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Citation to Publisher Ulanski, Emily & Wu, Zhen. (2011). Measurement of dwell times of spin polarized rubidium atoms on octadecyltrichlorosilane- and paraffin-coated surfaces. *Applied Physics Letters* 98(201115), 1-3. <http://dx.doi.org/10.1063/1.3591172>.

Citation to this Version: Ulanski, Emily & Wu, Zhen. (2011). Measurement of dwell times of spin polarized rubidium atoms on octadecyltrichlorosilane- and paraffin-coated surfaces. *Applied Physics Letters* 98(201115), 1-3. Retrieved from [doi:10.7282/T3D220P7](https://doi.org/10.7282/T3D220P7).

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Measurement of dwell times of spin polarized rubidium atoms on octadecyltrichlorosilane- and paraffin-coated surfaces

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(Received 25 January 2011; accepted 26 April 2011; published online 20 May 2011)

We report the measurement of dwell times of spin polarized Rb atoms on octadecyltrichlorosilane (OTS)- and paraffin-coated surfaces. We find that at a cell temperature of 72 °C the dwell times for OTS- and paraffin-coated surfaces are $0.9 \pm 0.1 \mu\text{s}$ and $1.8 \pm 0.2 \mu\text{s}$, respectively. Since the relaxation probability on paraffin is almost one order of magnitude smaller than that on OTS, the longer dwell time for paraffin indicates that the average strength of the interactions experienced by Rb atoms while they are inside paraffin is much weaker than while they are inside OTS. © 2011 American Institute of Physics. [doi:10.1063/1.3591172]

Long spin relaxation time of alkali atoms is important in many areas of atomic physics. One of the main spin relaxation mechanisms is surface interactions. Antirelaxation coatings have been used to reduce wall relaxation in atomic clocks,¹ magnetometers,^{2–4} fundamental symmetry studies,⁵ magneto-optical traps,^{6,7} electromagnetically induced transparency,⁸ slow light,⁸ spin squeezing,⁹ long-lived entanglement,¹⁰ and quantum memory.¹¹

The performance of antirelaxation coatings varies widely. For example, paraffin-coated surfaces can support about 10 000 polarization-preserving collisions with Rb atoms¹² whereas a spin polarized Rb atom can, on average, collide only about 1000 times with octadecyltrichlorosilane (OTS)-coated cell surfaces before it relaxes.^{13,14} Recently it was reported that an alkene-based coating $\text{CH}_2\text{CH}(\text{CH}_2)_{16}\text{CH}_2$ can support almost a million spin-preserving collisions.¹⁵ In view of the importance of antirelaxation coatings, many studies using different techniques have been undertaken to try to understand the antirelaxation property of various coating materials.^{7,14,16–19} Nonetheless, the antirelaxation property of the coatings is still not fully understood. The quality of the coatings in terms of preserving spin polarization is highly variable, even when they are made using the same procedure.¹⁴

The quality of antirelaxation coatings depends on the average dwell time τ_s of a spin polarized alkali atom, i.e., how long it stays in the coating without being relaxed, and on the average strength of the interactions experienced by the alkali atom while it is inside the coating. We note that τ_s can be significantly longer than the coherent interaction time if trapping occurs inside the coating.^{20,21} In this letter we report the direct measurement of τ_s of a spin polarized alkali atom (Rb) on the two most commonly used antirelaxation coatings, paraffin and OTS. Our results help answer the question whether the superior antirelaxation property of paraffin coating is due to a very short τ_s or because the average strength of the interactions experienced by Rb atoms while they are inside the coating is very weak or both. We find that at a cell temperature of 72 °C the dwell times of a spin polarized Rb atom on OTS and paraffin are $0.9 \pm 0.1 \mu\text{s}$ and $1.8 \pm 0.2 \mu\text{s}$, respectively.

