Optical Ramsey fringes induced by Zeeman coherence

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We utilize a two-photon polarization spectroscopy technique to observe Ramsey fringes that appear due to light scattering by coherently prepared rubidium atoms. We examine the effect in a vapor cell for spatially and temporally separated laser fields. We present a theoretical interpretation of the experimental results.

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I. INTRODUCTION

The Ramsey method of separated oscillatory fields is one of the most significant applications of atomic coherence [1]. The original idea of the method is based on an ability of a molecule or an atom, excited by a coherent electromagnetic field, to carry information about the field. This information can be transported by the molecule (atom) in space or in time, and be read by another electromagnetic field.

For example, when a molecule crosses two spatially separated regions with microwave fields, the first microwave field creates a coherence between molecular levels and the second field ''reads'' the coherence. The initial phase of the written coherence depends on the frequency and amplitude of the field as well as on the interaction time. After the interaction, the molecule travels between the interaction zones acquiring phase shift determined by the frequency of the molecular transition and the time of the free flight. Interaction of the molecule with the second coherent microwave field, i.e., the reading process, leads to the modulation of the absorption of the field depending on the phase difference between the molecular coherence and the field. The dependence of the modulation on the field frequency is similar to an interferometric picture and is called ''Ramsey fringes.'' The fringes also can be seen if the frequency of the separated fields stays unchanged while the separation distance is varied.

Transformation of the Ramsey method to the optical domain is complicated because an atom gets different phase shifts depending on the place and direction of its passage through the optical fields that washes out the interferometric picture. To solve this problem, two-photon Doppler-free spectroscopy can be used. The separated field experiments with atomic beams based on the Zeeman coherence of ground-state atomic sublevels demonstrated narrow spectral features both in absorption [2] and polarization rotation [3]. Ramsey fringes have been observed using stimulated Raman transitions in sodium [4] and cesium [5] atomic beams. The separated field method based on two-photon spectroscopy may be modified for atomic vapor cells. Two-photon Ramsey fringes in spatially separated fields were reported [6].

Instead of separate laser beams, a single cw beam may be used if atoms are allowed to interact with it several times. This idea has been realized for uncoated atomic cells and pulsed lasers [7], and for the cells with antirelaxation coating [8] and a cw laser. In the first case, short time-delayed laser pulses are used to introduce Doppler-free two-photon transitions in the same group of atoms in a vapor cell. If the pulses are not phase correlated or delay between them exceeds the coherence lifetime, the Ramsey fringes disappear. In the second case, atoms, after the first interaction with the laser field, freely move inside the cell eventually colliding with the cell walls. Subsequent interactions with the laser beam takes place after some time passes. Since the created coherence can survive many collisions with the walls, it leads to the appearance of Ramsey features in the field spectrum. Observation of the two-photon Ramsey interference effect in an atomic vapor due to collisional velocity diffusion was also demonstrated [9].

Coated atomic cells and atomic cells with buffer gas have much in common in terms of spectroscopy. For example, atomic diffusion in the velocity space mediated by wall collisions for coated cells [10] and collisions with buffer gas atoms for buffer gas cells [11] leads to the suppression of nonlinear Faraday rotation for particular tuning of the probe laser.

The goal of this paper is to study an impact of atomic motion occurring in atomic cells without antirelaxation coatings on Doppler-free spectroscopy. We show that the ideas of the separated field method is relevant here. We demonstrate experimentally that atomic diffusion results in the Ramsey effect for atomic cells containing buffer gas in the same way as it appears for coated cells [8]. This implies that theoretical models used for vacuum cells do not give a correct physical picture for cells with buffer gas. The assumption that the presence of buffer gas leads to an increase of the transient time of active atoms through the laser beam and, therefore, the effective coherence lifetime can be increased relative to that for the transit time of atoms across the laser beam in the absence of buffer gas, is insufficient. This assumption is based on a simplified model that an atom that has left the interaction region never returns. To complete the picture, the atoms that interact with the laser beam several times should also be taken into account. We propose a theoretical model that includes both effects.

The principles of the two-photon separated field method are closely related to the physics of recent experiments on coherent information storage, where the information was mapped into and retrieved out of long-lived atomic coherence [12–14]. The basic idea of the information storage via

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atomic coherence can be understood in terms of light interaction with Λ-type atoms. Light pulses, interacting with both transitions of the Λ system, propagate in an atomic vapor and excite a spatial profile of a long-lived coherence between ground states of the atoms. This coherence profile stores information about these pulses after they have left or have been absorbed by the medium. A subsequently reading pulse produces Raman scattering off the atomic coherence and generates a retrieved pulse.

Ideally, in the case of immobile atoms, the retrieved pulse can be identical to the signal pulse [15]. However, the atomic motion changes the picture significantly. If the writing pulses that excite the atomic coherence are long enough and/or the delay between the writing and reading pulses is long enough, too, a part of the atoms with excited coherence may leave the interaction region and enter this region after some time, determined by the properties of the cell, e.g., wall coating or concentration of a buffer gas. During this free motion, atomic coherence evolves freely as in the original method of separated fields and, as a result, one can observe Ramsey fringes. This effect can deteriorate statistical properties of the retrieved pulse compared with the signal pulse.

In the first part of this paper, we discuss in detailed experiments, where diffusion of the atoms in buffer gas is used to provide the repeated interaction of the atoms with the laser field within a single interaction region [16]. We excite the Zeeman coherence of Rb atoms by a linearly polarized light pulse and read the stored information by a delayed circularly polarized pulse, detecting the linear combination of the signal resulting from the scattering of the reading light on the coherently pumped atoms and the transmitted reading field. In the presence of a static magnetic field, the intensity of the measured signal is modulated with frequency determined solely by the magnetic splitting of Zeeman sublevels. Because the modulation frequency does not depend on the light intensity and the decay of the modulation feature is very slow, we conclude that it results from the atoms that enter and exit the interaction region several times.

Repeated atomic interactions with the laser field is not the only effect observed in our experiments. We find a trace of the modulation for vacuum cells, too. It is well known that wall collisions in atomic cells without antirelaxation coating destroy the atomic coherence. Therefore, a coherently prepared atom that has left the laser beam return to the interaction region with its coherence erased. The atom-atom collisions are negligible in a vacuum cell. Therefore, we explain the Ramsey-like modulation obtained in an uncoated vacuum cell as a consequence of the coherent information storage in the atomic vapor [12–14].

In the case of coherent information storage, the modulation appears because the radiation retrieved from the atomic vapor by stimulated Raman scattering of the reading pulse on the coherent atoms that had not left the interaction region has a frequency different from the frequency of the reading field resulting from magnetic splitting between Zeeman sublevels [17]. The modulations appearing due to repeated atomic interaction with the reading laser and the modulations appearing due to stimulated Raman scattering on the atoms left in the interaction region can be distinguished by their decay time. In the first case, the decay time is determined by atomic diffusion in the buffer gas, while in the second case, it is determined by the group velocity of the retrieved pulse. We show this by simple theoretical calculations.

