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Optical control of high-density alkali atom vapor in antirelaxation coated cells

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Abstract. In this work, we report on our investigations on LIAD (Light Induced Atomic Desorption) aimed at achieving high density of alkali atoms vapor in a coated cell at room temperature. The experimental results show the possibility to reach a density up to the limit when the medium becomes optically thick by applying highly efficient homogeneous illumination. The photon reabsorption mechanism prevents the precise evaluation of the density by measuring the absorption of a probe laser beam, but there is clear evidence that densities can be achieved higher by two orders of magnitude than the thermodynamic equilibrium value.

1. Introduction

Antirelaxation (AR) coatings are organic films (as, for example, paraffin, PDMS, OTS, SC-77 among others) used in optical absorption cells to maintain the laser-induced polarization of the atoms after collisions with the cell's walls. The long-lived ground state polarization in AR coated cells is a basis for the development of atomic clocks, optical magnetometers, quantum memory, slow light experiments and precision measurements of fundamental symmetries [1, 2].

At thermal equilibrium, the atomic density in a cell containing an alkali metal vapor is defined by the coldest point of the cell. Usually, this is the reservoir (the stem), which is kept some degrees colder to avoid a thin film deposition on the cell's walls, which contributes to making the relaxation properties very poor [3]. The main disadvantage of the thermal control for application in AR coated cells is the deterioration of the AR properties of the coating at high temperatures [4].

Another way to control the alkali metal vapor density in AR coated cells is the light-induced atomic desorption (LIAD) [2]. LIAD is a non-thermal process, whereby atoms are desorbed from the walls of AR coated cells under illumination. In the cases when high atomic densities at low temperatures and/or small dimensions are needed, LIAD is usually applied. Density control by LIAD is considerably faster than the thermal control and does not degrade the AR properties of the coatings over time [5]. Long-term LIAD density stabilization has been reported in a few papers, but due to the influence of the stem, it requires increasing light intensities to sustain the atomic density [6].

In this work we report on our investigations on optical control of the atomic density in an AR coated cell with the aim to achieve a high density at room temperature. The work is another step towards the development of new all-optically controlled and miniaturized vapor cell-based sensors with long-term stability.

2. Experimental setup

The experimental setup is given in figure 1. A PDMS (Polydimethylsiloxane) coated cell (6 cm long and 2.5 cm in diameter) filled with a natural mixture of Rb is used for the measurements. The cell is mounted in a sphere with a high-reflection coating on the inner surface to achieve homogeneous illumination when an internal light source is used [7]. A 60 W 460 nm LED irradiates the sphere from inside. When the desorbing light is kept off, the density of the vapor is defined by the temperature of the stem of the cell (in this case, $T = 24\ ^{\circ}$C). The density of the Rb atoms in the cell is measured by the transmission signal of a 780 nm probe free-running diode laser tuned in resonance with the D2 Rb transition. In order to make the optical pumping negligible, which is significantly enhanced in AR coated cells, the power of the laser light is reduced to 4 µW [8].
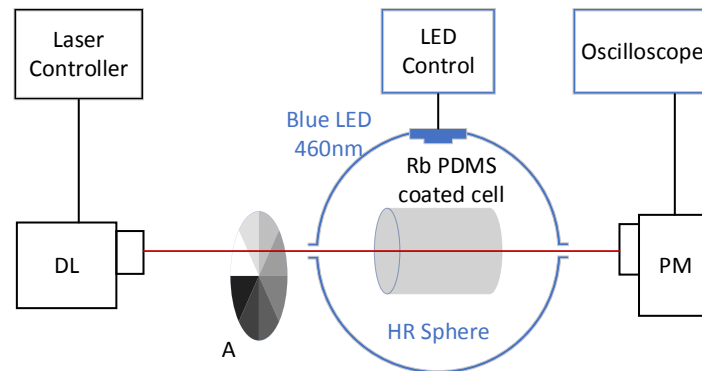
Figure 1. Experimental setup.

(DL – 780 nm diode laser, HR Sphere - high reflectivity sphere, PM - power meter,

A - attenuator).

For an optically thin medium, the increase in Rb density due to the LIAD effect is measured by the change of the frequency-dependent absorption coefficient $\alpha(\nu) = \alpha_\nu$. According to Beer's law, the transmission $T(\nu) = T_\nu$ is:

$$T_\nu = I/I_0 = \exp(-\alpha_\nu L) = \exp(-\sigma_\nu N L), \tag{1}$$

where I_0 is the beam intensity at the entrance of the cell, I, the beam intensity exiting the cell, $\alpha_\nu = \sigma_\nu N$, the absorption coefficient, σ_ν, the atomic absorption cross-section, N, the number density of the atomic vapor, L, the length of the cell and ν, the frequency of the incident light.

