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Spin randomization of light-induced desorbed Rb atoms

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Abstract. We present the first experimental observation of atomic spin randomization of Rb atoms released by light-induced atomic desorption (LIAD). A natural mixture of Rb atoms contained in paraffin and PDMS coated glass cells is irradiated by a free-running diode laser light tuned to the Rb D_2 resonance line. The transmission spectrum of the Rb vapor is thus modified and shows a strong enhancement of the hyperfine optical pumping as the light intensity is increased and the laser-frequency scanning rate is decreased. The D_2 line spectra are compared for two cases: without and with illumination of the walls of the cell by a UV lamp centered around the wavelength of 404 nm. A simple theoretical model based on the solution of the rate balance equations is introduced in order to analyze the experimental results.

1. Introduction

In order to suppress the spin randomization of vapor-phase atoms during their collisions with the cell walls, a coating procedure is frequently used [1,2]. Among the most common coatings, polysiloxane or paraffin films show particularly low atomic adsorption energies [1-3]. Furthermore, it has been found that the antirelaxation coating adsorbs a significant number of alkali atoms, which can be easily released by any low intensity, non-resonant light beam, as a consequence of a process called light induced atomic desorption (LIAD). Most of the papers devoted to this phenomenon mainly concern the vapor density dynamics in coated cells. Even if some works have indirectly addressed the effects of external illumination on the coatings' behavior [4,5], the question remains open concerning the details of the influence of LIAD effect on the key property of the coatings, namely their ability to significantly reduce the spin relaxation of atoms desorbed and introduced in the cell volume [1].

In this communication we present the results of studying the hyperfine optical pumping depending on the light intensity and the laser-frequency scanning rate, which made it possible to observe experimentally for the first the spin randomization of Rb atoms released by LIAD. A theoretical model

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based on the solution of the rate balance equations was developed in order to analyze the experimental results.

2. Experimental results

A natural mixture of Rb atoms contained in a paraffin or a PDMS coated pyrex cell is irradiated by a free-running diode laser light tuned to the Rb D_2 line ($\lambda = 780{,}24$ nm) and scanned around it by applying a triangular waveform to the input current. The transmission spectrum of Rb is measured for different laser intensities as well as for various laser-frequency scanning rates. The D_2 line spectra are compared for: (i) paraffin and PDMS coated cells and (ii) without or with illumination of the walls of the cell by a UV lamp at a wavelength around 404 nm and a bandwidth of 20 nm (figure 1). All experiments are performed at room temperature. The laser beam is split in two parts: the first beam is expanded and directed to the coated cell, while the second beam passes through an uncoated vacuum cell used as a reference. Linearly or circularly polarized light is used.

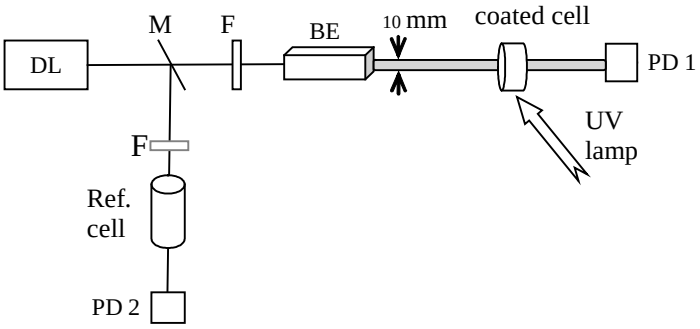

Figure 1. Experimental set up: DL - diode laser, M – mirror, F – neutral density filter, BE – beam expander, PD 1 and PD 2 – optical detectors.

In figure 2, the transmission spectra of both Rb isotopes are shown for a paraffin coated cell at a scanning frequency of 10 Hz and in both scanning directions. All measurements presented are performed by recording simultaneously the transmission signals from the reference and the coated cell. It is seen that the spectrum of the reference cell does not depend on the direction of the laser frequency scan.

