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# Effect of collisions on the shape of the coherent population trapping resonance detected by the Ramsey method

G.V. Voloshin, K.A. Barantsev, A.N. Litvinov

***Abstract.*** **We report on a theoretical model of the effect of coherent population trapping in alkali atoms in an optically thin gas cell with a buffer gas at a nonzero temperature in a pulsed laser field. The shape of the Ramsey resonance is analysed for two different temperature ranges, i. e. for a 'cold' atomic ensemble and for an ensemble of 'hot' atoms in a cell with a buffer gas. The influence of the hyperfine structure of the excited level on the shift of the central Ramsey resonance is investigated.**

***Keywords:*** *coherent population trapping, pulsed pumping, gas cell, alkali atoms, moving atoms.*

## 1. Introduction

The interaction of bichromatic laser radiation with atomic ensembles leads under certain conditions to the appearance of the coherent population trapping (CPT) phenomenon [1–4]. A distinctive feature of this effect is the possibility of observing narrow resonances that are free of Doppler broadening of the optical transition. The CPT resonance width can reach hundreds or even tens of hertz. This opens up wide possibilities for using the CPT phenomenon in various practical applications, including in optical magnetometers [5, 6], high-resolution spectroscopic devices [7, 8], as well as for the development of quantum-information recording and storing devices [9–11] and inversionless lasers [12–15]. A special place in the application of the CPT phenomenon is occupied by small-size quantum frequency standards [16–21].

Recently, the use of pulsed pumping to excite CPT resonance (Ramsey scheme) has attracted considerable interest [22]. The essence of this method is that the atomic ensemble interacts with two successive (pump and interrogation) pulses separated by a dark pause. It turns out that with such an interrogation scheme, the width of the CPT resonance is determined only by the dark pause, which makes it possible to obtain a much narrower CPT resonance [23].

The essence of this method, as well as of its various modifications as applied to the CPT effect, has been studied in detail in a number of works. Research in this area was initiated in the pioneering work by Zanon-Willette et al. [24], who reported the observation of Raman–Ramsey fringes using a double lambda scheme creating coherent population trapping in an atomic ensemble excited by pulsed optical radiation. Liu et al. [25] experimentally observed high-contrast and narrow CPT resonances with the Ramsey interrogation scheme in atomic vapours of $^{133}$Cs. In order to increase the stability of the atomic clock, Yano et al. [26, 27] suggested using the pulsed two-step method to excite CPT resonances. This method is a Raman–Ramsey scheme relying on the use of laser pulses with a lower light intensity, which makes it possible to provide a lower sensitivity of the CPT resonance shape to changes in the light intensity. The influence of the buffer gas pressure on the CPT resonance shape in the Raman–Ramsey scheme for $^{87}$Rb atoms was considered by Kuchina et al. [28]. High-contrast and narrow CPT resonances under pulsed pumping in atomic vapours of $^{133}$Cs were studied by Hafiz et al. [29], who demonstrated the stability of an atomic clock at a level of $2.3 \times 10^{-13}$ per 100 s. This paper also notes that the width of the central Ramsey resonance may be narrower than the expected width of the Ramsey line. Similar features were observed by Boudot et al. [30] when studying CPT resonances in the Raman–Ramsey scheme in microcells filled with Cs–Ne vapours. Experimental study of CPT resonances using the Raman–Ramsey technique in cells containing an $^{87}$Rb–Ar–Ne gas mixture for the lin$\|$lin polarisations was reported by Baryshev et al. [31]. Hafiz et al. [32, 33] proposed a scheme based on a symmetric autobalanced Ramsey pulse train, which made it possible to reduce the light shift by an order of magnitude. Using the proposed method, the authors have implemented a high-precision atomic clock based on $^{133}$Cs, with the contribution of the light shift to the frequency stability now varying within $10^{-16}$ at averaging times of $10^4$ s. Lenci et al. [34] observed Ramsey bands when studying the effect of light-induced transparency in a cell with a buffer gas.

The development of frequency standards requires an increase in the signal, which can be realised by increasing the temperature. In this case, the number of active atoms increases, and at a certain concentration of them we can speak of an optically dense medium. The influence of the optical density of a medium on the shape of the CPT resonance line in the implementation of the Raman–Ramsey scheme was considered theoretically in Refs [35–37].

Note that, despite a fairly large number of studies on the CPT effect in gas cells in the Raman–Ramsey scheme (see, for example, [28–33]), they are mostly experimental. The purpose of this work is to construct a theory of the CPT effect for a nonzero temperature in the presence of a buffer gas in the case of a Ramsey interrogation scheme; in this case, special attention is paid to the influence of the presence of the hyperfine structure of the excited level on the shift of the central Ramsey resonance.