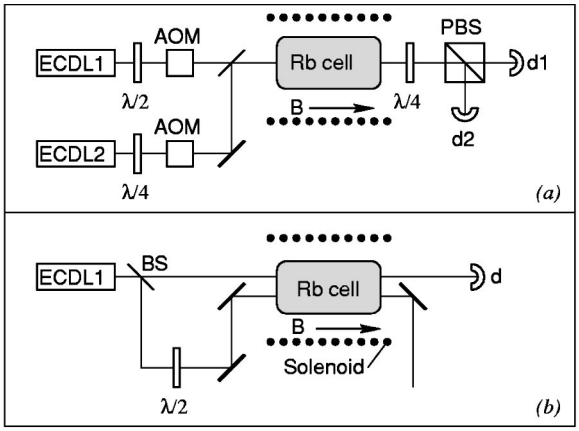

FIG. 1. Experimental setups used for observation of the Ramsey effect in (a) time (pulse regime) and (b) frequency (cw regime) domains.

We also present experimental data concerning Ramsey fringes in frequency domain, obtained for the vacuum atomic cells, as well as cells containing buffer gas. The experiment is performed for two spatially separated interaction zones.

In the second part of the paper, we propose a theoretical explanation of the experimentally observed effects and show that our results allow for a better understanding of a line narrowing mechanism for electromagnetically induced transparency (EIT) [19–21] in the presence of a buffer gas [18]. Moreover, our results may be used to connect concepts of coherent population trapping (CPT)/Hanle effect [19,22] and the phenomenon of alignment to orientation conversion [23] that both can be used to describe polarization rotation [24] in coherently prepared atomic vapors. Another potential application is the measurements of the decay time of atomic coherence that allows us to find the cross section of the velocity changing collisions in atomic cells with buffer gas [25].

II. EXPERIMENT

A. Experimental setup

Our experiment can be logically divided in two independent parts. In the first part of the experiment, we study the temporal evolution of light transmission through an atomic medium excited by the pulsed laser radiation and use two different lasers [see Figs. 1(a) and 2]. As the result of this experiment, we are able to observe temporal Ramsey fringes. In the second part, we study the change in the frequency spectrum of light transmitted through the atomic vapor, having two spatially separated interaction regions with the radiation, and use one continuous wave (cw) laser [see Fig. 1(b)

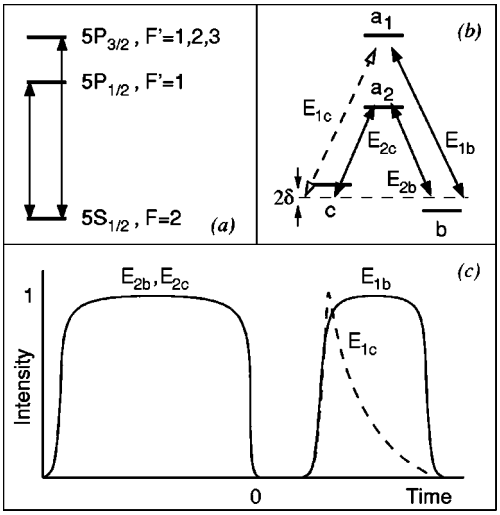

FIG. 2. (a) Level scheme of atomic ^{87}Rb used for studying the temporal Ramsey fringes [see Fig. 1(a)]. (b) Simplified level scheme of atomic ^{87}Rb. Levels $|a_1\rangle$ and $|a_2\rangle$ correspond to $P_{3/2}$, $F'=1,2,3$ and $P_{1/2}$, $F'=1$. Levels $|b\rangle$ and $|c\rangle$ correspond to Zeeman sublevels with $m=\pm 1$ of level $S_{1/2}$, $F=2$. (c) Temporal behavior of the fields. Writing pulses E_{2b} and E_{2c} generate low-frequency coherence ρ_{cb}. Reading pulse E_{1b}, delayed in time compared to the writing pulses, is scattered by the coherence that results in generation of the retrieved pulse E_{1c}. We assume that the origin of the time scale corresponds to the back edge of the writing pulses.

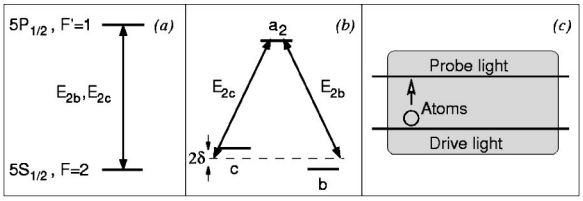

FIG. 3. (a) Level scheme of atomic ^{87}Rb used for studying the Ramsey fringes in the frequency domain (cw lasers) [see Fig. 1(b)]. Both lasers are tuned to the $D1$ line. (b) Simplified level scheme of atomic ^{87}Rb. Level $|a_2\rangle$ corresponds to $P_{1/2}$, $F'=1$. Levels $|b\rangle$ and $|c\rangle$ correspond to Zeeman sublevels with $m=\pm 1$ of level $S_{1/2}$, $F=2$. (c) Atomic cell with spatially separated weak probe and strong drive laser beams. The drive laser light excites the atomic coherence, after that, the atoms move toward the probe beam and eventually interact with it. This interaction influences the transmission of the probe field significantly.

and 3]. This experiment allows us to study the narrowing of the EIT resonance due to free atomic motion between the interaction regions.

The experimental setup for studying temporal response of the coherent medium is shown in Fig. 1(a). We perform the experiment in atomic ^{87}Rb vapor at 80°C, which corresponds to atomic density $N\sim 10^{12}$ cm^{-3}. We made our experiments with: (i) an atomic cell (the length 5 cm, the diameter 2.5 cm) containing isotopically pure ^{87}Rb and 3.0 Torr of Ne buffer gas; (ii) an atomic cell (the length 7.5 cm, the diameter 2.5 cm) containing isotopically pure ^{87}Rb and 0.3 Torr of Ne buffer gas; (iii) a vacuum atomic cell (the length 7.5 cm, the diameter 2.5 cm) containing isotopically pure ^{87}Rb. The cells are made of pyrex glass and have windows with small birefringence. Each cell is placed into a three layer μ-metal antimagnetic screen. A solenoid mounted inside the magnetic shield allows us to control the amplitude of the homogeneous magnetic field applied along the direction of the light propagation.

