Influence of spontaneous emission transfer on the resonances of interaction of laser radiation with alkali atoms confined in an extremely thin cell

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Abstract. The nonlinear absorption and fluorescence spectra of ^{133}Cs vapours confined to the extremely thin cell are computed in the framework of perturbation theory regarding to the intensity of the laser light illumination. The amplitude as well as the sign of the longitudinal alignment of the ground state is shown to be defined by the spontaneous decay of the upper level. The sign and the magnitude of the nonlinear absorption resonance on the closed optical transition are shown to be defined by this process as well.

1. Intoduction

The sub-Doppler spectroscopy of alkali atom vapour was strongly advanced by the invention of the Extremely Thin Cells (ETC) [1]. Using the conventional spectroscopic methods of registration, a number of peculiarities in the absorption, reflection and fluorescence spectra were observed in these cells, which are not present in commonly used (cm-size) cells. The main reason for the formation of the sub-Doppler structures in the spectra is the strong spatial anisotropy in the ETC environment. Collisions with the cell walls induce loss of atomic phase memory and excitation quenching. Hence, the atoms departing from the cell walls undergo a transient regime of excitation before they acquire the steady state polarization that corresponds to their velocity as well as to the laser field intensity and detuning. Nonlinear resonances in absorption and fluorescence are also influenced by velocity selective optical pumping [2].The present work gives a description of the interaction of resonant laser fields with alkali atoms in an ETC with the full account of the hyperfine sublevel degeneracy using iteration procedure over the laser field intensity.

The problem of determination of the nonlinear atomic polarization was solved for arbitrary values of the total angular momenta of the resonance levels for excitation with linearly polarized laser light. Using the previously developed methodology [3-6], the equations of motion for the statistical operator in the irreducible tensor operator (ITO) representation were solved, taking into account the transient processes started by the atomic collisions with the cell walls as well as the spontaneous transfer of coherence and population from the upper level to the ground state [7].

2. Theory

The developed theoretical approach is based on the full account of transient polarizations induced by the atom-wall collisions in extremely thin cells as well as nonlinear optical effects and magnetic sublevel degeneracy. Let us start with the introduction of the statistical operator ρ_q^κ ($\rho = f, \varphi, \xi$) that defines the populations (k=0) and alignments (k=2) of the upper (f_q^κ) and the lower (φ_q^κ) levels as well as the optical coherence

$$\xi_q^\kappa(x,v,t) = \xi_q^\kappa(x, v)\exp\{-i\,\omega_{las}\,t\}\,, \tag{1}$$

where x is the atomic coordinate along the propagation direction of the laser beam, which is perpendicular to the cell windows, t is time, v is the projection of the atomic velocity on the x axis, and ω_{las} is the laser frequency. In the rotating wave approximation one obtains time-independent equations of the form [8]

$$v\frac{\partial}{\partial x} f_q^\kappa + \gamma_f^\kappa f_q^\kappa = ih^{-1}(2\mathrm{F}_\varphi+1)^{-1/2}\sum_{\kappa' q' Q} E_{-Q}(x)\, \mathrm{C}_{qq'Q}^{\kappa\kappa'}\left[\mathrm{d}\xi_{q'}^{\kappa'}(x,v) + \mathrm{d}^*(\xi_{-q'}^{\kappa'}(x,v))^*(-1)^{\kappa+\kappa'+q'}\right]$$
$$+\left(2F_f+1\right)\gamma_f^\kappa N_f W(v)\delta_{\kappa 0} \tag{2a}$$

$$v\frac{\partial}{\partial x}\varphi_q^\kappa + \gamma_\varphi^\kappa \varphi_q^\kappa = (-1)^\kappa i h^{-1}(2\mathrm{F}_\varphi+1)^{-1/2}\sum_{\kappa' q' Q} E_{-Q}(x)(-1)^{\kappa'}\mathrm{B}_{qq'Q}^{\kappa\kappa'}\left[\mathrm{d}\,\xi_{q'}^{\kappa'}(x,v) + \mathrm{d}^*(\xi_{-q'}^{\kappa'}(x,v))^*(-1)^{\kappa+\kappa'+q'}\right]$$
$$+\left(2F_\varphi+1\right)\gamma_\varphi^\kappa N_\varphi W\left(v\right)\delta_{\kappa 0} + \Gamma_{F_f F_\varphi}^\kappa f_q^\kappa \tag{2b}$$

$$v\frac{\partial}{\partial x}\xi_q^\kappa(x,v) + \left[\gamma_\xi^\kappa + i(\omega_0 - \omega_{las})\right]\xi_q^\kappa(x,v) = ih^{-1}(2\mathrm{F}_f+1)^{-1/2}\mathrm{d}^* \times$$
$$\sum_{\kappa' q' Q} E_{-Q}(x)\left[\mathrm{S}_{qq'Q}^{\kappa\kappa'} f_{q'}^{\kappa'} + (-1)^{\kappa+\kappa'}\mathrm{R}_{qq'Q}^{\kappa\kappa'}\varphi_{q'}^{\kappa'}\right] \tag{2c}$$

