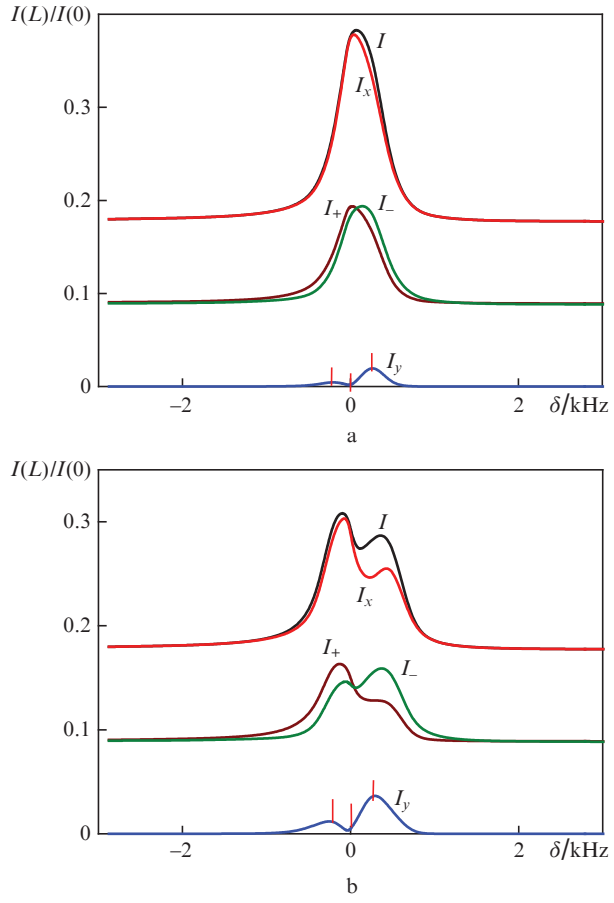

Figure 3. (Colour online) Shape of the CPT resonance when detecting different polarisation components for $\boldsymbol{B}$ = (a) 0.1 and (b) 0.2 Gs (notches on the curves indicate the points for which the polarisation ellipses are shown in Fig. 4). Input intensities, $I_1 = I_2 = 0.4$ mW cm^{-2}; polarisation, lin||lin; tuning to the level with $F_e = 1$; concentration of the buffer gas (nitrogen), $n_{buf} = 2.9\times10^{17}$ cm^{-3}; temperature, $T = 48.5$°C; cell length, $L = 6$ cm.

Figure 3 shows the CPT resonances when detecting the forward transmitted intensity of different polarisation radiation components. Detected are the intensities of the linear components, I_x and I_y, in the case of projections onto the Cartesian axes, as well as the intensities of the cyclic components, I_+ and I_-, found from the expressions:

$$I_{\pm} = \frac{c}{4\pi}\left(|p_1^{\pm}E_1^0|^2 + |p_2^{\pm}E_2^0|^2\right),$$

$$I_{x,y} = \frac{c}{4\pi}\left(|p_1^{x,y}E_1^0|^2 + |p_2^{x,y}E_2^0|^2\right), \qquad (18)$$

$$p_j^x = \frac{1}{\sqrt{2}}\left(p_j^- - p_j^+\right), \quad p_j^y = -\frac{\mathrm{i}}{\sqrt{2}}\left(p_j^- + p_j^+\right), \quad j = 1, 2.$$

$I = I_+ + I_- = I_x + I_y$ is the total intensity. In expressions (18), there are no interference terms determining the signal beating at the difference frequency, since it is assumed that at a large time τ, the signal accumulation at the beat photodetector is averaged. This assumption is fulfilled when the signal accumulation time on the photodetector is much greater than the beat period: $\tau \gg 2\pi/(\omega_1 - \omega_2)$. When the D_1 line of the ^{87}Rb atom is excited, the period of the beats is 0.15 ns, which is much shorter than the averaging time of the detectors used in the experiments.

Figure 4 shows the hodographs of the vector of the electric field strength of a laser wave in the xy plane at the cell output, normalised to a unit length. It is seen that after the passage through the gas cell, the radiation polarisation plane rotates and its ellipticity appears [Fig. 4, curves (*1*) and (*3*)]. The rotation is proportional to the magnitude of the magnetic field $\boldsymbol{B}$ and is due to the difference in the refractive indices for the left- and right-hand circular polarisation components of radiation (the Faraday effect). The appearance of ellipticity is due to the difference in the absorption coefficients for these components. When the two-photon detuning is zero, a CPT resonance arises, and the interaction of the atoms with the field weakens; therefore, the rotation of the polarisation plane and its ellipticity appear to a lesser extent [Figs 4a, 4b, curves (*2*)]. However, as the magnetic field increases, the CPT resonance splits into two peaks (see Fig. 3b), which is associated with the appearance of the so-called pseudoresonance with the shift of levels 1, 3, 5 and 7 in a magnetic field [14, 15]. In this case, when the two-photon detuning is zero, the atom begins to interact with radiation more strongly, and so a rotation occurs and the ellipticity of the polarisation arises, as seen in Figs 4c, 4d [curves (*2*)].

