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Faraday rotation density measurements of optically thick alkali metal vapors

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Abstract

We investigate the measurement of alkali number densities using the Faraday rotation of linearly polarized light. We find that the alkali number density may be reliably extracted even in regimes of very high buffer gas pressure, and very high alkali number density. We have directly verified our results in potassium using absorption spectroscopy on the second resonance ($4^2S \rightarrow 5^2P$). © 2001 Published by Elsevier Science B.V.

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1. Introduction

As linearly polarized light passes through a circularly birefringent medium, its plane of polarization rotates. If the birefringence is caused by the application of a longitudinal magnetic field, this rotation is known as the Faraday effect. In this paper, we explain a simple and reliable method to measure the density of an optically thick alkali vapor using the Faraday effect. We focus on alkali vapor density measurements because among other applications, high density alkali vapors are frequently used for spin-exchange optical pumping, and accurate knowledge of the vapor density is necessary to predict the effects of spin-exchange and spin-relaxation collisions (see Ref. [1]).

We report experiments using a relatively simple apparatus to obtain the alkali number density using the Faraday effect. In particular, we report the first verification of this technique on very high density ($> 10^{16}$ cm^{-3}) vapor and verify that it is applicable even at buffer gas densities as high as 7.8 amagats. The applicability of this technique at lower alkali density and in the absence of buffer gas has been demonstrated before (see Ref. [2]), and its use as a polarization monitor is discussed in Ref. [3]. We have focused on potassium density measurements in a He buffer gas because of the importance of polarized ^{3}He for magnetic resonance imaging [4,1], high-energy particle targets [5,6] and in other applications. Because of the small K–K and K–He depolarization cross-sections, we expect that spin-exchange optical pumping using K will be a very efficient way to produce large amounts of hyperpolarized ^{3}He [7]. We expect that this method is equally

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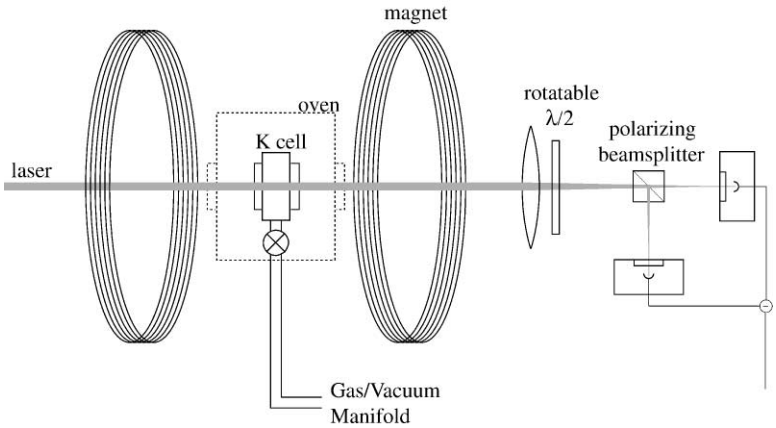

Fig. 1. Experimental apparatus.

valid for density measurements of other alkali species.

Faraday rotation complements the more commonly used technique of absorption spectroscopy, in which one extracts the atomic vapor density by comparing the measured absorption spectrum to $\exp(-[A]\sigma(v)l)$ ($[A]$ is the alkali density, l is the optical path length through the vapor and $\sigma = \pi r_e cfg(v - v_o)$ is the absorption cross-section in terms of the classical electron radius r_e, oscillator strength f, and normalized lineshape g centered at the atomic resonance v_o). Using absorption spectroscopy to measure a dense alkali vapor is challenging, however, because the vapor attenuates the probe light almost completely within many linewidths of resonance. Therefore, the above comparison requires either accurate knowledge of the lineshape g (to infer the absorption near line center from measurements in the wings), or accurate measurements of the very low transmittance near v_o. Even at lower alkali densities, an absorption spectroscopy measurement may be difficult if the lineshape is broadened and distorted by collisions with a high-pressure buffer gas. In contrast, the Faraday effect is insensitive to the above problems if the probe light is far from the atomic resonance, so this technique is valuable under conditions that make absorption spectroscopy difficult. In addition, laser vapor density measurements using the Faraday effect can be done at a fixed wavelength, eliminating the need for the more complex and time-consuming measurement of absorption as a function of wavelength. We have compared our results to absorption measurements where such comparisons are feasible, and find very good agreement between the two methods.

