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Polarization Limits in K-Rb Spin-Exchange Mixtures

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We present measurements of the optical absorption of K vapor at 795 nm due to the presence of high pressure He gas. The results set a limit on the polarization attainable in hybrid spin-exchange optical pumping of ^{3}He.

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Hybrid spin-exchange optical pumping [1] spin polarizes ^{3}He through spin-exchange collisions with an optically pumped mixture of K and Rb atoms. A dilute vapor of Rb is optically pumped in the usual manner [2], and polarizes a denser vapor of K by Rb-K spin-exchange collisions. ^{3}He nuclei then become polarized through K-^{3}He spin-exchange collisions. The method takes advantage of the high efficiency of spin-exchange between K and ^{3}He [3], while retaining the convenience of using 795 nm diode array bars for optical pumping of Rb. Current state-of-the-art neutron spin-filters [4, 5] and spin-polarized targets [6, 7] utilize hybrid spin-exchange.

As long as the pumping light is sufficiently intense, the polarization achieved in hybrid spin-exchange experiments should be virtually the same as for single-species pumping. However, several experiments [1, 8–10] have found that at high K/Rb density ratios $\mathcal{D}$ it is not possible to optically pump the alkali atoms to full polarization, even at very high optical pumping rates. As proposed in [1], the most natural explanation for this is that there is weak off-resonant optical pumping of the K atoms by the 795 nm pumping light. Assuming no spin-dependence to this rate, this acts as a light-induced spin relaxation mechanism that keeps the atoms from becoming fully polarized. If the alkali-metal atoms are in spin-temperature equilibrium, so that their electronic spin-polarizations P are equal, and ground state spin relaxation can be ignored, the optical pumping equation becomes

$$[\mathrm{Rb}]\frac{dF_R}{dt}+[\mathrm{K}]\frac{dF_K}{dt}=[\mathrm{Rb}]\frac{R}{2}(P_\infty-P)-[\mathrm{K}]\frac{R_K}{2}P \qquad (1)$$

which is basically a statement of angular momentum conservation. The total angular momentum density $[\mathrm{Rb}]F_R+[\mathrm{K}]F_K$ of the Rb and K atoms increases by optical pumping of the Rb atoms at a rate R, increasing P towards its maximum possible value P_∞ [11]. The angular momentum is also lost by light absorption at a rate R_K by the potassium atoms. The factors of 1/2 assume relaxation of the electronic angular momentum in the excited state, and that sufficient N_2 quenching gas is included in the cell so that the nuclear spin is conserved in the excited-state [12].

In steady-state, the polarization becomes

$$P=P_\infty\frac{R}{R+\mathcal{D}R_K} \qquad (2)$$

so the spin-polarization is significantly reduced when $\mathcal{D}R_K$ becomes comparable to R. Since both R and R_K are proportional to the pumping light intensity, the attainable polarization saturates at a value less than P_∞. As has been noted before [1, 11], the extreme optical depths of SEOP experiments make them particularly sensitive to such polarization limiting processes.

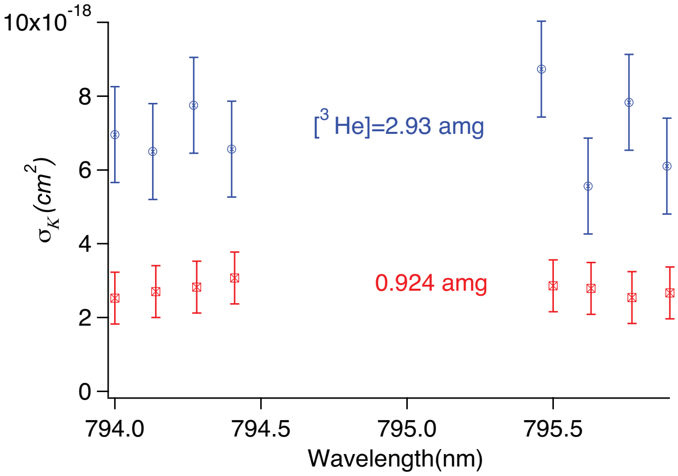

FIG. 1. Potassium absorption cross-section σ_K as a function of wavelength in the vicinity of the Rb resonance at 795 nm, for two different ^{3}He densities.

Using the apparatus described in Ref. [11], we have measured the absorption cross section σ_K for unpolarized K atoms near the Rb pumping wavelength of 795 nm. We observed the transmission $1-e^{-n\sigma l}$ of a weak (50 μW) linearly-polarized laser beam through 2 potassium cells, one l = 4.8 cm diameter sphere containing 0.063 amg of N_2 and 2.93 amg of ^{3}He and an l = 5.7 cm sphere with 0.083 amg N_2 and 0.924 amg of ^{3}He. The measured transmissions ranged from 99% to 99.9% depending on the cell and temperature used. The transmission of a second probe beam at 855 nm, spatially overlapped with the first, was also monitored to account for any drifts in the cell transmissions that occur due to K droplet formation on the cell walls. The potassium density n and the helium density [^{3}He] were deduced from K line-center absorption spectroscopy using the recently measured K-^{3}He lineshape [10] with an assumed value for the line-broadening asymmetry parameter of zero.