As the magnetic field decreases from 0.2 to 0.1 Gs, the pseudoresonance ceases to be resolved when the total intensity I is detected, but the splitting remains resolvable on the curve corresponding to the y-component [see Fig. 3a, curve (*1*)]. Thus, in this case, the observation of the intensity of an individual y-component of polarisation gives more complete information about the CPT-resonance structure than when observing the total intensity, although the signal level is significantly reduced.

In conclusion, we note that the appearance of polarisation ellipticity in pumping alkali atoms with bichromatic laser radiation is manifested as a change in the signal amplitude in the recording system when the light is transmitted through a polarisation plate. This feature must be taken into account when developing quantum frequency standards and quantum rotation sensors based on nuclear magnetic resonance [56, 57].

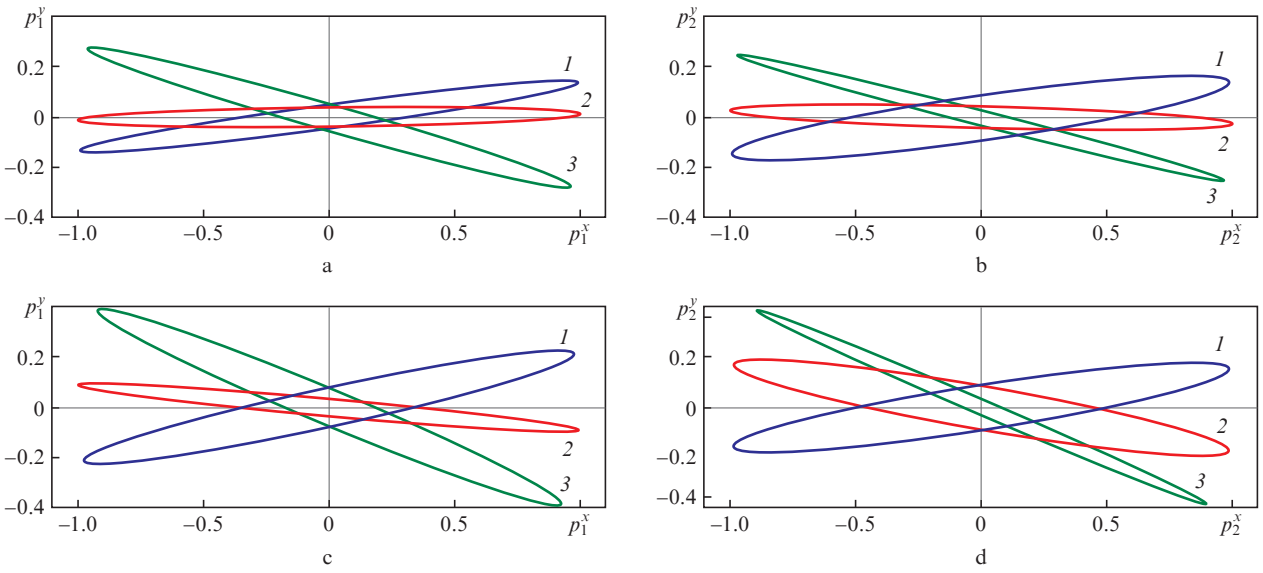

Figure 4. (Colour online) Polarisation ellipses of output radiation at detunings $\delta = (1)$ –208, (2) 0 and (3) 270 Hz (see the notches in Fig. 3). The magnetic field is B = (a, b) 0.1 and (c, d) 0.2 Gs. The parameters are the same as in Fig. 3.

4. Conclusions

In this paper, we have derived quantum kinetic equations for the density matrix in the case of excitation of alkali metal atoms by arbitrarily polarised bichromatic laser radiation in order to calculate the CPT signal. The equations are supplemented by a system of field transfer equations that describe the propagation of two frequency and two polarisation radiation components in an optically dense gas cell at above room temperature.

The presented mathematical model is a generalisation of the model constructed in [52, 53] to the case of the presence of complete hyperfine and Zeeman structures of the levels of an alkaline atom. The model makes it possible to correctly take into account the polarisation of laser radiation and its variation upon propagation along the cell, as well as the effect of a constant magnetic field.

An analysis is made of the CPT signal with respect to the intensity of various polarisation components of transmitted radiation. The polarisation plane rotation and the ellipticity appearance of the radiation linearly polarised along the x axis at the input are revealed. It is shown that the detection of the transverse y-component carries additional information about the structure of the main magneto-independent CPT resonance in comparison with the total-intensity signal.

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