Two extended cavity diode lasers [ECDL1 and ECDL2 in Fig. 1(a)] are used. Acousto-optical modulators chop the radiation of each laser to produce light pulses. We excite Zeeman coherence [19] of ^{87}Rb atoms [the coherence between levels $|b\rangle$ and $|c\rangle$ in Fig. 2(b)] by two circular components E_{2b} and E_{2c} of a linearly polarized light pulse (writing pulse), and read the stored information by a delayed circularly polarized reading pulse E_{1b}, detecting the signal (retrieved pulse) E_{1c}, which results from the scattering of the reading light on the coherently prepared atoms. In order to study the phase dependence of the signal light, we measure the power of a linear combination of the reading and retrieved pulses. The writing laser ECDL1, tuned to the ^{87}Rb D1 line, has 1 ms pulse duration, whereas the reading laser ECDL2, interacting with ^{87}Rb D2 line [Fig. 2(a)], has pulse duration about 500 μs. The fall and rise times for the pulse are about 3 μs in both cases [Fig. 2(c)].

The Doppler broadening of rubidium vapor (the full width at the half maximum is equal to $\sim$540 MHz) overlaps all hyperfine structures of the $^2P_{3/2}$ state, so the reading light interacts with all three hyperfine components of the $^2P_{3/2}$ ($F'=1,2,3$) states. This feature is important for the estimation of the achieved efficiency of the reading process.

The most efficient retrieving takes place when the reading pulse is applied to the same atomic transition as the writing one, i.e., $F=2\rightarrow F'=1$. There is CPT for this transition. The retrieved pulse, resulting from stimulated Raman scattering of the reading pulse on the Zeeman coherence of the $F=2$ atomic sublevel, is not absorbed by this transition. In turn, there is no CPT for $F=2\rightarrow F'=2$ and $F=2\rightarrow F'=3$ transitions. Hence, some part of the retrieved light is absorbed due to the interaction with these transitions and the total scattering efficiency decreases.

The $\lambda/4$ wave plate and polarizing beam splitter placed after the vapor cell separate the reading σ^+ pulse and the retrieved σ^- light, which are detected by the fast photodiodes d_1 and d_2. By rotating the $\lambda/4$ plate, we are able to measure a beatnote between the reading and retrieved pulses. The peak powers of the reading and writing lasers are 350 and 290 μW, the beam sizes inside the cell are 1.5 mm. Under these conditions, the cell transmission is only about 10% for the incident linearly polarized laser radiation at 795 nm.

We apply a longitudinal magnetic field to the system that leads to antisymmetric shift of Zeeman sublevels. This shift is equal to $\delta=aB$, where B is the amplitude of the magnetic field and $a=2\pi\times 0.7$ MHz/G is the magneto-optic constant for ^{87}Rb.

In the second part of the experiment, we study the influence of the atomic motion on the transmission of a cw laser radiation through an atomic cell. To do this, we create the

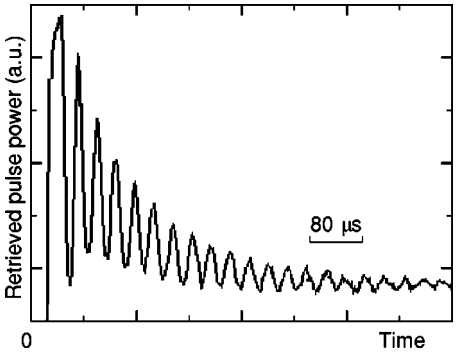

FIG. 4. Time dependence of the beat note power of the retrieved and reading pulses for the magnetic-field $B=23$ mG. The origin of the time scale corresponds to the back edge of the writing pulses. The front edges of reading and retrieved pulses coincide.

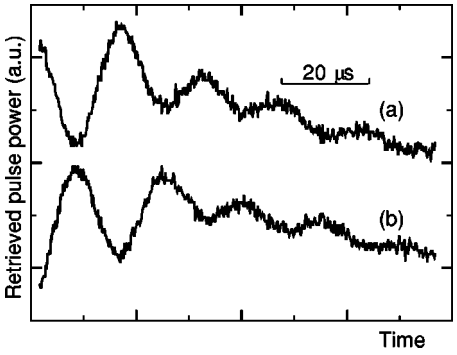

FIG. 6. Time dependence of the beat note power of the retrieved and reading pulses for various polarization directions of the writing fields. The 45° rotation of λ/2 plate [see Fig. 1(a)] leads to the 180° phase shift of the oscillations of the power of the retrieved radiation [compare curves (a) and (b)].

coherence between ground-state sublevels of ^{87}Rb vapor by cw linearly polarized light (unlike the previous case, where we use pulsed light). There are two spatially separated interaction regions in the cell [Figs. 1(b) and 3]. We measure the transmission of each separated in the space field as a function of the separation, static magnetic-field B, and mutual polarization of the fields.

For this experiment, we use only one external cavity diode laser ECDL1 tuned on the $F=2\rightarrow F'=1$ transition of ^{87}Rb D1 line. The laser beam, divided into two spatially separated beams, enters a cell. We call the stronger beam the drive, and the weaker beam the probe. A polarization direction of the driving beam is controlled by the λ/2 plate. We set the probe laser power at 0.1 mW and the beam diameter at 2.5 mm. The drive laser has a power of 0.2 mW and a beam diameter of 2.5 mm. The beam separation is 5 mm. The transmitted power of the probe beam is monitored by the photodiode *d*. The cell temperature is 80 °C.

B. Ramsey fringes in time domain

The typical time dependence of the beat note intensity of the retrieved and reading pulses is presented in Fig. 4. The retrieved signal appears simultaneously while switching on the circularly polarized reading laser ECDL2. To summarize the basic features of the signal, we present Figs. 5 and 6.

The frequency of the oscillations is determined solely by the magnitude of the magnetic-field B (Fig. 5). At zero magnetic field, no oscillations are observed. A general experimentally obtained relation between the period of the oscillations T and the magnetic-field B is $T\delta/\pi\approx 1$. The modulation depth of the oscillations depends on the rotation of the λ/4 plate. For the same experimental conditions except the magnetic field, the oscillation depth is smaller at higher frequencies of the oscillations (for larger magnetic field). The rotation of the writing pulse polarization using λ/2 plate changes the phase of the oscillations (Fig. 6).

The damping rate of the oscillations depends on the buffer gas pressure. For example, in a vacuum cell Fig. 7, for the same experimental conditions as described above, the oscillations are almost invisible and their damping rate equals to ~ 25 μs, while 3 Torr Ne cell reveals a ~ 800 μs damping rate.

A separation in space of the writing and reading pulses causes a shift of the maximum of the oscillation amplitude to later moments of time. The separation in 2 mm between the beams results in ~ 75 μs shift of the maximum (see Fig. 8).

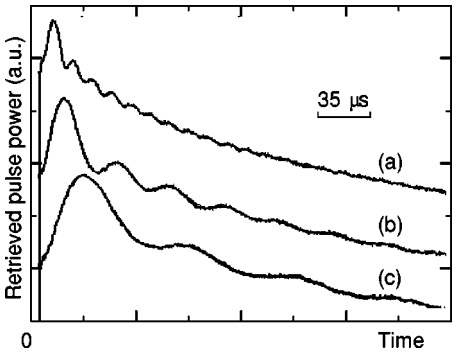

FIG. 5. Time dependence of the beat note power of the retrieved and reading pulses for various values of the magnetic field: (a) $B=56$ mG, the oscillation period is $T_a=12.6$ μs; (b) $B=21$ mG, the oscillation period is $T_b=34.8$ μs; (c) $B=10$ mG, the oscillation period is $T_c=68.8$ μs.

