Evanescent wave spectroscopy of sublevel resonances near a glass/vapor interface

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We report the observation of atomic sublevel coherence from a quasi-twodimensional region of atomic Na vapor near a glass surface. The ground state coherences are excited with a circularly polarized laser beam and detected with a linearly polarized probe beam that is incident on the interface under conditions of total internal reflection. The coherences of the atoms that are within the evanescent wave lead to selective absorption and phase shifts of the reflected beam which can be measured by a polarization-selective detection.

1. Introduction

High resolution optical spectroscopy of atoms near an interface has received considerable interest recently [1]. In these experiments, the modification of the reflectivity of a glass–atomic gas interface due to the presence of electronic transitions is utilized to obtain spectroscopic information on the atomic medium. Selective reflection experiments usually display spectral features with sub-Doppler resolution, indicating that different velocity-classes contribute differently to the signal. These experiments can therefore yield information on the kinetic behavior of atoms near the interface. Theoretical analyses of the situation [2] indicated that the connection between the optical polarization excited in the atomic medium near an interface and the optical field are nonlocal; the internal state of the atom depends therefore on the external degrees of freedom, such as the position and velocity.

The high sensitivity of these optical methods makes them also an attractive tool for the measurement of other quantities, e.g. for magnetic resonance spectroscopy. In a recent example, Lukac and Hahn [3] have shown that it is possible to obtain selective spectroscopic information on the magnetic resonance transitions of quadrupolar nuclei near the surface of a single crystal. They analyzed the reflected light from the sample, using a Stark modulation technique. The reflectivity of the crystal surface could be changed by applying a radio frequency field at the magnetic resonance transition. By scanning the radio frequency, the magnetic resonance spectrum could be recorded.

However, not only the optical resonances are influenced by the presence of the interface, but also sublevel transitions, such as magnetic resonance transitions [4]. These authors demonstrated that the magnetic resonance spectrum of a gaseous sample depends in general on the geometry of the container and developed a theory of the interactions responsible for the observed frequency shifts. While the experiments described in ref. [4] were performed on volume samples, no experiments using local methods have been described that investigate modifications of the magnetic resonance transitions by the presence of the interface.

In this letter we present an alternative possibility

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to obtain spectroscopic information on the Zeeman transitions of atoms located near a glass–gas interface. The method uses an optical pump field to excite the atomic resonances and a second optical field as a probe to observe the resonances excited in the atoms near the interface. By applying the technique to an interface glass–atomic gas, we demonstrate the high sensitivity of the method which should have a wide range of applications to other systems as well.

2. Theory

In order to calculate the signal expected in a reflection experiment, we model the atomic medium as a $J=1/2 \leftrightarrow J'=1/2$ optical transition (see fig. 1). The procedure for the calculation is as follows: we first determine the eigen-polarizations and the corresponding wavevectors of a homogeneous plane wave travelling through the atomic medium. We expand then the incident and reflected fields in terms of s- and p-polarization and the transmitted field in terms of the eigenpolarizations. The expansion coefficients can then be calculated from the condition that the field components parallel to the interface be continuous. Only the results of the calculation are summarized here; details will be published elsewhere [5].

To calculate the propagation of light in the atomic medium, we assume that the population of the excited state can be neglected; experimentally, this is achieved by the relatively low laser intensity of our cw laser and by the addition of buffer gas that leads to a pressure broadening of the optical transition of the atoms. On the other hand we assume that the system is optically pumped so that the ground state sublevels are unequally populated and/or the coherences between them are nonzero. Experimentally, this optical pumping is achieved with a polarized laser beam while a second, weaker test field is used to monitor the state of the atomic system. For this calculation, we take the orientation of the atomic ground state as given and assume that the effect of the probe beam on the system can be neglected.

