Temperature and excitation power influence on the velocity-selective optical pumping resonances of $^{133}$Cs atoms confined in an extremely thin cell

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Abstract. Linear and nonlinear absorption spectra of $^{133}$Cs vapor confined in an extremely thin cell were computed via iterations with respect to the resonance radiation intensity. When the incident radiation intensity is low, the transient polarization of the atoms that undergo frequent collisions with the cell walls leads to sub-Doppler features in the absorption spectra. Higher incident radiation intensities result in the appearance of velocity-selective optical pumping resonances. The theory developed agrees quantitatively with the experimental findings.

1. Introduction

The extremely thin layers of atomic vapors possess unique spectral properties due to the frequent collisions of the atoms with the walls of the optical cell containing the alkali vapor. The development of extremely thin cells (ETC) in 2001 [1] considerably widened the sub-Doppler spectroscopy capabilities. The main features of an atomic vapor confined in an ETC and subjected to resonant electromagnetic excitation are the strong anisotropy of the gas phase, the frequency selection of the atomic velocities and the dominant role of the polarization transients. As the width of the ETC is less than the mean-free-path, the atoms do not reach steady-state polarization. Acting together, these mechanisms lead to the appearance of narrow resonances in the resonance fluorescence excitation spectra, as well as in absorption and reflection. Such narrow resonances are of increasing interest from both fundamental and practical points of view. Many properties of these resonances and their dependence on the excitation conditions have been the object of numerous studies [e.g. 2-5].

In the present paper we present a comparative study of the influence of the temperature and the excitation power density on the absorption resonances of Cs in an ETC. The main new goal was to separate the effects of these experimental factors in order to suggest the possible mechanisms of interaction in the atomic layer and on the cell walls. In the experiments, the power was varied in a very wide range – from 1.25 mW/cm$^2$ to 125 mW/cm$^2$ for three values of the temperature, namely, 85 °C,
99 °C and 120 °C. The proposed theoretical description based on an iteration approach is only adequate for power densities in the order of threshold values of the saturation power not exceeding 3 mW/cm². This constraint notwithstanding, by varying the relaxation parameters in the numerical experiments, in the third-order solutions we were able to obtain good quantitative agreement with the experimental spectra for values of the power exceeding several times the saturation power density for this particular transition.

2. Theory
Our approach to the theoretical description of an ETC interacting with resonant laser radiation was detailed in [6]. The linear and nonlinear polarizations of atomic vapor sandwiched between transparent dielectric walls are computed for arbitrary values of the atomic levels’ full angular momenta. The master equations are written in the irreducible tensor operator (ITO) representation and solved while accounting for the transients induced by the atomic collisions with the ETC walls [4,7-11]. Also, the coherence and population transfer via spontaneous emission is accounted for [6]. The analytical solutions for the components of the density matrix are obtained up to terms of third order with respect to the laser field. The components of the density matrix in the ITO representation $\rho^K (\rho = f, \varphi, \xi)$, have the following meaning: $(f^K)$ characterizes the upper level population and alignment; $(\varphi^K)$ does the same for the lower level; while $(\xi^K)$ is the optical coherence directly connected with the polarization of the medium.

We derived solutions for the tensor components of the optical coherence $(s)\xi^l(x,\nu) = (s)\xi^l(x,\nu > 0) + (s)\xi^l(x,\nu < 0)$ in the first $(s = 1)$ and third $(s = 3)$ order of the iteration procedure and calculated the respective amplitudes of the transmitted field $(s)E^l$: 

$$(s)E^l = (\pm)^{F_f - F_\varphi} (2F_\varphi + 1)^{-1/2} 2\pi ikd \int_{-\infty}^{+\infty} W(\nu) \exp[-i\nu \gamma(s)\xi^l(x',\nu)] dx'; \ s = 1, 3.$$  (1)

We should emphasize that the numerical integration of the analytical solutions was performed by assuming that the relaxation parameters are different $\gamma^k (\rho = f, \varphi, \xi)$ and by varying their values.

Designating the results of the averaging over the Maxwellian velocity distribution as follows $(s)J^+ = \int_{0}^{\infty} du \left(\rho, \nu, \varphi, \xi\right)$ and $(s)J^- = \int_{-\infty}^{0} du \left(\rho, \nu, \varphi, \xi\right)$, the first-order amplitudes may be written in the following form 

$$(s)E^l_1 / E_0 = K(F_{\rho^s}, \varphi, \kappa, \nu \tau) \left[ (s)J^+ - (s)J^- \right]$$

with

$$K(F_{\rho^s}, \varphi, \kappa, \nu \tau) = \frac{2\pi^{1/2}}{3} \frac{d^2_{F_{\rho^s}, \nu \tau}}{\hbar(k\nu)} \left( \frac{\phi_0^0}{2F_\varphi + 1} - \frac{f_0^0}{2F_f + 1} \right).$$  (2)

In the third-order expression for the transmitted field, the last term describes the contribution of the coherence and population transfer via the spontaneous emission process:

$$(s)E^l = \beta_0 K(F_{\rho^s}, \varphi, \kappa, \nu \tau) E_0 \sum_{k=0,2} \left( S_{000}^{000} C_{000}^{000} F_\varphi (s)J^+ + R_{000}^{000} B_{000}^{000} (s)J^+ - C_{000}^{000} (s)J^+ \right) + \beta_0 C_{0}^{0} (F_{\rho^s}, F_\varphi) \Omega^{2} \left( \Gamma f^{k_s} \right)^2 \Gamma^{k_s} = \gamma^{k_s} / \kappa \nu \tau, \ s = f, \xi, \varphi.$$  (3)
Describing correctly the spontaneous emission is critical in establishing the connection between the nonlinear resonance sign-reversal on the $F_g = 4 \rightarrow F_f = 5$ closed transition of the $D_2$ line with the analogous sign reversal of the ground-state longitudinal alignment that depends on the transition principal quantum numbers, and on the cell thickness and temperature.