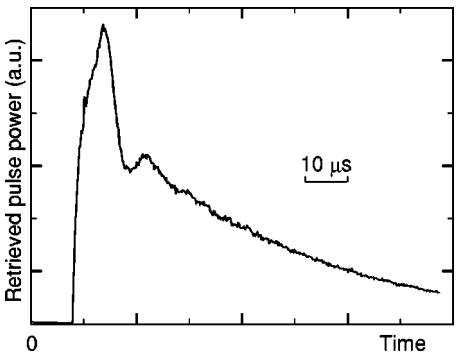

FIG. 7. Time dependence of the beat note power of the retrieved and reading pulses for the vacuum cell.

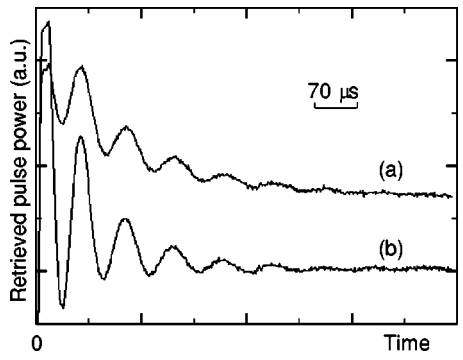

FIG. 8. Time dependence of the beat note power of the retrieved and reading pulses for spatially separated (2 mm) writing and reading beams [curve (a)], and overlapping writing and reading beams [curve (b)]. The maximum of the scattering efficiency shifts in time for the separated beams compared with the overlapping beams.

C. Ramsey fringes in frequency domain

Atomic motion significantly changes the frequency spectrum of the medium response. Using the setup shown in Fig. 1(b), we measure the transmission of the medium for a probe electromagnetic field as a function of magnetic-field B and study the behavior of the transmission spectra relative to the properties of the driving field, spatially separated from the probe field. The results of these measurements are shown in Figs. 9–11.

Because both the probe and the drive fields used in the experiment are strong enough, there are narrow transparency features in the vicinity of zero magnetic field for the probe field with no drive field and for the drive field with no probe field. These resonances result from EIT/CPT physics [19]. When both fields are present, one can see substantial narrowing for the transmission resonance (Fig. 9) along with total transmission increase (Fig. 11) compared with the case when only one field is present.

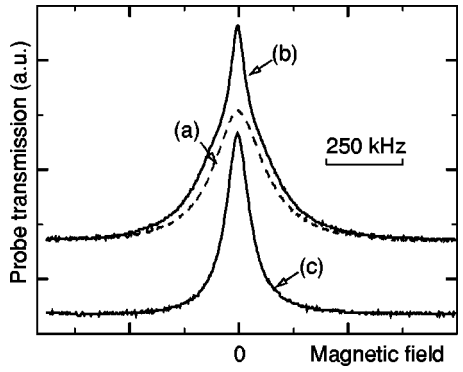

FIG. 9. Transmission of cw laser radiation resonant with the D1 line of ^{87}Rb vs magnetic field for the vacuum atomic cell and spatially separated laser beams. The magnetic field is shown in frequency units corresponding to the magnetic shift of Zeeman sublevels $2\delta = 2aB$, where B is the amplitude of the magnetic field and $a = 2\pi \times 0.7$ MHz/G is the magneto-optic constant for ^{87}Rb. The polarizations of the laser fields are the same. (a) Transmission of the probe field with no drive field. (b) Transmission of the probe field with drive field turned on. (c) Transmission of the drive field with no probe field.

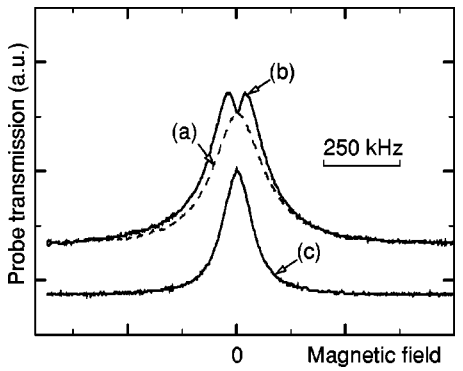

FIG. 10. The same as per Fig. 9, except the angle between the polarizations of the probe and drive fields is 90°.

Rotation of polarization of the drive field changes the transmission of the probe field and vice versa. For example, when the angle between the probe and drive polarizations is equal to 90°, the transmission resonance has a narrow absorption feature (see Figs. 10 and 11).

III. THEORY

The narrowest possible two-photon resonance in a coherent medium is determined by the relaxation time of the ground-state coherence. In experiments with vacuum atomic cells, this time may be modeled as the time of atomic passage through the laser beam. There are a number of methods to increase this time: expansion of the laser beam, usage of cells with antirelaxation coatings, or cells with buffer gas.

The spectroscopic properties of atomic vapor in the buffer gas atomic cells, however, may not be properly described by the equations valid for the vacuum cells. The reason for this complication is the atomic motion. In the presence of buffer gas, the interaction time of an atom with the laser beam is

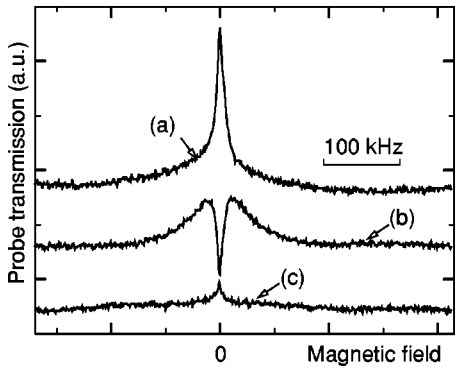

FIG. 11. Transmission of cw probe-laser radiation resonant with the D1 line of ^{87}Rb vs magnetic field for the atomic cell containing 0.3 Torr of Ne buffer gas. The magnetic field is shown in frequency units as per Fig. 9. The probe and drive beams are spatially separated (the separation is equal to 5 mm). (a) The polarizations of the probe and drive fields are the same. (b) The angle between the polarizations is equal to 90°. (c) The drive field is absent.

determined by the diffusion through the buffer gas. This is not the only effect. The coherence survives many collisions and some atoms leave the interaction region and return there after some time. These atoms should also be taken into consideration. We here propose a simple theoretical model that includes both the effect of increasing the coherence life time and the effect of multiple interaction of an atom and the laser beam.

Because an analytical solution of the exact problem including consideration of the Zeeman substructure of atomic levels is very complicated, we simplify it and substitute the exact scheme of atomic levels by an idealized three-level Λ scheme [Fig. 3(b)].

