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SUB-DOPPLER LASER SPECTROSCOPY IN THIN GAS CELLS

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Sub-Doppler Laser Spectroscopy in thin gas cells

(Review)

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1. Introduction

Usually the laser beam diameter D is much less than the length ℓ of a gas cell (Fig. 1a) at spectroscopic investigations [1]. Therefore the relaxation of particles (atoms, molecules) resulting from the finite time of their flight along a cell is ignored as a rule. However such relaxation can lead to qualitative new results in rarefied gases if $D \gg \ell$ (Fig. 1b). Indeed , for a Doppler-broadened spectral line of a resonant transition with the central frequency ω_0 , a variation of a travelling monochromatic wave with a frequency ω in a gas medium results mainly from its interaction with a group of particles, whose velocity projection v (on the wave vector $\vec{K}$) is close to $(\omega - \omega_0)/K$ [1]. At the same time, the relaxation rate $|v|/\ell$ of particles, connected with their flight between walls of the thin gas cell, also is determined by the value v . Therefore appearance of Sub-Doppler absorption (dispersion) resonances is possible for the running monochromatic wave at a scan of its frequency detuning $\delta = (\omega - \omega_0)$ near the value $\delta = 0$ because of dependences of a transient establishment of populations and coherences of quantum levels of the resonance transition on the velocity projection v . Two types of such Sub-Doppler resonances have been discovered and analyzed in the optical spectral region [2-8].

Thus long ago the Dicke effect was discovered in the microwave region at wave lengths $\lambda \gtrsim \ell$, when the narrowing of Doppler-broadened spectral lines takes place [9].

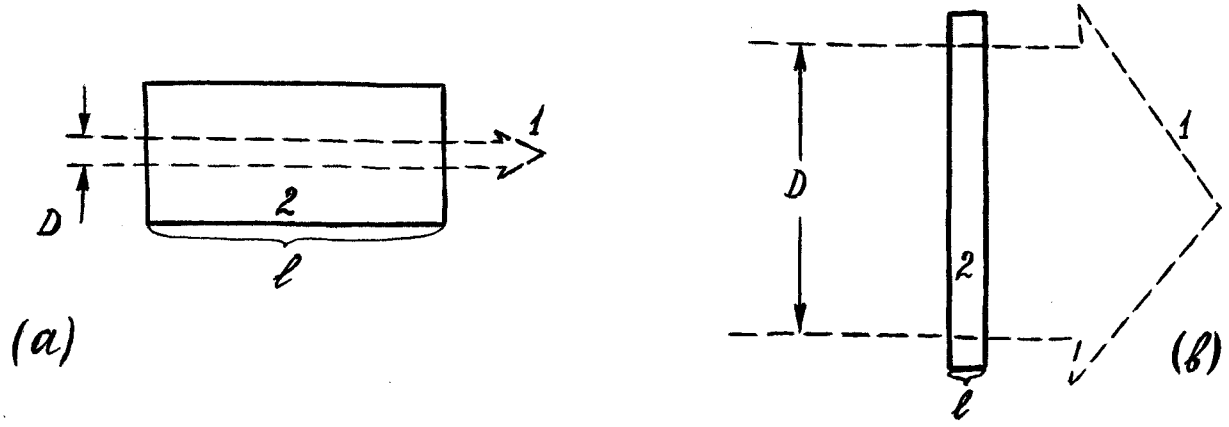

Fig.1. Spreading of the laser beam {1} with the diameter *D* through the gas cell {2} with the length *l* in cases *l*>>*D* *(a)* and *D*>>*l* *(b)*.

Recently manifestations of the Dicke effect were theoretically investigated by Nienhuis and Zambon for optical wave lengths $\lambda \leq 1\,\mu m$ [5,6]. In consequence, linear optical sub-Doppler absorption resonances were analyzed, which are caused by the build-up of the transient dipole polarization in ultra-thin gas cells. Then manifestations of given resonances were registered also at experiments [7,8].

The new types of sub-Doppler absorption and dispersion resonances were predicted by Izmailov [2,3]. Unlike Dicke effect, such resonances may arise even in comparatively lengthy cells (when $\ell \gg \lambda$) because of establishment processes of a population of the long-lived (ground or metastable) quantum level at the optical pumping. Later given resonances were registered and analyzed at experiments by Briaudeau, Bloch and Ducloy [4,8].

Sub-Doppler optical resonances in thin gas cells under consideration may be used in the ultrahigh resolution spectroscopy. Compact devices for frequency standards may be created on their basis. Moreover a motivation for thin gas cell studies is related with the analysis of atom-surface interaction, as well as the analysis of desorption and scattering processes [10].

2. Summary of theoretical results

To emphasize the specific effects of a thin gas cell, the theoretical descriptions for thin cell spectroscopy generally assume that interatomic collisions are negligible relative to atom-surface collisions. On the other hand, surface collisions are assumed to be strong enough to destroy all of the optical excitation. As a key to understanding the novel predicted sub-Doppler resonances one has to analyze the transient interaction of atoms with the resonant light. The transient regime combines a double velocity dependence: on one hand, the atomic resonance is Doppler-shifted according to the atomic velocity projection (along the light beam propagation axis); on the other hand, the transient effects depend on the atomic flight duration from wall to wall, i.e. on the atomic velocity along the normal to the cell. For these reasons, the expected effects are maximum under normal incidence (in the following, one assumes such a situation), while a residual Doppler broadening should be expected for other incidences.