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## 2. Mathematical model

Let us assume that an ensemble of atoms resides at a nonzero temperature in the field of a plane electromagnetic wave with two carrier frequencies (Fig. 1a). The wave propagates along the $z$ axis, and the electric field intensity is expressed as

$$\boldsymbol{E}(z,t) = \boldsymbol{e}_1 E_1 \exp[-\mathrm{i}(\omega_1 t - k_1 z)] + \boldsymbol{e}_2 E_2 \exp[-\mathrm{i}(\omega_2 t - k_2 z)] + \text{c.c.}, \quad (1)$$

where $E_j, \boldsymbol{e}_j$ and $k_j$ are, in the general case, the complex amplitude of intensity, the unit vector along the polarisation direction, and the wavenumber, respectively ($j = 1, 2$).

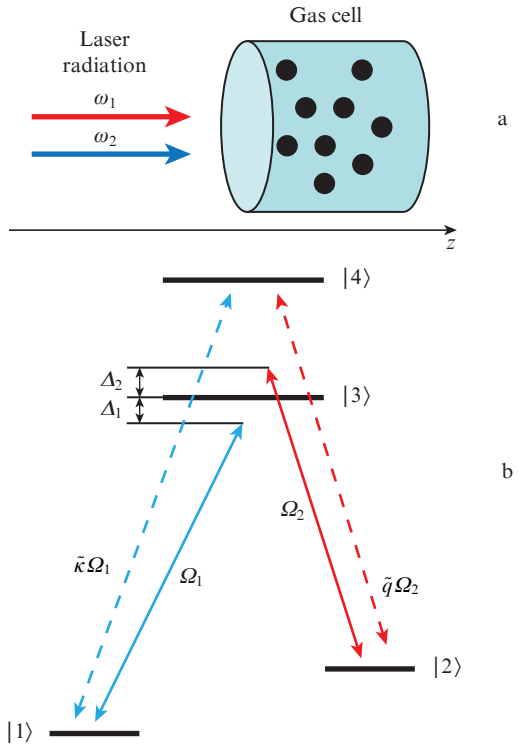


**Figure 1.** (a) Gas cell with active atoms and a buffer gas and the direction of propagation of two-frequency laser radiation, as well as (b) energy level diagram of active atoms and excited transitions.

Let us consider a model in which atoms have four energy levels: two ground levels ($|1\rangle$ and $|2\rangle$) corresponding to the hyperfine splitting of the s-state; and two excited levels ($|3\rangle$ and $|4\rangle$), corresponding to the hyperfine splitting of the p-state (Fig. 1b). The frequencies of the fields, $\omega_1$ and $\omega_2$, are close to the frequencies of the $|1\rangle \leftrightarrow |3\rangle$ and $|2\rangle \leftrightarrow |3\rangle$ transitions with detunings $\Delta_1$ and $\Delta_2$, respectively, and $\omega_{34}$ is the frequency of the hyperfine transition between the excited state levels $|3\rangle$ and $|4\rangle$.

The state of an ensemble of atoms will be described using a single-atom density matrix $\hat{\rho}(\boldsymbol{p},\boldsymbol{r},t)$, which in the Wigner representation in the translational degrees of freedom of the atom satisfies the following quantum kinetic equation:

$$\left(\frac{\partial}{\partial t} + \frac{\boldsymbol{p}}{m}\nabla\right)\hat{\rho}(\boldsymbol{p},\boldsymbol{r},t) = -\frac{\mathrm{i}}{\hbar}[\hat{H},\hat{\rho}(\boldsymbol{p},\boldsymbol{r},t)] + \hat{\hat{R}}\{\hat{\rho}(\boldsymbol{p},\boldsymbol{r},t)\} + \hat{\hat{S}}\{\hat{\rho}(\boldsymbol{p},\boldsymbol{r},t)\}, \quad (2)$$

where $\boldsymbol{p} = m\boldsymbol{v}$ is the momentum of the atom; $m$ is the mass of the atom; $\hat{H}$ is the Hamiltonian of the atom, which takes into account its interaction with the external field; $\hat{\hat{R}}$ is a superoperator that phenomenologically takes into account the spontaneous relaxation of atoms of the ensemble; and $\hat{\hat{S}}$ is a superoperator that takes into account collisions between active and buffer atoms.

The Hamiltonian can be represented as $\hat{H} = \hat{H}_0 + \hat{V}$, where

$$\hat{H}_0 = \sum_{n=1}^{4} \varepsilon_n |n\rangle\langle n| \quad (3)$$

is the Hamiltonian of the system in the absence of a field, and $\hat{V}$ is the operator of interaction with the field, which in the dipole approximation has the form

$$\hat{V} = -\hat{\boldsymbol{d}}\boldsymbol{E} = \hbar\Omega_1 \exp[-\mathrm{i}(\omega_1 t - k_1 z)]|3\rangle\langle 1| + \hbar\Omega_2 \exp[-\mathrm{i}(\omega_2 t - k_2 z)]|3\rangle\langle 2| + \text{h.c.} \quad (4)$$

Here $\hat{\boldsymbol{d}} = \boldsymbol{e}_\mathrm{d}\hat{d}$ is the atomic dipole moment operator, $\Omega_j = E_j d_{3j}/\hbar$ are the Rabi frequencies ($j = 1, 2$), and it is also assumed that the polarisation vectors of the incident waves are codirectional with the dipole moment vector $[(\boldsymbol{e}_\mathrm{d}, \boldsymbol{e}_{1,2}) = 1]$, and elements of the matrix of the dipole moment $d_{12} = 0$ due to the fact that the electric dipole transition $|1\rangle \leftrightarrow |2\rangle$ is forbidden; and $\tilde{\kappa} = d_{41}/d_{31}$ and $\tilde{q} = d_{42}/d_{32}$ are the ratios of the matrix elements of the dipole moment.