In the case of the experiments with pulsed lasers, the relevant simplified level scheme is four-level double-Λ scheme [Fig. 2(b)]. However, because the writing and reading procedures are separated in time, we may consider these processes independently, using a simple three-level Λ scheme. On the first stage, we create low-frequency coherence on the dipole forbidden transition $|b\rangle \rightarrow |c\rangle$. This process may be described using levels $|c\rangle$, $|b\rangle$, and $|a_2\rangle$. And, on the second stage, we read out the coherence. This process may be described using levels $|c\rangle$, $|b\rangle$, and $|a_1\rangle$.

A. Basic equations

Let us consider the interaction of two copropagating plane electromagnetic waves $E_{2b}(z,t)\exp[-i(\nu t-kz)]$ and $E_{2c}(z,t)\exp[-i(\nu t-kz)]$ with a gas of three-level Λ-type atoms shown in Fig. 3(b), where $E_{2b}(z,t)$ and $E_{2c}(z,t)$ are the slowly varying envelops, ν is the carrier frequency, and k is the wave number of the fields. Field $E_{2b}(z,t)$ interacts with the transition $|a_2\rangle \rightarrow |b\rangle$, while field $E_{2c}(z,t)$ interacts with the transition $|a\rangle \rightarrow |c\rangle$. We consider the case of the near resonant tuning, i.e., $\nu-\omega_{ac}=\omega_{ab}-\nu=\delta \ll \gamma$, where γ is the natural decay rate.

To describe the propagation of the light pulses through the atomic vapor, we use Maxwell-Bloch equations in the slowly varying amplitude and phase approximation. The Maxwell equations may be written as

$$\left(\frac{\partial}{\partial z}+\frac{\partial}{\partial ct}\right)\Omega_{2b}=i\eta\sum_j P_j(\vec{r},t)\rho_{2b}^j, \tag{1}$$

$$\left(\frac{\partial}{\partial z}+\frac{\partial}{\partial ct}\right)\Omega_{2c}=i\eta\sum_j P_j(\vec{r},t)\rho_{2c}^j, \tag{2}$$

where $\Omega_{2b}=\wp_{ab}E_{2b}/\hbar$ and $\Omega_{2c}=\wp_{2c}E_{2c}/\hbar$ are the Rabi frequency of the fields, $\wp_{2b}$ and $\wp_{2c}$ are the dipole moments, ρ_{2b}^j and ρ_{2c}^j are the matrix elements of the corresponding atomic transitions, c is the speed of the light in the vacuum, V is the volume of the interaction region, and $P_j(\vec{r},t)$ is the probability to find jth atom in the point with coordinates $\vec{r}$ at the moment of time t. The summation is over the atoms that are in the interaction region. The coupling constant is $\eta=\sqrt{2\pi\nu\wp^2/\hbar c^2 V}$. We assume that $\wp_{2c}\simeq\wp_{2b}=\wp$, and $\lambda_{2c}\simeq\lambda_{2b}=\lambda$.

The Bloch equations for the matrix elements of the populations and polarizations of the atoms are

$$\dot{\rho}_{cc}^j=2\gamma_{2c}\rho_{22}^j+i(\Omega_{2c}^*\rho_{2c}^j-\Omega_{2c}\rho_{c2}^j), \tag{3}$$

$$\dot{\rho}_{bb}^j=2\gamma_{2b}\rho_{22}^j+i(\Omega_{2b}^*\rho_{2b}^j-\Omega_{2b}\rho_{b2}^j), \tag{4}$$

$$\dot{\rho}_{2b}^j=-(\gamma_{2b}+\gamma_{2c})\rho_{2b}^j-i\Omega_{2b}(\rho_{22}^j-\rho_{bb}^j)+i\Omega_{2c}\rho_{cb}^j, \tag{5}$$

$$\dot{\rho}_{c2}^j=-(\gamma_{2b}+\gamma_{2c})\rho_{c2}^j+i\Omega_{2c}^*(\rho_{22}^j-\rho_{cc}^j)-i\Omega_{2b}^*\rho_{cb}^j, \tag{6}$$

$$\dot{\rho}_{cb}^j=-(\gamma_{cb}+2i\delta)\rho_{cb}^j-i\Omega_{2b}\rho_{c2}^j+i\Omega_{2c}^*\rho_{2b}^j, \tag{7}$$

where $\gamma_{ac}\simeq\gamma_{ab}=\gamma/2$. These equations are to be supplemented by the population conservation law

$$\rho_{22}^j+\rho_{bb}^j+\rho_{cc}^j=1. \tag{8}$$

Generally speaking, it is not enough to have Eqs. (1)–(8) to describe the Ramsey fringes observed in the experiment. The motion of the atoms should be taken into account. In the case of a cell containing Ne buffer gas, the probability distribution for the jth atom in the cell $P_j(\vec{r},t)$ may be described by the diffusion equation [27]

$$D\nabla^2 P_j(\vec{r},t)=KP_j(\vec{r},t)+\frac{\partial P_j(\vec{r},t)}{\partial t}, \tag{9}$$

where D is the diffusion coefficient, K is a quantity related with the coherence decay due to collisions between Rb and Ne atoms. The diffusion coefficient may be estimated as $D\simeq l_f v_T$, where l_f is the mean free path for a Rb atom in the cell, and v_T is the average velocity of the atom in the cell.

B. Solution

1. Temporal modulation

Let us describe the temporal modulation of the retrieved pulses. Similar to [23], we divide the interaction process by three independent stages: preparation of the atomic coherence, free evolution of the atomic coherence, and the reading process.

In the first stage, a part of the atoms is optically pumped in the dark state

$$\rho_{cb}^{0\,j}\simeq-\frac{\Omega_{2c}^*\Omega_{2b}}{|\Omega_{2b}|^2+|\Omega_{2c}|^2}, \tag{10}$$

as it follows from the steady-state solution of Eqs. (3)–(8) under the condition $|\Omega_{2b}|^2+|\Omega_{2c}|^2 \gg \gamma\gamma_{cb},\gamma\delta$. Increasing of the magnetic field leads to the increasing of the detuning δ and, eventually results in the decreasing of the amplitude of the coherence.

The distribution of these aligned atoms is defined by the solution of Eq. (9). If the duration of the writing pulses is long enough compared with the time of diffusion of an atom through the laser beam, the distribution will have a spatial

size larger than the beam diameter because some atoms left the interaction region during the coherence preparation.

In the second stage, the atoms continue to diffuse through the cell and more atoms leave the interaction region. Because of the magnetic field, the phase of the atomic coherence changes as $\rho_{cb}^{j}(t_j)=(\rho_{cb}^{0})^{j}\exp[-(\gamma_{cb}+2i\delta)t_j]$, where $(\rho_{cb}^{0})^{j}$ is the atomic coherence at the moment of start of the free evolution of the atom. Here, we take into account the coherence decay rate γ_{bc}, which appears due to atomic collisions.