Let us consider the steady state propagation of the following plane monochromatic light wave perpendicularly to walls of the thin gas cell:

$$\vec{\mathcal{E}}(z,t) = \vec{e}\sqrt{I}\exp[i(\omega t - \kappa z)] + c.c., \qquad (1)$$

where I and $\vec{e}$ are the intensity and the unit polarization vector of the wave; ω is the frequency, $\kappa = \omega/c$ is the wave number. It is assumed that the wave frequency ω is close to

the central frequency ω_0 of the electric dipole transition $a \rightleftarrows b$ between the long-lived (ground or metastable) non-degenerate lower state a and the excited level b of atoms (molecules). The sufficiently rarefied gas medium in the cell is considered where collisions between particles are negligible. The interaction of the wave (1) with particles of the gas was analyzed on the basis of equations for density matrix elements [11] with corresponding boundary conditions on walls of the plane cell for optical coherence and populations of levels a and b [2-8]. Then the following expression may be received for the absorption coefficient α of the wave (1) in the gas cell:

$$\alpha(\delta, I, \ell) = 4\pi\omega |(\vec{e}\vec{d})|^2 N \hbar^{-1} c^{-1} \int_{-\infty}^{+\infty} F(v)\, Q(\delta, I, v, \ell)\, dv, \qquad (2)$$

where $\delta = (\omega - \omega_0)$ is the frequency detuning, $\vec{d}$ is the matrix element of the dipole moment for the resonance transition $a \rightleftarrows b$, N is the density of particles in the gas cell, $F(v) = \pi^{-\frac{1}{2}} u^{-1} \exp(-v^2 u^{-2})$ is the Maxwell distribution of particles on the velocity projection v with their most probable velocity u. At the linear optical regime in the thin cell, the function Q in Eq.(2) has the form [5,6]:

$$Q = \frac{\gamma}{\gamma^2 + \Delta^2} + \frac{|v|}{\ell} \operatorname{Re}\left\{ \frac{\exp[-(\gamma - i\Delta)\ell / |v|^{-1}] - 1}{(\gamma - i\Delta)^2} \right\}, \qquad (3)$$

where $\Delta = (\delta - kv)$, γ is the homogeneous half-width of the spectral line of the transition $a \rightleftarrows b$. The first term in Eq.(3) don't depend on the cell length ℓ and corresponds to steady-state Doppler-broadened absorption in the coefficient α (2) expected in a usual cell [1]. At the same time the second term in Eq.(3) includes the interferometric dependence upon the cell length ℓ, which is connected with the build-up of the transient dipole polarization of particles in the cell and its sudden de-excitation on walls of the cell. Exactly this term describes manifestations of the Dicke effect [9] for optical wave lengths $\lambda \geq \ell$. Fig.2 presents dependences of the linear absorption coefficient α (2) with the function Q (3) on the frequency detuning δ at different lengths ℓ of the gas cell in case of the large Doppler broadening $ku \gg \gamma$ of the spectral line. The value α in Figs.2 and 3 is expressed in fractions of the linear absorption coefficient α_0 of the wave for the usual (macroscopic) cell at the zero frequency detuning $\delta = 0$. One can see that the dependence $\alpha(\delta)$ on Fig.2 has the sub-Doppler peak near the detuning $\delta = 0$ at the sufficiently small cell length ℓ. Indeed the wave absorption sharply decreases for particles, whose transit time $\tau = \ell / |v|^{-1}$ between walls of the cell is less than the characteristic time γ^{-1} of establishment of the optical coherence for the transition $a \rightleftarrows b$. According to the Doppler effect, frequency detunings $|\delta| > \gamma(k\ell)$ correspond to such velocity projections v. Therefore sub-Doppler peaks under consideration may arise at