2. Apparatus

Our apparatus is shown schematically in Fig. 1. We use K vapor contained in a right circular cylindrical cell (1.91 cm in length and 5.72 cm in diameter). The vapor pressure is varied between 10^{15} and 2×10^{16} cm^{-3} by varying the cell temperature. The pyrex windows of the cell are 0.95 cm thick and are affixed with a knife-edge gasket as described in Ref. [8]. The cell is attached to a gas manifold so that the buffer gas pressure can be varied up to about 15 atm. The cell also contains potassium (99.95% pure) from a glass ampoule which is broken under a dry N_2 atmosphere. We mount the cell between the pole faces of an electromagnet which can generate fields of up to 12 000 G.

We normally use inexpensive diode lasers to measure alkali vapor densities.[1] However, since the

[1] Our diode lasers are of the type described by MacAdam et al. [9]. We note that the tunability range of many commercially available semiconductor diode lasers coincides with the D_2 resonance of rubidium.

experiments we describe here were aimed at verifying this technique over a wide range of laser detunings, we found it convenient to use a broadly tunable Ar^+-pumped Ti:Sapphire laser. The linearly polarized Ti:Sapphire laser beam passes through holes in the pole faces and through the K vapor cell. In order to avoid any optical pumping effects, the beam intensity is attenuated to approximately 10^{-4} W/cm^2. As shown in Fig. 1, the beam emerges from the magnet and passes through a 25 cm focal length lens, a $\lambda/2$ plate (mounted on a rotation stage accurate to 10 min of arc) and a polarizing beam splitter. The resulting two beams are focused through pinhole openings in light-tight boxes and are detected by photodiodes contained inside. The photodiodes are further isolated from stray light by covering the pinholes with 2 mm thick broadband red filter glass.

We use the apparatus to make measurements as follows: first, we rotate the $\lambda/2$ plate so that the difference signal between the photodiodes is nulled. This indicates that the plane of polarization of the light incident on the beam splitter is at 45° to the plane of Fig. 1. Next, we apply a magnetic field and measure the difference signal from the photodiodes. By calibrating the difference signal to a mechanical rotation of the $\lambda/2$ plate, we arrive at the field-induced Faraday rotation angle. Alternatively, for large rotation angles, we mechanically rotate the $\lambda/2$ plate to re-null the difference signal each time the field is increased. The Faraday rotation angle is then twice the angle through which the $\lambda/2$ plate was rotated.

We note that the glass windows have a field-induced birefringence, which causes about 8×10^{-5} degrees of rotation per Gauss of applied field. We account for this by measuring field-induced rotation at extremely low K vapor pressure ($[K] \ll 10^{10}$ cm^{-3}). We find that the rotation due to the windows is independent of the laser wavelength and is proportional to magnetic field strength over the wavelength range used for our experiments. The rotation angle due to the windows can therefore be described by a single measured parameter. We subtract off the effects of the windows in the analysis of our data.

3. Measurements and interpretation

We have measured the Faraday rotation angle for three different K densities in the absence of any buffer gas. Fig. 2 shows the Faraday rotation angle as a function of laser wavelength over a wavelength range from well below the $4S_{1/2} \leftrightarrow 4P_{3/2}$ (766.8 nm) to well above the $4S_{1/2} \leftrightarrow 4P_{3/2}$ (770.1 nm) transitions. The fits shown in Fig. 2 use the theoretical form of Wu et al. (Ref. [2]), including the paramagnetic terms proportional to $1/k_B T$. Thus, the rotation angle of near resonant, linearly polarized light with optical path length l through

