Wall relaxation of spin-polarized sodium measured by reflection spectroscopy

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Using reflection spectroscopy of optically pumped sodium vapor, we have performed what is to our knowledge the first direct measurement of the spatially inhomogeneous spin polarization near a glass surface. On the basis of a theoretical description of the reflection at an inhomogeneous, anisotropic medium, we deduce the magnitude of the small residual magnetization at the surface from an analysis of the optical line shape. This allows us to specify the depolarizing properties of the surface. © 1995 Optical Society of America

When an atom comes into contact with a solid surface it may become adsorbed for a certain time that depends on the interaction energy. In the case of spin-polarized atoms, fluctuating magnetic fields present at the surface tend to diminish the polarization, depending on the dwell time on the surface, the surface diffusion, the presence of paramagnetic constituents on the surface, and so on. Hence the study of surface-induced spin relaxation provides information about properties such as adsorption energies, surface diffusion, cluster formation, and surface contamination. Detailed studies of wall relaxation date back to the 1960's, and they focused on the investigation of organic wall coatings such as paraffin that prevent disorientation to a large extent.[1,2] In recent times such studies have found renewed interest in the context of polarized ion sources in nuclear physics.[3–5] Few data on relaxation at uncoated glass surfaces are available. In many cases the interpretation of experimental data is based on the assumption that a glass surface is completely disorienting and that the spin polarization is zero at the surface.[6] In this Letter we demonstrate how reflection spectroscopy can be used for studying the polarization of an atomic vapor in the vicinity of a solid surface and how a theoretical analysis provides quantitative information about the depolarization probability. In contrast to earlier experiments, in which the polarization was measured in the volume, our method provides a surface-selective probe and allows us to measure the small residual polarization present at a glass surface.

The system investigated consists of Na atoms in a buffer gas close to a glass surface. The atoms are spin polarized by optical pumping with a circularly polarized laser beam tuned to the sodium D_1 ($\lambda = 590$ nm) line. To study the wall relaxation of atoms reaching the surface by diffusion through the buffer gas, we selectively probe the atoms close to the surface by reflecting a probe laser beam at the glass–vapor interface.[7] The spin polarization corresponds to a macroscopic magnetization and makes the atomic vapor optically anisotropic. Because this lead to a change of the polarization of the probe beam, we obtain information about the spin state of the atoms close to the surface from the polarization of the reflected beam.

The experimental setup is depicted in Fig. 1. Sodium vapor together with 230 mbars Ar is contained in a glass cell heated to 540 K that carries a Pyrex glass prism. The linearly polarized probe beam is incident upon the interface between the prism and the atomic medium at an angle close to the critical angle of total internal reflection, θ_c. A polarization-sensitive detector measures the intensity difference between two orthogonally polarized components of the reflected light. The pump beam passes through the interface at normal incidence. A magnetic field is applied perpendicular to both beams, and an electro-optic modulator modulates the polarization of the pump beam between left and right circular. This results in a forced precession of the magnetization around the magnetic field direction at the modulation frequency.[8] The modulation frequency is set to the Larmor frequency (~300 kHz at a magnetic field of 42 μT), which results in a resonant enhancement of the magnitude of the precessing magnetization. The precession causes a modulation of the optical anisotropy and a corresponding modulation of the polarization of the reflected probe beam, which allows us to extract the signal with a lock-in amplifier. The angle of incidence of the probe beam can be varied around the critical angle θ_c in steps down to ~0.1 mrad.

In the steady state, diffusion of polarized atoms through the buffer gas to the glass surface, relaxation

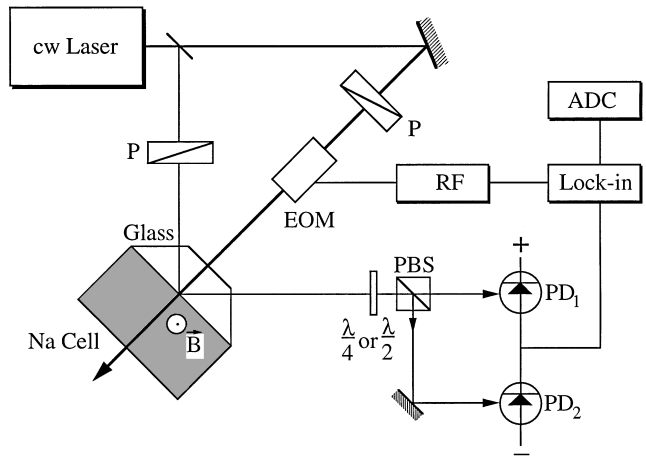

Fig. 1. Experimental setup: P's, polarizers; EOM, electro-optic modulator; RF, frequency synthesizer; ADC, analog-to-digital converter; $\lambda/4$ or $\lambda/2$, retardation plate; PBS, polarizing beam splitter; PD's, photodiodes.