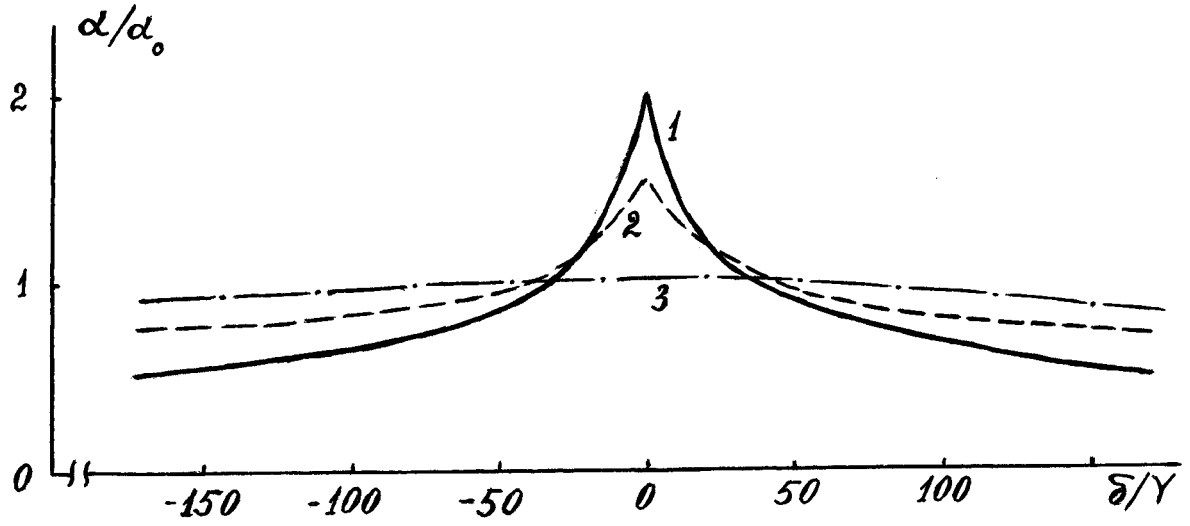

Fig.2. Theoretical dependences of the absorption coefficient α of the running wave on the frequency detuning δ at the linear optical regime in the thin gas cell, when $\gamma = 0.002\ ku$, $kl = 1$ (1), 10 (2), and 100 (3).

the condition $\gamma(kl) < ku$ that is at the cell length $l < u\gamma^{-1}$. We can directly see the decrease of the contrast and broadening of sub-Doppler absorption resonances (near the value $\delta = 0$) at the growth of the cell length l (Fig.2). Such narrow resonances were theoretically investigated in papers [5,6] and their manifestations were registered at experiments [7,8].

At the same time, appearance of nontrivial sub-Doppler absorption and dispersion resonances is possible even at the cell length $l \gg u\gamma^{-1}$ due to the optical pumping effects [2-4,8]. Indeed let us consider the noncycling optical transition $a \rightleftharpoons b$, when the excited level b may radiatively decay not only on the lower state a but also on other long-lived states, which don't interact with the wave (1). In this case a light induced repumping of atoms from the state a may be essential [12]. Such repumping is selective on the projection v of the atomic velocity and increases with a growth of the transit time $\tau = l/|v|^{-1}$ of atoms between cell walls [13]. At the approach of the frequency detuning δ to zero, the wave (1) effectively interacts with atoms having lesser velocity projections $|v|$ and hence characterized by greater times τ of the transit relaxation. In this connection we may expect a resonance weakening of the wave absorption in a sub-Doppler neighborhood of the quantity $\delta = 0$ because of the decrease of the population of the state a . Atoms (molecules) have time to transit macroscopic distances during a sufficiently large relaxation time of pumped long-lived levels [12,13]. Therefore, unlike the Dicke effect, such sub-Doppler resonances of the optical pumping may arise also at the cell length $l \gg u\gamma^{-1}$ [2-4,8]. Really, at the small saturation parameter $I|(\vec{e}\vec{d})|^2\hbar^{-2}\gamma^{-2} \ll 1$ and cell length $l \gg u\gamma^{-1}$, it is possible to receive the expression for the absorption coefficient α (2) with the following function $Q(\delta, I, v, l)$ [2,3]:

$$Q = \frac{k|v|}{S\gamma^2}\left\{1 - \exp\left[-\frac{S}{k|v|}\frac{\gamma^3}{[\gamma^2 + (\delta - kv)^2]}\right]\right\}, \qquad (4)$$

where

$$S=2(1-B_{\ell a})(k\ell)I/(\vec{e}\vec{d})^2\hbar^{-2}\gamma^{-2}, \qquad (5)$$

$B_{\ell a}<1$ is the probability of the radiative decay from the level ℓ to the state a ; dimensionless parameter S (5) describes the optical pumping rate. It is not difficult to show from Eq.(4), that at the zero frequency detuning $\delta=0$ the essential decrease of the population of the long-lived state a occurs for atoms with velocity projections $|v|\leq S\gamma k^{-1}$. For example, in the case of spectroscopic characteristics of transitions $^2S_{1/2}-{}^2P_{1/2}$ and $^2S_{1/2}-{}^2P_{3/2}$ of alkali atoms [14] for the cell length $\ell\sim 1\ mm$ the parameter $S\sim 1$ (5) if the wave intensity $I\sim 10^{-6}\div 10^{-5}\ W/cm^2$. According to Fig.3, the dependence of the absorption coefficient α (2), (4) on the frequency detuning δ has the resonance dip near the point $\delta=0$ on the Doppler-broadened background. At the small parameter $S\leq 1$ (5), the width of this narrow resonance is determined by the homogeneous width 2γ of the spectral line (Fig.3). The amplitude and width of given resonance increase at rise of the wave intensity, because of the intensification of the repumping of atoms from the state a . We can obtain the following asymptotic expression for the absorption coefficient α (2), (4) in the limit of the small parameter $S\to 0$ (5) [2,3]:

$$\frac{\alpha}{\alpha_0}=\exp\left[-\left(\frac{\delta}{ku}\right)^2\right]+\frac{S}{\pi}\left(\frac{\gamma^2}{\gamma^2+\delta^2}\right)^2 ln\left[\left(\frac{\gamma S}{ku}\right)\frac{\gamma^2}{(\gamma^2+\delta^2)}\right]. \qquad (6)$$

The structure of the resonance, described in formula (6) by the second term, essentially differs from known earlier Doppler-free absorption resonances [1].

