Mapping surfaces using regionally specific hyperfine polarization

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We show that the hyperfine polarization in the vicinity ($\sim 10^{-5}$ cm) of cell surfaces can be used as a way to quantitatively and map the regional surface property inside optical pumping cells. In silicone-coated cells our method allows us to image the regional quality of the coatings, revealing the existence of areas that have strikingly different qualities.

It is well known that coatings have a very dramatic effect in optical pumping experiments. For example, in a typical uncoated Pyrex glass cell, a single wall collision destroys the spin polarization of Rb atoms whereas it takes thousands of wall collisions to relax the spin polarization of Rb atoms on coated walls [1,2]. Similar dramatic effect of coatings is observed for nuclear spin polarized Xe atoms. In a typical uncoated Pyrex glass cell that contains 20 Torr N$_2$ gas, the wall relaxation rate of $^{129}$Xe is $3 \times 10^{-3}$ s$^{-1}$ whereas it is $5 \times 10^{-4}$ s$^{-1}$ in a coated but otherwise similar cell [3]. The long wall relaxation time on the coatings can greatly enhance the atomic polarization that can be achieved and produce extremely narrow linewidth in atomic spectroscopy. Consequently coatings have the potential of being of great use in many fields such as precision atomic measurements, frequency standards, polarized sources and targets, medical imaging [4], etc. For example, recently they have been successfully used in atomic magnetometers to achieve a sensitivity comparable to that of superconducting quantum interference devices (SQUIDs) [5]. In spite of their great usefulness in many fields, however, coatings are not widely used. This is mainly because the reproducibility of coating quality is poor. For example, the relaxation rate for $^{129}$Xe was found to vary widely among similarly coated cells [6]. This mysterious behavior of coatings is not fully understood. Several studies of coatings in optical pumping cells have been reported [2,3,6,7]. In all these studies, however, the physical observables that are measured are affected by the average surface interactions, i.e., surface interactions averaged over the entire cell surfaces. The main difficulty in studying coatings is the lack of a simple method to quantify the regional quality of coatings, thus making it hard to determine whether a coating is regionally damaged or defective.

In this study we show that the hyperfine polarization in the vicinity ($\sim 10^{-5}$ cm) of cell surfaces can be used as a way to quantify and map the regionally specific surface property inside optical pumping cells. The hyperfine polarization near the surfaces is very sensitive to surface interactions, and therefore can be used to characterize their property. We use a pump beam to create hyperfine polarization in the Rb vapor. The evanescent wave of a weak $p$-polarized probe beam is used to probe the hyperfine polarization near the surfaces [8,9].

Although the Pyrex glass cells used in this study contain Rb metal of natural abundance ($72.2\% ^{85}$Rb and $27.8\% ^{87}$Rb), our study focuses on the hyperfine polarization $\langle \mathbf{S} \cdot \mathbf{I} \rangle$ of $^{85}$Rb atoms, where $h \mathbf{S}$ and $h \mathbf{I}$ ($I=5/2$) are, respectively, the spins of the electron and the nucleus. The hyperfine polarization $\langle \mathbf{S} \cdot \mathbf{I} \rangle$ is a measure of the deviation of the populations of the Rb atoms in the two ground state hyperfine levels from their thermal equilibrium values. It is given by

$$\langle \mathbf{S} \cdot \mathbf{I} \rangle = \text{Tr}(\mathbf{S} \cdot \mathbf{I} \rho) = \frac{I(I+1)}{N_a + N_b} \left(\frac{g_a}{g_a} \frac{N_a}{N_a + N_b} - \frac{g_b}{g_b} \frac{N_b}{N_a + N_b}\right),$$

where $\rho$ is the density operator of the ground state $^{85}$Rb atom, $N_a$ and $N_b$ are, respectively, the population densities of

[Diagram: Experimental setup. PD1 and PD2, silicon photodiodes; LP, Glan-Thompson linear polarizer. Inset: A representative attenuated total internal reflection signal $S(\nu)$. The cell temperature is 126 °C and Rb density $2.76 \times 10^{13}$ cm$^{-3}$. The incidence angle of the probe beam corresponds to a penetration depth of 0.51 μm. The signal is averaged 10 times. The dashed line corresponds to no absorption and therefore is equal to $C_1/C_2$. The reflectivity $R(\nu)$ is obtained by dividing the signal $R(\nu)C_1/C_2$ by the dashed line $C_1/C_2$.]

