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OPTICAL PUMPING AT HIGH TEMPERATURES*

L. C. Balling, R. H. Lambert, J. J. Wright, and R. E. Weiss
Department of Physics, University of New Hampshire, Durham, New Hampshire
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Optical pumping signals with Rb have been detected at temperatures ranging from 50 to 850°C. Spin-exchange signals have been detected with Ag (at 750°C), Li (at 400°C), and with electrons, hydrogen, and nitrogen over the entire temperature range. A preliminary value for the Li^6 hfs separation is given.

We report here a significant extension of optical pumping technique[1-5] that will greatly increase its utility in radio-frequency spectroscopy. With the use of this technique one can perform optical pumping experiments over an extended temperature range. We have used spin-exchange collisions between oriented rubidium and silver at 750°C to detect the Zeeman transitions in the silver[6] in order to demonstrate the potential of the technique. Measurements of the hfs and its pressure shifts for the lithium isotopes are in progress and we report here preliminary results. The technique has also been successfully applied to spin-exchange polarization of electrons, atomic hydrogen, and atomic nitrogen at temperatures up to 850°C. Besides considerably increasing the number of elements that can be polarized, this relatively simple technique enables one to investigate the energy dependence of both spin-exchange cross sections, spin-relaxation times, and hyperfine pressure shifts.

For the sake of illustration we describe the silver experiment in detail. The basic optical pumping arrangement is shown in Fig. 1. Circularly polarized Rb-$D1$ radiation is used to polarize the Rb atoms in a 300-cm^3 flask containing a buffer gas and atomic silver. The absorption flask is in an oven whose temperature varied from 700 to 750°C during the course of the measurements. At this temperature silver has a vapor pressure of approximately 10^{-5} Torr. Spin-exchange collisions between the silver atoms and the polarized Rb atoms orient the silver atoms. The equilibrium polarization of the Rb, and thus the amount of light absorption, is altered when a resonant rf magnetic field is applied to the silver atoms. The rf field is amplitude modulated and the signal is detected in a phase-sensitive detector.

Because of the exponential temperature dependence of the alkali vapor pressure, optical pumping has been possible in the past only over a very

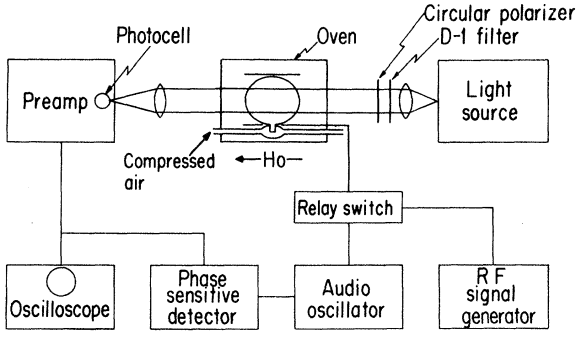

FIG. 1. Basic optical-pumping arrangement. Helmholtz coils are not shown.

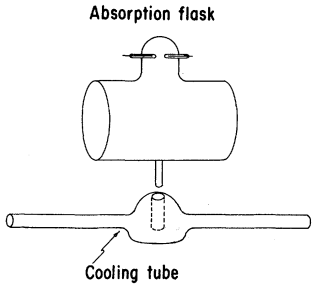

FIG. 2. Pyrex optical-pumping flask. The quartz flask used in the Ag experiment was spherical, had no discharge electrodes, and had an additional small side arm for the Ag.

limited temperature range. In order to optically pump Rb in a flask at high temperature, one must control the Rb density independently of the temperature of the flask.

The method used here[7] for simultaneously controlling the vapor density of both the Rb and Ag can be understood from Fig. 2. The silver is deposited in the absorption flask, and the silver vapor pressure is controlled by the temperature of the flask. The Rb is contained in a side arm which is air cooled. The density of the Rb in the flask is controlled by the amount of air flowing through the cooling tube.

For the silver experiment a quartz flask was used. For optical pumping experiments below 550°C, Pyrex is more convenient. The flasks are prepared on a vacuum system in the conventional way except that during the bakeout (at least 50°C higher then the maximum operating temperature) the Rb is thoroughly driven out of the flask walls and into the side arm which is cooled for this purpose. The flasks are then removed from the vacuum system. If they have been prepared in this way the flasks can be heated to any desired temperature in the optical-pumping apparatus while the Rb density is controlled by the cooling air to maximize the optical pumping signal. Control of the Rb density is possible regardless of the pressure or type of buffer gas used. We have successfully used flasks with pressures ranging from 20 to 500 Torr. The flasks can be stored and reused again just like conventional optical-pumping flasks.