where

$$C_{qq'Q}^{\kappa\kappa'} = (-1)^{2\mathrm{F}_\varphi+q'}(2\mathrm{F}_f+1)^{1/2}(2\kappa'+1)\begin{Bmatrix}\kappa' & 1 & \kappa\\ F_f & F_f & F_\varphi\end{Bmatrix}\begin{pmatrix}\kappa' & 1 & \kappa\\ -q' & Q & q\end{pmatrix} \tag{3a}$$

$$\mathrm{R}_{qq'Q}^{\kappa\kappa'} = (-1)^{2\mathrm{F}_\varphi+q'}(2\mathrm{F}_f+1)^{1/2}(2\kappa'+1)\begin{Bmatrix}\kappa' & 1 & \kappa\\ F_f & F_\varphi & F_\varphi\end{Bmatrix}\begin{pmatrix}\kappa' & 1 & \kappa\\ -q' & Q & q\end{pmatrix} \tag{3b}$$

while $\mathrm{B}_{qq'Q}^{\kappa\kappa'}$ and $\mathrm{S}_{qq'Q}^{\kappa\kappa'}$ may be obtained out of $C_{qq'Q}^{\kappa\kappa'}$ and $\mathrm{R}_{qq'Q}^{\kappa\kappa'}$ via substitution $F_f \to F_\varphi$. $d \equiv (F_f\|d\|F_\varphi) \equiv \left\|d_{F_f F_\varphi}\right\|$ is the reduced dipole moment of the transition.

The populations ρ_0^0 and the alignments ρ_0^2 ($\rho = f, \varphi$) of both the ground and the excited states were obtained in the second order of the perturbation theory and integrated over the cell thickness

$${}^{(2)}\rho_0^{\kappa_2}(v) = \frac{1}{l}\int_0^l \,[{}^{(2)}\rho_0^{\kappa_2}(x, v>0) + {}^{(2)}\rho_0^{\kappa_2}(x, v<0)]dx,\;\; \rho = f,\varphi\;. \tag{4}$$

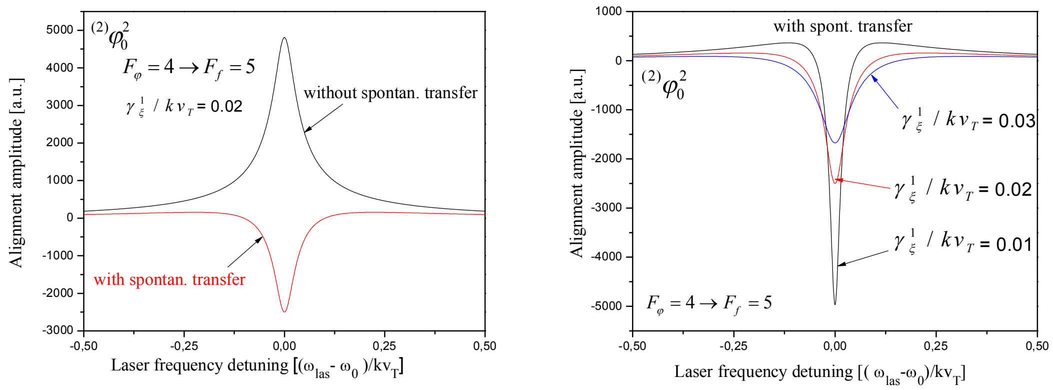
The linearly polarized laser light induces the population difference of magnetic sublevels of the ground state that results in the longitudinal alignment φ_0^2. Spontaneous emission from the upper level [7-9] accounted for by the term $\Gamma_{F_f F_\varphi}^\kappa f_q^\kappa$ in Eq. (2b) partially returns the atoms back and leads to the changes of alignment as well. According to the results of detailed computations, accounting for the spontaneous emission leads to the sing reversal of the alignment on the closed transition

$F_\varphi = 4 \to F_f = 5$. Open transitions are affected as well but less strongly. The amplitudes of the resonances depend on the values of the relaxation constants (Fig. 1).

Numerical integration of ${}^{(2)}f_0^{\kappa_2}(v)$ over the Maxwell velocity distribution $W(v)$ with the thermal velocity v_T gave the intensity of the unpolarized fluorescence in the given direction $\vec{n}$

$$ {}^{(2)}I_{f\varphi}(\vec{n}) = K_0 \left\| d^2_{F_\varphi F_f} \right\| \left[\frac{{}^{(2)}f_0^0}{3(2F_f+1)^{1/2}} + 5(-1)^{F_f+F_\varphi} \begin{Bmatrix} 1 & 1 & 2 \\ F_f & F_f & F_\varphi \end{Bmatrix} \left[\sum (-1)^q \, {}^{(2)}f_q^2 \, T_{-q}^2(\vec{n}) \right] \right] \quad (5) $$

Here $T_{-q}^2(\vec{n})$ is the observation tensor [10].