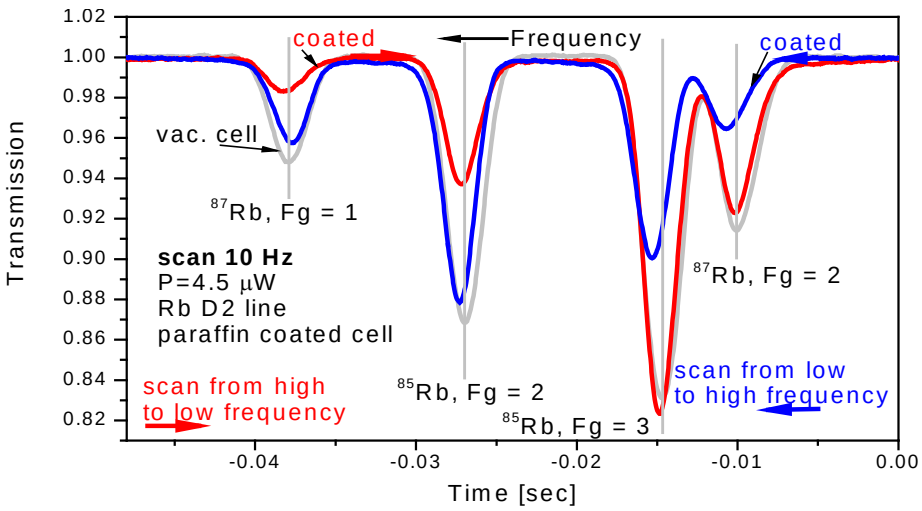

Figure 2. Transmission spectra of the D_2 line of Rb, for linear polarization, low laser power and laser-frequency scanning rate of 10 Hz (grey trace) – the reference spectrum of Rb atoms contained in an evacuated glass cell; (**blue** trace) – the spectrum of Rb in a paraffin-coated cell at increasing laser frequency; (**red** trace) – the spectrum of Rb at decreasing laser frequency; the frequency difference between the $F_g = 2$ and $F_g = 3$ levels (^{85}Rb) is 3036 MHz. The difference in the transmission spectra for frequency scanning in the two opposite directions is clearly seen in each line profile.

In the case of the coated cell, however, the experimental study shows that the scanning direction strongly influences the transmission of each absorption line. For example, in the case of ^{85}Rb, the frequency scan over the $F_g = 2$ line produces atomic accumulation on the $F_g = 3$ level, which is measured while scanning the frequency over the $F_g = 3$ set of transitions (figure 2, **red** trace). In the opposite direction (figure 2, **blue** trace), however, the absorption of the $F_g = 2$ line is larger than that of the $F_g = 3$ line. Reducing the laser frequency scanning rate results in a stronger optical pumping of the ground state hyperfine levels not interacting with the laser light. Due to this process, in addition to the difference in the transmission amplitudes for the blue and red spectra, a well pronounced shift of the transmission profile maximum occurs for each line (figure 3). The shift of the profile maximum depends on the laser frequency scanning direction, as recently reported in ref. [6]. It is attributed to the competition between the hyperfine optical pumping that depletes the atomic population of one ground level through the laser light excitation, followed by decay to the other ground level and the absorption enhancement during the scan when the laser frequency approaches the center of the absorption line. Thus, the maximum of the absorption profile is formed before the center of the hyperfine transition, i.e. the absorption maximum shifts to the starting point of the frequency scan.