In the third stage, the atoms interact with the reading field. To describe the reading procedure we rewrite equations (3)–(7) for the reading Ω_{1b} and retrieved Ω_{1c} pulses taking in consideration that the reading occurs on the other optical transition [see Fig. 2(b)], so we change the index ″2″ by ″1″ in set (3)–(7). We introduce collective variables instead of considering the evolution of each particular atom, and consider an *open* system with external injection to take into account the diffusion.

$$\dot{\rho}_{cc}=-\tilde{\gamma}_{bc}[\rho_{cc}-\mathcal{P}(t)\rho_{cc}^{0}]+\gamma\rho_{11}+i(\Omega_{1c}^{*}\rho_{1c}-\Omega_{1c}\rho_{c1}), \tag{11}$$

$$\dot{\rho}_{bb}=-\tilde{\gamma}_{bc}[\rho_{bb}-1+\mathcal{P}(t)\rho_{cc}^{0}]+\gamma\rho_{11}+i(\Omega_{1b}^{*}\rho_{1b}-\Omega_{1b}\rho_{b1}), \tag{12}$$

$$\dot{\rho}_{1c}=-\gamma\rho_{1c}-i\Omega_{1c}(\rho_{11}-\rho_{cc})+i\Omega_{1b}\rho_{bc}, \tag{13}$$

$$\dot{\rho}_{b1}=-\gamma\rho_{b1}+i\Omega_{1b}^{*}(\rho_{11}-\rho_{bb})-i\Omega_{1c}^{*}\rho_{bc}, \tag{14}$$

$$\dot{\rho}_{bc}=-(\gamma_{bc}+\tilde{\gamma}_{bc}-2i\delta)\rho_{bc}+\tilde{\gamma}_{bc}\mathcal{P}(t)\rho_{bc}^{0}e^{-(\gamma_{bc}-2i\delta)t}-i\Omega_{1c}\rho_{b1}+i\Omega_{1b}^{*}\rho_{1c}, \tag{15}$$

where $\tilde{\gamma}_{bc}$ is determined by the average time of atomic diffusion through the reading laser beam. If the atomic beam has radius r, then $\tilde{\gamma}_{bc}\approx D/r^{2}$.

For the sake of simplicity, we assume here that during all the time the population of the state $|b\rangle$ is about unity. This is possible if all atoms were optically pumped in this state and $\Omega_{1c}\gg\Omega_{1b}$ as well as $\Omega_{2c}\gg\Omega_{2b}$.

The diffusion of the oriented atoms into the interaction region is described by means of a source function $\mathcal{P}(t)\rho_{bc}^{0}e^{-(\gamma_{bc}-2i\delta)t}$, where $\rho_{cb}^{0}(z)$ is the initial atomic coherence, $\mathcal{P}(t)$ describes the atomic flow into the interaction region from outside, and $e^{2i\delta t}$ stands for the free evolution of the atomic coherence in the magnetic field.

The function $\mathcal{P}(t)$ can be found from Eq. (9). For the sake of simplicity, we find $\mathcal{P}(t)$ using simple reasoning. Let us assume that during the first two stages of the experiment, we have excited the coherence ρ_{cb}^{0} of all the atoms within a cylinder coaxial with the laser beam and having radius $R\gg r$. In the absence of the reading field, the decay of this coherence due to the atomic diffusion does not depend on r, and can be described as $\sim\rho_{cb}^{0}\exp[-(\tilde{\gamma}_{bc}tr^{2}/R^{2})]$. This expression is the solution of Eq. (15) if

$$\mathcal{P}(t)\simeq\frac{R^{2}-r^{2}}{R^{2}}e^{-r^{2}/R^{2}\tilde{\gamma}_{bc}t}. \tag{16}$$

Assuming $\rho_{bb}\simeq 1$, while $\rho_{11}=\rho_{22}=\rho_{cc}=0$, $|\Omega_{1b}|^{2}\gg\gamma_{bc}\gamma,\delta\gamma$, and

$$\rho_{1c}=\frac{i\Omega_{1c}}{\gamma}+\frac{i\Omega_{1b}}{\gamma}\rho_{bc}, \tag{17}$$

we derive equations

$$\left(\frac{\partial}{\partial z}+\frac{\partial}{\partial ct}\right)\Omega_{1c}=-\kappa[\Omega_{1c}+\Omega_{1b}\rho_{bc}], \tag{18}$$

$$\frac{\partial}{\partial t}\rho_{bc}=-\left[\gamma_{bc}+\tilde{\gamma}_{bc}+\frac{|\Omega_{1b}|^{2}}{\gamma}-2i\delta\right]\rho_{bc}-\frac{\Omega_{1b}^{*}}{\gamma}\Omega_{1c}+\mathcal{P}(t)\rho_{bc}^{0}e^{2i\delta t}, \tag{19}$$

where $\kappa=3N\lambda^{2}/8\pi$ is the coupling constant.

The approximate solution of Eqs. (18) and (19) may be presented in form

$$\Omega_{1c}(L,t)\simeq-\Omega_{1b}\rho_{bc}^{0}(L-v_g t)e^{-(\gamma_{bc}+\tilde{\gamma}_{bc}-2i\delta)t}-\kappa\frac{\gamma\mathcal{P}(t)}{\Omega_{1b}^{*}}e^{2i\delta t}\int_{0}^{L}\rho_{bc}^{0}(z)dz, \tag{20}$$

where $v_g=|\Omega_{1b}|^{2}/\kappa\gamma\ll c$ is the group velocity for the probe light pulse. The first term in the right-hand side (r.h.s.) of Eq. (20) is the radiation that appears due to stimulated Raman scattering of the reading light on the atoms that where in the channel of the reading beam Ω_{1b} when the leading edge of the pulse entered the medium. This is the retrieved radiation widely discussed in the literature concerning quantum information storage. As was shown in [15], this radiation may possess the same properties as the pulse Ω_{2c} under appropriate conditions. The second term in the r.h.s. of Eq. (22) results from the diffusion of the atoms in the atomic cell. This term decays slower in time than the first term. In the experiment, it leads to the long ″tail″ that appears on the retrieved pulse.

It is easy to see that $\Omega_{1c}(L,t)$ is modulated with the frequency corresponding to the ground level splitting due to the magnetic field, i.e., it contains the time-dependent term $\exp(2i\delta t)$. Reading fields, in turn, have no modulation. Therefore, if one measures the intensity of a linear combination of the reading and retrieved fields, one sees the oscillations of the power of the signal as the function of time. The frequency of these oscillations is equal to 2δ. The phase of the oscillation depends on the phase of the written coherence and the phase of the reading field.