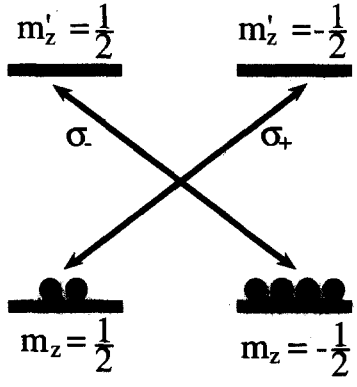

Fig. 1. $J=1/2 \leftrightarrow J'=1/2$ model system used to calculate the reflectivity from the interface. The dots represent the given ground state orientation, the arrows the right and left circularly polarization components of the optical probe beam coupling to σ_+ and σ_- transitions and thereby monitoring the ground state populations.

In order to calculate the effect of the ground state orientation on the optical probe beam, we write the optical field as a homogeneous plane wave with frequency ω whose wavevector is parallel to the z-direction (i.e. the quantization axis). The two-level ground state system can then be described by a magnetization vector $\boldsymbol{m}=(m_x,\ m_y,\ m_z)=(\rho_{12}+\rho_{21},\ -\mathrm{i}(\rho_{12}-\rho_{21}),\ \rho_{22}-\rho_{11})$ where ρ represents the density operator of the system. Due to the presence of the magnetization in the sample, the atomic medium becomes anisotropic; and electromagnetic field propagating through it experiences a dielectric susceptibility tensor

$$\chi=\chi_0\begin{pmatrix} 1 & \mathrm{i}m_z & -\mathrm{i}m_y \\ -\mathrm{i}m_z & 1 & \mathrm{i}m_x \\ \mathrm{i}m_y & -\mathrm{i}m_x & 1 \end{pmatrix}, \tag{1}$$

where χ_0 represents the susceptibility of the unpolarized medium (i.e. $\boldsymbol{m}=0$). In the atomic vapors of interest here, with densities of the order of 10^{11} cm^{-3}, the susceptibility χ_0 is of the order of 10^{-5}. For most purposes it is therefore possible to neglect all but the lowest order terms in χ_0. For a wavelength parallel to the z-axis, the (slowly varying) amplitudes of the E-fields of the eigenpolarizations are

$$\begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}_{\pm} = E_0 \begin{pmatrix} 1+\chi_0\left(1+\mathrm{i}\dfrac{m_x m_y}{m_z}\right) \\ \pm\mathrm{i}\left(1\pm\chi_0\dfrac{m_x^2-m_y^2}{2m_z}\right) \\ \chi_0(\pm m_x-\mathrm{i}m_y) \end{pmatrix}, \tag{2}$$

where E_0 describes the overall field amplitude. The length of the corresponding wave vectors is

$$|k_z|_{\pm} \approx i\omega\left(1+\chi_0\frac{1\mp m_z}{2}\right). \quad (3)$$

In the limit $\chi_0 \to 0$, the two eigenpolarizations corresponding to the two different signs obviously become left and right circular polarization and the wavevectors become equal. The susceptibility of the medium and therefore its index of refraction depends therefore on the direction of propagation of the field as well as on its polarization; in the limit considered here, the complex index of refraction is determined only by the magnetization component parallel to the wavevector, as it is usually assumed [6,7]. As can be seen from eq. (2), the optical field in the polarized medium is in general not transverse, but has also a longitudinal component; the laser beam is therefore in general displaced by a propagation through a polarized atomic medium.

Using these results on the propagation of the field in the atomic medium, we can now analyze the situation near the interface. The relevant fields are shown schematically in fig. 2: the shaded area represents the isotropic medium with refractive index n_1 from which a harmonic plane wave with wavevector k_1 is incident on the interface under an angle θ_i. The positive y-axis points into the atomic medium and the z-axis is parallel to the interface and the plane of incidence. If the angle of incidence is near the critical angle θ_c for total internal reflection,