Our experimental method is similar to the one used in Ref. 21. We use evanescent waves to optically pump and

probe the Rb atoms in the vicinity ($\sim 10^{-4}$ cm) of cell surfaces. When a Rb atom is in the evanescent pump beam, its Larmor frequency is shifted (light shift). The observed Larmor frequency depends on the average dwell time τ_s of a spin polarized Rb atom and the average time τ_b it stays in the bulk between two consecutive wall collisions, where the latter can be varied by adjusting the length of the cell. We measure the Larmor frequencies corresponding to σ^+ and σ^- evanescent pump beams for a number of different cell lengths. We show that the inverse of the difference of these Larmor frequencies depends linearly on the cell length, from which one obtains the dwell time τ_s of Rb atoms.

Experiments are performed inside two layers of magnetic shielding (Fig. 1). Three orthogonal pairs of Helmholtz coils inside the shielding cancel the residual field and provide a homogeneous magnetic field along the positive x direction. A cylindrical Pyrex glass cell of one inch in diameter contains isotopically enriched Rb (98.3 at. % ^{87}Rb) in the stem and nitrogen buffer gas of 5 Torr. The pressure is measured at 25 °C. A moveable glass prism is inside the cell and acts as the adjustable back wall of the cell, thus the effective cell length (L) can be varied from 60 to 700 μm by tapping the stage on which the cell sits. A HeNe laser is used to measure the cell thickness by retroreflection with an accuracy of 20 μm .²² The inner walls of the cell and moveable prism are coated with OTS or paraffin. The coating procedure for OTS is similar to that in Ref. 23 and that for paraffin is similar to that in Refs. 24 and 25 with a baking time of

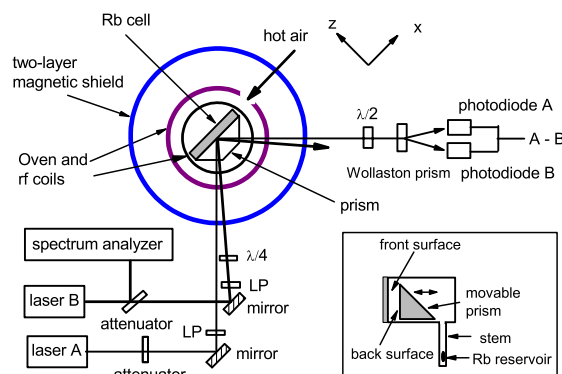


FIG. 1. (Color online) Experimental setup. The pump laser is circularly polarized using a Glan–Thompson linear polarizer (LP) and a quarter-wave plate ($\lambda/4$). The probe laser is s -polarized using a Glan–Thompson LP. Inset: a Rb cell of adjustable length.

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1 h at 300 °C. The paraffin used in the present experiment is pentacontane $[\text{CH}_3(\text{CH}_2)_{48}\text{CH}_3]$ with a purity of 97% and a melting point of 93 °C. Paraffin-coated cells, after being made, are cured in an oven at about 70 °C for three days. Curing, albeit for a shorter period of time (one or two days at 70 °C), is also needed for OTS-coated cells. Measurements are done at a cell temperature of 72 °C in order to be sufficiently far away from the melting point of paraffin. The temperature is stabilized to within 0.5 °C throughout the experiment.

The glass cell is held inside a peek oven. The cell is heated by hot air such that the stem, where the Rb metal is, is kept at a temperature a few degrees lower than the cell body so as to prevent Rb condensation on the coatings. Two diode lasers, A and B, are used to provide probe and pump beams. The linewidth of the lasers is 45 MHz. The probe beam A is s -polarized and the pump beam B is σ^\pm polarized. Both beams are incident on the same spot of the cell window with slightly different incidence angles, and undergo total internal reflection at the front surface of the cell, with penetration depths of 1.16 μm and 0.82 μm for the pump and probe beams, respectively. The Zeeman polarization in the Rb vapor that contributes to the signal, is in the $F=2$ hyperfine multiplet, and is produced by the repopulation pumping of the pump beam, the frequency of which is tuned to the $F=1 \rightarrow F'=1, 2$ transitions of the ^{87}Rb D1 line. The Zeeman polarization is monitored by the rotation of the polarization plane (Faraday rotation) of the probe beam A, the frequency of which is tuned to be slightly off the $F=2 \rightarrow F'=1, 2$ transitions to maximize the signal. The Faraday rotation is measured as an imbalance in the outputs of two identical silicon photodiodes, which are first balanced for unpolarized Rb vapor using the combination of a half wave plate and a Wollaston prism. The evanescent pump beam B causes a frequency shift for the probed Zeeman transitions through virtual transitions.²⁶ A spectrum analyzer is used to monitor the frequency of the pump beam so that its frequency does not drift by more than 40 MHz. An amplitude-modulated radio frequency (rf) field $2B_1 \cos(\omega t)$ is applied along the y axis to measure the Larmor frequency of Rb atoms. Thus, if a spin polarized Rb atom is relaxed upon leaving the coated surface, it will not be detected. The scan of the frequency of the oscillating magnetic field across the ^{87}Rb Larmor frequency yields a magnetic resonance line of ^{87}Rb atoms.