When the medium is optically thick, the laser light intensity is reduced through the optical cell and in this case, the average absorption coefficient is measured as:

$$\alpha_\nu = (1 - T_\nu)/L. \tag{2}$$

3. Results and discussion

Figure 2 shows the frequency dependent transmission of Rb D2 line at different intensities of the desorbing 460 nm LED light. The spectrum was obtained by linearly scanning the probe laser current at a low repetition rate by the output of a waveform generator. This corresponds to a frequency scan over several GHz, in such a way that the spectrum reveals four Doppler broadened profiles. In fact, in the transition to the 5P$_{3/2}$ excited state, the ground state hyperfine levels F$_g$=1 and F$_g$=2 for [87]Rb and F$_g$=2 and F$_g$=3 for [85]Rb can be resolved, while the energy distances of the excited hyperfine levels are much smaller than the Doppler width and cannot be distinguished. Due to the optical pumping, the transmission of the absorption lines strongly depends on the repetition rate of the scanning, its direction (from the lower to the higher frequencies or vice versa), and on the intensity of the laser light, though

LIAD reduces the optical pumping by desorbing fresh atoms from the coated walls [9]. Therefore, all parameters are selected so that the influence of the optical pumping is reduced, and the spectrum is very similar to this in an uncoated cell (orange dash-dot line) [10].
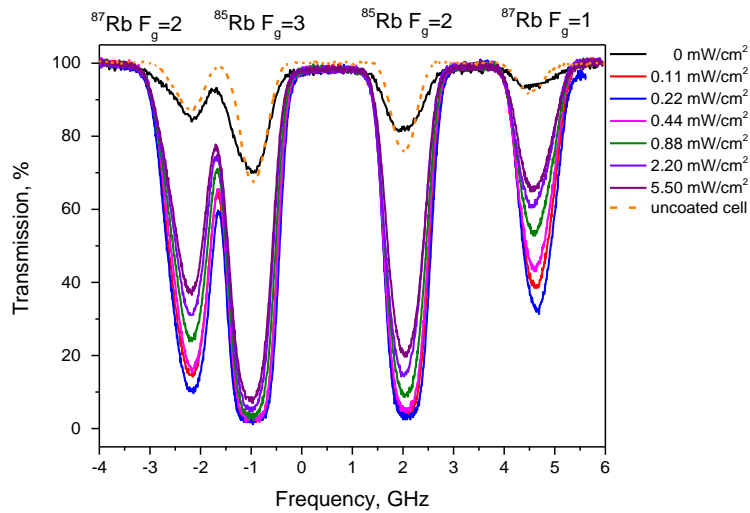
Figure 2. Transmission of the Rb D2 line in PDMS coated cell at different intensities of 460 nm desorbing light.
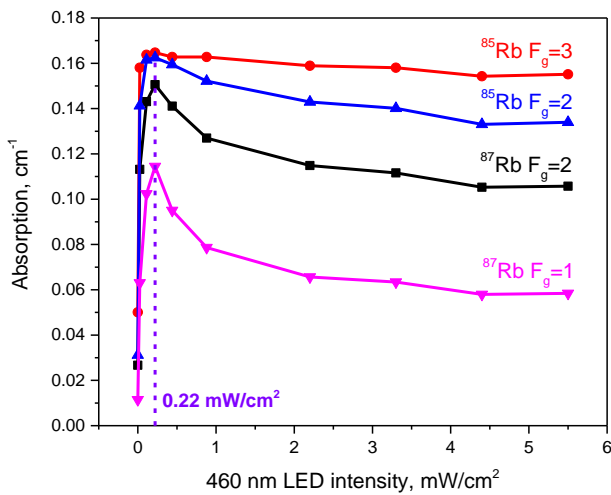
Figure 3. The absorption coefficients vs desorbing light intensity as measured by the transmission spectrum which is shown in figure 2.
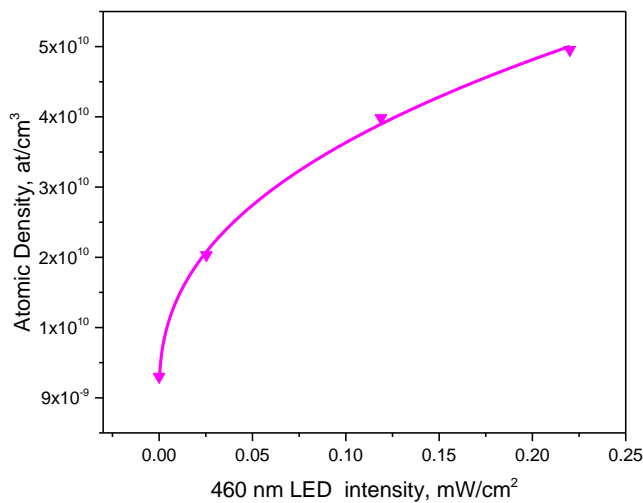
Figure 4. Atomic density is derived from the transmission of ^{87}Rb F_g=1 only to the point when the medium becomes optically thick. The line is only to guide the eyes.

Increasing the intensity of the desorbing illumination at 460 nm leads to enhanced absorption due to the increased density of the desorbed Rb atoms. The transmission profiles are saturated for the F_g=2 and F_g=3 ^{85}Rb at just 0.22 mW/cm^2 as a result of the efficient homogeneous illumination and the vapor becomes optically thick for these two lines. Further increasing the LED intensity leads to decreasing all four Doppler profiles, although the density continues to grow.

