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Skew light propagation in optically thick optical pumping cells

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Circularly polarized light propagating at an angle to the external magnetic field does not optically pump atoms to full transparency under conditions common to spin-exchange optical pumping experiments. This causes excess absorption of the pumping light that can significantly reduce the efficiency of the optical pumping process even for angles of a few degrees.

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In current applications of spin-exchange optical pumping [1], it is common that optically very thick alkali-metal atom samples are optically pumped with high-power laser sources such as broadband diode laser arrays [2], frequency narrowed diode arrays, and diode-array bars [3–5] or Ti:Sapphire lasers [6,7]. As circularly polarized light propagates through the cell parallel to an applied magnetic field, the atoms are optically pumped into states that are nearly transparent to the light. This allows the light to deeply penetrate optically thick cells, as demonstrated in the pioneering work of Bhaskar *et al.* [8]. Imperfect circular polarization of the light can even be tolerated, since the strong circular dichroism of the atoms rapidly attenuates the undesired polarization component near the entrance to the vapor cell.

When circularly polarized light propagates at an angle to the external magnetic field, however, the electron spins rapidly precess about the magnetic-field direction, so that the time-averaged spin polarization is reduced. Thus, even in the absence of spin relaxation the atoms do not attain full polarization, and continue to absorb light. Equivalently, the atoms experience an effection light polarization that is less than 100%. Due to the required transversality of the pumping light, the atomic circular dichroism does *not* correct for small polarization imperfections as the light traverses the cell. Even at very small propagation angles, dramatic changes in the cell transparency can occur for optically thick cells. This effect, which we discovered in the course of systematic studies of the spin-exchange process, is theoretically and experimentally analyzed in this paper.

For the case of near-resonant D_1 circularly polarized light propagating along the direction $\hat{z}$ of the external magnetic field, under the typical conditions of spin-exchange optical pumping the photon spectral flux density $\phi_0(\nu)$ [photons/(sec cm Hz)] obeys the simple propagation equation [1]

$$\frac{d\phi_0(\nu)}{dz} = -n\sigma(\nu)\phi_0(\nu)(1-P), \tag{1}$$

where P is the electron spin polarization of the alkali-metal atoms, $\sigma(\nu)$ the absorption cross section at frequency ν, and n the alkali-metal vapor density. The spin polarization is a balance between the pumping rate $R=\int d\nu\sigma(\nu)\phi_0(\nu)$ and the spin-relaxation rate Γ:

$$P = \frac{R}{R+\Gamma}. \tag{2}$$

Using this, and integrating Eq. (1) over frequency, we get for the photon flux density $\phi_0 = \int d\nu\phi_0(\nu)$,

$$\frac{d\phi_0}{dz} = -n\left(\frac{R\Gamma}{R+\Gamma}\right) = -n\Gamma P, \tag{3}$$

which simply expresses the fact that photons are removed from the laser beam at the rate ΓP per atom.

When the light propagates at an angle θ to the external magnetic field, we show below that the propagation obeys

$$\frac{d\phi_\theta}{d\zeta} = -n\left(\frac{R(R\sin^2\theta+\Gamma)}{R+\Gamma}\right), \tag{4}$$

where ζ is the position along the propagation direction of the light. Comparison with Eq. (3) indicates that light absorption exceeds the $\theta=0$ case by a factor

$$\Upsilon = 1+\frac{R\sin^2\theta}{\Gamma}. \tag{5}$$

To see the importance of this effect, consider optical pumping with a 50-W, 1-THz-broad 795-nm laser with a cell area of 40 cm^2. Since the laser linewidth is much broader than the atomic linewidth, we estimate

$$R \approx \phi(\nu)\int d\nu\sigma(\nu) = \phi(\nu)\pi r_e f c = 4.4\times 10^4\ \mathrm{s}^{-1}. \tag{6}$$

A typical relaxation rate for ^{3}He might be 500 s^{-1}, implying $R/\Gamma=90$, so that an angle of only 0.11 rad between the light propagation direction and the magnetic field doubles the light absorption rate at the entrance to the cell. Imperfect cancellation of typical stray laboratory magnetic fields or divergence of the laser beam can easily give angles of this size.

Alternatively, in the absence of skew propagation the minimum flux density required to spin polarize a cell of length l is $\phi_{0\,\mathrm{min}}=n\Gamma l$. Skew propagation increases this, by a factor of Υ, to $nl\Upsilon\Gamma \approx nlR\sin^2\theta \sim nl\sigma\phi_{0\,\mathrm{min}}\sin^2\theta$. Thus the light absorption roughly doubles at a propagation angle of $\sim 1/\sqrt{nl\sigma}$, which can be quite small for highly optically thick samples.