It is obvious that sub-Doppler resonances of the optical pumping under consideration may arise not only in the absorption of the wave (1) but also in the fluorescence of the gas medium.

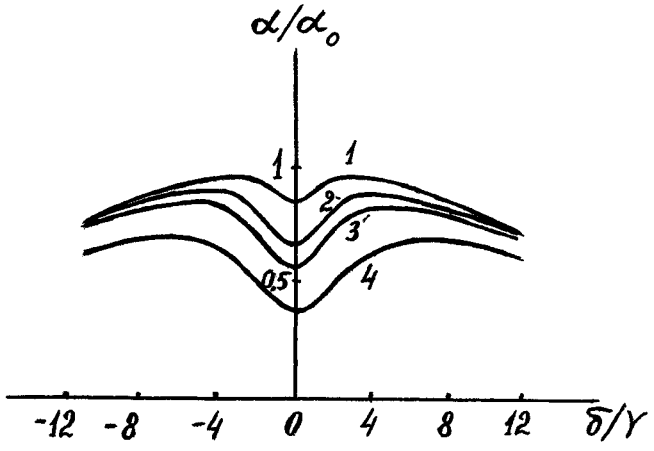

Fig.3. Absorption coefficient α of the running wave as the function of the frequency detuning δ at the optical pumping regime in case of $\gamma=0.05\ ku$ for $s=0.2$ (1), 1 (2), 2 (3), , and 6 (4).

Moreover under the analogous conditions in the thin gas cell, sub-Doppler dispersion resonances may arise owing to the light induced redistribution of populations and coherences of the Zeeman sublevels of the degenerate ground (or metastable) level of particles with small velocity projections v. Such nontrivial resonances in polarization characteristics of the running monochromatic wave were predicted and theoretically investigated in paper [3] for angular momenta ½ of levels of the optical transition. Then corresponding sub-Doppler polarization resonances were detected also at experiments with thin gas cells [8].

Detailed experimental investigations of sub-Doppler absorption and polarization resonances in thin gas cells under consideration were carried out in the Laboratory of "Laser Physics" in the Galilee Institute of the University Paris-Nord [4,7,8].

3. Description of experimental apparatus

Corresponding experiments consist of performing the transmission spectroscopy of a single laser beam through a thin vapor cell, irradiated under normal incidence [4,7,8]. The setup includes a tunable narrow-linewidth laser diode (optically locked to an off-axis confocal FP [15]) which can be frequency-modulated (FM), a thin cell of Cs vapor, heated in oven and (most often) located in a magnetic shield, and a sensitive photodetector (generally an avalanche photo-diode). At the output of the diode laser and following an optical isolator, several optical elements are used for the attenuation of the incident irradiation and the control of the laser beam diameter and polarization, as well as for the attenuation of the overall intensity reaching the photodetector. In addition, a beam splitter permits to conduct simultaneously auxiliary experiments, namely Cs saturated absorption (SA) (with an applied FM) in a "macroscopic" cell (i.e., length~2 *cm* largely exceeding the light beam diameter). The experiments [4,7,8] were performed with commercial quartz cell, filled up with Cs and sealed after a prior outgassing. Different cells were used, of nominal lengths 10, 20, 50, and 100 μm, and 1 *mm*.

Methods of the frequency modulated (FM) spectroscopy also were used at experiments [4,7,8]. Then the frequency modulation applied to the light source generates an induced variation on the signal beam when it passes through a frequency discriminator. The obtained line shape is hence the frequency derivative of the nonmodulated (direct) signal. This is true as long as (i) the FM amplitude is weak enough, i.e. much smaller than the desired resolution, and (ii) the frequency of the modulation is slow relative to the time constants of the physical mechanism involved in the signal. Such a frequency derivative technique is particularly appealing for discriminating a narrow signal from a broader background, as the overall line-shape contrast is enhanced.

Experimental investigations [4,7,8] were carried out on the $6S_{1/2}-6P_{3/2}\ D_2$ resonance line of Cs [14], whose energy diagram is shown on Fig.4.

