

HIGH-RESOLUTION LASER SPECTROSCOPY BASED ON OPTICAL PUMPING AND TRANSIT OF ATOMS IN A THIN CELL

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 The paper presents the brief description of principles and results of sub-Doppler laser spectroscopy based on non-trivial narrow absorption and dispersion resonances, which arise because of the optical pumping and specific transit relaxation of atoms (molecules) in thin gas cells.

 Basic part of our knowledge about structure of a matter on the atomic-molecular level is obtained mainly from data of the optical spectroscopy. Therefore it is very important to elaborate effective methods of highresolution spectroscopy which allow to analyze the structure of spectral lines hidden by the Doppler broadening because of a thermal motion of atoms or molecules [1]. In 1992-1993 I suggested new methods of sub-Doppler laser spectroscopy based on optical pumping and transit of atoms in thin gas cells [2-5]. Later given methods were successfully realized at experiments in laboratories of different countries for the precision spectral analysis of atoms and the laser frequency stabilization. The present paper is the brief description of basic principles and achievements of this new direction of the high-resolution laser spectroscopy.

Usually the laser beam diameter *d* is much less than the length *l* of a gas cell (fig.1a) at spectroscopic investigations. Therefore the relaxation of particles (atoms, molecules) resulting from the finite time of their flight along the cell is ignored as a rule. However such relaxation can lead to qualitative new results in rarefied gases if *d>>l* (fig.1b). Indeed, for a Doppler broadened spectral line of a resonant transition with the central frequency Ω*,* a variation of traveling monochromatic wave with the frequency ω in a gas medium results mainly from its interaction with a group of particles, whose velocity projection v (on the wave vector k) is close to $(\omega \cdot \Omega)/k$ [1].

Let us consider the noncycling resonant optical transition $a \leftrightarrow b$, when the excited level *b* may radiatively decay not only on the lower quantum state *a* but also on other long-lived states, which don't interact with the monochromatic wave. In this case a light induced repumping of atoms from the state *a* may be essential [1].

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).*

Such repumping is selective on the projection ν of the atomic velocity and increases with a growth of the transit time $\tau = l/|v|$ of atoms between cell walls. At approach of the frequency detuning $\delta = (\omega \cdot \Omega)$ to zero, the monochromatic wave 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*. Such non-trivial absorption and polarization sub-Doppler resonances were predicted and theoretically analyzed for the first time in papers [2,3]. Later given resonances were registered and investigated at xperiments by French scientists [6,7] in case of the D line (852 nm) of Cs for a series of thin gas cells with different lengths (from $10\mu m$ to $1000 \mu m$). The structure of such resonances essentially depends on the laser beam diameter *d* [2,3,6-8]. Recently Chinese scientists demonstrated a simple method of stabilizing an external cavity diode laser frequency to these resonances in thin cells with the cesium vapor [9].

It is important to note that, unlike the known methods of laser spectroscopy in gas cells [1], described above sub-Doppler absorption and polarization resonances are induced and registered by means of only one running monochromatic wave. However such sufficiently narrow resonances may be observed only for the one-quantum transition spectroscopy , under rigid restrictions on laser beam parameters, and for very thin gas cells (with the length *l*<0.1mm at the usual beam diameter *d*∼1mm).

Therefore the more universal method of sub-Doppler spectroscopy was proposed by the author in papers [4,5], which also is 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 of the thin cell by the broadband radiation. Then the populations of the longlived particles levels will relax to their equilibrium values primarily in collisions with the cell walls. Such process is determined by the wall-to-wall transit time $\tau = l/|\nu|$, where ν is the particle velocity component along the cell. The optical pumping of the particles will be efficient if

 $Wl \ge |v|$, (1) where *W* is the probability of a population redistribution among the long-lived particle quantum states through the 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 ν for long-lived quantum states in the region of sufficiently small values $|\nu|$ defined by Eq.(1). Under certain conditions this nonequilibrium distribution will create sub-Doppler resonances in frequency dependences of absorption (dispersion) of the probe radiation in the medium and of its induced fluorescence. Indeed we assume that the optically pumped gas medium under consideration to be probed along the cell by a traveling monochromatic wave with the frequency ω and the wave vector k (fig.2). We also assume that this wave induces the direct n-quantum (n≥1) transition $a \leftrightarrow c$ to an excited level *c* from a long-lived state *a*, where the pump radiation drives an electric dipole transition $a \leftrightarrow b$ (fig.3) so that the atomic velocity projecttion ν on the wave vector k satisfies the condition (1) .