The data of Fig. 1 clearly illustrate the effect. We used a 15-W frequency narrowed diode laser [4] sent along the axis of a 5-cm-long, 4.5-cm-diameter cylindrical Rb vapor cell

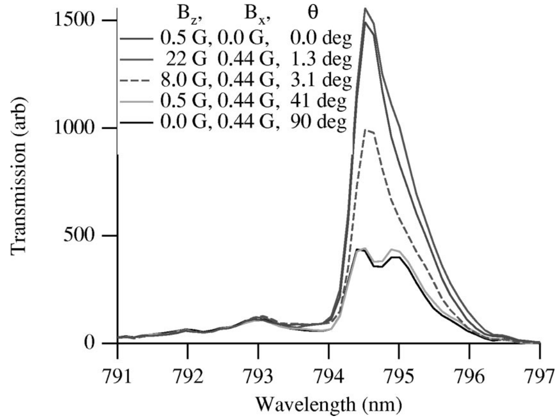

FIG. 1. Light transmission for various magnetic-field directions and magnitudes. The transverse field of 0.44 G is simply the Earth's field that is cancelled for the 0.5-G data.

containing 0.85 bar of ^{3}He and 65 mbar of N_2. The cell is heated to 185 °C, corresponding to a line center optical depth for unpolarized atoms of about 600. We used a low-resolution spectrometer to indicate the laser spectrum transmitted by the cell under various magnetic-field conditions. When the magnetic field is parallel to the light propagation direction, we observed high transmission of the light, which hardly differs for fields varying from 0.5 G to 20 G. When the magnetic-field direction was rotated by a mere 3.1°, nearly half the light is attenuated and for larger angles the attenuation becomes even more severe.

As a second illustration of the effect, we show in Fig. 2 how the addition of a collimating lens just in front of the optical pumping cell highly improves the Rb polarization under conditions where the pump light is being nearly depleted by absorption. The cell is heated to 200 °C, with an optical depth over 1000, nearly fully depleting the 17-W laser. Using light with a radius of curvature of 75 cm at the cell gives a Rb polarization of about 45% as deduced using electron paramagnetic resonance (EPR) spectroscopy [9,10] The addition of a collimating lens increases the polarization to 80%.

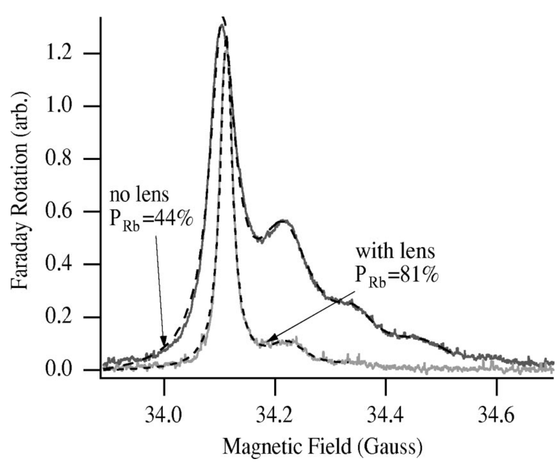

FIG. 2. Rb EPR spectra taken with divergent and collimated light. The main peak is the $F=3,\ m=3 \rightarrow F=3,\ m=2$ transition in ^{85}Rb, while the second peak is the unresolved $F=3,\ m=2 \rightarrow F=3,\ m=1$ and $F=2,\ m=2 \rightarrow F=2,\ m=1$ pair.

The propation of plane light waves with electric field amplitude $\mathbf{E}$ and propagation vector $\mathbf{k}$ through an anisotropic vapor with susceptibility tensor $\overleftrightarrow{\chi}$ obeys

$$(k^2 - \mathbf{kk}\cdot)\mathbf{E} = k_0^2(1 + 4\pi\overleftrightarrow{\chi}\cdot)\mathbf{E}. \tag{7}$$

The vapor is dilute $(4\pi\chi \ll 1)$, so to an excellent approximation the electric field is transverse. We therefore write the electric field as the superposition

$$\mathbf{E} = E_a\hat{a} + E_b\hat{b}, \tag{8}$$

where $\hat{a} = \hat{z}\times\hat{k}/\sin\theta$ and $\hat{b} = \hat{k}\times\hat{a}$ are unit basis vectors orthogonal to $\hat{k}$.