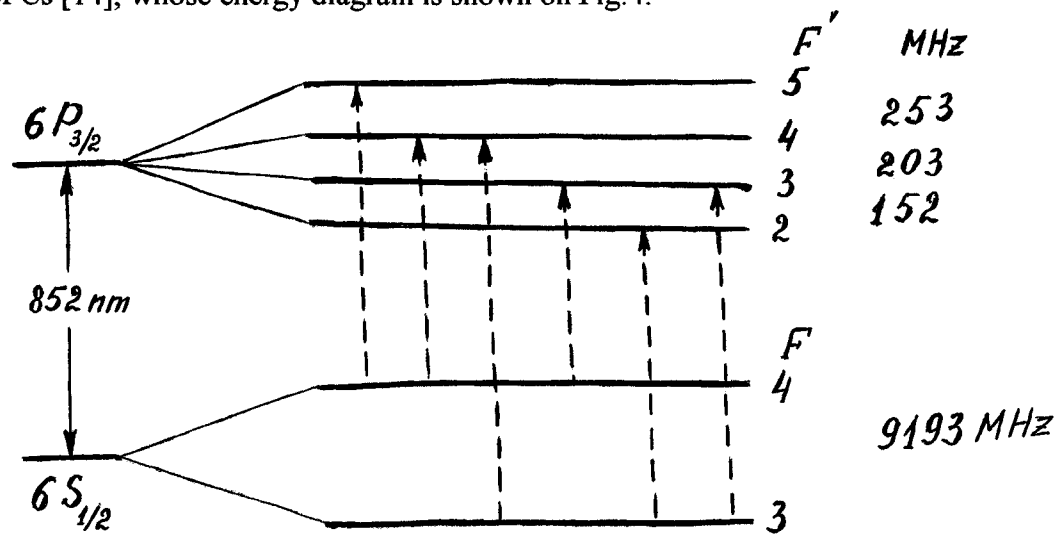

Fig.4. Energy levels diagram of the cesium *D2* line showing the allowed electric dipole transitions between hyperfine sublevels.

4. *Single beam transmission experimental results*

Fig.5a shows a typical transmission spectrum through an ℓ=10 μm cell in the absence of FM [4,8]. Narrow peaks are applied on the background associated with the Doppler-broadened absorption. As shown by comparison with a reference saturated absorption (Fig.5b), these structures showing a reduced absorption are centered onto the hyperfine components of $6S_{1/2}(F=3)-6P_{3/2}\ D_2$ resonance line (Fig.4). Sub-Doppler resonances on Fig.5a are caused by the optical pumping. Therefore at the decrease of the radiation intensity, these

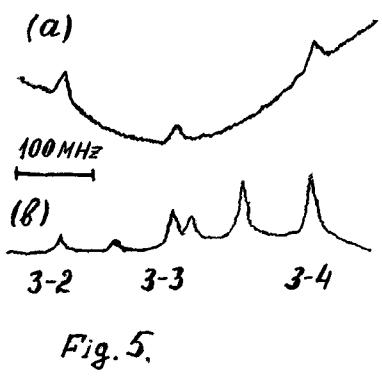

Fig. 5.

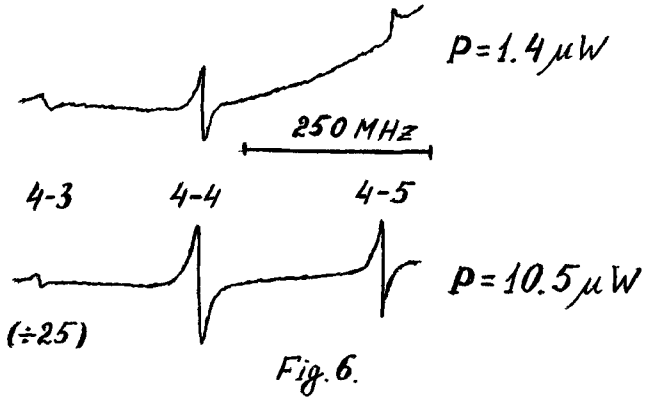

Fig. 6.

Fig.5.- a) Transmission spectrum through a *10μm* Cs cell, on the $6S_{1/2}(F=3)-6P_{3/2}\ D_2$ resonance line. Incident power *6 μW*, beam diameter: *4 mm*, cell temperature: *90°C*. b) Reference saturated absorption spectrum in an auxiliarly cell.

Fig.6.-FM transmission spectra at *852 nm* in a *10μm* Cs cell on the $6S_{1/2}(F=4)-6P_{3/2}\ D_2$ resonance line for various incident powers **P** (beam diameter *4mm*, σ polarization, FM: *7 kHz*).

resonances tend to vanish relative to the Doppler broadened background, which correspond to the linear absorption.

Through the direct detection of the transmission signal, the sub-Doppler features mentioned above appear superimposed on a much larger Doppler background, and analyzing the details of the line shapes of these narrow structures would turn out to be a difficult task. In the FM version of the above experiments the enhanced contrast authorizes both a quantitative measurement of the sub-Doppler structure and the observation of narrow structures even at very low radiation intensities. Thus Fig.6 presents the FM transmission spectra, which yields the first derivative of the corresponding transmission profile [4,8].We can see, that unlike Fig.5a, a series of high-contrast sub-Doppler resonances (having a dispersion form) appear on the comparatively low Doppler-broadened background (Fig.6).

In the range of extremely low incident intensities ($\sim 10^{-9} \div 10^{-6}$ W/cm^2) the linear optical regime in the thin gas cell is effectively reached [7,8]. Then directly narrow absorption resonances appear on the Doppler broadened background in the FM transmission spectrum, because of the Dicke effect (Fig.7). One can see good agreement between experimental and calculated dependences on Fig.7. Experimental data on Fig.7 were obtained on the shortest cell available ($\ell \sim 10$ μm). At the cell length $\ell \sim 100$ $\mu m \gg \lambda$ Dicke effect was not manifest for D_2 Cs line and sub-Doppler structures did not appear even in FM transmission spectra at the linear optical regime.