The observed Larmor frequency of the Rb atoms can be interpreted as the ensemble average of the average Larmor frequencies of Rb atoms and is given by²⁷

$$\omega^{(\pm)} = \omega_0 + \frac{\delta\phi_s + \delta\phi_e^{(\pm)}}{\tau_s + \tau_b}. \quad (1)$$

Here ω_0 is the Larmor frequency of a Rb atom in the bulk, $\delta\phi_s$ is the average phase shift of the Rb atom during its dwell time τ_s on the surface, $\delta\phi_e^{(\pm)}$ is the average phase shift of the Rb atom during its interaction with the evanescent $\sigma^{(\pm)}$ pump beam due to the light shift (Fig. 2). Defining $\delta = \omega^{(-)} - \omega^{(+)}$, we have from Eq. (1)

$$\delta = \frac{\delta\phi_e^{(-)} - \delta\phi_e^{(+)}}{\tau_s + \tau_b} = \frac{\delta\phi_e^{(-)} - \delta\phi_e^{(+)}}{\tau_s + \sqrt{3\pi/2}L/v_{\text{rms}}}. \quad (2)$$

Here we have used $\tau_b = \sqrt{3\pi/2}L/v_{\text{rms}}$, with v_{rms} being the root-mean-square velocity of Rb atoms.^{21,28} Equation (2) im-

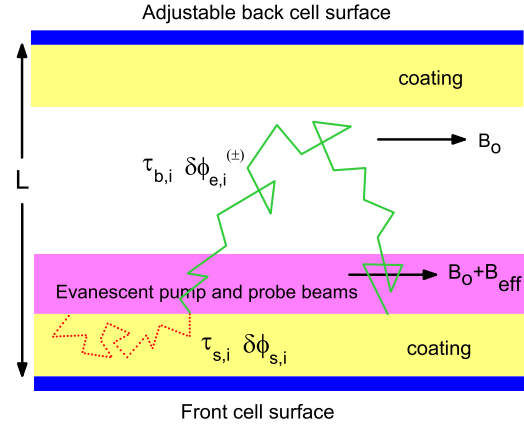


FIG. 2. (Color online) A representative i th segment of the diffusion path of a Rb atom between two consecutive wall collisions. The part of the segment in the coating (dotted line) is associated with the surface induced phase shift $\delta\phi_{s,i}$ and dwell time $\tau_{s,i}$ and the part of the segment in the bulk (solid line) is associated with a phase shift $\delta\phi_{e,i}^{(\pm)}$ due to the light shift in the evanescent pump beam and a time $\tau_{b,i}$, which is the time the Rb atom spends in the bulk between two consecutive wall collisions. Note that $\delta\phi_{e,i}^{(\pm)}$ is zero if the segment does not cross the pump beam. B_0 is the uniform holding magnetic field along the x direction, and B_{eff} is the effective magnetic field due to the light shift of the pump beam. The figure is not drawn to scale.

plies a linear relationship between the cell length L and $1/\delta$

$$\sqrt{\frac{3\pi}{2}} \frac{L}{v_{\text{rms}}} = (\delta\phi_e^{(-)} - \delta\phi_e^{(+)}) \frac{1}{\delta} - \tau_s. \quad (3)$$