Fig. 1 shows the absorption cross-sections in the two cells found over a range of wavelengths near 795 nm.

Due to easily observable 1/10000 Rb contamination of these nominally pure K cells, we avoided measurements directly on the Rb resonance. At the detunings used, Rb absorption was negligible. At each wavelength, the absorption was measured at several temperatures, corresponding to several potassium densities. A plot of the optical depth versus potassium density was made, and the cross section for absorption deduced from dividing the slope of a linear fit by the length l of each cell.

The absorption cross section has no discernable frequency dependence, so the values at each wavelength were averaged to get the absorption cross section at 795 nm for each cell. The ratio of the absorption cross sections is, within uncertainty, equal to the ratio of the buffer gas densities in the two cells. Since the ^{3}He densities greatly exceed the N_2 densities, the dominant absorption process must be K-^{3}He collisions. Combining the results for the two cells we obtain the K-^{3}He and K-N_2 cross sections

$$\sigma_{\mathrm{K-He}} = (2.19 \pm .39) \times 10^{-18} \frac{\mathrm{cm}^2}{\mathrm{amg}} [^3\mathrm{He}] \tag{3}$$

and

$$\sigma_{\mathrm{K-N_2}} = (8.8 \pm 7.6) \times 10^{-18} \frac{\mathrm{cm}^2}{\mathrm{amg}} [\mathrm{N_2}] \tag{4}$$

The large uncertainty in the N2 cross-section is due to its small abundance in the two cells. The uncertainty in the measurements arises mainly from etalon effects on the transmission of the 795 nm and 850 nm probe beams. Etalon effects contributed an uncertainty of 0.11% to the transmission of both beams in the high ^{3}He cell, and 0.07% to the transmission in the low ^{3}He cell. In the low ^{3}He density cell there is also a 0.10% percent uncertainty in correction of transmission changes due to migration of K droplets on the cell wall. Since the measured transmissions were in excess of 99%, these etalon effects dominate the final cross section uncertainties. There are also small contributions from uncertainty in the path length (2.5% in the high ^{3}He cell and 2.1% in the low ^{3}He cell).

The cross section being measured here corresponds to absorption in the quasistatic wings of the K resonance line[13]. The spin-dependence of the absorption is discussed in our previous work [11]. Since the light is detuned by about 8 times the K fine-structure splitting, the absorption cross section is nearly spin-independent. Recent theoretical investigation of K-^{4}He far wing line broadening in the context of understanding the spectra of cool brown dwarfs [14] gives $\sigma_{\mathrm{K-^4He}} = 2.7 \times 10^{-18} \frac{\mathrm{cm}^2}{\mathrm{amg}} [^4\mathrm{He}]$ at T=500K, within 2σ of our result.

The observed cross section is in reasonable agreement with the value $R_K/R = 2.2 \times 10^{-3}$ inferred by Babcock *et al.* from the $\mathcal{D}$ dependence of P_{max}, the alkali-metal polarization extrapolated to infinite pumping power[1]. Those results were obtained using an unnarrowed diode array bar as the pumping source for $[^3\mathrm{He}] = 8$ amg cells. Solving Eq. 2 under those conditions with very large pump power, P_∞ from [11] and our measured K-^{3}He cross section, produces a curve in good agreement with the experiment at high $\mathcal{D}$ (Fig. 2). (A linewidth of 1000 GHz was chosen for the pump laser, as an estimate of the unknown linewidth of the laser used in that experiment.) Note that without potassium absorption, the model predicts high maximum alkali-metal polarization even at high $\mathcal{D}$. At low $\mathcal{D}$, the observed polarizations were smaller than our simulations suggest, as discussed in detail previously [11].

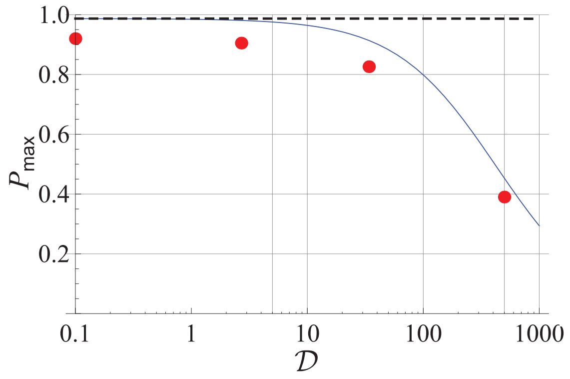

FIG. 2. Comparison of measured [1] and modeled maximum achievable alkali polarization as a function of K/Rb density ratio $\mathcal{D}$ in a $[^3\mathrm{He}]$=8.0 amg hybrid cell pumped by a 1000 GHz bandwidth source. Modeling was done with (solid blue) and without (dashed black) off-resonant potassium absorption.

