Ultra-narrow spectroscopic cells in atomic spectroscopy: reflection, transmission, fluorescence, and nonadiabatic transitions at the walls

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Abstract. Ultra-narrow cells with the thicknesses in the range from several wavelengths to the small fractions of the wavelength brought a number of new opportunities for atomic spectroscopy. Depending on the cell thickness, spectral lines recorded in ultra-narrow cells are either Doppler-free or Doppler-broadened. With careful selection of the cell thickness hyperfine structure may be easily resolved without resorting on the multibeam nonlinear optical techniques. Moreover, frequent collisions with the walls leads to the important modifications of velocity selective optical pumping resonances. Finally, ultra-narrow cells provide with the unique opportunity to study collisions of the excited atoms with the solid surfaces. In this contribution several examples of the use of the ultra-narrow spectroscopic cells filled with the alkali atomic vapour is presented. First, we discuss general aspects of the transient polarisation that defines all peculiarities of an ultra-narrow cell as a spectroscopic tool. Second, we demonstrate the resolution of the magnetic sublevels in the transition from Zeeman to Paschen-Back regime in the Cs hyperfine structure. Third, new aspects of velocity selective optical pumping resonances in reflection and transmission of resonant radiation by the 6 wavelengths thick cell filled with Cs are discussed. Forth, the experimental evidences of the nonadiabatic transitions between excited states of Rb atoms in the course of collisions with the sapphire surface are presented.

1. Transients in the atomic spectroscopy with ultra-narrow cells

Transient behavior of the atomic polarization under resonance electromagnetic excitation is a very well understood process that gave rise to a number of well-known time-resolved spectroscopic techniques. The role played by the transients in the stationary spectroscopy is much less appreciated although it was first clearly mentioned in 1954 by Cojan who observed sub-Doppler features in the reflection spectrum of resonant radiation from the mercury vapor. His interpretation [1] of this unexpected result was based on the statement that the atoms departing from the surface are in a transient state rather than the steady state. Hence, there are no reasons to describe their response by the steady state polarization corresponding to their Doppler-shifted resonance frequency. In the following year the first “ultra-narrow” pill-box shaped cell was built by Romer and Dicke [2] for ammonia interacting with microwave radiation. It was 6.2 mm thick, exactly one halve of the resonant...
wavelength. In this cell sub-Doppler line widths were obtained. Although the transient behavior of atomic polarization was not calculated in Ref. [2], it contains an important statement that would the cell be thicker than one half of the wavelength the phase of the field acting on the moving atoms will have opposite phase in different parts of the cell.

The role of transients in reflection spectroscopy was further elucidated in the linear regime in Ref. [3]. It was stressed that due to their transient behavior after collision with the wall departing atoms mimic the contribution of arriving atoms that have same absolute value of their velocity, thus effectively reducing the vapor polarization to the doubled contribution of arriving atoms. Nonlinear theory of selective reflection with the full account of transient effect was given in Ref. [4].

Theoretical treatment of reflection and transmission of the narrow cell filled with the resonant atomic vapors with the full account for the transients was given in Ref. [5]. It was shown that the line shapes depend periodically on the cell thickness. The period of these changes is twice as large as compared to the period of Fabri-Perrot resonances.

First experimental realization of a really ultra-narrow cell suited for optical spectroscopy was reported in Ref. [6]. It has variable thickness of the Cs atomic vapor layer in the range of 150-300 nm and enables full resolution of hyperfine transitions that are usually covered by Doppler broadening. Thickness dependence of the narrowing effect was studied in more detail in Ref. [7].

2. Giant modification of atomic transitions probabilities induced by magnetic field

Ultra-narrow cell filled with Cs vapour was used to study the magnetic field-induced giant modification of probabilities for seven components of $6S_{1/2}, F_g=3 \rightarrow 6P_{3/2}, F_e=5$ transition of Cs D2 line forbidden by selection rules. Figure 1 plots the recorded absorption spectrum of Cs in an ultra-narrow cell. The fourteen transition components appear with $\approx 100$ MHz linewidth, thus being completely frequency resolved except for transitions 5, 6 and 7′ resolved partially, and 6′ and 7, which are fully overlapped. As it is seen from the inset, the amplitudes of $F_g=3, m_e=-3 \rightarrow F_e=5, m_e=-2$ and $F_g=3, m_e=-2 \rightarrow F_e=5, m_e=-1$ transitions (labeled 7′ and 6′, respectively) are the largest ones among the group of fourteen lines.

Figure 1. Absorption spectrum of Cs cell with $L=\lambda/2=426$ nm for magnetic field $B=920$ G and $\sigma+$ laser excitation. Seven transitions $F_g=3 \rightarrow F_e=5$ are labeled 1′-7′, while other seven transitions $F_g=3 \rightarrow F_e=4$ are labeled 1-7.

