Optical characterization of antirelaxation coatings for photonics applications

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Optical characterization of antirelaxation coatings with regard to their applications in coherent spectroscopy and LIAD experiments is presented. A simple method, based on the recording of the fluorescence intensity of the Rb alkali atoms during resonant light pulse irradiation is used for comparison of the antirelaxation properties of the coatings. The LIAD yield and dynamics are measured by registration of the 780 nm Rb line transmission. The comparison of the parameters of PDMS coatings prepared with two different solutions in ether (PDMS 2% and PDMS 5%) shows that when illuminating with such LED intensity at which the LIAD efficiency is equal in the two cells, the light induced Rb vapor density changes are about an order of magnitude slower in the PDMS 2% cell, and the antirelaxation properties of the two cells are equal.

Keywords: antirelaxation coating; light-induced atomic desorption (LIAD); optical characterization, atom-surface interaction, light-surface interaction

INTRODUCTION

Antirelaxation coatings (ARC) are organic films (as, for example, paraffin, PDMS, OTS, SC-77 etc.) used in optical cells containing alkali metal vapor, which reduce the depolarization of alkali atoms after collisions with the cell's walls [1]. The long-lived ground state polarization is a basis for development of atomic clocks, magnetometers, quantum memory, slow light experiments, and measurements of fundamental precision symmetries. The antirelaxation properties of the coatings are characterized by the number of collisions of a single atom with the walls without spin randomization. A simple method for ARC characterization was proposed in [2], which comprises recording of the time dependence of the fluorescence intensity of alkali atoms during exposure of the cell to resonant radiation pulses.

Light-induced atomic desorption (LIAD) is a non-thermal process in which atoms are desorbed from the walls (coated or uncoated) of a vapor cell under illumination. It is applied mostly for realization of optical atomic dispensers in cases when high atomic densities at low temperature and/or small dimensions are needed – for example, for loading atomic devices such as atomic magnetometers, atomic clocks, magneto-optical traps and their miniaturization [3,4 and references therein]. However, as desorption depends on the atom-surface interaction, it can be applied for optical characterization and manipulation of alkali metal nanoparticles [5], too. For the first time LIAD was reported in polydimethylsiloxane (PDMS) coated cell with Na vapor [6]. Since then LIAD in PDMS coated cells has been observed with Rb, Cs, K and radioactive Fr alkali atoms as well.

Polydimethylsiloxane CH₃[Si(CH₃)₂O]_nSi(CH₃)₃ is a polymer, which includes inorganic and organic parts, the backbone consists of silicon and oxygen atoms, where each silicon has attached two organic groups to it. PDMS is viscoelastic and has a good stability after dehydration. The glass transition temperature is 144 K and the melting temperature is 232 K. In our experiments it is close to the room temperature. Although the structure of PDMS is suitable for alkali metal atoms adsorption, it causes problems such as bubble formation and sample evaporation [7].

The AFM measurement of coatings prepared with different concentrations of PDMS in ether showed that the surface roughness increases from 20 nm to 50 nm as the PDMS concentration is

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raised from 2% to 5%, and the thickness of the PDMS film increases with the concentration [8]. The X-ray photoelectron spectroscopy investigations of the glass surface condition of alkali metal vapor cell have shown that Rb atoms diffuse into the Pyrex glass [9].

In this work we report on methods for optical characterization of ARC from point of view of their application in coherent spectroscopy and LIAD experiments. The yield of LIAD and its dynamics, and the number of atom-wall collisions without spin randomization in two PDMS coated cells prepared with 2% (cell PDMS2) and 5% (cell PDMS5) concentration of PDMS in ether are compared.

EXPERIMENTAL

LIAD and its dynamics are characterized by the vapor density and its changes in time n(t). For an optical medium, the change of the Rb density due to the LIAD effect is measured by the absorption coefficient κ_{ω} of Rb vapor. According to the Beer's law the transmission is

$$T_{\omega} = I/I_0 = \exp(-\kappa_{\omega}L) = \exp(-\sigma_{\omega}NL), \quad (1)$$

where I_0 is the 780 nm laser power entering the vapor sample; I, the transmitted power; κ_{ω} , the frequency dependent absorption coefficient; σ_{ω} , the non-saturated, frequency dependent absorption cross-section; N, the atomic density and L, the cell length.