According to the Doppler effect, the probe wave is efficiently absorbed by particles whose velocity projections *v* satisfy the relationship:

$$
|\delta - nkv| \leq \gamma , \qquad (2)
$$

where $k=|\mathbf{k}|$, $\delta=(n\omega-\Omega)$ is the frequency detuning with respect to the n-quantum resonance transition $a \leftrightarrow c$, which is characterized by the central frequency Ω and the homogeneous spectral line half-width γ substantially smaller than the corresponding Doppler broadening. We can see from Eqs. (1) and (2) , that at sufficiently low pumping intensity, when *Wl*[≤] ^γ */(kn)*, the probe wave efficiently interacts with particles having a nonequilibrium velocity distribution in the state *a* only at small frequency detunings $|\delta| \leq \gamma$. Hence the amplitude and polarization characteristics of the wave as functions of the detuning δ may exhibit Doppler-free resonances in the region $|\delta| \leq \gamma$.

Later given method of sub-Doppler spectroscopy was realized at experiments by Japanese scientists [10-14]. At first they used two independent laser beams for optical pumping and probing [10, 11]. As a result, a hyperfineresolved sub-Doppler spectrum of the D line of the cesium was observed even with a 10-mm cell (with the inner diameter 34 mm). Compared to the method with the single running wave in ultra-thin gas cells [6, 7], such thicker cells are easier to fabricate and the longer interaction length gives a better signal to noise ratio. Therefore, in particular, the effective frequency stabilization of a diode laser was achieved on the hyperfine component of the Cs D line using the elaborated spectroscopic method [11].

Fig.2. Scheme of the experiment: {1} is the gas cell, {2} is the pump radiation, {3} is the monochromatic probe wave.

Fig.3. Diagram of atomic levels and transitions: $a \leftrightarrow b$ is the pumping transition, $a \leftrightarrow c$ is the n-quantum probe transition from the long-lived state *a.*

Then a more simple and convenient experimental configuration was realized [12], where a single laser beam, split into two paths, pumps and probes atoms in perpendicular directions. In this case hyperfine components of the Cs D line also were clearly resolved in a series of glass cells with lengths from 0.5 mm to 5 mm [12-14]. The obtained results may be used in highresolution spectroscopy and for the effective laser frequency stabilization, for example, in downsizing frequency standards on the basis of thin vapor cells.

The specific transit relaxation of atoms (molecules) in thin gas cells causes also another non-trivial optical phenomena which may be interesting for specialists on laser spectroscopy and quantum electronics. Thus the experimental work [15] demonstrates features of the degenerate four wave mixing (DFWM) in thin gas cells. Indeed the resonant DFWM signals usually are 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 [15]. Such technique extends the utilization of DFWM to noncycling transitions in atoms or molecules.

Saturated absorption (SA) in thin gas cells also is interesting. Thus the experimental research was carried out of the interaction of probe and pump monochromatic beams (from the same laser) in such Cs vapor cells [7]. 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.

Interesting magneto-optical phenomena in thin gas cells were theoretically analyzed in papers [16, 17]. Thus non-trivial sub-Doppler resonances may appear in amplitude and polarization characteristics of the running monochromatic wave due to the Hanle effect in the longlived degenerate atomic state [16]. Given resonances may be used in quantum magnetometry because their positions strongly depend on a sufficiently weak magnetic field even when the Zeeman splitting of the spectral line of the resonance optical transition is negligibly small.

Multi-quantum transitions also may strongly changes in thin gas cells. Thus in paper [18] the theoretical investigation was carried out of the interaction of the twofrequency laser radiation with the three-level atomic (molecular) Λ-system (fig.4a) between the excited quantum state $/3$ and long-lived lower states $/1$ and $/2$. The relaxation rate of this coherence is determined by the transit time $\tau = l/|v|$ 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 [18]. Later interesting features of such coherence between lower states /1> and /2> (fig.4a) were observed and analyzed at experiments [19] with thin Cs vapor cells.