We measure δ for a number of cell lengths L , and plot $\sqrt{3\pi/2}L/v_{\text{rms}}$ against $1/\delta$. The intercept is equal to the negative of the average dwell time τ_s . Even though the slope depends on the pump beam intensity, the intercept does not. The representative data showing this linear relationship between L and $1/\delta$ for OTS- and paraffin-coated cells are, respectively, shown in Figs. 3 and 4. Averaging over all the experimental data, we find that at a cell temperature of 72 °C the dwell time for OTS-coated cells is $\tau_s = 0.9 \pm 0.1 \mu\text{s}$, and that for paraffin-coated cells is $\tau_s = 1.8 \pm 0.2 \mu\text{s}$.²⁹

It is interesting to compare the dwell times for OTS and paraffin coatings. The dwell time for OTS is shorter than that for paraffin whereas the relaxation probability for OTS is larger than that for paraffin by almost one order of magnitude. This implies that the superior antirelaxation property of paraffin is not because of an extremely short dwell time of

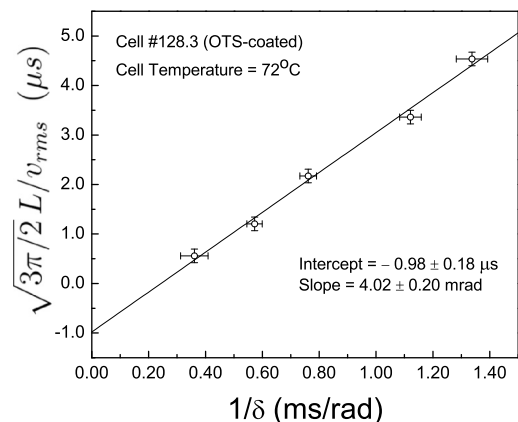


FIG. 3. A representative plot of cell length L versus $1/\delta$ for a cell coated with OTS and filled with 5 Torr N_2 buffer gas.

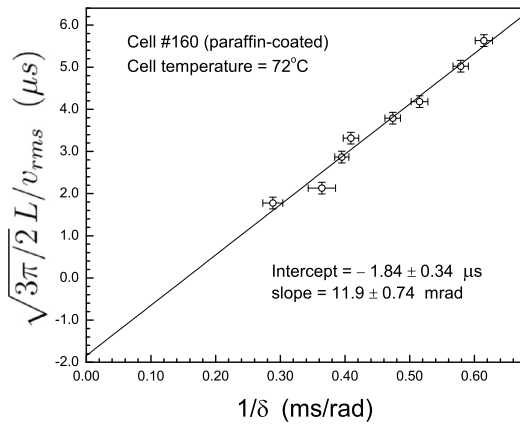


FIG. 4. A representative plot of cell length L versus $1/\delta$ for a cell coated with paraffin and filled with 5 Torr N_2 buffer gas.

spin polarized Rb atoms in paraffin, but because the average strength of the interactions experienced by them while they are inside paraffin is much weaker than while they are inside OTS. The OTS coating, unlike paraffin coating, which is coated with vapor and tends to be thick, is only several nanometers thick.¹⁸ Thus, for wall collisions of longer dwell time, spin polarized Rb atoms may diffuse to the sites near the interface between the glass surface and OTS and become depolarized. These Rb atoms, upon leaving the OTS coating, will not contribute to the signal. Thus the long dwell time tail of the dwell time distribution of Rb atoms is truncated, resulting in a shorter average dwell time. It was reported that the OTS coating does not completely cover the bare glass surface.¹⁶ The Rb atoms can occasionally collide with these bare glass patches and become depolarized. The relaxation of Rb atoms near the interface and on bare glass patches can help explain the larger relaxation probability of Rb atoms on OTS coating.