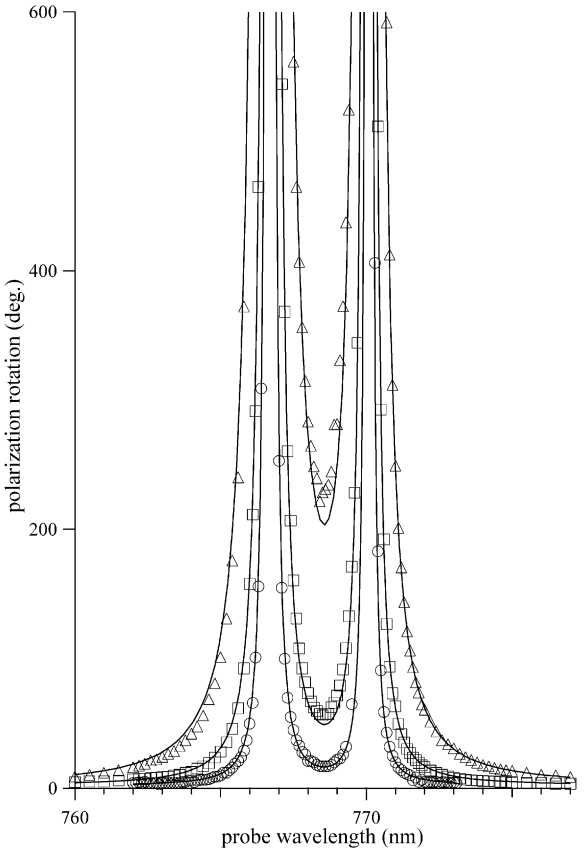

Fig. 2. Faraday rotation angle from a field of 12 000 G at K densities of 1.03 (circles), 3.24 (squares) and 1.40×10^{16} cm^{-3} (triangles). The fits are to a functional form derived from Ref. [5]. The figure shows no significant departure from the theoretical curves, even at high K density.

a K vapor of density [K] is

$$\theta = \frac{1}{6}[\mathrm{K}] l r_e \lambda_0^2 \mu_B B/h \left[\frac{\lambda^2}{3c} \left(\frac{4}{(\Delta\lambda)_{1/2}^2} + \frac{7}{(\Delta\lambda)_{3/2}^2} \right) \pm \frac{h}{k_B T} \left(\frac{1}{(\Delta\lambda)_{1/2}} - \frac{1}{(\Delta\lambda)_{3/2}} \right) \right], \quad (1)$$

where $(\Delta\lambda)_{1/2}$, $(\Delta\lambda)_{3/2}$ are the probe detuning from the $P_{1/2}$ and $P_{3/2}$ resonances, respectively, and we approximate the oscillator strengths by $f_{1/2} = \frac{1}{3}$ and $f_{3/2} = \frac{2}{3}$. In order to investigate rotation at large detunings, these rotation measurements were performed at 12 000 G, although such high fields are not necessary – in a typical spin-exchange optical pumping cell with $l = 10$ cm and an alkali density of 10^{15} cm^{-3}, several tens of Gauss would be adequate for high accuracy measurements.

The sign of the paramagnetic terms in Eq. (1) depends on the direction of the applied field. The paramagnetic corrections are on the order of a few percent, and result in a significantly better fit. The fits are performed allowing only the K number density to vary. We note that there is minimal departure from the theoretically predicted curve, even at these large alkali number densities. This shows that no significant rotation is produced by colliding pairs of K atoms, or by K_2 molecules.

This point requires a brief discussion. In order for measurements of Faraday rotation to accurately reflect alkali density, we must be assured that no significant rotation angle arises from any other species, including molecular alkali species. It is clear from thermodynamic considerations that only singlet alkali dimers are present in the vapor with appreciable number density. Using thermodynamic arguments,[2] we estimate that the singlet dimer density is approximately 0.35% of the atomic number density for an alkali density of 10^{15} cm^{-3}, and increases by a factor of three over the range of alkali densities shown in Fig. 2. In addition, the dimer resonances are at significantly different wavelengths (at least 80 nm from the atomic resonances for the 0–0 dimer transitions [10]). We therefore expect that Faraday rotation from molecules would show up as a distortion of the curves shown in Fig. 2 away from the theoretical shapes, and that the distortion should increase with alkali number density. We expect similar behavior for Faraday rotation caused by colliding pairs of K atoms. Since no density-dependent distortion is present in our data, we conclude that the observed Faraday rotation is determined almost entirely by the atomic number density.

We have also measured the Faraday rotation angle for K vapor in the presence of He buffer gas at He densities of 0, 2.1, 3.7 and 7.8 amagats. These measurements are shown in Fig. 3, scaled by the derived number density. We do not find significant buffer gas-induced distortion of the Faraday rotation lineshape over this range of pressures.

To understand the observed robustness of this measurement, we briefly consider the effects of large amounts of buffer gas on the observed relationship between the Faraday rotation angle and the wavelength detuning. The underlying absorption lineshape for the alkali resonance lines exhibits a dispersive asymmetry [13,14] due to the finite collisional interaction time. This asymmetry, which comprises a larger fraction of the overall lineshape as one probes farther detunings and higher pressures, will dominate the value at the far wings of the absorption. This is one of the reasons that it is very difficult to extract useful information from an optically thick vapor. Because of the fundamental relationship between absorption and dispersion, one would correctly expect the wavelength dependence of the index of refraction to exhibit some asymmetry as well. However, unlike the case with absorption, where the asymmetries become more important at large wavelength detunings, the asymmetry for the index of refraction becomes less important as the detuning increases. This can be qualitatively understood by noting that the Faraday rotation angle arises from the difference between the indices of refraction for the two helicities of light, and that the lineshape asymmetries cancel out to lowest order.