To see these modulations in our experiment, we do not compensate the reading field exactly, so that the intensity on the photodetector is proportional to $|\xi\Omega_{1b}+\Omega_{1c}(L,t)|^{2}$, where $\xi\ll 1$ is the compensation rate. The best modulation appears when $\xi\Omega_{1b}\simeq\Omega_{1c}$, then the power is proportional to

$\cos^2(\delta t+\Phi)$, where phase Φ depends on the phases of all the fields involved in the experiment.

Separation of the reading and writing laser beams will lead to a shift of the fringes because initially, the atoms in the reading region are not polarized, so the first term in the r.h.s. of Eq. (20) is zero, and it takes some time for the atoms diffusing through the cell to reach the interaction region with the reading field. This result is similar to the results reported in [27].

2. Changing of the CW EIT resonance

Let us now turn to the problem of the narrowing of the transparency resonance of the linearly polarized probe field in the presence of a linearly polarized drive field spatially separated with the probe [Fig. 1(b)]. The propagation of the probe field through atomic vapor can be described by the steady-state solution of the Maxwell-Bloch equations. The Bloch equations may be written in the form

$$\dot{\rho}_{cc}=-\tilde{\gamma}_{bc}(\rho_{cc}-1/2)+\gamma\rho_{11}+i(\Omega_{1c}^{*}\rho_{1c}-\Omega_{1c}\rho_{c1}), \tag{21}$$

$$\dot{\rho}_{bb}=-\tilde{\gamma}_{bc}(\rho_{bb}-1/2)+\gamma\rho_{11}+i(\Omega_{1b}^{*}\rho_{1b}-\Omega_{1b}\rho_{b1}), \tag{22}$$

$$\dot{\rho}_{1c}=-\gamma\rho_{1c}-i\Omega_{1c}(\rho_{11}-\rho_{cc})+i\Omega_{1b}\rho_{bc}, \tag{23}$$

$$\dot{\rho}_{b1}=-\gamma\rho_{b1}+i\Omega_{1b}^{*}(\rho_{11}-\rho_{bb})-i\Omega_{1c}^{*}\rho_{bc}, \tag{24}$$

$$\dot{\rho}_{bc}=-(\tilde{\gamma}_{bc}-2i\delta)\rho_{bc}+\tilde{\gamma}_{bc}\tilde{\mathcal{P}}\rho_{bc}^{0}e^{2i\delta\tau}-i\Omega_{1c}\rho_{b1}+i\Omega_{1b}^{*}\rho_{1c}. \tag{25}$$

Unlike the set (11)–(15), $\tilde{\mathcal{P}}$ here is a constant that describes the probability of the interaction of the same atom with the drive and the probe fields. The time τ is the time of atomic diffusion between the driving and reading beams. The initial populations of the ground states for all the atoms are equal to each other, which is taken into account by introducing pump rate 1/2 in the equations for the populations. We neglect by the coherence decay due to the collisions with the buffer gas atoms and assume that the decoherence rate is determined by the diffusion time of atoms through the interaction area. Assuming that the population of the excited state $|a\rangle$ is always much less than the populations of each ground state, we write

$$\rho_{1c}=\frac{i\Omega_{1c}}{2\gamma}+\frac{i\Omega_{1b}}{\gamma}\rho_{bc}, \tag{26}$$

$$\rho_{b1}=-\frac{i\Omega_{1b}^{*}}{2\gamma}-\frac{i\Omega_{1c}^{*}}{\gamma}\rho_{bc}. \tag{27}$$

Substitution of Eqs. (26) and (27) into Eq. (25) yields

$$\rho_{bc}=-\frac{\Omega_{1b}^{*}\Omega_{1c}+\tilde{\gamma}_{bc}\gamma\tilde{\mathcal{P}}\rho_{bc}^{0}e^{2i\delta\tau}}{\gamma(\tilde{\gamma}_{bc}-2i\delta)+|\Omega_{1b}|^2+|\Omega_{1c}|^2}. \tag{28}$$

Substituting Eq. (28) into Eqs. (26) and (27), we find the polarizations ρ_{1c} and ρ_{1b} and write Maxwell equations

$$\frac{\partial\Omega_{1c}}{\partial z}=-\frac{\kappa}{2}\frac{\gamma(\tilde{\gamma}_{bc}-2i\delta)-|\Omega_{1b}|^2+|\Omega_{1c}|^2}{\gamma(\tilde{\gamma}_{bc}-2i\delta)+|\Omega_{1b}|^2+|\Omega_{1c}|^2}\Omega_{1c}+\frac{\kappa\tilde{\gamma}_{bc}\gamma\tilde{\mathcal{P}}\rho_{bc}^{0}e^{2i\delta\tau}}{\gamma(\tilde{\gamma}_{bc}-2i\delta)+|\Omega_{1b}|^2+|\Omega_{1c}|^2}\Omega_{1b}, \tag{29}$$

$$\frac{\partial\Omega_{1b}}{\partial z}=-\frac{\kappa}{2}\frac{\gamma(\tilde{\gamma}_{bc}+2i\delta)+|\Omega_{1b}|^2-|\Omega_{1c}|^2}{\gamma(\tilde{\gamma}_{bc}+2i\delta)+|\Omega_{1b}|^2+|\Omega_{1c}|^2}\Omega_{1b}+\frac{\kappa\tilde{\gamma}_{bc}\gamma\tilde{\mathcal{P}}\rho_{cb}^{0}e^{-2i\delta\tau}}{\gamma(\tilde{\gamma}_{bc}+2i\delta)+|\Omega_{1b}|^2+|\Omega_{1c}|^2}\Omega_{1c}. \tag{30}$$

In the approximation of a strong probe field $|\Omega_{1b}|^2+|\Omega_{1c}|^2=|\Omega|^2\gg\gamma\tilde{\gamma}_{bc},|\gamma\delta|$, we write the equation for the sum of the probe fields $|\Omega|^2$, which is proportional to the probe-field intensity measured in our experiment for zero magnetic field B,

$$\frac{\partial}{\partial z}|\Omega|^2=-\kappa\gamma\tilde{\gamma}_{bc}\left[1+\frac{2\tilde{\mathcal{P}}}{|\Omega|^2}(\Omega_{1c}^{*}\Omega_{1b}\rho_{bc}^{0}e^{2i\delta\tau}+\Omega_{1b}^{*}\Omega_{1c}\rho_{cb}^{0}e^{-2i\delta\tau})\right]. \tag{31}$$

This equation means that if the coherence ρ_{bc}^{0} coincides with the ''dark state'' coherence, generated by the probe fields Ω_{1c} and Ω_{1b}, i.e., $\rho_{bc}^{0}\simeq-\Omega_{1b}^{*}\Omega_{1c}/|\Omega|^2$, then the absorption of the probe field decreases. It may completely vanish if all the atoms that have interacted with the drive field also interact with the probe field, i.e., $\tilde{\mathcal{P}}=1$. Phase shift in the coherence ρ_{bc}^{0}, resulting from the phase shift of the drive field, inevitably leads to changes in absorption of the probe field, observed in our experiment.