- [1]. Demtroder W., "Laser Spectroscopy", (Heidelberg, Springer, Berlin) 2003.
- [2]. Izmailov A.Ch., Laser Physics,1992, v.2, N5, pp.762- 763.
- [3]. Izmailov A.Ch., Optics and Spectroscopy, 1993, N1, pp.25-29, (Opt. i Spektrosk., 1993, v.74, pp.41- 48).
- [4]. Izmailov A.Ch., Laser Physics, 1993, v.3, N2, pp.507-508.
- [5]. Izmailov A.Ch., Optics and Spectroscopy, 1993, v.75, N3, pp.395-398, (Opt. i Spektrosk., 1993, v.75, pp.664-669).
- [6]. Briaudeau S., Bloch D., Ducloy M., Europhysics Letters, 1996, v.35, N5, pp.337-342.
- [7]. Briaudeau S., Bloch D., Ducloy M., Physical Review A, 1999, v.59, N5, pp.3723-3735.
- [8]. Tajalli H., Ahmadi S., Izmailov A.Ch., Laser Physics, 2001, v.11, N12, pp.1256-1261.
- [9]. Zhao YT., Zhao JM., Huang T., et al , Journal of Physics D: Applied Physics,2004, v.37, pp.1316- 1318.
- [10]. Tachikawa M., Fukuda K., Hayashi S., Kawamura T., Jpn. J. Appl. Phys., 1998, v.37, N12B, L1556- L1559.

Fig.4. Λ (a) and cascade (b) systems of electric dipole transitions $(1) \leftrightarrow (3)$ and $(3) \leftrightarrow (2)$ between quantum levels.

In paper [20] the theoretical research was carried out of the interaction of the probe and pump monochromatic waves with the resonant cascade system of levels $(1 \rightarrow 3 \rightarrow 4)$ (fig.4b) from the ground state $(1 \rightarrow 6)$ 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 $(1) \leftrightarrow (2)$ and on the optical pumping of the transition $/1$ > \leftrightarrow $/3$ > (fig.4b). In consequence, qualitatively new features were established in the absorption spectrum of the probe wave in comparison with a "macroscopic" gas cell [20].

Now research on optics of thin gas cells is carried out in laboratories of different countries and we may expect new interesting results of this direction of the highresolution spectroscopy.

- [11]. Fukuda K., Furukawa M., Hayashi S., Tachikawa M., IEEE Trans. Ultrason. Ferroelectr. Freq. Control, 2000, v.47, N2, pp.502-505.
- [12]. Otake M., Fukuda K., Tachikawa M., Apllied Physics B, 2002,v.74, pp.503-508.
- [13]. Fukuda K., Kinoshita M., Tachikawa M., Applied Physics B, 2003, v.77, pp.823-827.
- [14]. Izmailov A.Ch., Fukuda K., Kinoshita M., Tachikawa M., Laser Physics, 2004, v.14, N1, pp.30- 38.
- [15]. Ai B., Glassner D.S., Knize R.J., Physical Review A, 1994, v.50, N4, pp.3345-3348.
- [16]. Izmailov A.Ch., Optics and Spectroscopy, 1996, v.80, N3, pp.321-324, (Opt. i Spektrosk., 1996, v.80, pp.365-368).
- [17]. Tajalli H., Ahmadi S., Izmailov A.Ch., Laser Physics, 1998, v.8, N6, pp.1223-1227.
- [18]. Namdar A., Tajalli H., Kalafi M., Izmailov A.Ch., Laser Physics, 1999, v.9, N2, pp.476-480.
- [19]. Fukuda K., Toriyama A, Izmailov A.Ch., Tachikawa M., accepted in "Applied Physics B", 2005.
- [20]. Kalafi M., Tajalli H., Namdar A., Izmailov A.Ch., Laser Physics, 2000, v.10, N2, pp.553-556.