Since we assume the front of the incident wave to be plane and infinite, the problem can be reduced to one-dimensional one in coordinate. In this case, the density matrix will depend only on the projection of the atomic velocity along the radiation propagation direction and on the $z$ coordinate: $\hat{\rho}(\boldsymbol{p},\boldsymbol{r},t) = \hat{\rho}(v,z,t)$, where $v = v_z$.

Let us make a substitution that selects rapidly oscillating factors in the off-diagonal elements of the density matrix:

$$\tilde{\rho}_{ge} = \rho_{ge}\exp[-\mathrm{i}(\omega_g t - k_g z)], \quad g = 1,2, \quad e = 3,4, \quad (5)$$

$$\tilde{\rho}_{12} = \rho_{12}\exp[-\mathrm{i}(\omega_1 - \omega_2)t + \mathrm{i}(k_1 - k_2)z], \quad (6)$$

where $\rho_{nm}(v,z) = \langle n|\,\hat{\rho}(v,z)\,|m\rangle$, after which we use the rotating wave approximation.

We will assume that the medium is optically thin along the wave propagation direction, i.e. the mean free path of a photon is much greater than the length $L$ of the atomic ensemble in this direction ($n_\mathrm{a}\sigma L \leqslant 1$, where $n_\mathrm{a}$ is the concentration of active atoms and $\sigma$ is the effective cross section for photon scattering by an atom of the ensemble). Thus, neglecting the dependence on $z$, we use the model of strong collisions [38, 39] to write system (2) in the form [37]

$$\begin{aligned}\dot{\rho}_{11}(v) = {} & -\mathrm{i}\Omega_1\tilde{\rho}_{13}(v) + \mathrm{i}\Omega_1^*\tilde{\rho}_{31}(v) - \mathrm{i}\tilde{\kappa}\Omega_1\tilde{\rho}_{14} + \mathrm{i}\tilde{\kappa}^*\Omega_1^*\tilde{\rho}_{41} \\ & + \frac{\gamma}{2}(\rho_{33}(v) + \tilde{\kappa}^2\rho_{44}(v)) - \nu\rho_{11}(v) \\ & + \nu_{11}M(v)\int\rho_{11}(v')\,\mathrm{d}v' + \nu_{21}M(v)\int\rho_{22}(v')\,\mathrm{d}v' \\ & + \nu_{31}M(v)\int\rho_{33}(v')\,\mathrm{d}v' + \nu_{41}M(v)\int\rho_{44}(v')\,\mathrm{d}v', \end{aligned} \quad (7)$$

$$\dot\rho_{22}(v) = -i\Omega_2\tilde\rho_{23}(v) + i\Omega_2^*\tilde\rho_{32}(v) - i\tilde q\,\Omega_2\tilde\rho_{24} + i\tilde q^*\Omega_2^*\tilde\rho_{42} + \frac{\gamma}{2}(\rho_{33}(v) + \tilde q^2\rho_{44}(v)) - v\rho_{22}(v) + v_{22}M(v)\int\rho_{22}(v')\,dv' + v_{12}M(v)\int\rho_{11}(v')\,dv' + v_{32}M(v)\int\rho_{33}(v')\,dv' + v_{42}M(v)\int\rho_{44}(v')\,dv', \quad (8)$$

$$\dot\rho_{33}(v) = -i\Omega_1\tilde\rho_{13}(v) - i\Omega_1^*\tilde\rho_{31}(v) + i\Omega_2\tilde\rho_{23}(v) - i\Omega_2^*\tilde\rho_{32}(v) - \gamma\rho_{33}(v) - v\rho_{33}(v) + v_{33}M(v)\int\rho_{33}(v')\,dv' + v_{43}M(v)\int\rho_{44}(v')\,dv', \quad (9)$$

$$\dot\rho_{44}(v) = i\tilde k\Omega_1\tilde\rho_{14}(v) - i\tilde k^*\Omega_1^*\tilde\rho_{41}(v) + i\tilde q\,\Omega_2\tilde\rho_{24}(v) - i\tilde q^*\Omega_2^*\tilde\rho_{42}(v) - \frac{\gamma}{2}(\tilde k^2 + \tilde q^2)\rho_{44}(v) - v\rho_{44}(v) + v_{44}M(v)\int\rho_{44}(v')\,dv' + v_{34}M(v)\int\rho_{33}(v')\,dv', \quad (10)$$

