Spectroscopy of the atom-wall interactions in a nanocell

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Abstract. The optical spectroscopy of rubidium atoms confined to a nanocell was employed to study the quenching and energy transfer processes in the course of atom-wall collisions. The distance between the sapphire windows of the cell varies between 150 and 500 nm forming a vapour wedge. At moderate pressures, the frequency of the atom-wall collisions greatly exceeds the frequency of atom-atom collisions. Hence, the line shapes and intensities recorded in a nanocell provide valuable information about interactions of excited atoms with the surface of the window material.

1. Introduction

The atom-solid surface interactions play an important role in many applications and are of fundamental interest for atomic physics as well as for solid state physics. In this contribution we report on the results of our experimental investigations of the energy transfer processes and quenching of the excitation of rubidium atoms due to collisions with the monocrystalline sapphire surface.

An extremely thin sapphire cell with the diameter of 24 mm and the smoothly changing thickness in the range of 150-500 nm, thus, forming a vapour wedge, was used in order to increase the number of atomic collisions with the sapphire surface. Rubidium vapours were excited by means of two cw diode lasers with the powers of $I_P \sim 20$ mW and $I_D \sim 40$ mW. The wavelengths of the lasers were adjusted to $5S_{1/2}-5P_{3/2}$ and $5P_{3/2}-5D_{5/2}$ transitions, respectively. Figure 1 plots the rubidium levels and the transitions involved in the study.

The pumping rates of the atomic states $5P_{3/2}$ and $5D_{5/2}$ exceeded the relaxation rates of the corresponding levels, $\gamma_P$ and $\gamma_D$. On the other hand, the pumping rates were smaller than the inverse time of the free flight between the cell walls as well as the Doppler width. Under these conditions, a very anisotropic distribution of the excited atoms over the cell volume is established. The atoms, slowly moving in the direction to the walls, interact with the resonant radiation more effectively than those atoms that move faster [1].

The populations of the atomic level, $n_P$ and $n_D$, depend on the pumping intensities $I_P$ and $I_D$ in a specific way: $n_P \sim I_P^{1/2}$ and $I_D \sim n_P I_D^{1/2}$. This dependence is due to the gradual saturation of the atoms with different velocities that correspond to different detunings of the atomic transition relative to the pump [2].
In the experiment we have measured the fluorescence intensities of the rubidium vapour on the above mentioned transitions as well as on the wavelength of 420 nm that corresponds to the transition \(6P_{3/2} - 5S_{1/2}\). The fluorescent light was analyzed by a wide-aperture monochromator and registered by a photo-multiplier tube connected to a lock-in amplifier.

2. Methods and results
In an extremely thin cell that was used in our experiments the fluorescence intensity was substantially reduced due to the effective quenching of the excited atoms on the cell walls. Nevertheless, we were able to measure and to compare the fluorescence intensities on the transitions \(5P_{3/2} - 5S_{1/2}\) and \(5D_{5/2} - 5P_{3/2}\), although their intensities were several times smaller than the corresponding intensity of the pump scattered by the walls. This goal was achieved by a special modulation technique: instead of the pump light modulation, the population of the atomic levels was modulated. For instance, while measuring the fluorescence intensity on \(5S_{1/2} - 5P_{3/2}\) transition, only the intensity of the laser that is resonant to \(5D_{5/2} - 5P_{3/2}\) transition was modulated, and vice versa. A lock-in amplifier was employed to reject the scattered light and detect the weak fluorescence on the studied transitions. In all case dc component of the scattered light was not detected.

The detection part of the experimental set-up consists of a diffraction grating monochromator and a photo-multiplier tube. It was calibrated in a range of 400-800 nm with Xenon discharge lamp.

The main outcome of these measurements is that the fluorescence intensity ratio of the \(5P_{3/2} - 5S_{1/2}\), \(5D_{5/2} - 5P_{3/2}\) and \(6P_{3/2} - 5S_{1/2}\) transitions is appeared to be 500:2:1.