However at the higher range of wave intensities, sub-Doppler resonances induced by the optical pumping (Figs.5a, 6) have been observed with the whole set of different cells, including a 1 *mm* thick cell [4,8]. Indeed, characteristic half-width $\Gamma \geq \gamma$ of sub-Doppler optical pumping resonances in thin gas cell under consideration for a real laser beam with the diameter D is

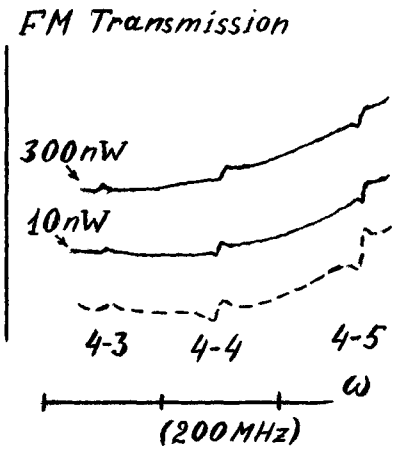

Fig.7. FM absorption spectrum *(F=4 – F'={3,4,5})* at the linear optical regime as obtained for various low incident power on a *20μm* cell. Beam diameter is *4 mm*; the dashed line corresponds to theoretical calculations. Horizontal frequency axis is *200 MHz* per division.

restricted by the condition:

$$\Gamma \geq \frac{K\ell}{T}, \qquad (7)$$

when the time T of atoms transit across the beam is more than the time $\ell/|v|^{-1}$ of atoms transit along the wave vector with the velocity projection $|v| \sim \Gamma/K$. By substitution of the characteristic transit time $T \sim D/u$ of atoms with the most probable velocity u, we receive from (7) the following condition of application of the theoretical model of the plane light wave up to the half-width $\Gamma \simeq \gamma$:

$$\frac{\gamma}{Ku} > \frac{\ell}{D}. \qquad (8)$$

Indeed, at such small cell length ℓ (8), the structure of registered at experiments [4,8] optical pumping absorption resonances is well described by theoretical relationships of papers [2,3]. However at a breach of the condition (8) it is necessary to carry out theoretical calculations for the real spatially restricted laser beam.

5. Sub-Doppler polarization spectroscopy with a running laser beam

As an alternate to the applied frequency modulation, which singles out those atoms slow enough to be optically pumped, it was established theoretically [3], that polarization spectroscopy of a single beam in a thin cell can exhibit a response specific to the atoms flying nearly parallel to the surface. More generally, for an elliptically polarized beam, the axes of the elliptical polarization are the natural quantization axes of the atomic system (e.g. π and σ). When the beam is saturating, a population redistribution takes place between the sublevels, so that the two principal polarizations finally exhibit different coupling strength with the atomic system [1,12,13]. This leads to a differential absorption and phase shift according to the polarization, i.e., to dichroism and birfringence, that can be observed in polarization spectroscopy, notably through the resonant transmission across a normally (nearly) blocking polarization analyzer. In a macroscopic cell, such a single beam propagation effect is expected to be Doppler-broadened [1]. Alternatively, in a thin cell, these effects related to optical pumping and saturation are efficient only for those atoms with a long time of flight, and polarization spectroscopy of a single beam should yield sub-Doppler resonances.

The novel type of the single beam polarization spectroscopy in thin gas cells has been demonstrated on a 10 μm cell with incident elliptical polarizations [8]. Doppler-free resonances are directly observed on the light transmitted through a cross-polarizer, without the help of any FM modulation techniques (Fig.8). Because of unperfect extinction ($\sim 2*10^{-4}$), one still observes

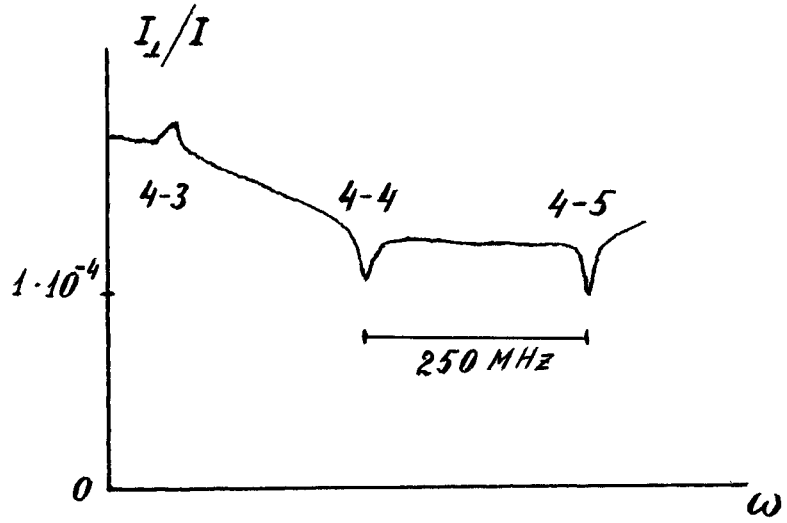

Fig.8. Running laser beam polarization spectra monitored with an elliptic polarization on the *10 μm Cs* cell (vertical scale yields the intensity transmitted $I_\perp$ through the cross-polarizer in units of the incident intensity I).

the linear Doppler-broadened absorption on the stray background light, but the very good contrast of the Doppler-free peaks is noteworthy (Fig.8).