[2] We calculated the singlet dimer ro-vibrational energies $E(n,j)$ using the ab initio potentials of Ref. [11], adjusted to fit the measured binding energy of Ref. [12]. The fraction of singlet molecules in thermodynamic equilibrium is then

$$\frac{[A_2]}{[\mathrm{A}]} = \frac{[A]}{8} \left[\frac{h^2}{\pi m k_B T} \right]^{3/2} \sum_{n,j} e^{-E(n,j)/k_B T} (2j+1). \quad (2)$$

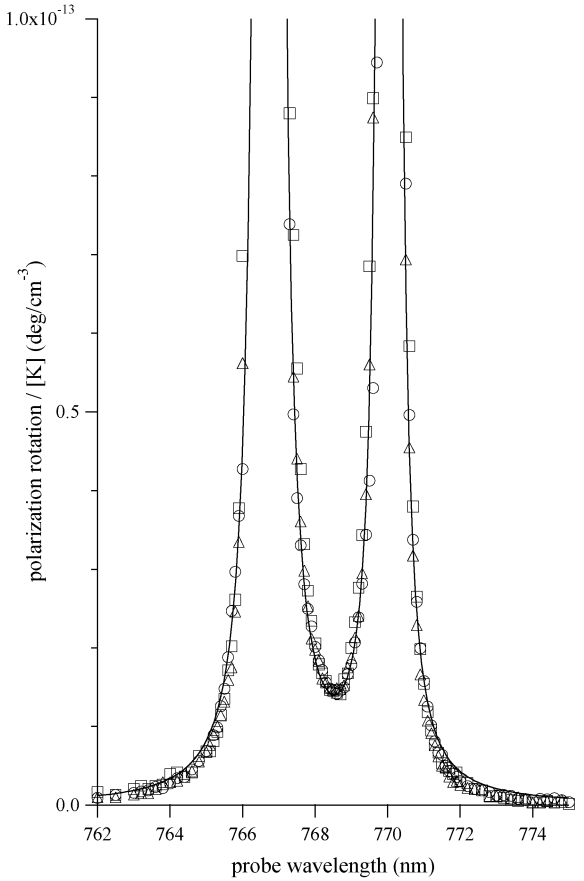

Fig. 3. Faraday rotation angle divided by derived K density at He buffer gas pressures of 0 (diamonds), 2.1 (circles), 3.7 (squares) and 7.8 (triangles) amagats. The fits are to a functional form derived from Ref. [5]. The figure shows that there is no significant broadening, shift, or asymmetry introduced by the buffer gas.

More quantitatively, we use the notation of [2] and represent the pressure-induced contribution to the Faraday rotation angle in terms of an asymmetry parameter T_d as

$$\Delta\theta(\lambda) = 0.106 T_d \frac{[A] l r_e f \mu_B B \gamma^2 \kappa \lambda_o^6}{hc^2(\lambda - \lambda_o)^3}, \quad (3)$$

where μ_B is the Bohr magneton, γ is the (pressure-broadened) width of the transition, λ_o is the center of the alkali resonance and the and κ is $\frac{4}{3}$ for the D_1 line and $\frac{7}{6}$ for the D_2 line. For the Rb D_2 line and He buffer gas, Romalis et al. [13] measured $T_d = (-0.44 \pm 0.1) \times 10^{-13}$ s.

In spite of the increased importance of the asymmetry term with width γ, which is directly dependent on the third body number density, this term is unimportant even in optical pumping cells with many tens of atmospheres of buffer gas. For instance, the expected error in inferred number density caused by 10 amagats of He is less than 1%, as long as probe detuning is at least 0.5 nm.

We note that Kristensen et al. [15] have observed changes in the Faraday rotation spectrum of Rb in Xe at small detunings and large Xe pressures. We do not observe similar distortions in our experiment, which is expected because of the relatively large probe detuning. This alkali density measurement technique therefore requires no modification to account for the presence of a buffer gas, as long as Faraday rotation measurements are made at sufficiently large detuning. The small (< 0.02 nm) pressure shifts caused by the buffer gas are not visible at the scale of Fig. 3, and cause negligible error in determining the alkali vapor density.