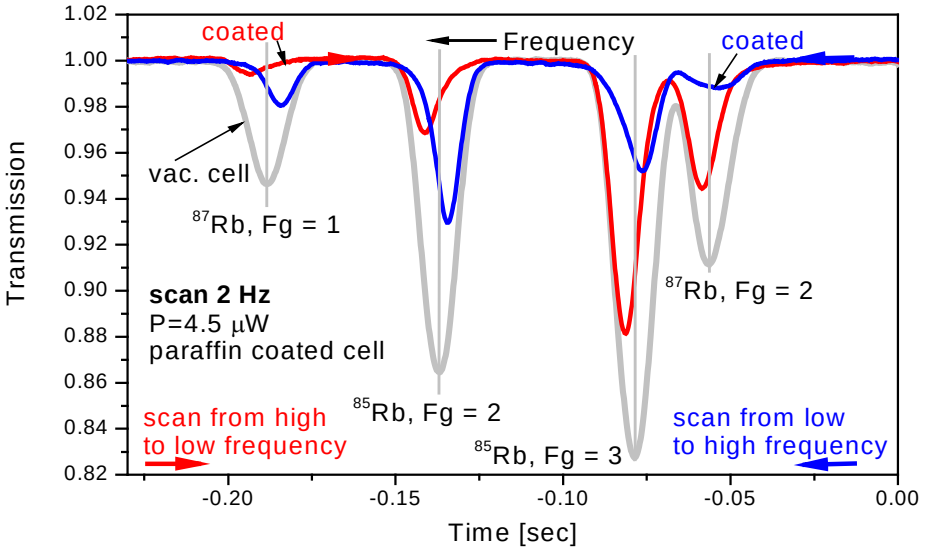

Figure 3. Transmission spectra for linearly polarized light and lower laser-frequency scanning of 2 Hz. (grey trace) – the reference spectrum of Rb atoms contained in an evacuated cell; (**blue** trace) – the spectrum of Rb in a paraffin coated cell at increasing laser frequency; (**red** trace) – the spectrum of Rb in a paraffin coated cell for frequency scanning in opposite direction.

Our experimental investigation shows that both types of coated cells (paraffin and PDMS) exhibit a significant hyperfine optical pumping (figure 4). Thus, the first irradiated ground state level of each Rb isotope is strongly depleted and its absorption is almost cancelled (figure 4, **red** line). The fact that absorption from the level interacting with the light vanishes represents a proof of the strong orientation of the atomic spins caused by the laser light. Under such experimental condition, it is important to know if the atoms released by the LIAD effect will also be oriented or their spins will suffer some randomization of the orientation due to the time spent within the cell wall coating.

Our experiment shows that the flow of LIAD atoms results in a reduction of the hyperfine optical pumping. Such a reduction of the Rb atoms orientation in the cell volume is attributed to the random spins distribution of the atoms desorbed from the cell wall. In the case of LIAD involvement, the measured shift of the absorptions profile maximum is smaller, and both the absorption ratio of the ^{85}Rb ($F_g = 2$) set of transitions to the ^{85}Rb ($F_g = 3$) set and that of the ^{87}Rb ($F_g = 1$) set of transitions to the

^{87}Rb ($F_g = 2$) set change. The spin randomizing of atoms desorbed by LIAD is illustrated for the PDMS coated cell that shows a stronger LIAD effect than the paraffin coated one.

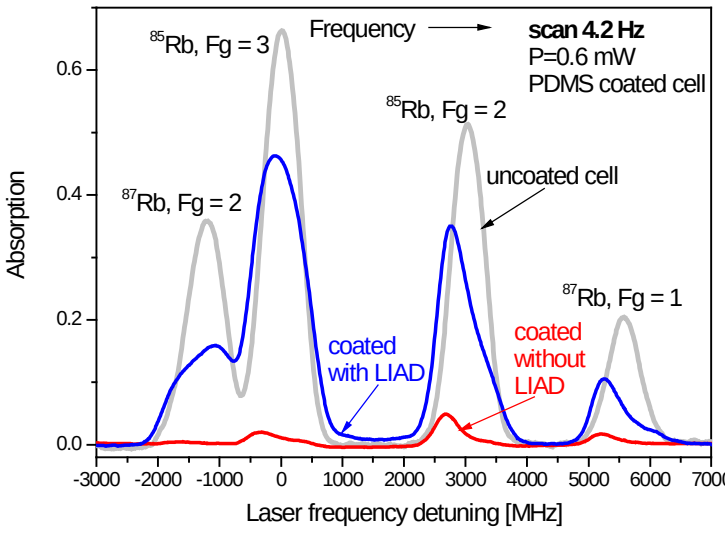

Figure 4. Absorption spectra for circular polarization and without/with modification by the LIAD effect; (grey trace) – reference spectrum of Rb atoms; (**red** trace) – the spectrum of Rb in the PDMS coated cell without LIAD effect, exhibiting strong hyperfine optical pumping: (i) the population of the $F_g = 2$ level of ^{87}Rb is almost depleted and the absorption of the $F_g = 3$ level of ^{85}Rb is strongly reduced; (**blue** trace) – the spectrum of Rb in the PDMS coated cell, in the case of a strong LIAD effect; the enhancement of the absorption at each line due to LIAD is clearly seen.