The absorption coefficients as deduced from the transmission spectrum are shown in figure 3. At low intensities of the homogeneous LED illumination, the absorption coefficients of the four Doppler broadened transitions increase rapidly and reach their maxima at 0.22 mW/cm^2. The absorption increases up to a point when the medium becomes optically thick on the length scale of the atomic cell size and the process of photon reabsorption becomes significant [11,12]. The threshold atomic density for radiation trapping can be calculated from the following condition:

$$\frac{3}{8\pi} N \lambda^2 D \frac{\gamma_r}{W_d} > 1, \qquad (3)$$

where D is the diameter of the cell, N, the atomic density, γ_r, the radiative decay rate, λ, the transition wavelength. In our case, the evaluated threshold value for radiation trapping is 5×10^{10} atoms/cm^3. This estimation corresponds to the experimental results in figure 4, where the maximum is about 5×10^{10} atoms/cm^3. After surpassing the maximum at 0.22 mW/cm^2 in figure 3, the absorption coefficient decreases slowly with the intensity of illumination.

Long exposure to homogeneous LIAD depletes the coating, not only on the surface layer, but also in the bulk of the organic film. Moreover, all ejected atoms enter the vapor phase in such a way that the reservoir starts to heavily counterbalance the excess respect to the equilibrium value – the so called "reservoir effect" [13]. Solutions of the problem with the influence of the stem are a lockable stem [14] and an internal atomic vapor source [15]. As the adsorbed atoms serve as a reservoir for the LIAD effect the cell needs to "rest" for a long time or to be reheated and the coating replenished with atoms [16]. In fact, a diffusion process is needed to refill all the free sites of the coating, and in the dark and at room temperature the diffusion proceeds very slowly. In cases of open stem cells, this problem might be resolved by a balanced illumination of the stem and the cell wall to avoid accumulation of metal on the walls.
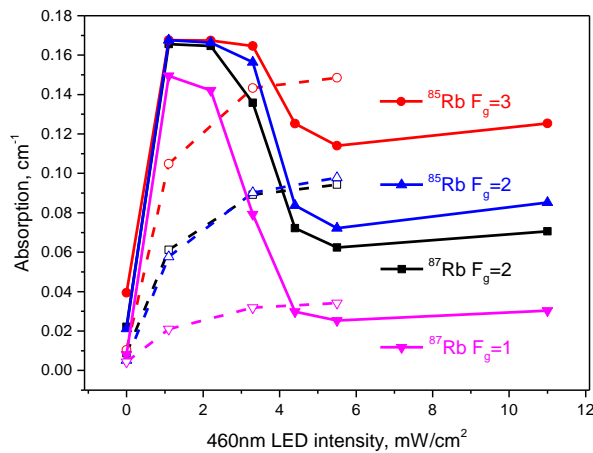
Figure 5. Absorption coefficient as a function of desorbing light intensities for homogeneous (solid lines) and inhomogeneous (dashed lines) illumination when the coating is well saturated with atoms.

Figure 5 shows the absorption coefficient values for homogeneous and inhomogeneous (without the sphere) illumination when the coating is well saturated with atoms. For homogeneous illumination (solid lines), increasing the blue light intensity from 0 to 1.1 mW/cm^2 increases the absorption coefficient. From 1.1 mW/cm^2 to 4.4 mW/cm^2 it saturates and then decreases to a second plateau. Indeed, increasing the illumination from 5.5 mW/cm^2 to 11 mW/cm^2, leads to an increase in the absorption coefficients by approx. 15%. This behavior can be explained based on such high rubidium vapor densities that the photon reabsorption becomes significant inside the laser beam. In our case, the ratio of the laser beam size to the cell diameter being about 0.1, the atomic density can be estimated as being higher than 5×10^{11} atoms/cm^2, which means two orders of magnitude higher than the density defined by the temperature.

At inhomogeneous illumination (dashed lines), the absorption coefficient increases with the desorbing light intensity and simply saturates, but at lower values and in a region where the previous curves have already started to decrease. This can be considered as evidence of radiation trapping at high densities for homogeneous illumination.

4. Conclusions

In this work, the capability was demonstrated of LIAD applied to a PDMS coated cell to achieve atomic densities two orders of magnitude higher than the thermodynamic equilibrium value when a homogeneous illumination of the whole cell surface is performed,. The obtained density is beyond the point when the photon reabsorption becomes a relevant process in the volume of the cell, as well as when photon reabsorption is observed in the volume of the laser beam. This might be useful in applications when one needs to maintain high density at low temperatures to both protect the coating from degradation and to limit the atomic velocities. The drawbacks include a depletion of atoms inside the coating, but this problem might be resolved when balanced illumination of the stem and the cell wall is used. This work could be useful in the future for longtime stable all-optical devices in applications like atomic clocks, atom and molecule cooling and trapping, sensor miniaturization, or other high-density spectroscopy experiments.

Acknowledgments

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