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The procedure for coating the cells is described in Refs. [3] and [6]. In addition to Rb metal, the cells are also filled with 5 Torr N$_2$ buffer gas. The pressure refers to 25 °C. The experimental setup is shown in Fig. 1. Single mode diode lasers operated in the free-running mode with a linewidth of 45 MHz and followed by a Glan-Thompson linear polarizer (LP) (extinction ratio of $\sim 10^{-5}$) provide pump and probe beams. Both the pump and probe beams are $p$ polarized. The $xz$ plane is the plane of incidence, with the $z$ axis being normal to the cell surface and pointing into the cell. The pump beam is incident perpendicularly on the cell surface and tuned to transitions $c_2, d_2$ [see Fig. 2(a)]. The line profiles of these two transitions overlap as a result of collisional and Doppler broadening. In spite of the narrow linewidth of the pump beam, all velocity groups can be pumped due to collisional broadening and velocity-changing collisions [10]. The pump beam depletes the population of the lower hyperfine level $b$ of the ground state, causing an accumulation of the $^{85}$Rb atoms in the upper hyperfine level $a$ of the ground state. A weak probe beam, which is incident at the same spot where the pump beam is and at an angle slightly larger than the critical angle $\bar{\theta}_c=\sin^{-1}(1/n_1)$, where $n_1$ is the index of reflection of Pyrex glass, undergoes total absorption near the cell surface and tuned to transitions $c_1, d_1$ in the probe beam. The probe beam also produces a small amount of $^{87}$Rb hyperfine polarization, which under our experimental conditions is negligible in uncoated cells and about 10% in coated cells. Therefore, for hyperfine pumping near the cell surfaces in an uncoated cell, the population of the ground state is given by
to obtain a good fit between the measured reflectivity and the calculated one. The Rb density is $2.76 \times 10^{13}$ cm$^{-3}$ in the uncoated cell and $2.92 \times 10^{13}$ cm$^{-3}$ in the coated cell. The incidence angle of the probe beam corresponds to a penetration depth of 1.06 μm in (a) and 0.98 μm in (b). The circles and squares are the experimental data and the lines are numerical calculations. From the data we obtain $(S \cdot I)=0.47\pm 0.01$ near uncoated and $1.20\pm 0.03$ near coated surfaces.

The cells are cylindrical in shape, about 30 mm in diameter, and between 20 and 50 mm in height. The inner surfaces of some of the cells are coated with SurfaSil (a siliconizing fluid). The procedure for coating the cells is the $^{85}$Rb ground state hyperfine multiplets of angular momenta $a=1+1/2=3$ and $b=1-1/2=2$, with $g_a=7$ and $g_b=5$ being their respective statistical weights. When all of the $^{85}$Rb atoms are in the hyperfine multiplet $a$, we have $(S \cdot I) = 1.25$.

FIG. 2. (Color) Hyperfine pumping near the cell surfaces in an uncoated (No. 30) and a coated (No. 49) cell. We note that because the transitions $c_1, d_1$ and $c_2, d_2$ overlap in the far wing, the pump beam also produces a small amount of $^{87}$Rb hyperfine polarization. The Rb density is $2.9 \times 10^{13}$ cm$^{-3}$. The penetration depth of the evanescent wave is 1.4 μm. (a) Images of an area (8 mm×10 mm) of the front and back surfaces of a cell coated according to the standard procedure. The probe beam size is 2 mm×2 mm. The average hyperfine polarization, while having vastly different values, is quite uniform across the mapped areas on both the front and back surfaces. (b) Image of an area (7 mm×13 mm) on a good coated surface that is intentionally damaged by high voltage discharge (a few tens kV at 500 kHz), using a Tesla coil, which has a tip in the shape of a knife edge more or less parallel to the $x$ direction. The probe beam size is 1.2 mm×1.2 mm. The hyperfine polarization measured at the same penetration depth on the coated surface, before it was damaged, was 1.20±0.03 across the mapped area. Thus, one sees that the lower right part of the coating in the mapped area is completely damaged, whereas the top part of the coating is only slightly damaged.

FIG. 3. (Color) Mapping silicone-coated surfaces using regionally specific $^{85}$Rb hyperfine polarization. The Rb density is 2.9×10$^{13}$ cm$^{-3}$. The penetration depth on the coated surface, before it was damaged, was 1.20±0.03 across the mapped area. Thus, one sees that the lower right part of the coating in the mapped area is completely damaged, whereas the top part of the coating is only slightly damaged.
internal reflection at the interface between the glass surface and Rb vapor [11]. The size of the probe beam is smaller than that of the pump beam. The intensity of the pump beam is 1.3 W/cm² and that of the probe beam 6 μW/cm². The frequency of the probe beam is scanned across the Rb D1 line and its reflectivity \(R(\nu)\) is measured. The probe beam is modulated by a chopper at 1900 Hz. To cancel laser intensity fluctuations, the intensity of the reflected probe beam and that of the laser are monitored by two photodiodes, the outputs of which are fed into a lock-in amplifier to yield a signal ratio \(S(\nu)=C_1R(\nu)/C_2\), where \(C_1\) and \(C_2\) depend on the reflectivity and transmissivity of various optical components. A typical total internal reflection signal is shown in the inset of Fig. 1, the caption of which explains the procedures for obtaining the reflectivity \(R(\nu)\) from the data.