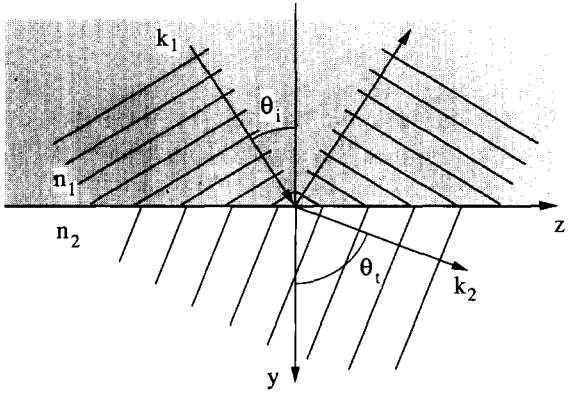

Fig. 2. Schematic summary of the optical fields involved in the experiment: incident, transmitted and reflected waves.

the wavevector of the transmitted wave is approximately parallel to the z-direction. As pointed out above, the index of refraction of the atomic medium depends in general on the direction of the wavevector; however, for the situation of interest here, the angle between the wavevector and the z-axis is so small that we can write the index of refraction of the atomic medium as

$$n_{2\pm} = 1+\chi_0\,(1\mp m_z)/2\,, \quad (4)$$

where the opposite signs refer to the two eigenpolarizations. We can now calculate the angle between the wavevector of the transmitted wave and the interface normal in the usual way as

$$\theta_{t\pm} = \sin^{-1}\{n_1 \sin(\theta_i)/[1+(\chi_0(1\pm m_z))/2]\}. \quad (5)$$

In the regime of total internal reflection, where the argument of the $\sin^{-1}$ function exceeds 1, the angle becomes imaginary, indicating that the transmitted wave is exponentially damped in the direction of the surface normal (y in our coordinate system). The amplitude of the reflected wave can now be calculated from the condition that the field components parallel to the surface be continuous at the boundary. Since the amplitude as well as the wavevector of the transmitted field depend on the magnetization in the atomic medium, the reflected beam contains information about the state of the atomic system.

Different experimental setups for the measurement of this information are possible. As an example we calculate the signal for the case that p-polarized light is incident on the interface and the reflected light is analyzed at $\pm 45°$; the difference between the two signals is then

$$\Delta S = I_0 \frac{2n_1}{\sqrt{n_1^2-1}}\,\mathrm{Im}[\cos(\theta_{t+})-\cos(\theta_{t-})]\,, \quad (6)$$

where I_0 represents the probe beam intensity. Since the experiment is performed in the vicinity of total internal reflection, it is useful to introduce the variable $\delta_i = \theta_i - \theta_c$ as the difference between the angle of incidence θ_i and the critical angle θ_c.

In the linear regime ($|\chi_0| \ll 1$, $|\delta_i| \ll 1$), this expression can be simplified to

$$\Delta S \approx -I_0\sqrt{2}\,n_1(n_1^2-1)^{-3/4}\,\mathrm{Re}(\chi_0/\sqrt{\delta_i})\,m_z. \quad (7)$$

The apparent divergence of the signal near the critical angle, i.e. for $|\delta_i| \to 0$ is absent in the exact form;

the linearized form is therefore invalid roughly in the region of $|\delta_i| < |\chi_0|$, a range which is not tested in our experiment. The signal is therefore directly proportional to the magnetization component parallel to the direction of propagation of the evanescent wave, to the atomic susceptibility and indirectly proportional to the square root of the deviation of the angle of incidence from the critical angle. Since this square root becomes imaginary at the critical angle, the dependence on the laser frequency detuning, which is contained in χ_0 in the above formula, changes from absorptive in the regime of total internal reflection ($\delta_i > 0$) to a dispersive behavior in the transmissive range ($\delta_i < 0$), if the other parameters remain constant.

3. Experimental results

For the actual experiment, we used a circularly polarized pump beam to create the magnetization in the atomic medium. The experimental setup is summarized in fig. 3: the pump beam traverses the atomic medium, thereby polarizing the atoms not only in the vicinity of the interface, but throughout the whole system. Alternatively, the angle of incidence of the pump beam can also be chosen near the critical angle, in which case only the atoms near the interface are pumped. A magnetic field is applied perpendicular to the plane of incidence of the test beam, so that the magnetization generated in the sample is forced to precess in the plane of incidence. In fig. 1, this precession of the magnetization corresponds to an oscillatory exchange of population between the two ground state sublevels and therefore to a modulation of the index of refraction; the modulation frequency is given by the Larmor frequency. The unmodulated probe beam was polarized parallel to the plane of incidence and the polarization-selective detection measured the signal at $\pm 45°$, as assumed in the theoretical section.