$$\dot{\tilde\rho}_{12}(v) = \dot{\tilde\rho}_{21}^*(v) = i\Omega_1^*\tilde\rho_{32}(v) - i\Omega_2\tilde\rho_{13}(v) + i\tilde k^*\Omega_1^*\tilde\rho_{42} - i\tilde q\,\Omega_2\tilde\rho_{14} + [i((\Delta_2 - \Delta_1) + (k_1 - k_2)v) - \Gamma_{12} - v]\tilde\rho_{12}(v) + v_{\rm coh}^{vv'}M(v)\int\tilde\rho_{12}(v')\,dv', \quad (11)$$

$$\dot{\tilde\rho}_{13}(v) = \dot{\tilde\rho}_{31}^*(v) = -i\Omega_1^*\rho_{11}(v) - i\Omega_2^*\tilde\rho_{12}(v) + i\Omega_1^*\rho_{33}(v) + [-i(\Delta_1 - k_1v) - \Gamma - v]\tilde\rho_{13}(v), \quad (12)$$

$$\dot{\tilde\rho}_{14}(v) = \dot{\tilde\rho}_{41}^*(v) = -i\tilde k^*\Omega_1^*\rho_{11}(v) - i\tilde q^*\Omega_2^*\tilde\rho_{12}(v) + i\tilde k^*\Omega_1^*\rho_{44}(v) + [-i(\Delta_1 - \omega_{34} - k_1v) - \Gamma - v]\tilde\rho_{14}(v), \quad (13)$$

$$\dot{\tilde\rho}_{23}(v) = \dot{\tilde\rho}_{32}^*(v) = -i\Omega_1^*\tilde\rho_{21}(v) - i\Omega_2^*\rho_{22}(v) + \Omega_2^*\rho_{33}(v) + [-i(\Delta_2 - k_2v) - \Gamma - v]\tilde\rho_{23}(v), \quad (14)$$

$$\dot{\tilde\rho}_{24}(v) = \dot{\tilde\rho}_{42}^*(v) = -i\tilde k^*\Omega_1^*\tilde\rho_{21}(v) - i\tilde q^*\Omega_2^*\rho_{22}(v) + i\tilde q^*\Omega_2^*\rho_{44}(v) + [-i(\Delta_2 - \omega_{34} - k_2v) - \Gamma - v]\tilde\rho_{24}(v). \quad (15)$$

The argument $t$ in formulae (7)–(15) is omitted for brevity. Here $\gamma$ is the rate of spontaneous decay of the excited state; $\Gamma$ is the decay rate of optical coherences; $\Gamma_{12}$ is the decay rate of low-frequency coherence (all decay rates are due to the interaction of an atom with a vacuum thermostat); $M(v) = (\sqrt{\pi}\,v_{\rm T})^{-1}\exp(-v^2/v_{\rm T}^2)$ is the Maxwell distribution function over the velocity projection; $v_{\rm T} = \sqrt{2kT/m}$ is the most probable velocity; $v$ is the total collision frequency; $v_{ij}$ are the frequencies of inelastic collisions with the transitions from the state $(|i\rangle$ to the state $|j\rangle$; and $v_{\rm coh}^{vv'}$ is the frequency of collisions at which low-frequency coherence is maintained. The collision frequencies are calculated based on the gas-kinetic formula $v_\alpha = (n_{\rm a} + n_{\rm buf})\sigma_\alpha\bar u$, where $n_{\rm a}$ and $n_{\rm buf}$ are the concentrations of active atoms and buffer gas atoms; $\sigma_\alpha$ is the section of the corresponding process; $\bar u = \sqrt{8kT/\pi\mu}$ is the average thermal velocity; and $\mu = m_{\rm buf}m_{\rm a}/(m_{\rm a} + m_{\rm buf})$ is the reduced mass. Note that the concentration $n_{\rm buf}$ in the ensemble is much higher than the concentration $n_{\rm a}$. Because of this, the temperature dependence of the frequency $v$ can be neglected, since only the concentration $n_{\rm a}$ depends on the latter.

The system of equations (7)–(15) is written in the approximation when the optical pump rate is much lower than the rates of gas kinetic processes and processes of spontaneous relaxation of the excited state, $|\Omega_{12}| \ll v,\gamma$. In this case, the populations of the excited state are small $[\int\rho_{ee}(v,t)dv \ll 1]$; therefore, the coherence $\rho_{34}(v,t)$ can be neglected.