With these experiments, one demonstrates that the general advantages of polarization spectroscopy (detection at null frequency on an arbitrary small background, relatively high contrast with no need of a sophisticated electronic lock-in detection) [1] do apply to the sub-Doppler resonances associated with the optical pumping in a thin cell.

6. Peculiarities of optical phenomena in thin gas cells and their possible applications

According to presented above results, non-trivial optical effects take place in thin gas cells even in the case of one-quantum transition between nondegenerate quantum states. Thus it was shown in papers [2-8], that unlike the known methods of nonlinear spectroscopy [1], sub-Doppler resonances under consideration may be induced and registered by means of only one running monochromatic wave in the cell. However these results relate only to one-quantum transition spectroscopy and can be achieved under sufficiently rigid restrictions on the laser beam parameters [2-8]. The more universal method of sub-Doppler spectroscopy was proposed in papers [16,17], which also based on time-of-flight effects in thin gas cells, but involves different waves for pumping and probing. Let us assume, that atoms (or molecules) of a rarefied gas to be pumped throughout the volume of the thin cell by the broadband radiation. Then the populations of the long-lived particles levels will relax to their equilibrium values primarily in collisions with the cell walls.

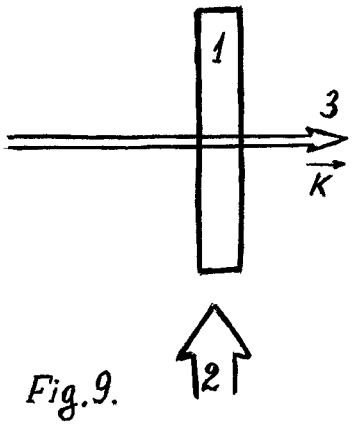

Fig.9.

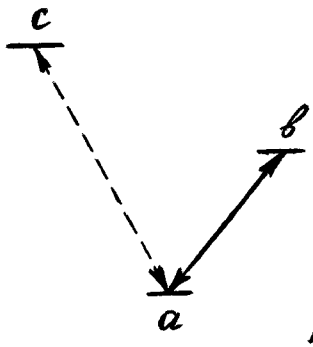

Fig. 10.

Fig.9. Scheme of the experiment: 1-gas cell, 2-pump radiation, 3-monochromatic probe wave.

Fig.10. Diagram of atomic levels and transitions: $a \rightarrow b$ is the pumping transition, $a \rightarrow c$ is the n-quantum probe transition from the long-lived state a.

Such process is determined by the wall-to-wall transit time $\tau = \ell/|v|^{-1}$, where v is the particle velocity component along the cell. The optical pumping of the particles will be efficient if $W\tau \geq 1$ i.e. when

$$W\ell \geq |v|, \tag{9}$$

where W is the probability of population redistribution among the long-lived particle quantum states through light-induced excitation from these states with a subsequent radiative decay of the excited levels. Thus, the transit time effects in a gas cell can be used to produce a nonequilibrium distribution of particles in the velocity component v for long-lived quantum states in the region of sufficiently small values $|v|$ defined by Eq.(9). Under certain conditions this nonequilibrium distribution will create sub-Doppler resonances in the frequency dependences of absorption (dispersion) of the probe radiation in the medium and of its induced fluorescence. Indeed, we assume the optically pumped gas medium under consideration to be probed along the cell by a travelling monochromatic wave with the frequency ω and the wave vector $\vec{K}$ (Fig.9). We also assume that this wave induces the direct resonant n-quantum ($n \geq 1$) transition $a \rightleftarrows c$ to an excited level c from a long-lived state a, where the pump radiation drives an electric dipole transition $a \rightleftarrows b$ (Fig.10) so that the atomic velocity projection v onto the wave vector satisfies the condition (9). According to the Doppler effect, the probe wave is efficiently absorbed by particles whose velocity projections v satisfy the relation:

$$|\delta - nKv| \leq \gamma, \tag{10}$$

where $K = |\vec{K}|$, $\delta = n\omega - \omega_0$ is the frequency detuning with respect to the n-quantum resonance transition $a \rightleftarrows c$, which is characterized by the central frequency ω_0 and the homogeneous

spectral line half-width γ substantially smaller than the corresponding Doppler broadening. As seen from Eqs.(9) and (10), at pump intensities low enough that $W\ell \lesssim \gamma(kn)^{-1}$, the probe wave efficiently interacts with particles having a nonequilibrium velocity distribution in the state a only at small frequency detunings $|\delta| \lesssim \gamma$. Hence the amplitude and polarization characteristics of the wave (and of the medium fluorescence) as functions of the detuning δ may exhibit Doppler-free resonances in the region $|\delta| \lesssim \gamma$. The suggested method of the ultra-high resolution spectroscopy in thin gas cells [16,17] may be used for analysis of a sub-Doppler structure of spectral lines not only of the electronic shell of atoms (ions, molecules) but also their nuclei. It is caused by the possibility of the orientation (alignment) of atomic nuclei by an optical radiation [12]. In this case the probe wave must be resonance to the quantum nuclear transition under study.