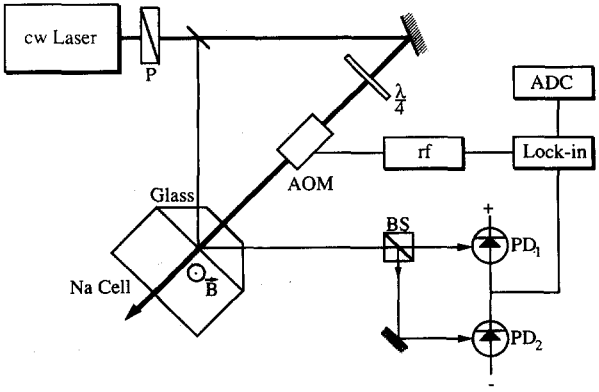

Fig. 3. Schematical representation of the experimental setup. P=polarizer, AOM=acoustooptic modulator, rf=frequency synthesizer, ADC=A/D converter, $\lambda/4$=retardation plate, BS=polarizing beamsplitter, PD=photoiode. The magnetic field $\boldsymbol{B}$ is applied perpendicular to the plane of the drawing.

In order to obtain the highest sensitivity, we used a modulation scheme described in detail elsewhere [8]. The pump beam was modulated sinusoidally with a frequency near the Larmor frequency of the spin system, thereby driving the precession of the magnetization in the sample. Since the signal expected from the reflection experiments is proportional to the component of the magnetization parallel to the surface, we expect it to oscillate sinusoidally at the modulation frequency. It is therefore possible to detect the signal via lock-in detection, using the modulation frequency as the reference. As shown in ref. [8], the magnetization induced in the sample is maximized if the modulation frequency is equal to the Larmor frequency; in general, it has a lorentzian dependence on the frequency difference, with the width of the resonance line determined by the optical pump rate and the relaxation rate.

The measurements described here were performed on the Zeeman sublevels of the $3s\ ^2S_{1/2}$ sodium ground state. The Na was contained in a glass cylinder with a prism at one end, as shown in the figure. The prism was heated to a temperature of some 500 K with the cell temperature slightly lower in order to avoid condensation of Na droplets on the glass surface; the resulting Na number densities were of the order of 10^{11} cm^{-3}, as measured by a transmission experiment. Argon buffer gas was added to the sodium vapor at a pressure of 210 hPa; the measured width of the pressure-broadened D_1-line was $\Gamma/2\pi = 4.2$ GHz (fwhm). As discussed elsewhere [7], it is possible to describe Na under these experimental conditions as a $J = 1/2 \leftrightarrow J' = 1/2$ optical transition, as assumed in the theoretical section.

A single-mode cw ring dye laser (short term line-

width ≤ 500 kHz) was used as the light source. The laser beam was split into a circularly polarized pump beam (thick line in fig. 3, average intensity ~ 300 $\mu W/mm^2$) and a linearly polarized probe beam (dashed line, intensity ~ 100 $\mu W/mm^2$). The pump laser intensity was kept low in order to avoid excessive power broadening of the magnetic resonance transition which occurs when the optical pump rate exceeds the relaxation rate due to diffusion; as a consequence of that, the difference between the pump beam intensity and the probe beam intensity is relatively small. The laser frequency was set on resonance at the Na-D_1 line ($\lambda = 589.6$ nm). The modulation of the pump beam was achieved with an acoustooptic modulator (AOM).