(a) (b)

Figure 1. (a) Inversion of the longitudinal alignment resonance with the account for the spontaneous decay on the closed transition $F_\varphi = 4 \to F_f = 5$, (b) influence of the spontaneous relaxation rate on the amplitude of the inverted resonance.

In the first and the third orders of perturbation theory the computed values of the optical coherences ${}^{(1)}\xi_0^1(x,v) = {}^{(1)}\xi_0^1(x,v>0) + {}^{(1)}\xi_0^1(x,v<0)$ and ${}^{(3)}\xi_0^1(x,v) = {}^{(3)}\xi_0^1(x,v>0) + {}^{(3)}\xi_0^1(x,v<0)$ were used to obtain the transmitted field amplitudes [4,5]

$$ E_{tr}^1 = (-1)^{F_f - F_\varphi} (2F_\varphi + 1)^{-1/2} 2\pi ikd \int_0^l \exp[-ikx'] \int_{-\infty}^{+\infty} W(v) [{}^{(1)}\xi_0^1(x,v) + {}^{(3)}\xi_0^1(x,v)] \, dv \, dx' \quad (6) $$

Finally, the intensity of the transmitted light was obtained as the squared modulus of the sum of the incident light and the fields ${}^{(1)}E_{tr}^1$ and ${}^{(3)}E_{tr}^k$ induced by the vapor polarization

$$ {}^{(\Sigma)}I_{tr} \propto |E_0|^2 \left[1 + 2\,\mathrm{Re}\left(\frac{{}^{(1)}E_{tr}^1}{E_0} + \frac{{}^{(3)}E_{tr}^\kappa}{E_0} \right) \right] \quad (7) $$

Let us stress that contrary to the previous publications, relaxation rates γ_ρ^k $(\rho = f, \varphi, \xi)$ were assumed to be different. They were varied to reach the best fit to the experimental data.

3. Comparison with the experiment

The results of the computations were used for interpretation of the experimental findings. Fig. 2 plots the fluorescence spectra of ^{133}Cs vapor confined to the extremely thin cells of the thickness l=(3/2)λ, where $\lambda = 852{,}35$ nm corresponds to the cesium D_2 line $6s\ ^2S_{1/2} \to 6s\ ^2P_{3/2}$.

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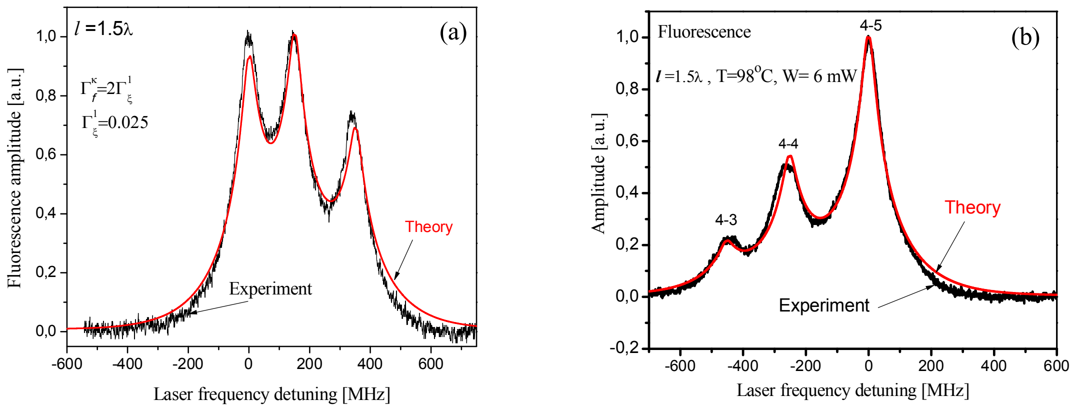

Figure 2. Theoretical and experimental fluorescence spectra of ^{133}Cs vapor confined to the extremely thin cells of the thicknesses l=(3/2)λ. (a) hyperfine transitions $F_\varphi = 3 \to F_f = 2, 3, 4$, (b) hyperfine transitions $F_\varphi = 4 \to F_f = 3, 4, 5$.

Fig. 3 plots the absorption spectra of cesium vapor in the same extremely thin cells. Saturated absorption resonances are well reproduced by the calculated nonlinear term ${}^{(3)}E^k_{tr}$. The enhanced absorption that is clearly seen in the case of l=(3/2)λ is due to the Dicke-like transient polarizations [4]. This linear effect dominates and obscures the influence of the nonlinear resonances and of the spontaneous transfer.