The width of the absorptive feature is determined by the diffusion time between the interaction regions τ. Therefore, if the separation between the beams is large enough, the resonant feature is narrow, which corresponds to our observations.

IV. DISCUSSION

Therefore, we see the following explanation of our experiments. The signal appears due to coherent Raman-like scattering process. The linearly polarized writing pulse creates a coherence among Zeeman sublevels of $5S_{1/2},F=2$ level, that scatters the circularly polarized reading pulse into a pulse with the carrier frequency shifted on the value equal to the ground-state splitting 2δ and the opposite circular polarization. The phase of the retrieved field is determined by the phase of the atomic coherence, as well as the phase of the reading field. Because the frequency of the generated field is shifted, the beating between the retrieved and reading fields appears in the experiment [see Figs. 4–8]. This proves that our experiment is different from the four-wave mixing experiments [26], where all the fields interact with atoms simultaneously. In that case, the frequency of the generated

field coincides with the frequency of the reading field. The magnetic field leads to a decrease of the transformation efficiency only.

The long oscillating ''tail'' of the beat note of the retrieved and reading fields may be explained in terms of the separated field method and atomic diffusion in the buffer gas [27,28]. The atoms prepared by the writing pulses (the atoms in a coherent superposition of $|b\rangle$ and $|c\rangle$ states) leave the interaction region, then move chaotically in the cell bouncing off the buffer gas atoms elastically, and return into the interaction zone after some time delay. Each atom acquires a phase shift depending on the time of ''free flight.'' This phase shift changes the scattering efficiency of the reading field, similar to the Ramsey method of separated fields. As a result, the temporal intensity profile of the beat note of the pulses is modulated.

As follows from Eq. (20), the retrieved radiation contains two basic parts: the field that is generated in the channel of the reading laser beam just after the field switching on and the field that appears due to atoms arriving to the interaction region with some time delay. The atomic coherence decays very fast after switching on the reading pulse ~ 10 ns if the atoms do not leave the interaction zone. This results from the radiation broadening. During this short time, the first part of the retrieved field is generated [the first term in the r.h.s. of Eq. (20)]. This pulse leaves the atomic cell with the group velocity v_g, that is $\sim 10^5$ cm/s in our experiment. Therefore, this part of the signal has a duration less than $L/v_g \sim 100$ μs, while the duration of the observed beat note signal is about 1 ms. Hence, in our experiments, the damping rate of the coherence is defined by diffusion processes in the buffer gas. This explains the long ''tail'' of the retrieved pulse.

In a vacuum cell with uncoated walls, the atomic coherence decays due to atomic collisions with the walls. This leads to washing out the scattering effect observed in the cell with buffer gas. The residual beating (7) may appear either due to the initial retrieved radiation, not connected with the diffusion process (even in the vacuum cell, the group velocity of light is less than 10^6 cm/s), or due to incomplete coherence destruction by the wall collisions.

The modulation of the signals appears only for nonzero magnetic fields because degenerate Zeeman coherence has zero frequency and does not evolve in time. The magnetic field removes this degeneracy and leads to the light modulation. In the spirit of separated field experiments, the modulation frequency is independent of the intensity of the reading field, but it does depend on the eigenfrequency of the spin coherence determined by the Zeeman splitting of atomic sublevels (Fig. 5).

Because our scheme is in the Doppler-free configuration, it is insensitive to the atomic velocity. The main reason of the Ramsey interference effect in our experiment is the atomic diffusion in the real space, not the velocity diffusion of atomic coherence within an optical Doppler distribution, originated from velocity-changing collisions with the buffer gas [9].

The phase sensitivity of the oscillations of the intensity of the retrieved pulse to the writing light polarization also confirms the concept of atomic diffusion used in our explanation. The atomic coherence depends on the product of the amplitudes of the writing fields $E_{2b}^* E_{2c}$. A rotation of the $\lambda/2$ plate by angle ϕ leads to the polarization rotation of the writing fields by the angle 2ϕ. In other words, the polarization of the writing pulse changes the phases of the circular components $E_{2b} \rightarrow E_{2b} \exp(2i\phi)$ and $E_{2c} \rightarrow E_{2c} \exp(-2i\phi)$. This changes the phase of the written coherence $E_{2b}^* E_{2c} \rightarrow E_{2b}^* E_{2c} \exp(-4i\phi)$ and, therefore, changes the scattering efficiency. This is exactly what we see in Fig. 6

The shift of the maximum of the oscillation amplitude in the case of spatially separated writing and reading beams is related to the time of diffusion of the coherent atoms between writing and reading zones. The mean free path of Rb atoms in 3 Torr of Ne is ~ 0.01 cm [28], so it takes ~ 400 collisions with the buffer gas atoms to pass 2 mm distance. This corresponds to ~ 100 μs time delay, which is in a good agreement with the experimental result.

To see how the atomic motion changes the properties of the light transmission, we perform the second part of our experiment, where the transmission of the probe field depends on the spatially separated drive field. We use uncoated cells to be sure that atoms that have collided with the wall are nearly incoherent. Only the atoms that interact with the driving field and after that enter the probe field are able to change probe transmission.

The experimental results (Figs. 9 and 10) are in good agreement with our theoretical predictions. Namely, the presence of the driving field increases transmission of the probe field and makes the transmission line narrower. It is worth to mention here, that if the drive and the probe fields are not spatially separated, the transmission increases too, but the resonance becomes broader.

Change of the phase of the prepared coherence by rotation of the $\lambda/2$ plate leads us to change the sign of the narrow feature in the transmission. We have performed the same experiment with a cell containing buffer gas and found similar behavior of the transmission (Fig. 11).

It is interesting that this spatial coherence transfer mechanism may explain some important features of the experiments on the nonlinear magneto-optic rotation [24]. Usually the presence of a buffer gas in an atomic cell results in narrow features in the transmission and polarization rotation in the vicinity of a zero magnetic field (substantial narrowing for the transmission resonance, and the unusual kinklike shape in the rotation). We explain these features as consequences of the atomic diffusion. In a sense of the method of separated fields, the diffusing atoms make the resonance feature narrower similar to the effect of line narrowing that occurs in the atomic cells with anti-relaxation coating [8].

V. CONCLUSION

We have shown that diffusion of Rb atoms in a cell with buffer gas changes the atomic response on resonant excitation by laser light. These changes may be qualitatively described as a result of repeated, and separated in time, inter

action of the light with the same atoms. In between these interactions, the atoms freely diffuse through the cell as in the method of separated oscillatory fields. This leads to a narrowing of the resonant features. The effect may find applications in the study of atomic diffusion in the buffer gas, as well as in high-precision metrology, e.g., frequency standards and magnetometry.

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