3. Theoretical model and discussion

The theoretical model presented is based on a system of rate balance equations describing the accumulation and loss of atomic population from energy levels and is used to analyze the optical pumping processes, considering for simplicity only the D_2 line of one isotope (^{85}Rb). A slightly modified energy levels scheme is shown in figure 5. The two sets of hyperfine transitions, separated by 3036 MHz, are depicted in such a way to emphasize the scanning frequency direction as realized in the experimental approach. The two systems are connected through spontaneous emission from excited levels to a ground level not interacting with the laser light, namely the open transition $F_g = 3 \rightarrow F_e = 2', 3'$ and $F_g = 2 \rightarrow F_e = 2, 3$. Note that the $F_e = 2', 3'$ and $F_e = 2, 3$ are the same excited levels.

The system of rate balance equations describes the evolution of the energy levels' atomic population as a result of the spontaneous and stimulated transitions [7,8]. The system of rate equations is formulated taking the laser frequency as a variable and is solved for the steady-state case as follows:

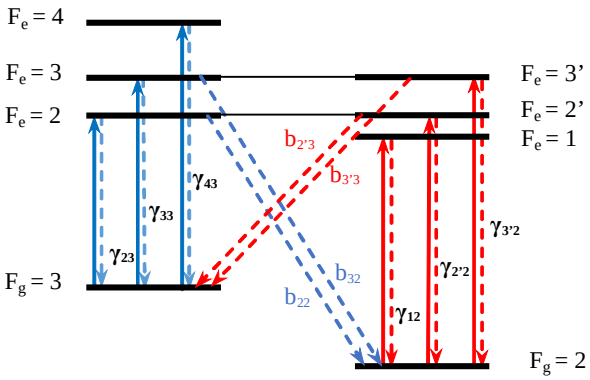

Figure 5. Energy level diagram of ^{85}Rb on D_2 line, the hyperfine ground state splitting is 3036 MHz.

$$\frac{\partial N\{e_2\}}{\partial t} = \left(N\{g_3\} - N\{e_2\}\right) * \sigma_{32} - \gamma_{23} * N\{e_2\} - b_{22} * N\{e_2\}, \quad (1)$$

$$\frac{\partial N\{e_3\}}{\partial t} = \left(N\{g_3\} - N\{e_3\}\right) * \sigma_{33} - \gamma_{33} * N\{e_3\} - b_{32} * N\{e_3\}, \quad (2)$$

$$\frac{\partial N\{e_4\}}{\partial t} = \left(N\{g_3\} - N\{e_4\}\right) * \sigma_{34} - \gamma_{43} * N\{e_4\}, \tag{3}$$

$$\begin{aligned}\frac{\partial N\{g_3\}}{\partial t} = &\left(N\{e_2\} - N\{g_3\}\right) * \sigma_{32} + \gamma_{23} * N\{e_2\} + \left(N\{e_3\} - N\{g_3\}\right) * \sigma_{33} + \gamma_{33} * N\{e_3\} + \\ &+ \left(N\{e_4\} - N\{g_3\}\right) * \sigma_{34} + \gamma_{43} * N\{e_4\} + b_{2'3} * N\{e_{2'}\} + b_{3'3} * N\{e_{3'}\} + \\ &+ \left[n_0 G(\upsilon) - N\{g_3\}\right] * \Gamma\end{aligned}, \tag{4}$$

$$\frac{\partial N\{e_1\}}{\partial t} = \left(N\{g_2\} - N\{e_1\}\right) * \sigma_{21} - \gamma_{12} * N\{e_1\}, \tag{5}$$