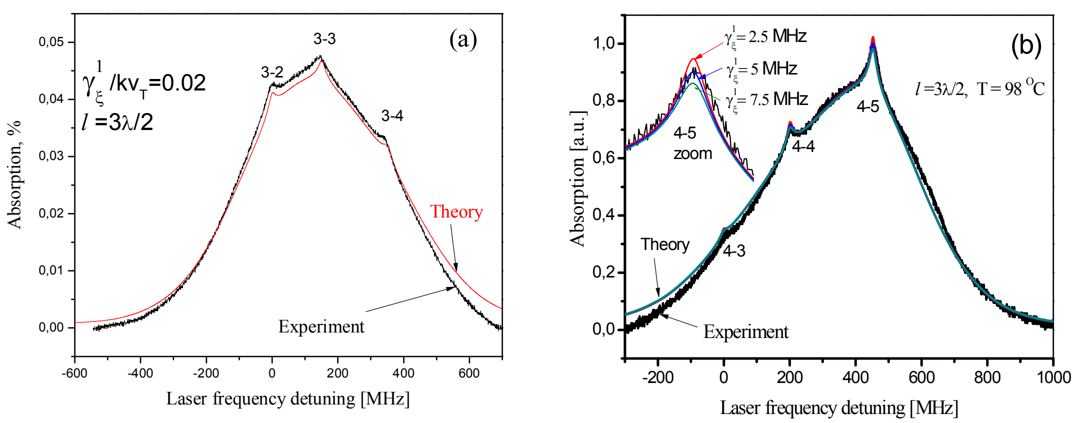

Figure 3. Theoretical and experimental absorption spectra of ^{133}Cs vapour confined to the extremely thin cells of thicknesses l=(3/2)λ.(a) hyperfine transitions $F_\varphi = 3 \to F_f = 2, 3, 4$, (b) hyperfine transitions $F_\varphi = 4 \to F_f = 3, 4, 5$

The role of the spontaneous transfer is shown in pure form in Fig. 4 for the cell that is 6λ thick. The detailed theoretical considerations have shown that absorption is enhanced only on the closed transition $F_\varphi = 4 \to F_f = 5$. This effect is the direct consequence of the sign reversal of the longitudinal alignment. The amplitude of this resonance depends on the relaxation constants and may be used for their estimations by comparing with the experimental data.

4. Conclusion

Steadily increasing demand for miniaturized optical devices leads to the growing importance of the theories that are able to account for atom-wall collisions and to predict the spectral lines shapes in the extremely thin cells. Conventional approaches based on simplified models failed to give quantitative description of the optical processes in the extremely thin cells. Simultaneous account for the transient polarizations and optical pumping effects are known to be the indispensible ingredients of the theoretical models.

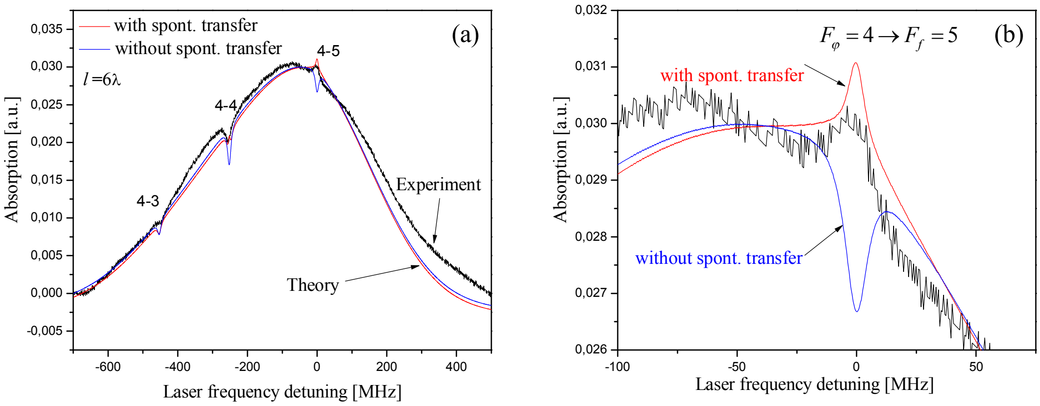

Figure 4. Theoretical and experimental absorption spectra of ^{133}Cs vapours confined to the extremely thin cell of the thicknesses l=6λ; (a) overview of hyperfine transitions $F_\varphi = 4 \to F_f = 3, 4, 5$; (b) detailed absorption spectrum associated with the closed transition $F_\varphi = 4 \to F_f = 5$.

In this contribution we showed that accounting for the alignment in the ground state induced by the spontaneous emission transfer is also of crucial importance. In particular, the sing of the nonlinear absorption resonance on the closed transition can not be obtained in accord with the experimental data unless the theory accounts for the spontaneous emission transfer and the right sing of alignment in the ground state.

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