Figure 3 is a tracing of the silver Zeeman lines, all of which overlap in the small horizontal components of the earth's field used. The oven was relatively unstabilized for the silver measurements, and the readings were taken while it cycled between 750 and 700°C. In a weak rf field the linewidth of the silver Zeeman line (1400 Hz/mG) was approximately 145 Hz.[8] This compared with the Rb^{87} line (700 Hz/mG) which had a width of approximately 75 Hz, indicating that most of the width was caused by magnetic-field inhomogeneity. The high signal-to-noise ratio in this spin-exchange signal is comparable to the signal-to-noise ratio typically obtained in conventional optical pumping experiments.

Both silver isotopes 107 and 109 were present in approximately equal abundance, and each has $I=\frac{1}{2}$ and $J=\frac{1}{2}$. In order to confirm that the resonances were in fact silver, a larger horizontal magnetic field was added (3 G) and the expected two pairs of lines were observed with the correct splittings expected for the known silver splittings.[9] The size of the split signals indicate that there will be no difficulty measuring the hyperfine splittings.

The lithium measurements are being carried out at 400°C (determined by the lithium vapor pressure required for optimum spin-exchange signals with Rb). The apparatus and procedures for preparing the flasks are basically the same as that described for the silver experiment. The measurements are being made with a variety of buffer gases, and both temperature and pressure shifts of the hfs will be determined. The preliminary value of the hfs separation constant for Li^6 is $\Delta\nu=\frac{3}{2}A=228\,205\,263(15)$ Hz.

The application of the technique to electrons, nitrogen, and hydrogen differ only in that electrodes were added to produce the second species. We have not gone higher than 850°C because some

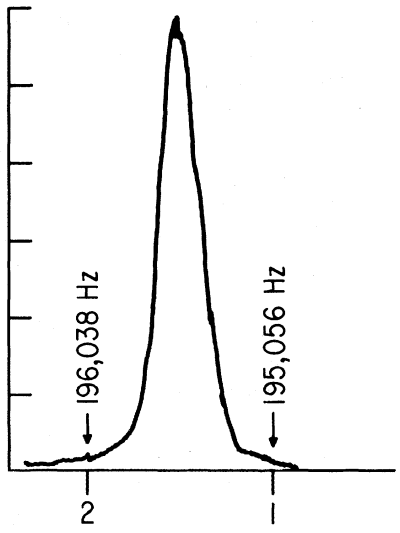

FIG. 3. Tracing of Ag Zeeman signal. The phase-sensitive detector had a 0.03-sec time constant.

of the components in the oven are Pyrex—there does not appear to be any physical reason why the technique could not be extended to high temperatures.

Optical pumping utilizing the alkalis has in the past provided very precise values for free-atom hfs and g_J values. The high precision is especially useful in looking for effects caused by higher order multipole interactions. The present technique will make possible the pumping of a considerable number of new elements—As, Sb, Bi, Cr, Mn, Mo, Tc, Re, Cu, Ag, Au, Eu, and Am all have orbital S states with $J \neq 0$ and therefore can potentially be polarized by spin-exchange collisions.

Several aspects of interatomic interactions have been studied with optical pumping. These include pressure shifts of hfs[10] and g_J values,[11] spin-exchange cross sections,[2-5] spin-relaxation cross sections, and diffusion coefficients.[12] The ability to measure these quantities over an extended temperature range should greatly assist in their understanding. Of particular interest is the energy dependence of the electron-alkali spin-exchange cross section.[13] It is also possible that the utility of optical pumping in chemistry may be increased.

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DETERMINATION OF MUONIUM HYPERFINE STRUCTURE INTERVAL THROUGH MEASUREMENTS AT LOW MAGNETIC FIELDS*

P. A. Thompson, J. J. Amato, P. Crane, V. W. Hughes, R. M. Mobley, G. zu Putlitz, and J. E. Rothberg
Gibbs Laboratory, Physics Department, Yale University, New Haven, Connecticut
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The ground-state hyperfine structure interval $\Delta\nu$ of muonium has been remeasured with significantly improved accuracy through the observation of transitions between hfs states at low magnetic fields. The formation of muonium has been observed for the first time in krypton and the fractional pressure shift of $\Delta\nu$ has been measured in this gas. The average value of $\Delta\nu$ obtained from these measurements is $\Delta\nu = 4463.26 \pm 0.04$ Mc/sec.

The hyperfine structure interval $\Delta\nu$ of the muonium atom (μ^+e^-) in its ground state has been remeasured with improved accuracy through the observation of transitions between magnetic sublevels at weak magnetic field (about 3 G)[1] and at very weak magnetic field (about 10 mG). Compared with the earlier high-field experiment,[2,3] these measurements involve some different experimental techniques and thus provide an independent determination of $\Delta\nu$. The more accurate value for $\Delta\nu$ can be used to obtain a value for the fine structure constant α, or to determine a value for the muon magnetic moment, μ_μ, free from uncertainties of magnetic shielding.[4]

In a low external static magnetic field H, the relative energies of the sublevels of muonium are