Solving this system of equations is a challenging task. In order to simplify it, we will use the reduced density matrix

$$\rho_{ij}(t) = \int\rho_{ij}(v,t)\,dv, \quad i,j = 1,2,3,4. \quad (16)$$

To this end, we integrate equations (7)–(15) over the velocity, assuming the velocity distributions of the populations of the ground states and low-frequency coherences to be Maxwellian:

$$\rho_{jj}(v,t) = M(v)\rho_{jj}(t), \quad j = 1,2, \quad (17)$$

$$\tilde\rho_{12}(v,t) = M(v)\tilde\rho_{12}(t). \quad (18)$$

Then, we write down Eqns (7)–(11) for the reduced elements of the density matrix (relations for the collision frequencies were considered in [39]):

$$\dot\rho_{11} = -2{\rm Re}\{i\Omega_1[\tilde\rho_{13} + \tilde\kappa\tilde\rho_{14}]\} + \frac{\gamma}{2}(\rho_{33} + \tilde\kappa^2\rho_{44}) + (v_{11} - v)\rho_{11} + v_{21}\rho_{22} + v_{31}\rho_{33} + v_{41}\rho_{44}, \quad (19)$$

$$\dot\rho_{22} = -2{\rm Re}\{i\Omega_2[\tilde\rho_{23} + \tilde\kappa\tilde\rho_{14}]\} + \frac{\gamma}{2}(\rho_{33} + \tilde q^2\rho_{44}) + (v_{22} - v)\rho_{22} + v_{12}\rho_{11} + v_{32}\rho_{33} + v_{42}\rho_{44}, \quad (20)$$

$$\dot\rho_{33} = 2{\rm Re}\{i[\Omega_1\tilde\rho_{13} + \Omega_2\tilde\rho_{23}]\} - \gamma'\rho_{33} + v_{43}\rho_{44}, \quad (21)$$

$$\dot\rho_{44} = 2{\rm Re}\{i[\Omega_1\tilde\kappa\tilde\rho_{14} + \Omega_2\tilde q\,\tilde\rho_{24}]\} - \frac{\gamma}{2}(\tilde\kappa^2 + \tilde q^2)\rho_{44} + (v - v_{44})\rho_{44} + v_{34}\rho_{33}, \quad (22)$$

$$\dot{\tilde\rho}_{12} = i[\Omega_1^*\tilde\rho_{32} - \Omega_2\tilde\rho_{13} + \tilde k^*\Omega_1^*\tilde\rho_{42} - \tilde q\,\Omega_2\tilde\rho_{14}] + [i\delta - \Gamma_{12}']\tilde\rho_{12}, \quad (23)$$

where $\gamma' = \gamma + v - v_{33}$ and $\Gamma_{12}' = \Gamma_{12} + v - v_{\rm coh}^{vv'}$ are the rates of decay of the excited level and low-frequency coherence modified due to collisions; and $\delta = \Delta_2 - \Delta_1$ is the two-photon detuning. In our case, assumptions (17) and (18) are only used in obtaining equation (23).

For optical coherences, the velocity distribution cannot be considered Maxwellian, since they are rapidly destroyed in collisions. Therefore, it is necessary to find these distributions and integrate them in velocity. To this end, we find solutions to equations (12)–(15) in quadratures by the method of variation of an arbitrary constant. Let us take into account that in the approximation of weak fields the adiabatic approximation

is valid, in which $\rho_{33,44} \ll \rho_{11,22}$. This allows us to neglect the third terms on the right-hand sides of equations (12)–(15). Using expressions (12)–(15), we write

$$\tilde{\rho}_{j3}(t) = -\mathrm{i}\int_0^t \mathrm{d}t'[\Omega_2^*(t')\tilde{\rho}_{j2}(t') + \Omega_1^*(t')\tilde{\rho}_{j1}(t')] \times \int_{-\infty}^{\infty} \mathrm{d}v M(v)\exp[-(\mathrm{i}(\Delta_j - k_j v) + \Gamma')(t-t')], \quad (24)$$

$$\tilde{\rho}_{j4}(t) = -\mathrm{i}\int_0^t \mathrm{d}t'[\tilde{q}^*\Omega_2^*(t')\tilde{\rho}_{j2}(t') + \tilde{\kappa}^*\Omega_1^*(t')\tilde{\rho}_{j1}(t')] \times \int_{-\infty}^{\infty} \mathrm{d}v M(v)\exp[-(\mathrm{i}(\Delta_j - \omega_{34} - k_j v) + \Gamma')(t-t')], \quad (25)$$

where $\Gamma' = \Gamma + \nu$ is the collision-modified decay rate of optical coherences. The velocity integrals in (24) and (25) are real in the sense of the principal value and can be calculated analytically:

$$\tilde{\rho}_{j3}(t) = -\mathrm{i}\int_0^t \mathrm{d}t' \exp\left[\frac{v_\mathrm{T}^2 k_j^2}{4}(t-t')^2 - (\mathrm{i}\Delta_j + \Gamma')(t-t')\right] \times [\Omega_2^*(t')\tilde{\rho}_{j2}(t') + \Omega_1^*(t')\tilde{\rho}_{j1}(t')], \quad (26)$$

$$\tilde{\rho}_{j4}(t) = -\mathrm{i}\int_0^t \mathrm{d}t' \exp\left[\frac{v_\mathrm{T}^2 k_j^2}{4}(t-t')^2 - (\mathrm{i}(\Delta_j - \omega_{34}) + \Gamma') \times (t-t')\right][\tilde{q}^*\Omega_2^*(t')\tilde{\rho}_{j2}(t') + \tilde{\kappa}^*\Omega_1^*(t')\tilde{\rho}_{j1}(t')]. \quad (27)$$

In formulae (26) and (27), the first term in the exponent describes a decrease in the interaction of atoms with radiation due to Doppler detuning, while the second denotes the homogeneous broadening of absorption lines due to collisions.