The effect of K-^{3}He absorption on optical pumping of $[^3\mathrm{He}]$=8 amg cells with realistic pump power is shown

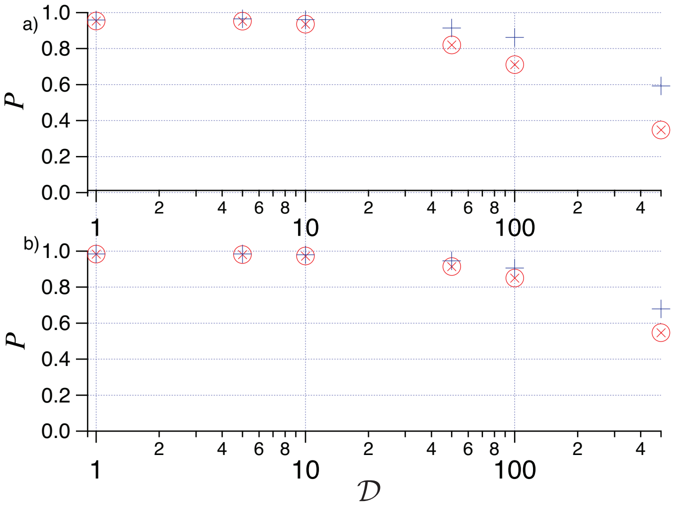

FIG. 3. Modeled average alkali polarization as a function of K/Rb density ratio $\mathcal{D}$ in a 7.9 cm long $[^3\mathrm{He}]$=8.0 amg hybrid pumping cell at T=210°C with (red) and without (blue) K-^{3}He absorption, for a) a 100 W broadband 800GHz pumping source and b) a 50 W narrowband 125 GHz pumping source.

$\mathcal{D}$	P(Expt)	Theory, $R_K = 0$	Theory, $R_K \neq 0$
6.2	∼.99	.987	.986
46	.77	.960	.954
155	.62	.838	.825

TABLE I. Comparison of measured [8] and predicted alkali polarizations P in [^{3}He] ~ 1.5 amg cells of increasing $\mathcal{D}$. The low polarizations in high $\mathcal{D}$ cells at these helium densities are not explained by potassium absorption.

in Fig. 3. The average alkali polarization under typical conditions is calculated with and without potassium absorption for a range of values of $\mathcal{D}$ using an optical pumping model that includes the effects of the ground state spin relaxation, spin relaxation at the cell walls, $\sigma_{\mathrm{K}-^3\mathrm{He}}$, P_∞, excited state nuclear spin relaxation [12], ground state hyperfine splitting, and pump laser propagation. For a 100 W broadband source at $\mathcal{D}$ above 5 the contribution from K-^{3}He absorption becomes significant, and above $\mathcal{D}$=10 the alkali polarization quickly drops below 0.90. It should be noted that the alkali polarization is limited to ~ 0.95 even at low $\mathcal{D}$, largely due to the Rb-^{3}He P_∞ effect. With a 50 W narrowband source, the K-^{3}He absorption noticeably reduces the alkali polarization, but P_{Rb} is above 0.90 up to a $\mathcal{D}$ of 50.

Alkali spin-polarization limits in high D cells of modest helium density(1-2 amg), pumped with narrowband ($\sim$ 100 GHz) laser sources, were reported in [8]. Chen *et al.* observed a decrease in the measured ^{3}He polarization with increasing $\mathcal{D}$ in three cells of $\mathcal{D}$=6.2, 46 and 155, and [^{3}He]=1.4, 1.9, and 1.1 amg respectively. From measurements of the ^{3}He polarizations, they inferred alkali polarizations of only 0.77 in the $\mathcal{D}$=46 cell and 0.62 in the $\mathcal{D}$=155 cell. There is an expectation that, with a fixed amount of laser power, the alkali-metal polarization should decrease with increasing $\mathcal{D}$ due to increased effective relaxation rate for the Rb atoms. For the conditions described in [8], our optical pumping simulations indicate this effect only accounts for a fraction of the observed drop in P (Table I). However, potassium absorption also has little effect in narrowband pumping of low [^{3}He] cells, and does not help explain the observed low alkali polarizations. Although narrowband pumping gives better performance than broadband pumping, it is not as big an improvement as modeling would suggest [8].

Our measurement of the off-resonant pumping rate for K atoms explains the reduced performance of hybrid spin-exchange optical pumping at high K/Rb ratios, but only for gas densities of several amagat. Our simulations predict that the use of lower densities and narrower laser linewidths should greatly reduce off-resonant pumping effects. Nevertheless, it is well-documented experimentally [8, 10] that the polarization still drops in high density ratio, low pressure hybrid cells pumped by narrowband light. Thus there must be another as yet unknown mechanism at work, perhaps associated with the signficantly higher optical pumping rates in low pressure, narrowband experiments.

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