The lower curve is absorption spectrum of the reference cell with $L=\lambda/2$, showing the positions of the Cs $F_g=3 \rightarrow F_e=2,3,4$ transitions for $B = 0$. The inset shows the result of the fitting of the spectrum fragment enclosed in a dashed rectangle, where atomic transitions are partially overlapped.

3. Comparison of reflection and transmission spectra in Cs vapor

High resolution laser spectroscopy of Cs vapor confined in optical cell with longitudinal dimension $L=6\lambda$ ($\lambda=852$ nm) provides good opportunity to study the sub-Doppler-width resonances like Velocity Selective Optical Pumping (VSOP) resonance by single beam spectroscopy. For such a cell, the mean time of atomic flight between the cell windows is close to the lifetime of the excited atomic levels. In addition, the slight enhancement of the cell thickness from $L=0.5 \lambda$ to $L=6 \lambda$ provides with the possibility for strong reduction of the used laser light intensity and allows working at lower atomic source temperature [8]. Both are of significant importance for the VSOP resonance study.
Here, we present experimental results related to some peculiarities in VSOP resonance behavior observed for closed hyperfine transition on the D$_2$ line of $^{133}$Cs. Most of the optical transitions involved in this line are open, suffering atomic population loss due to the fluorescence decay to hyperfine or Zeeman ground-state levels, which do not interact with the exciting laser light. Only the F$_g$=4→F$_e$=5 transition is a closed one, i.e. it exhibits fluorescence decay only to ground state levels that interact with the laser light. The two types of transitions demonstrate different saturation behaviors [9].

Figures 2 and 3 illustrate the transmission and reflection spectra, respectively. Surprisingly, it was observed that for the same (very low) light intensity and similar temperature interval, in the absorption spectrum observed by the reflected light the VSOP resonance sign at the closed transition is opposite, namely absorption dip, with centre at the F$_g$ = 4 → F$_e$ = 5 transition (Fig.3). Moreover, the contrast of the VSOP dip is very good, even at the lowest used atomic density. Note that the shape of the resonance at the closed optical transition differs from those formed at both open transitions. More particularly in the first case, the resonance profile is asymmetric.

The difference in the absorption spectra of transmitted and reflected beams could be related to the fact that in the reflected light the contribution of atoms flying within the two layers with thickness L=$\lambda$, which are located right next to the cell window surfaces is larger than for the case of transmitted light beam. It could be assumed that such atoms undergo stronger depolarization of the excited F$_e$=5 state due to the influence of dielectric surface. This can result in registration of reduced absorption VSOP resonance even at lower temperature, for the absorption spectrum measured by the reflected beam.

Figure 2. Absorption spectra measured by the laser beam transmitted through the L=6 $\lambda$ cell, for different Cs source temperatures. VSOP resonances of reduced absorption occur for both open F$_g$=4 → F$_e$=3,4 transitions, while absorption peak is present at the closed F$_g$=4 → F$_e$=5 transition.

Figure 3. Absorption spectra observed by measuring the signal reflected from both inner surfaces of cell windows. Here all 3 resonances exhibit reduced absorption features with sub-Doppler spectral widths. Curves (1,2) present the full absorption profiles, while the (3,4) are zooms registered at related lower Cs source temperatures.

4. Nonadiabatic transitions between excited states of rubidium atoms

Ultra-narrow cell filled with Rb vapour was excited by two diode lasers at the wavelengths resonant to the transitions between the ground state 5$^2$S$_{1/2}$, the first excited state 5$^2$P$_{3/2}$ and the upper excited state 5$^2$D$_{5/2}$ in sequence. An intense luminescence at the wavelength of 420 nm corresponding to the transition from 6$^2$P$_{3/2}$ state to the ground state was observed. The luminescence intensity at the
A wavelength of 420 nm was found to be proportional to the product of the intensities of both laser sources. In macroscopically thick cells, this luminescence follows after the radiative transition between $5^2\text{D}_{5/2}$ and $6^2\text{P}_{3/2}$ [10] at the wavelength of 5.2 μm. Taking into account that this radiative process takes 707 ns, that the time of flight of an atom between the walls in the ultra-narrow cell is only 1 ns, and that the probability of quenching of electronic excitation upon collision with the solid surface is very near to unity [11], one concludes that the nonadiabatic transition takes place in the course of the atom-wall collision. Fig. 4 displays luminescence intensity versus vapor thickness.

**Figure 4.** The thickness dependence of the intensity of luminescence at 420 nm measured for several densities of rubidium vapor. The data may be approximated by the power low dependence with the exponent of about 2.3. This highly nonlinear dependence on the layer thickness is in accord with the large probability of quenching at the surfaces and the transient character of the excitation process.

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