at the wall, and the restoration of the polarization of atoms leaving the wall by optical pumping lead to a gradient of the magnetization close to the wall. We calculate the magnetization m as a function of the distance z from the wall from a one-dimensional diffusion equation:

$$\frac{\partial m}{\partial t} = D\frac{\partial^2 m}{\partial z^2} + P(1 - m) - \gamma m\,, \tag{1}$$

where D denotes the diffusion coefficient, P the optical pump rate, and γ the relaxation rate in the buffer gas. The optical pumping (P) represents a source of polarization and drives the system toward complete polarization ($m = 1$) but is counteracted by the relaxation (γ). When the modulation technique described above is applied, m has to be identified with the magnetization in a frame of reference rotating at the modulation frequency around the direction of the magnetic field. In this frame the magnetization vector has a fixed direction and may be described as a scalar m. The stationary solution of Eq. (1) reads as

$$g(z) = m(z)/m_\infty = 1 - (1 - g_0)\exp(-\mu z)\,, \tag{2}$$

with the bulk limit $m_\infty = m(z \to \infty) = P/(P + \gamma)$, the boundary condition $m(0) = m_\infty g_0$, and the inverse decay length $\mu = [(P + \gamma)/D]^{1/2}$. Because of the z dependence of the magnetization, the atomic medium is optically inhomogeneous close to the surface. For a pump rate of $5 \times 10^4\ \mathrm{s}^{-1}$ and a diffusion coefficient of $2 \times 10^{-4}\ \mathrm{m}^2/\mathrm{s}$, which represent typical numbers for our experiment, the thickness $1/\mu$ of the inhomogeneous layer is of the order of 100 μm. On the other hand, the depth probed by the reflected probe beam is comparable with the optical wavelength, so the experiment is sensitive to the magnetization $m_\infty g_0$ in the immediate neighborhood of the surface.

The quantitative extraction of g_0 from the experiment requires a theoretical understanding of the reflection at the interface between an isotropic homogeneous medium (glass) and an anisotropic inhomogeneous medium (spin-polarized Na vapor). The atomic vapor is stratified in the sense that its tensor of susceptibility $\overleftrightarrow{\chi}$ depends only on the coordinate z perpendicular to the interface. With a perturbative treatment on the basis of a matrix formalism applicable to stratified media,[9] we calculated the reflection matrix, which relates the s- and p-polarized amplitudes of the electric field of an incident plane wave to the corresponding amplitudes of the reflected wave. Because the susceptibility of the sodium vapor is small ($\sim 10^{-5}$), it was sufficient to consider only terms up to first order in the tensor elements χ_{ij}.

The result may be summarized as follows: The electric field $\mathbf{E}_t$ of the transmitted part of the electromagnetic wave excites an optical polarization $\mathbf{P}(z) = \varepsilon_0 \overleftrightarrow{\chi}(z)\mathbf{E}_t$. The induced electric dipoles contribute to the reflected field by reradiating an electro-magnetic wave in the direction of the reflected light. The contribution to the s-polarized reflected wave is due to the component P_s of the polarization perpendicular to the plane of incidence and results from a coherent superposition of the fields that are reradiated by the induced dipoles in different depths z. It can be written as the sum of a contribution $P_s(0)$ of the interface itself and an integral over z, where the net contributions of the deeper layers are superposed coherently with an appropriate phase factor:

$$\delta E_{rs} \propto P_s(0) + \int_0^\infty \exp(i2k_0 z\ \cos\ \theta_t)\left[\frac{\mathrm{d}}{\mathrm{d}z}P_s(z)\right]\mathrm{d}z\,. \tag{3}$$

Here k_0 is the vacuum wave vector and θ_t is the angle of refraction as given by Snell's law with the anisotropic medium replaced by vacuum. A similar expression is obtained for the p-polarized component of the reflected wave.

To evaluate the reflection matrix for the spin-polarized sodium vapor we treat the D_1 transition as a transition between two states with angular momentum 1/2. This represents a proper description in the presence of a buffer gas that causes a pressure broadening exceeding the hyperfine structure splittings. The change of polarization that is detected in the experiment is due to the off-diagonal elements of $\overleftrightarrow{\chi}$, which follow directly from the magnetization.[10] With the aid of the reflection matrix it can be shown that the modulated part of the measured intensity difference (normalized to the total intensity I_0 of the incident probe beam) is theoretically given by

$$S = \Delta I_{\mathrm{mod}}/I_0 = \mathrm{Re}(\chi_0 v)\,, \tag{4}$$

where v is a complex-valued function of the angle of incidence of the probe beam and χ_0 is the complex susceptibility of the unpolarized atomic medium, which has a Lorentzian dependence on the laser frequency. The function v can be written as

$$v = v_{\mathrm{hom}}\left\{g_0 + \frac{1 - g_0}{1 - i2k_0\ \cos\ \theta_t/\mu}\right\}, \tag{5}$$

with v_{hom} characterizing the case of a homogeneous, z-independent magnetization corresponding to $g_0 = 1$. The expression in braces stems from the evaluation of the reflection matrix and follows from $g(z)$ by an integration of the type given by relation (3). The signal as a function of the laser frequency represents a mixture between an absorptive and a dispersive line shape with a mixing angle given by the phase φ of v, which depends on the angle of incidence. In the region of total internal reflection, cos θ_t is purely imaginary, and the expression in braces in Eq. (5) becomes real. Therefore g_0 has no influence on the line shape in this case. However, in the region of partial transmission, where cos θ_t is real, the mixing angle φ depends on the value of g_0, and this quantity becomes experimentally accessible through the observed line shape.