Fig. 1. Experimental setup for LIAD characterization. (DL – 780 nm diode laser; PM – powermeter; PD – photodiode; Att - attenuator; OSC – oscilloscope).

The experimental setup for LIAD and its dynamics characterization is given in Fig. 1. It is described in details in [10, 11]. A 780 nm free running diode laser (DL) is used for measuring the absorption coefficient of the D2 Rb resonance line. The D2 absorption line has 4 Doppler broadened sets of hfs transitions, related to the ground state $F_g=1$ and $F_g=2$ of the 87 a.m.u. isotope and $F_g=2$ and $F_g=3$ of the 85 a.m.u. isotope of the natural mixture. All LIAD measurements refer to the ⁸⁵Rb $F_g=2$ set of lines. A special sphere with diffusion reflectance inner surface is used to improve the efficiency of the blue light illumination from a 460 nm light emitting diode (LED) [11, 12].

All measurements are performed at temperature of 25° C. In order to minimize the optical pumping and deduce the corresponding absorption features, the transmission spectra are measured at relatively low 780 nm light intensity (around 3-4 μ W).

The PDMS2 and PDMS5 cells are with radius $R_c=1.3$ cm and lengths 6.0 cm and 4.7 cm respectively. This results in only 5% difference of the surface to volume ratio S/V and the LIAD yield for the two cells.

The dynamics of desorption and adsorption in different coated cells are compared following the model proposed in [3]. The time constants are determined by fitting the experimental data for the time evolution of the atomic density in the volume of the cell n(t) with the following equation:

$$n(t) = \begin{cases} n_0 + N_\tau \left(1 - e^{-(t - t_0)/\tau_1} \right) e^{-(t - t_0)/\tau_2}, t_0 \le t \le t_{off} \\ n_0 + \left[n_{off} - n_0 \right] e^{-(t - t_{off})/\tau_3}, \quad t > t_{off} \end{cases}$$
(2)

where t_0 and t_{off} are the times when the desorption light is switched on and off; n_0 , the density before illumination; $n_{off} = n(t_{off})$; τ_1 , characterizes the exponential density growth after the cell is illuminated; τ_2 , the density decrease when the illuminating light is still on; τ_3 , the density decrease when the light is off; $\Delta n = n_{max} - n_0$ and $N_{\tau} = \Delta n (1 + \tau_1/\tau_2) (1 + \tau_2/\tau_1)^{\tau_1/\tau_2}$.

As the LIAD density increase depends on the time of illumination, all measurements are performed with equal time intervals, more specifically by 75 sec during which the cell is illuminated, followed by 75 sec without illumination.

A simple model for characterization of antirelaxation coating of optical cells is proposed in [2]. The method is based on the registration of the temporal dependence of the fluorescence intensity from irradiated alkali atoms by pulsed light at resonance. It was found that in order to find the number of atom-wall collisions without spin randomization N, it is enough to have the value of the fluorescence decay rate β and the ratio between the intensities of the fluorescence at the beginning I_i and at the end of the decay I_f and the characteristic time between atom collisions with the cell's walls $\tau_c=R_c/2v_T=27 \ \mu s$, where v_T is the atom's thermal velocity

$$1/N = \beta \tau_c I_f / I_i \tag{3}$$



Fig. 2. Experimental setup for ARC characterization. (DL – diode laser; PM – powermeter; PD – photodiode;Ch - chopper; OSC – oscilloscope)

In Fig. 2 the experimental setup for ARC characterization is given. The absorption and the fluorescence of the 780 nm diode laser light, interrupted by a chopper at 5 Hz, are detected by a powermeter (PM) and a photodiode (PD). The measurements are performed on the the ⁸⁵Rb $F_g=3$ set of hfs lines at 40 μ W 780 nm diode laser power.