![Figure 1. Rubidium levels and transitions involved in the study. Two diode lasers operate at the wavelengths of 780 nm and 776 nm.](image)

3. Discussion
The first part of the ratio quoted above, 500:2, is in a fair agreement with the estimations of the corresponding levels populations based on the oscillation strengths, lifetimes and light intensities in use. Such agreement supposes that the quenching probabilities of \(5P_{3/2}\) and \(5D_{5/2}\) states are close to...
each other. Note, that even the order of magnitudes of these probabilities is not known. As far as we
know the only one precise measurement of the excited alkali atom quenching rate under collisions
with the sapphire surface was performed with cesium [3]. The probability of the cesium 6P\textsubscript{3/2} excited
state deactivation after collision with the wall was reported to be 0.98±0.02. If we suppose that
the quenching efficiency is of the same order for rubidium atom, we could expect that quenching
probability is close to 100% for both 5P\textsubscript{3/2} and 5D\textsubscript{5/2} states as well. Unexpectedly, the observed
fluorescence intensities of 6P\textsubscript{5/2}–5S\textsubscript{1/2} and 5D\textsubscript{5/2}–5P\textsubscript{3/2} transitions differ only twice. This is difficult to
explain, because spontaneous transition from 5D\textsubscript{5/2} to 5P\textsubscript{3/2} state should occur during the time of flight
between cell walls. For our thin cell at room temperatures this time is about 1 ns, while spontaneous
decay lifetime is around 700 ns. Then, without any other processes, which lead to population of 6P\textsubscript{3/2}
state, each atomic collision with the wall should deactivate both states with probability close to 100%.
As a result the total population of 6P\textsubscript{3/2} state cannot exceed 1/700-th part of the population of 5D\textsubscript{5/2}
state. If we additionally take into account that the 5D\textsubscript{5/2}–5P\textsubscript{3/2} transition is approximately two times
faster than the 6P\textsubscript{3/2}–5S\textsubscript{1/2} transition and that the 6P\textsubscript{3/2} state has also a nonzero probability to decay to a
lower lying 6S\textsubscript{1/2} state, the expected fluorescent intensity of 6P\textsubscript{3/2}–5S\textsubscript{1/2} transition must be 2000 times
weaker than that of 5D\textsubscript{5/2}–5P\textsubscript{3/2} transition. Contrary to that, the measured values differ only by a factor
of two.

Two alternative explanations of the observed effect may be considered. First, extremely small
quenching efficiency of 6P\textsubscript{3/2} state may be the reason. But it is quite improbable, that 6P\textsubscript{3/2} state is
quenched in a substantially different way than 5P\textsubscript{3/2} state. Another possibility is an “incomplete
quenching” of 5D\textsubscript{5/2} state, which implies that during the collisions with the wall the atoms are not
quenched completely, but are transferred into 5P\textsubscript{3/2} state with a high enough probability. In order to
check both alternatives, additional measurements were performed.

We have studied the fluorescence intensity ratio of 5D\textsubscript{5/2}–5P\textsubscript{3/2} and 6P\textsubscript{1/2}–5S\textsubscript{1/2} transitions in its
dependence on the thickness of the cell. It was observed that the fluorescence intensity ratio does not
depend on the thickness. This excludes the first alternative and testifies an effective incomplete
quenching of 5D\textsubscript{5/2} state as a main process in atom-wall interaction. A plausible explanation of this
large efficiency of the population transfer from 5D\textsubscript{5/2} to 6P\textsubscript{1/2} level is that the energy gap between these
levels is small (it corresponds to an IR transition at the wavelength of 5.5 \textmu m), and lies in the range of
the phonon absorption spectrum of the sapphire substrate. Hence, high efficiency of this “incomplete
quenching” may be due to matching between the sapphire phonon spectrum and the energy difference
between the 5D\textsubscript{5/2} and 6P\textsubscript{1/2} states of rubidium [4].

One can expect that the “incomplete quenching” should manifest itself also in other spectroscopic
features. In particular, a distant wing of the fluorescence spectrum of the excited atomic states may be
suppressed or disappear provided the initial state has an opportunity to make a transition to a lower
lying states with high probability.

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