In the experiment we measured the signal as a function of the laser detuning for a number of different angles of incidence around the critical angle θ_c. We extracted the mixing angle φ by fitting a mixed absorptive–dispersive line shape to the experimental line profiles. Figure 2 shows the experi-

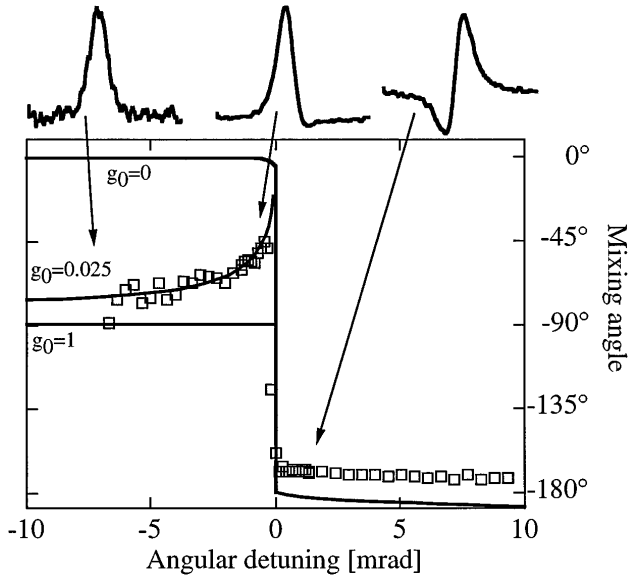

Fig. 2. Mixing angle between the absorptive and dispersive components of the optical line shape as a function of the detuning of the angle of incidence from the critical angle of total reflection. The squares indicate the experimental data points, and the solid curves represent theoretical predictions for three different values of g_0. Furthermore, three examples of experimental line shapes are given.

mental values versus the angular detuning from θ_c, together with theoretical curves for three different values of g_0. In the calculation, a value of $\mu = 0.018\ \mu\mathrm{m}^{-1}$ was used, which follows from the diffusion coefficient[11] and an estimated pump rate of $2\pi \times 10$ kHz. Experimentally, we find essentially an inverted dispersion line in the regime of total reflection. At θ_c there is an abrupt jump toward a noninverted dispersion line, and with increasing angular detuning in the region of partial transmission the line shape approaches an absorptive character. All three theoretical curves coincide in the case of total reflection, where they predict an inverted dispersion line, in close agreement with the experiment. The small deviation can well be due to an asymmetry of the line caused by the unresolved hyperfine structure. In the region of partial transmission the different values of g_0 lead to distinctly different line shapes. In the case of a homogeneous, z-independent magnetization ($g_0 = 1$) the theory predicts an absorption line for all angles of incidence below θ_c. On the other hand, if the magnetization vanishes completely at the surface ($g_0 = 0$), a dispersion line is expected. The experimental observations clearly deviate from these two cases but are well described by $g_0 = 0.025$. Hence the experiment clearly indicates that the magnetization at the wall is much smaller than in the bulk but does not vanish totally.

This experimental result can be interpreted in terms of the depolarization probability per wall collision on the basis of the following arguments: One half of the atoms at the wall are atoms arriving from the volume. They have traveled a mean distance L in the z direction since their last collision with a buffer-gas atom that is given by 2/3 of the mean free path in the buffer gas.[12] Therefore their degree of polarization should correspond to $g(z = L)$. The other half consists of atoms that originally came from the same mean distance but have made a wall collision and returned to the gas phase from the surface. These atoms have been partly depolarized, so that their polarization is reduced by a factor of $1 - \varepsilon$, where ε denotes the probability of depolarization. Within this model g_0 is related to $g(z = L)$ by

$$g_0 = \frac{1 + (1 - \varepsilon)}{2} g(L). \tag{6}$$

From the diffusion coefficient, the mean free path can be estimated to be 0.9 μm in our experiment, so $L = 0.6\ \mu$m. Then a value $\varepsilon = 0.6$ follows from Eqs. (2) and (6). It should be pointed out that, although the value of g_0 depends on the assumption made about μ, the resulting ε is almost independent of μ. However, the knowledge of L is crucial for a reliable determination of ε and must be considered the main uncertainty.

In conclusion, we have demonstrated a new approach to the quantitative study of spin-relaxation phenomena at surfaces. Reflection spectroscopy was applied as a surface-selective probe, and information about the extent of spin relaxation was extracted from the analysis of the optical line shape.

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