RESULTS AND DISCUSSION

In Fig. 3 the dynamics of desorption and adsorption in PDMS2 and PDMS5 cells when the blue light is switched on and off are compared. The measurements are at low intensity (0.7 mW/cm²) where the heating from the LED is negligible. The LIAD yield depends on the LED intensity. At low LED intensities (less than 2.5 mW/cm²) the LIAD yield in the PDMS2 and PDMS5 cells is almost equal, while at intensities higher than 2.5 mW/cm², the yield is higher in the PDMS2 cell [12].

The shape of the transmitted light response is quite different for the two cells and in PDMS5 cell the changes in the Rb vapor density are about an order faster than in PDMS2 cell. The dynamic response in PDMS2 cell is like in the "weak desorbing light regime", when the light induces negligible change in the Rb atom density near the coating surface. The dynamic response in PDMS5 cell is like at the "high desorbing light intensity" regime, when all atoms in the region close to the surface are desorbed into the cell volume [13,14].



Fig. 3. Comparison of LIAD dynamics in PDMS2 and PDMS5 cells: the measured normalized transmission at 0.7 mW/cm² LED intensity.

An explanation of the difference in the adsorption and desorption rates (Fig. 3) in PDMS2 and PDMS5 could be the different probability for alkali atoms to find adsorption sites in substrates with different volume density and surface morphology.

Comparing the dynamics of different cells the influence of the stem has to be considered, too. Difference of one order was measured with open and closed stem in Rb paraffin coated cell [3]. In our case, the comparison of the PDMS2 and PDMS5 cells shows that there is more metal in the PDMS5 cell stem, which can influence the dynamics of the processes.





In Fig. 4 the experimental fluorescence decay of Rb atoms in the PDMS2 (black) and PDMS5 (red)

coated cells, when irradiated by 50 ms square 780 nm laser light pulse are compared. The ratios $I_{f}/I_i=0.6$, $\tau_c=27\mu s$, and the fluorescence decay rates are equal for the two cells as it is seen from Fig. 4. Following Eq. 3 the number of atom-wall collisions without randomization of the atom spin in PDMS2 and PDMS5 cells (Fig. 4) differ of the order of 3.5%, which is less than the experimental error: 1602(72) in PDMS5 and 1657(266) in PDMS2 cell.

CONCLUSIONS

The comparison of the absorption in two ARC cells, prepared with different solutions of PDMS in ether, from point of view of their application in LIAD experiments, have shown that in PDMS2 the desorption and adsorption rates are about an order lower than in PDMS5, which can be explained by the different morphology of the surface, thickness of the coating, and volume density.

The application of the simple method for optical characterization of ARC for coherent application by registration of the temporal dependence of the intensity of the fluorescence from irradiated alkali atoms by pulsed light at resonance [2] has shown that the number of the atom-wall collisions without a spin depolarization is comparable in the two PDMS cells.

The performed investigation and the results are interesting for the better understanding of lightatom-surface interactions, for development of new optical elements for application in photonics, LIAD-loaded atomic devices and their miniaturization, and new methods for surface and coating diagnostics and monitoring.

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ОПТИЧНО ХАРАКТЕРИЗИРАНЕ НА АНТИРЕЛАКСАЦИОННИ ПОКРИТИЯ ЗА ПРИЛОЖЕНИЯ ВЪВ ФОТОНИКАТА

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(Резюме)

Представени са изследвания за оптично характеризиране на антирелаксационни покрития от гледна точка на приложението им в кохерентната спектроскопия и за светлинно индуцирана десорбция на атоми (СИАД). За анализ на антирелаксационните свойства на покритията е използван прост метод, при който се регистрира интензитетът на флуоресценцията на Rb атоми при облъчване с резонансна светлина. Скоростта и ефективността на СИАД са измерени чрез регистриране на преминалата светлина на линията на Rb 780 нм. Сравняването на параметрите на PDMS покритията направени с два различни разтвора в етер (PDMS 2% и PDMS 5%) показва, че при интензитет на облъчване, при който ефективността на СИАД е равна, процесите на адсорбция и десорбция в PDMS 2% клетката са приблизително 1 порядък по-бавни, а антирелаксационните свойства са еднакви в двете клетки.