$$\frac{\partial N\{e_{2'}\}}{\partial t} = \left(N\{g_2\} - N\{e_{2'}\}\right) * \sigma_{22'} - \gamma_{2'2} * N\{e_{2'}\} - b_{2'3} * N\{e_{2'}\}, \tag{6}$$

$$\frac{\partial N\{e_{3'}\}}{\partial t} = \left(N\{g_2\} - N\{e_{3'}\}\right) * \sigma_{23'} - \gamma_{3'2} * N\{e_{3'}\} - b_{3'3} * N\{e_{3'}\}, \tag{7}$$

$$\begin{aligned}\frac{\partial N\{g_2\}}{\partial t} = &\left(N\{e_1\} - N\{g_2\}\right) * \sigma_{21} + \gamma_{12} * N\{e_1\} + \left(N\{e_{2'}\} - N\{g_2\}\right) * \sigma_{22'} + \gamma_{2'2} * N\{e_{2'}\} + \\ &+ \left(N\{e_{3'}\} - N\{g_2\}\right) * \sigma_{23'} + \gamma_{3'2} * N\{e_{3'}\} + b_{22} * N\{e_2\} + b_{32} * N\{e_3\} + \\ &+ \left[n_0 G(\upsilon') - N\{g_2\}\right] * \Gamma\end{aligned}. \tag{8}$$

In the rate equations system, the terms σ_{ij} describe the cross-sections of optical transition and light-induced emission and absorption, while γ_{ij} and b_{ij} represent the spontaneous decay rates from the excited state to the ground state interacting and non-interacting with the laser field, respectively [7-9]. The last terms in the equations for the ground levels introduce the non-radiative population relaxation Γ; in a coated cell such a process is caused by collisions of atoms with the coating. Γ assumes different values depending on the type of coating and the magnitude of spin randomization after atoms collide with the coated wall of the cell. In our case, we set Γ = 10 MHz, including velocity-changing collisions of Rb atoms with the cell wall without spin randomization [6].

We assume that initially, before irradiation (I = 0), the population densities of both ground states are equal and the excited states are not populated: $N\{g_3\} = n_0 G(\upsilon)$, $N\{g_2\} = n_0 G(\upsilon')$, where $G(\upsilon)$ and $G(\upsilon')$ are the Doppler profiles with $\Delta\upsilon_D$ = 400 MHz. The spontaneous decay rate is taken as γ = 60 MHz, involving also the spectral width of the free-running diode. The saturation parameter is assumed to be s = 50, as it includes also the hyperfine transition saturation due to the optical pumping.

In the steady state, the analytical solution of the system of rate-balance equations in this regime yields the following expressions:

$$N\{g_3\} = n_0 \frac{B\left[G(\upsilon) + G(\upsilon')\right] + \Gamma * G(\upsilon)}{A + B + \Gamma} \quad \text{and} \quad N\{g_2\} = n_0 \frac{A\left[G(\upsilon) + G(\upsilon')\right] + \Gamma * G(\upsilon')}{A + B + \Gamma}, \tag{9}$$

where A and B are $$A = \frac{0.42\gamma * \sigma_{23}}{\sigma_{23} + 0.54\gamma} + \frac{0.34\gamma * \sigma_{33}}{\sigma_{33} + 0.77\gamma} \quad \text{and} \quad B = \frac{0.12\gamma * \sigma_{22'}}{\sigma_{22'} + 0.55\gamma} + \frac{0.43\gamma * \sigma_{23'}}{\sigma_{23'} + 0.78\gamma}. \tag{10}$$

The model is illustrated by its first result describing the ground levels populations, at a very low laser-light intensity and in the strongly saturated regime. For low saturation, the theoretical result is in agreement with the experimental spectrum obtained in an uncoated vacuum cell where the optical pumping effect is very small (figure 6). At high saturation, the theoretical modelling shows a difference between the populations of the ground levels due to the hyperfine optical pumping.