According to Eq. (1), the hyperfine polarization of \(^{85}\text{Rb}\) atoms in the vicinity of cell surfaces is determined by the values of \(N_a\) and \(N_b\) near the surfaces, which we deduce from the measured reflectivity of the probe beam as follows [8]. When the pump beam is off, the reflectivity \(R(\nu)\) is measured and fitted to the calculated one [11]. The fitting parameters are Rb number density \(N\) and homogeneous linewidth \(\gamma\), which includes natural broadening, collisional broadening, etc. The best fit yields the values of \(N\) and \(\gamma\). When the pump beam is on, the population densities \(N_a\) and \(N_b\) of \(^{85}\text{Rb}\) in the vicinity of cell surfaces are functions of the distance \(z\) from the surface due to surface interactions. If we ignore the dependence of \(N_a\) and \(N_b\) on \(z\) and replace \(N_a(z)\) and \(N_b(z)\) by their average values \(\bar{N}_a\) and \(\bar{N}_b\), we can obtain the average hyperfine polarization \((\mathbf{S} \cdot \mathbf{l})\) by fitting \(R(\nu)\) to the calculated reflectivity with \(\bar{N}_a\) and \(\bar{N}_b\) as fitting parameters, using the same \(N\) and \(\gamma\) as determined when the pump beam is off [8]. The \(z\) dependence of the actual population densities \(N_a\) and \(N_b\) manifests itself in the dependence of the average hyperfine polarization \((\mathbf{S} \cdot \mathbf{l})\) on the penetration depth \(d\) or incidence angle \(\theta\) of the probe beam [8]. The penetration depth \(d\) is defined by

\[
d = \frac{\lambda_0}{2m_1} \frac{1}{\sqrt{\sin^2 \theta - \sin^2 \theta_c}},
\]

where \(\lambda_0\) is the wavelength of the beam in the vacuum. For fixed penetration depth, the value of the \(^{85}\text{Rb}\) hyperfine polarization \((\mathbf{S} \cdot \mathbf{l})\) can therefore be used to map the regional surface properties. Shown in Figs. 2(a) and 2(b) are the typical data and fit of hyperfine pumping in the vicinity of cell surfaces in a coated and an uncoated cell. It is seen that the average hyperfine polarization near uncoated surfaces is significantly smaller than that near coated ones.

By measuring the average \(^{85}\text{Rb}\) hyperfine polarization \((\mathbf{S} \cdot \mathbf{l})\) at micron or submicron distance from the cell surfaces we have been able to image or map the regional property of surfaces inside optical pumping cells. This imaging of regional surface property is especially interesting in coated cells commonly used in atomic physics experiments. The widely different spin relaxation rates of \(^{129}\text{Xe}\) in similarly coated cells are usually attributed to the possible existence of the so-called “hot spots,” where the coating is bad. The existence of such “hot spots,” however, has never been verified. Our imaging method makes it possible to directly observe these “hot spots.” Shown in Fig. 3 are some representative 2D images of surfaces in silicone-coated optical pumping cells. The images are obtained by translating the stage on which the cell is mounted horizontally and vertically in a step size equal to that of the probe beam. Figure 3(a) displays the images of two surface areas in a cell that is coated according to the standard procedure, but happens to contain spots where the coating is defective. One sees that the average hyperfine polarization \((\mathbf{S} \cdot \mathbf{l})\) on the front and back surfaces are strikingly different, being between 0.57 and 0.59 on the front and between 0.97 and 1.02 on the back surfaces. That is, while the coatings on the back surface are quite good, the front surface is only slightly better than uncoated surfaces, on which the average hyperfine polarization is between 0.46 and 0.52 under the same experimental conditions. Shown in Fig. 3(b) is the image of a good coated surface that has been intentionally damaged using a Tesla coil.

The fundamental limit of the spatial resolution of the 2D images, using our mapping technique, is due to the Goos-Hänchen effect [12], and is on the order of the penetration depth of the probe beam. This is because, according to the Goos-Hänchen effect, the probe beam, which undergoes total internal reflection, travels in the Rb vapor a distance on the order of the penetration depth. The spatial resolution of the 2D images is also affected by the lateral diffusion of polarization, which blurs the boundary between regions of different polarizations. The resolution limit due to this diffusion process in the Rb vapor is also on the order of the penetration depth. For the 2D images displayed in Fig. 3, however, the spatial resolution is limited by the size of the probe beam. By using CCD area detectors and expanding the probe and pump beam size, not only the imaging time can be greatly reduced, the spatial resolution can also be improved.