We do note, however, that the theory consistently predicts a larger rotation angle than was observed at extremely large detuning. Although we do not know the source of this discrepancy, it does not depend on the alkali density or the buffer gas pressure over the ranges we have studied. We believe that this is a minimal source of error at smaller detunings and therefore recommend that probe detuning be kept between 0.5 and 2 nm whenever possible, where the difference between theory and experiment is always less than 10%.

In order to further verify that this alkali density measurement technique is accurate as well as robust, we have compared the number density obtained using Faraday rotation to that obtained from absorption spectroscopy on the $4^2S \rightarrow 5^2P$ resonance under nearly identical conditions.[3] Sample spectra are shown in Fig. 4. Near this resonance, the transmitted intensity is related to

[3] In some cases, the He buffer gas pressure was changed to simplify the absorption measurement and to avoid the large optical thickness from an unbroadened line

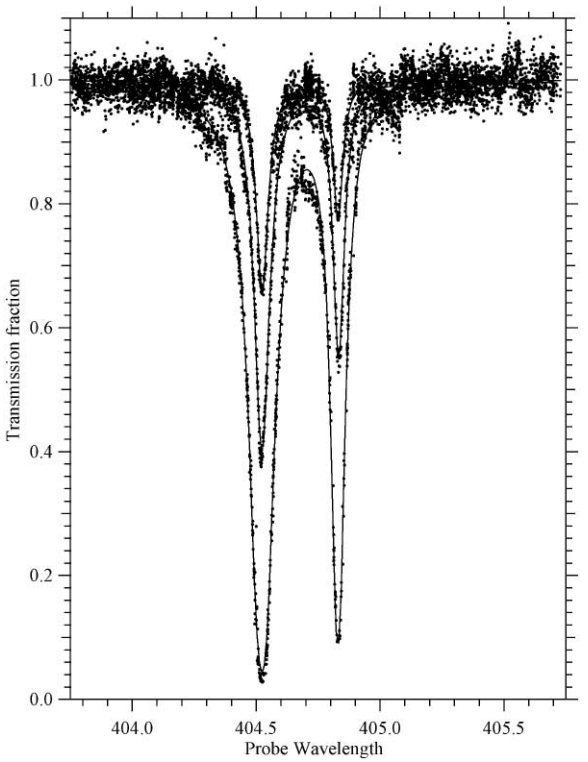

Fig. 4. Absorption of blue light by the K second resonance, taken with He buffer gas density of 2.7 amagats. The fits shown are to the theoretical absorption profile, allowing only the K number density to vary. The implied K densities are 0.37, 1.01, and 2.18×10^{15} cm^{-3}. Comparison of these implied densities to those obtained by measuring Faraday rotation (such as those in Fig. 2) show that the two methods agree to better than 10%.

K number density [K] as

$$I = I_0 e^{-([K]\sigma_1 l + [K]\sigma_2 l)}, \quad (4)$$

where l is the optical path length through the alkali sample and σ_1 and σ_2 are the absorption cross-sections for the $4^2S_{1/2} \rightarrow 5^2P_{1/2}$ and $4^2S_{1/2} \rightarrow 5^2P_{3/2}$ transitions, respectively. The absorption cross-sections can then be rewritten in terms of our normalized lineshape and previously measured oscillator strengths $(g_k/g_i)f_1 = 0.0124$ and $(g_k/g_i)f_2 = 0.0248$ [16] as $\sigma = \pi r_e c f g(v - v_0)$ in order to obtain K number density.

The number density derived from such spectra is always within 10% of the number density derived from a Faraday rotation measurement, as long as the Faraday probe light detuning is kept between 0.5 and 2 nm. We measured probe absorption across the second resonance to overcome the problems associated with very high absorptance near the $4^2S \rightarrow 4^2P$ transitions, although the requirements of a tunable light source and a frequency doubling crystal make this an unattractive general method.

4. Conclusions

In this paper, we have presented data that indicate the reliability of alkali vapor density measurement using Faraday rotation of linearly polarized light. We find that this technique is insensitive to problems associated with number density measurements using absorption spectroscopy, and is therefore particularly useful at high alkali density and in the presence of a buffer gas. In addition, vapor density measurements using the Faraday effect eliminate the need for a tunable light source and significantly simplify measurement and analysis.

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