Theoretical research showed, that non-trivial manifestations of the Hanle effect in the long-lived degenerate atomic state a may take place in thin gas cells [18]. Indeed a running monochromatic wave induces polarization moments (orientation, alignment) of the level a for a group of atoms whose velocity projection v (on the wave vector $\vec{k}$) is close to the value δ/k, where δ is the frequency detuning with respect to the center of the resonance optical transition $a \rightleftharpoons b$. In the presence of an external magnetic field $\vec{H}$, oscillations of the polarization moments of particles interacting with the wave take place [1,12]. The phase increment of such oscillations in the time of flight relaxation $\tau = \ell/|v|^{-1}$ is characterized by the quantity $\Omega\tau = \Omega\,\ell/|v|^{-1}$, where Ω is the Zeeman splitting of the level a. As the given quantity $\Omega\ell/|v|^{-1}$ is related (through v) to the frequency detuning δ, a series of nontrivial sub-Doppler resonances appear in dependences of pumping wave parameters on δ [18]. Given resonances may be used in quantum magnetometry because their positions depends strongly on a sufficiently weak magnetic field even when the Zeeman splitting of a spectral line and the upper level b of the optical transition $a \rightleftharpoons b$ is negligibly small. Because of the low optical density of a rarefied gas medium in a thin cell, it is appropriate to study experimentally the magneto-optical phenomena under consideration by using highly sensitive polarimetric methods. Therefore the theoretical analysis of these phenomena was performed in [18] at the Faraday effect resulting from reorientation of the light-induced quadrupole moment of the ground (metastable) level of particles in a weak magnetic field. Theoretical research of sub-Doppler resonances in polarization characteristics of the running monochromatic light wave at the Faraday effect in the thin gas cell was carried out also in papers [6,19].

Experimental investigations of Knize et al. [20] demonstrated interesting peculiarities of the degenerate four-wave mixing (DFWM) in thin gas cells. Indeed the resonant DFWM signals usually not observed at noncycling transitions in alkali-metal vapors due to population depletion from optical pumping. However frequent atom-wall collisions can overcome the effects of optical pumping when atomic vapors are contained in thin cells. Then significantly enhanced DFWM

signals are observed at noncycling transitions, and they are comparable in magnitude to the signals observed at cycling transitions in thin cells [20]. Such technique extends the utilization of DFWM to noncycling transitions in atoms or molecules.

Saturated absorption (SA) spectroscopy in thin gas cells also is interesting. Thus experimental research was carried out of the interaction of probe and pump monochromatic beams (from the same laser) in such Cs vapor cells [8]. It was shown that SA spectra of complex atomic systems may be essentially simplified, because sub-Doppler crossover resonances are suppressed in the thin gas cell.

Multi-quantum processes also may strongly changes in thin gas cells. Thus in paper [21] the theoretical investigation was carried out of the interaction of the two-frequency laser radiation with three-level atomic (molecular) Λ-system (Fig.11a) between the excited quantum state $|3\rangle$ and long-lived lower states $|1\rangle$ and $|2\rangle$. Then, unlike the case of the two level system, the coherence is essential between levels $|1\rangle$ and $|2\rangle$ [1,11]. The relaxation rate of this coherence is determined by the transit time $\ell/|v|^{-1}$ of atoms (molecules) between walls of the thin cell. Therefore non-trivial narrow two-quantum resonances arise in the absorption of the radiation, which may be used for the stabilization of both frequencies of waves and the difference of these frequencies [21]. Interesting manifestations of the electromagnetically induced transparency for the Λ-system of atomic levels in thin gas cells were theoretically analyzed also in paper [22].

In paper [23] the theoretical research was carried out of the interaction of the probe and pump laser waves with the resonant cascade system of levels $|1\rangle - |3\rangle - |2\rangle$ (Fig.11b) from the ground state $|1\rangle$ of atoms (molecules) in the thin gas cell. It is shown the possibility of the essential influence of the transit relaxation of atoms on processes of the two-quantum excitation $|1\rangle - |2\rangle$ and on the optical pumping of the transition $|1\rangle - |3\rangle$ (fig.11b). In consequence, qualitatively new peculiarities were established in the absorption spectrum of the probe wave in comparison with a "macroscopic" gas cell [23].

Thus the specific transit relaxation of atoms (molecules) in thin gas cells leads to a number of non-trivial optical phenomena which may be used in laser spectroscopy and quantum electronics.

Fig.11. Λ *(a)* and cascade *(b)* systems of electric dipole transitions $|1\rangle - |3\rangle$ and $|3\rangle - |2\rangle$ between quantum levels $|1\rangle, |2\rangle$ and $|3\rangle$.

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