Under typical spin-exchange optical pumping conditions (rapid alkali-alkali spin exchange, strong pressure broadening, rapid electron randomization in the excited state, and negligible ground-state coherences) the alkali-metal susceptibility can be written in the simple form [11]

$$\overleftrightarrow{\chi} = \chi_0(1 - iP\hat{z}\times) \tag{9}$$

and so we have the matrix elements

$$\hat{a}\cdot\overleftrightarrow{\chi}\cdot\hat{a} = \hat{b}\cdot\overleftrightarrow{\chi}\cdot\hat{b} = \chi_0; \quad \hat{b}\cdot\overleftrightarrow{\chi}\cdot\hat{a} = -iP\chi_0\cos\theta \tag{10}$$

and the eigenvectors and eigenvalues of $\overleftrightarrow{\chi}$ are

$$\mathbf{e}_\pm = \frac{\hat{a}\pm i\hat{b}}{\sqrt{2}}, \quad \chi_\pm = \chi_0(1 \mp P\cos\theta). \tag{11}$$

For a highly polarized vapor $P\sim 1$, the $\mathbf{e}_-$ mode is rapidly attenuated at the entrance to the vapor cell so that in the interior of the cell the light is primarily in the $\mathbf{e}_+$ mode. Note, however, that the transparency of this mode is not perfect even for $P=1$ due to the $\cos\theta$ factor; the attenuation of the light obeys

$$\frac{d\phi_\theta(\nu)}{d\zeta} = -n\sigma(\nu)\phi_\theta(\nu)(1 - P\cos\theta). \tag{12}$$

We now consider how the optical pumping is affected by the imperfect polarization of the light along the magnetic-field direction. The $\mathbf{e}_+$ light can be resolved into σ^+, π, and σ^- polarization components containing fractions

$$f_+ = \cos^4(\theta/2),$$
$$f_\pi = 2\cos^2(\theta/2)\sin^2(\theta/2), \tag{13}$$
$$f_- = \sin^4(\theta/2),$$

of the light flux. In steady state, the rate at which angular momentum is added to the alkali-metal atoms by depopulation pumping from the $m_s = -1/2$ state must be balanced by the rate at which angular momentum is removed by depopulation pumping from the $m_s = 1/2$ state and by spin relaxation

$$R\left(\frac{f_\pi}{2}+f_+\right)\left(\frac{1}{2}-\frac{P}{2}\right)=R\left(\frac{f_\pi}{2}+f_-\right)\left(\frac{1}{2}+\frac{P}{2}\right)+\frac{\Gamma P}{2}, \quad (14)$$

which gives

$$P=\frac{R(f_+-f_-)}{R+\Gamma}=\frac{R\cos\theta}{R+\Gamma}. \quad (15)$$

Combining this with Eq. (12) gives us Eq. (4), which is the main result of this paper.

Normally the existence of a small amount of improperly polarized light is not a cause of worry for optical pumping of optically very thick cells, since the "wrong" polarization components are rapidly attenuated at the cell entrance. One might think that this would also hold for light propagating at a small angle to the magnetic-field direction. However, the requirement of transversality of the electric field means that the stable polarization mode for light that is not propagating along the magnetic-field direction is imperfectly polarized. This means that the atoms do not become completely transparent to the light even in the absence of spin relaxation and therefore the attenuation of the light can be substantially greater than that required to simply compensate for spin relaxation, and the efficiency of the optical pumping process is correspondingly reduced. Even when the light beam is carefully aligned along the magnetic-field direction, care should be taken in order to minimize excess absorption due to the divergence of the light and magnetic-field inhomogeneities. Note that optical pumping of potassium, with spin-relaxation rates typically ten times smaller than Rb [12], should be correspondingly more sensitive to transverse magnetic fields. Therefore to realize the predicted gains in K-He spin exchange as compared to Rb-He spin exchange [12] it will be necessary to carefully align the magnetic field and laser propagation directions.

We have shown that unless careful attention is paid to the optical train used to couple laser light to the cell, and to the direction of the magnetic field in the cell, it is easy to greatly increase the amount of light required to pump a given sample of alkali atoms to high polarizations under optically thick conditions. This effect is most important for optical pumping of ^{3}He, where slow spin-relaxation allows for optically very thick samples, and where unnecessary heating of the cell should be avoided due to convection and increased Rb-He spin-relaxation rates [12,13].

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