The transmitted field intensity is determined by the squared modulus of the sum of the incident field and the polarization-induced fields $(1) E^I_\nu$ and $(3) E^X_\nu$, and can be written as:

$$I_\nu \propto |E_\nu|^2 \left( 1 + 2 \Re \left( \frac{(1) E^I_\nu}{E_\nu} + \frac{(3) E^X_\nu}{E_\nu} \right) \right).$$

(4)

3. Numerical modeling and comparison with the experimental findings

Figure 1 plots the results of the numerical modeling and demonstrates the influence of the field intensity and the atomic source temperature. Only the region of the Cs $D_2$ line spectrum is shown that involves the $F_g = 4 \rightarrow F_f = 4, 5$ hyperfine transitions (figure 1a).

![Figure 1. Absorption of an ETC filled with Cs vapor. (a) demonstrates the temperature dependence of the absorption in a $3\lambda/2$-thick ETC, where $\lambda$ is the resonance wavelength, while (b) shows the $4-5$ transition absorption dependence on the relaxation parameters.](image)

As figure 1a shows, a rise of about 10% of the atomic source temperature leads to a slight but noticeable rise of the resonance amplitude. On the other hand, a twofold rise of the optical coherence relaxation rate $\Gamma_\xi^1$ reduces the resonance amplitude considerably. The calculation of the nonlinear absorption leads to the conclusion that a fivefold increase of $\Gamma_\xi^1$ leads to the disappearance of the resonance on the $F_g = 4 \rightarrow F_f = 5$ transition. In addition, we found that raising the relaxation parameters can be used to simulate the power broadening. Indeed, in a strong laser field, the role of the spontaneous transitions diminishes and the resonance absorption follows the same trend (figure 1 b).

In the case of an ETC of thickness $L = 3\lambda/2$, we compared the experimentally-observed nonlinear narrow-width absorption resonances with the respective theoretical simulations (figure 2), for two different atomic source temperatures: $85^\circ\text{C}$ and $120^\circ\text{C}$. It can be seen that for the lower laser beam intensity (figure 2a), the experimentally-observed nonlinear resonance is very well reproduced by an optical coherence relaxation rate $\Gamma_\xi^1 = 0.03$. For the same two temperatures and following the theoretical approach developed, the laser-light-intensity enhancement is reproduced by means of a higher optical coherence relaxation rate, namely $\Gamma_\xi^1 = 0.064$ (figure 2b). Thus, the nonlinear resonance broadening could be related to optical coherence relaxation. The computed absorption resonances agree well with the experimental results.
We further considered the case of a cell thickness equal to an integer number of optical wavelengths [6] (figure 3a, b). For \( L = \lambda, 3\lambda \), narrow resonances with reduced absorption are observed for all \( F_\nu = 4 \rightarrow F_i = 3,4,5 \) transitions. Figure 3 compares the absorption resonances for two different light intensities and atomic source temperature \( T = 120 \degree \text{C} \). It is important to note that even for incident intensity as high as 15 mW/cm\(^2\), qualitative agreement with the experimental spectra is achieved (figure 3b).

![Figure 2. Temperature dependence of the absorption of an ETC filled with Cs vapor:](image)

**Figure 2.** Temperature dependence of the absorption of an ETC filled with Cs vapor:

![Figure 3. Field intensity dependence of the absorption of an ETC filled with Cs vapor with thickness \( L = \lambda \) (a) and \( L = 3\lambda \) (b).](image)

**Figure 3.** Field intensity dependence of the absorption of an ETC filled with Cs vapor with thickness \( L = \lambda \) (a) and \( L = 3\lambda \) (b).

### 4. Conclusions

We performed comparative studies of the roles played by the cell temperature and the incident light intensity in the formation of sub-Doppler resonances in an ETC filled with Cs vapor. In agreement with the experimental findings, the computed absorption spectra demonstrated a reduction in the amplitudes of the sub-Doppler resonances as the optical coherence relaxation rate \( \Gamma_\xi = \gamma_\xi / k\gamma_T \) increased. On the other hand, the amplitude of the resonance rose with the rise of the cell temperature. The simulation proposed of the power broadening via an artificial increase of the relaxation parameters \( \Gamma_\rho = \gamma_\rho / k\gamma_T, \rho = f, \varphi, \xi \) in the third-order terms of the perturbation series resulted in a good qualitative agreement with the experimental spectra, even for incident intensities several times larger than the saturation threshold.
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