Next, substituting expressions for $\tilde{\rho}_{j3}$ and $\tilde{\rho}_{j4}$ from (24) and (25) into system (19)–(23), we arrive at a system of Volterra integro-differential equations of the first kind, which can be represented in the general form as

$$\dot{y}_i(t) = \int_0^t K_{ij}(t,t')y_j(t')\,\mathrm{d}t' + A_{ij}y_j(t), \quad (28)$$

where $y_i(t)$ is the function to be found; $K_{ij}(t,t')$ is the kernel of the integral equation; and $A_{ij}$ is a constant matrix (here the silent summation rule is used).

To solve the system of equations (28), we applied a method based on the use of a difference scheme. The essence of this method is to replace the integral with its approximate value using the sum over the integration interval partition points $t_m \in [0,t]$ by the trapezoid method. As a result, for the step $h$ and the number of partitions $l$, expression (28) will be written in the form

$$\dot{y}_i(t) = \frac{h}{2}\sum_{m=1}^{l-1}[K_{ij}(t,t_m)y_{j,m} + K_{ij}(t,t_{m+1})y_{j,m+1}] + A_{ij}y_j(t), \quad y_{i,m} = y_i(t_m). \quad (29)$$

System (29) was solved numerically using Euler's method. The collision rates in calculations were assumed to be the following:

$$\begin{aligned} &\nu_{11} = \nu_{22} = \nu, \\ &\nu_{33} = \nu(1-\eta), \\ &\nu_{44} = \nu[1 - \eta(\tilde{\kappa}^2 + \tilde{q}^2)/2], \\ &\nu_{31} = \nu_{32} = \eta\nu/2, \\ &\nu_{41} = \nu_{14} = \eta\nu\tilde{\kappa}^2/2, \\ &\nu_{42} = \nu_{24} = \eta\nu\tilde{q}^2/2, \\ &\nu_{12}^{vv'} = \nu. \end{aligned} \quad (30)$$

Here $\eta$ is a parameter that determines the fraction of collisions leading to the decay of the excited state.

## 3. Pulsed pumping

The system of equations (19)–(23), (26), and (27) describes the behaviour of an atomic system in a laser field, whose amplitude arbitrarily changes in time. Let us consider a scheme of pulsed pumping by the Ramsey method, which makes it possible to significantly narrow the CPT resonance line (Fig. 2). The first pump pulse of duration $\tau_\mathrm{p}$, when interacting with atoms, transfers them to the CPT state. The duration of the pump pulse should be longer than the CPT settling time: $\tau_\mathrm{p} > \gamma'/\Omega^2$, where $\gamma'$ is the rate of decay of the excited state; and $\Omega$ is the characteristic Rabi frequency.

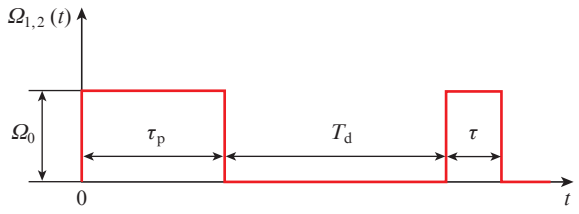


**Figure 2.** Sequence of laser pulses with the Ramsey scheme of CPT resonance interrogation: $\Omega_0$ is the Rabi frequency of the incident fields; and $\tau_\mathrm{p}$, $T_\mathrm{d}$, and $\tau$ are the durations of the pump pulse, dark pause, and interrogation pulse, respectively.

The duration of the dark pause lies within $1/\gamma' \ll T_\mathrm{d} < 1/\Gamma'_{12}$. In this case, the populations of excited levels and the optical coherences of atoms completely decay, and the low-frequency coherence freely evolves.

An interrogation pulse of duration $\tau_\mathrm{p} < \gamma'/\Omega^2$, depending on the phase incursion between low-frequency coherence and laser radiation, leads to different degrees of atomic excitation after the action of radiation, which makes it possible to detect the Ramsey resonance.

## 4. Results and discussion

### 4.1. Three-level model (lambda atom)

Let us consider the excitation of CPT resonances in atomic ensembles within the framework of a three-level model (we neglect the splitting of the excited state) for two temperature ranges: at low temperatures of 1 mK–1 K, corresponding to the case of cold atomic ensembles, the absorption line width of which is mainly affected by natural and Doppler broadening, and collisional broadening has a weak effect; and at tem-

peratures of 30–60 °C, corresponding to the temperature of a gas cell filled with saturated vapour of an alkali metal. In such a gas, the Doppler and collisional broadenings are comparable in magnitude and are two orders of magnitude higher than the natural one.