In order to simulate the atomic spectra's dependence on the scanning frequency direction, two types of calculations are performed: (i) starting from the $F_g = 3$ set of transition and going to the $F_g = 2$ set (figure 6, **blue** curve) and (ii) vise versa, i.e. starting from the $F_g = 2$ set of transitions (figure 6, **red** curve). The calculations are in a qualitative agreement with the experimental results, showing that the maximum population of each ground state level depends on the initial laser frequency. As in the experiment, different shifts of the atomic population profile maximum are demonstrated depending on the beginning of the calculation. Again, the results of the model are in a qualitative agreement with the experimentally observed spectra. Note that the dip in the atomic population of the $F_g = 3$ level (figure 6, **blue** curve) is not observed experimentally, which can be attributed to an additional broadening of the hyperfine transitions in the coated cell due to velocity-changing collisions of atoms with the cell walls taking place without disorientation of the atomic spin. This effect is not included in our theoretical description.

As a next step, the model proposed will be developed further in order to explain other peculiarities of the experimental spectra, particularly the spin randomization of the atoms desorbed by LIAD.

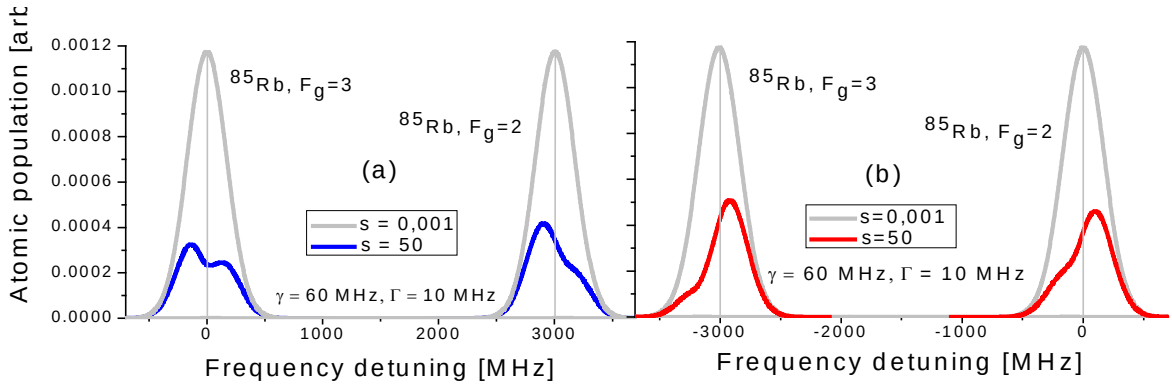

Figure 6. Theoretical atomic population of the $F_g = 3$ and $F_g = 2$ levels, for low and high saturation of the hyperfine transitions. (grey trace) – atomic population at very low saturation intensity; **(blue** trace) – atomic population at high saturation intensity and starting calculation from the Fg = 3 level; (**red** -trace) – atomic population at high saturation intensity and starting calculation from the $F_g = 2$ level. Now the $F_g = 2/F_g = 3$ population ratio is opposite to this of the previous case. Note also the shift of the atomic population profile maximum of the respective set of transitions at high saturation compared to the low saturation.

4. Conclusions

Our study shows that, in both types of coated cells, the Rb atoms undergo a significant hyperfine optical pumping in the saturated regime. The ^{87}Rb ($F_g = 1$) and ^{85}Rb ($F_g = 2$) ground state levels can be almost completely depleted. The maxima of the spectral profiles of the four absorption lines are frequency-shifted from the respective reference lines that do not suffer optical pumping. No significant difference is obtained between irradiation with linear and circular polarizations The absorption maximum shift observed for Rb atoms confirms the one obtained in the very recent work [6] for the case of Cs atoms.

Based on the effect of optical pumping, we demonstrate experimentally that the flow of fresh LIAD atoms results in a reduction in the atomic spin orientation due to the random spin distribution of the desorbed atoms. In the case of LIAD involvement, the measured shift of absorptions profile is smaller,

and the absorption ratio of the ^{85}Rb ($F_g = 2$) set of transitions to the ^{85}Rb ($F_g = 3$) set changes approaching the result obtained in an evacuated cell.

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