In order to observe the magnetic resonance transition, we scanned the modulation frequency over the Larmor frequency, keeping the strength of the magnetic field at 86 μT, corresponding to a Larmor frequency of 594 kHz. The result is shown in fig. 4: A well-defined resonance appears at the Larmor frequency, with a width of 60 kHz (fwhh). This width is clearly larger than the corresponding width of 19 kHz obtained in transmission experiments. The difference may be due to power broadening or to lifetime effects, especially the diffusion of atoms out of the laser beam. A more detailed analysis of the center frequency and the line shape is not possible with the data currently available; the shoulders in the resonance line as well as the details of the baseline are not reproducible and are therefore interpreted as noise. Clearly, more experiments are needed to study the dependence of the magnetic resonance signal on, e.g., the laser intensity and laser detuning and the angle of incidence of the test beam.

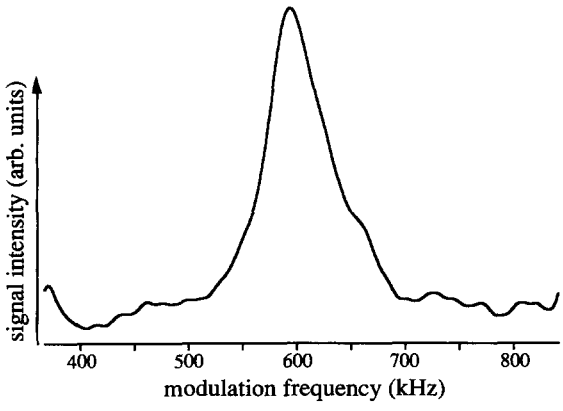

Fig. 4. Experimental spectrum obtained by scanning the modulation frequency. Experimental parameters: scan time 2 s; $B = 86$ μT; pump beam intensity 0.3 mW/mm^2; test beam intensity 0.1 mW/mm^2.

The signal shown in fig. 4 was obtained in a single scan with a scan time of ~ 2 s and has a signal to noise ratio of ~ 40. This clearly demonstrates that the experimental setup described here is suitable for the measurement of atomic sublevel coherence in a quasi-twodimensional system. Under the experimental conditions used here, the sensitive volume has the approximate dimensions 1 mm $\times$ 1 mm $\times$ 1 μm; with the atomic density given above, the number of atoms in this volume is therefore of the order of 10^5. This number shows not only the high sensitivity of the method, but also indicates that the system is indeed quasi-twodimensional: the average distance between atoms is ~ 2 μm, larger than the thickness of the sensitive volume.

4. Conclusion

The experiment described here demonstrates that it is possible to perform spectroscopy of atomic sublevel transitions on atoms in an evanescent wave. The method used here utilizes a laser beam to polarize the atoms and a second laser beam for optical detection of the magnetization generated in the sample. The detection process is based on the modification of the reflectivity of the glass–gas interface due to the presence of the atomic polarization, thereby modifying the reflected beam. This reflectivity change depends on the polarization of the probe beam so that it is possible to perform sensitive difference experiments via polarization-selective detection of the reflected beam.

In contrast to earlier experiments [3], the method described here requires no time-dependent electric or magnetic fields apart from the laser beam. Only a static magnetic field is applied perpendicular to the plane of incidence of the test beam in order to make the Zeeman sublevels non-degenerate. The presence of this magnetic field is primarily an experimental convenience, not a necessity: the experiment could equally well be performed in the absence of a magnetic field, in which case the magnetic resonance

would be observed at zero modulation frequency, in close analogy to the Hanle effect.

The experiment described here used a probe beam whose polarization was parallel to the plane of incidence and the polarization-selective detection measured the difference between the intensities of light with polarization at $\pm 45°$. Other geometries yield additional information about the atomic medium, especially different magnetization components. Experiments with different experimental setups are currently being prepared.

Not only the probe beam, but also the pump beam can be applied differently: e.g. it is possible to optically pump the atomic medium with the angle of incidence of the pump beam also near the critical angle. In such a geometry, only the atoms that stay within the evanescent wave for a longer period, i.e. those atoms that have a small velocity-component perpendicular to the surface should be excited. In addition, the dependence on the various parameters, such as optical detuning, angle of incidence, optical pump rate etc. could be investigated. Of special interest will be similar experiments on the surface of solid materials, where such experiments might be useful for the study of adsorption/desorption processes.

Acknowledgement

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