Alkali atoms are known to diffuse into coatings.¹² From the dwell time τ_s , we can estimate the total number of alkali atoms N_c trapped in the coating per square centimeter in equilibrium to be $N_c = n l \tau_s / \tau_b$, where n is the alkali vapor number density and l the characteristic dimension of the cell. Using $\tau_b \sim l / \bar{v}$, where $\bar{v} = \sqrt{8kT/\pi m}$ is the mean speed of Rb atoms, the estimated number of alkali atoms trapped in the coating per square centimeter is $N_c \sim n \tau_s \bar{v}$. For example, for paraffin-coated cells with a cell temperature of 72 °C, we have $\tau_s \sim 2 \mu\text{s}$, $n = 8.0 \times 10^{11} \text{ cm}^{-3}$, and $\bar{v} = 2.9 \times 10^4 \text{ cm/s}$. Thus $N_c \sim 5 \times 10^{10} \text{ cm}^{-2}$.

We would like to thank Kaifeng Zhao and Martin Schaden for helpful discussions. This work was supported by the ONR under Contract No. N00014-08-1-0483.

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- ²⁷Consider the diffusion path of a spin polarized Rb atom from the time it is polarized to the time it relaxes. The path can be divided into N segments as depicted in Fig. 2, with each segment corresponding to the path between two consecutive wall collisions. The average Larmor frequency shift for this Rb atom before it relaxes is given by

$$\omega^{(\pm)} - \omega_0 = \left(\sum_{i=1}^N \delta\phi_{s,i} + \sum_{i=1}^N \delta\phi_{e,i}^{(\pm)} \right) / \left(\sum_{i=1}^N \tau_{s,i} + \sum_{i=1}^N \tau_{b,i} \right),$$
 which becomes, upon dividing the numerator and denominator by N ,

$$\omega^{(\pm)} - \omega_0 = \left(\overline{\delta\phi_s} + \overline{\delta\phi_e^{(\pm)}} \right) / \left(\overline{\tau_s} + \overline{\tau_b} \right),$$
 where the bar indicates the average over the N segments. If N is sufficiently large, as is the case for good coatings such as OTS and paraffin, the distribution of the frequency shifts $\omega^{(\pm)} - \omega_0$ for all the Rb atoms will be sufficiently narrow to allow for a well-defined ensemble average frequency shift given by Eq. (1).
- ²⁸The expression for τ_b can be derived from the well known gas kinetic formula $n\bar{v}/4$ for the total number of collisions per second of Rb atoms on the front surface of area A . Thus the total number of collisions per second of Rb atoms on the front or back surface is $n\bar{v}A/2$, which can be written as $\bar{v}N/2L$, where N is the total number of Rb atoms in the vapor. Thus the rate of a single Rb atom hitting the front or back surface is $\bar{v}/2L$, the inverse of which yields the average time τ_b a Rb atom spends in the bulk between two consecutive wall collisions: $\tau_b = 2L/\bar{v} = \sqrt{3\pi/2} L/v_{rms}$. This derivation for τ_b ignores the finite relaxation time $1/\Gamma$ of the polarized Rb atoms. It is valid when $\pi^2 D/L^2 \gg \Gamma$, where D is the diffusion constant of Rb atoms in N_2 , i.e. when the decay rate of the first diffusion mode is larger than the total relaxation rate Γ of the polarized Rb atoms. Under this condition, we can concern ourselves only with the lowest diffusion mode (constant mode), which is the reason why τ_b does not depend on D . When D is very small (high N_2 density) or L is very large, one can no longer neglect the first diffusion mode, and the linear relationship between τ_b and L breaks down. Physically, this corresponds to the fact that the Rb atoms that collide with the front surface do not reach the back surface before relaxing. Note that the Rb polarization under our experimental conditions is what Masnou-Seeuws and Bouchiat called unconfined polarization [F. Masnou-Seeuws and M. A. Bouchiat, *J. Physique* **28**, 406 (1967)], for which the wall induced relaxation rate $\xi/(\tau_b + \tau_s)$, ξ being the probability of depolarization by a wall collision, is independent of buffer gas pressure.
- ²⁹The N_2 molecules in the cell could adsorb on the coating surface, the effect of which on the dwell time remains to be studied. The fact, however, that the measured dwell times depend on the type of coatings indicates that Rb atoms are interacting with atoms beneath the top layer of the surface where the N_2 molecules might reside.