Figure 3a shows a change in the CPT resonance detected using continuous radiation (dashed curves) and the Ramsey method (solid curves) in the low temperature region for the case of cold atoms. As the temperature increases, the Doppler broadening increases and the number of atoms in resonance with radiation decreases. As a consequence, the optical pump rate decreases with decreasing population of the excited level. This leads to a significant drop in the amplitude of the central Ramsey resonance. In addition to a decrease in population, the envelopes narrow down, which is due to the fact that the width of the CPT resonance is determined by two terms: $\Gamma'_{12} + \Omega^2/\gamma'$. The first term is associated with the decay of low-frequency coherence due to collisions of atoms with each other, with the buffer gas, and with the cell walls, and the second, with light broadening under the action of laser radiation. At low temperatures, the first term is close to zero, since it is caused only by collisions of atoms with each other, and the term $\Omega^2/\gamma'$ is of the order of $10^3$–$10^5$ s$^{-1}$ for the considered Rabi frequencies. With rising temperature, the decay rates of the low-frequency coherence $\Gamma'_{12}$ and the excited level $\gamma'$ begin to increase due to the presence of inelastic collisions. For Rb atoms with a concentration of $10^{11}$ cm$^{-3}$, when temperature rises from 0.001 to 0.01 K, the increment in $\Gamma'_{12}$ is $\sim 0.1$ s$^{-1}$, and the increment in $\gamma'$ is $10^3$ s$^{-1}$, which in total leads to a decrease in the resonance width.

In the case of hot atoms (Fig. 3b) in the temperature range of 30–60 °C, at which gas cells are used, the amplitude of the Ramsey resonance decreases with increasing temperature, and its change is much less than in the case of cold atoms.

### 4.2. Account for the hyperfine structure of the excited level

Let us now consider the influence of the additional upper level caused by the hyperfine structure on the peculiarities of the Ramsey resonance. We assume that the value of hyperfine splitting is in the range $60\gamma \leqslant \omega_{34} \leqslant 160\gamma$. When the fields are tuned to the $|1\rangle - |3\rangle$ transition, fast atoms, due to the Doppler broadening, interact with the level $|4\rangle$, which leads to light shifts of optical transitions and, as a consequence, to a shift of the low-frequency $|1\rangle - |2\rangle$ transition and CPT resonance. Figure 4a shows the dependences of the light shift $S$ of the central Ramsey resonance on the temperature $T$. One can see that the position of the central Ramsey resonance cha-

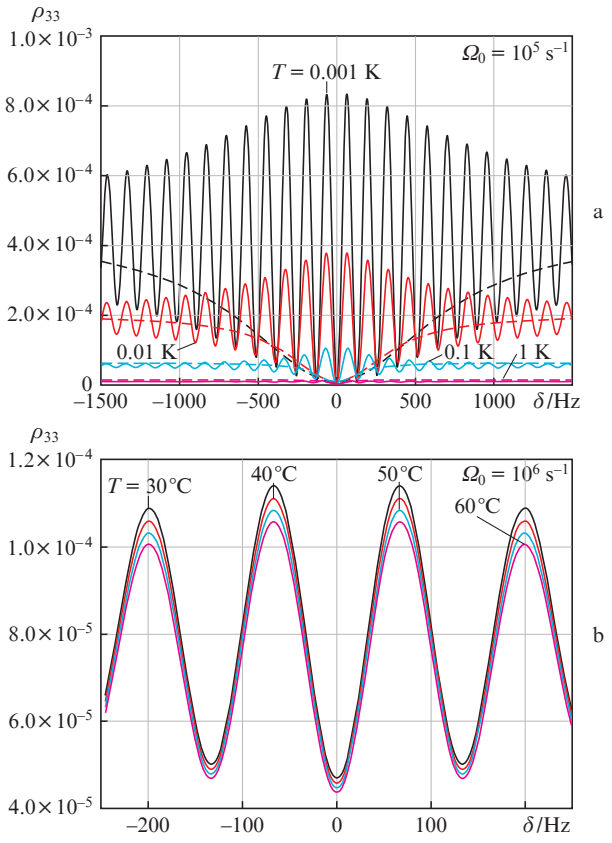


**Figure 3.** Forms of CPT resonances (a) in cold atoms and (b) in a hot gas cell (solid and dashed curves are resonances detected by the Ramsey method and continuous radiation, respectively). The duration of the interrogation pulse is $\tau = 10\gamma^{-1}$, the duration of the pump pulse $\tau_p$ corresponds to reaching a stationary state, the duration of the dark pause is $T_d = 0.8$ ms; it is also assume that $m_a = 87$ a.m.u, $m_{buf} = 40$ a.m.u, $n_a = 0.5 \times 10^{11}$ cm$^{-3}$, $n_{buf} = 10^{19}$ cm$^{-3}$, $\gamma = 10^7$ s$^{-1}$, $\Gamma = \gamma/4$, $\Gamma_{12} = 200$ s$^{-1}$, $\Delta_1 = \delta/2$, $\Delta_2 = -\delta/2$, and $\eta = 0.01$.

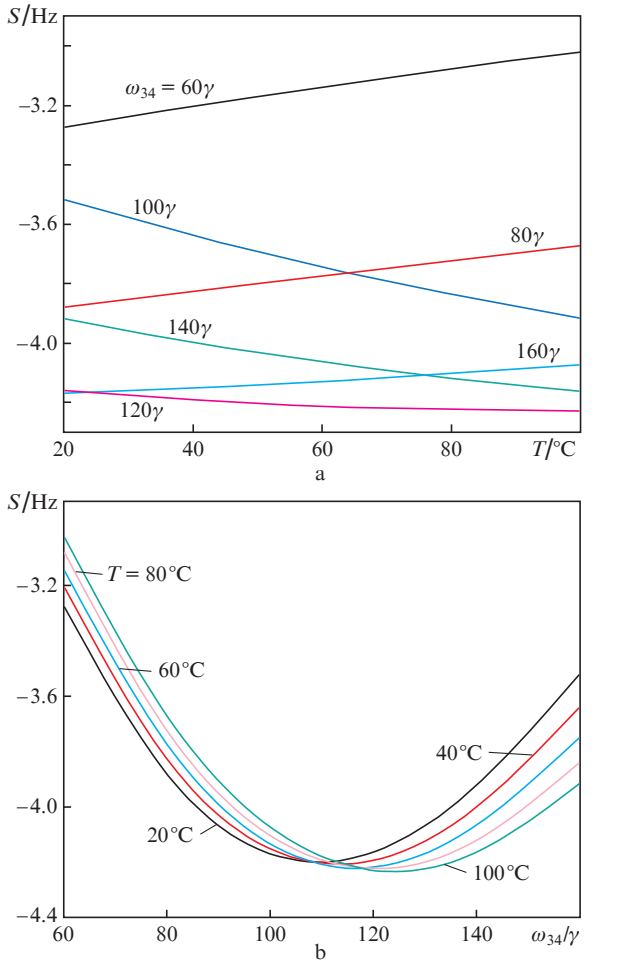


**Figure 4.** Dependences of the light shift of the central Ramsey resonance on (a) temperature for different values of hyperfine splitting $\omega_{34}$ and on (b) hyperfine splitting $\omega_{34}$ for different temperatures. The amplitude of fields is $\Omega_0 = 10^5$ s$^{-1}$, $\tilde{\kappa} = 1$, and $\tilde{q} = 0.5$; other parameters are the same as for Fig. 3.

nges. This change depends on the magnitude of the splitting of the excited state and has a nonmonotonic character. The nonmonotonicity is due to the fact that the dependence of the light shift on the detuning coincides with the shape of the dispersion contour [40]. As the temperature changes, the width of the dispersion contour changes, which is equivalent to scanning the $\omega_{34}$ splitting. Thus, if the dispersion contour of atoms is broadened consistently with the magnitude of the splitting of the excited state, the temperature dependence of the light shift can be either increasing or decreasing. The contours of the excited levels of atoms overlap only in a small group of atoms; therefore, the magnitude of the shift is units of hertz. This is in complete agreement with the experimental results [29]. It should be noted that in [17] the value of the light shift increases with increasing temperature. We believe this to be due to the fact that the effect of an optically dense medium, in which radiation pulses with different wavelengths propagate, also contributes to this shift [20, 36].

## 5. Conclusions

A theory is constructed that describes the excitation of CPT resonance by a pulsed laser field in an optically thin atomic ensemble with allowance for the motion of atoms. A system of equations is derived for the velocity-integrated elements of the atomic density matrix, in which the optical coherences are expressed in terms of quadratures. A method for solving such systems of equations is briefly described. The numerical solution of this system makes it possible to calculate the shape of the CPT resonance for the Ramsey interrogation scheme.

The shape of the Ramsey resonance is analysed for two temperature ranges, namely, for a cold atomic ensemble, in which the Doppler broadening is comparable to the natural one, and for an ensemble of hot atoms in a gas cell with a buffer gas (the Doppler and collisional broadening is much larger than the natural one). It is shown that with an increase in temperature in the case of cold atoms, the amplitude of the Ramsey resonance decreases with a simultaneous narrowing of the envelope, and in the case of hot atoms, the resonances shift to the region of lower populations with a simultaneous decrease in the amplitude. It is found that in the presence of a hyperfine structure of the excited state, a light shift of the Ramsey resonance takes place. The dependence of the light shift of the central Ramsey resonance on temperature and the magnitude of hyperfine splitting is analysed.

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