Enhanced polarization of electron spins in ^{4}He by simultaneous optical pumping with circularly and linearly polarized resonance radiation

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The polarization of helium ($2\,^3S_1$) metastable atoms saturates at 50% when the ensemble is optically pumped with the D_0 ($2\,^3S_1$–$2\,^3P_0$) transition. We have obtained polarizations approaching 100% with a simple, straightforward modification to the conventional experimental arrangement. The addition of a small amount of π-polarized D_0 light incident on the cell perpendicular to the defining field direction doubles the polarization produced by circularly polarized σ^+ light along the field direction. A simple rate-equation model illustrates the enhancement.

INTRODUCTION

Conventional optical pumping of helium metastable atoms uses a discharge lamp as a source of the resonance radiation for the pumping process.[1,2] The pumping radiation consists of three lines connecting the $2\,^3S_1$ metastable level to the $2\,^3P_{0,1,2}$ levels. A simple rate-equation analysis of the pumping equations shows that the optical pumping process is very inefficient with all-line pumping. Since the transitions are all within 0.1 nm, it is not possible to efficiently isolate one of them; thus, when high polarizations of the electron-spin system are desired, the use of a discharge lamp is unsatisfactory.

With the current availability of tunable lasers at the helium resonance transitions, it is now possible to optically pump ensembles of ^{4}He metastable atoms with a much greater efficiency than previously. One can show theoretically and substantiate experimentally that electron-spin polarizations of 50% and 100% are available when samples are optically pumped with D_0 ($2\,^3S_1-2\,^3P_0$) and D_1 ($2\,^3S_1-2\,^3P_1$) radiation, respectively. D_2 ($2\,^3S_1-2\,^3P_2$) light, although it is the most strongly absorbed transition, is generally ineffective in optical pumping and under certain conditions can even orient the spins in the opposite direction to the other two components.[3]

The Doppler width of the absorbing helium atoms at room temperature is, unfortunately, comparable to the separation of the D_1 and D_2 lines and the two lines are incompletely resolved. Thus, even with a narrow line laser, the D_1 and D_2 transitions cannot be effectively isolated. Figure 1, for example, is the fluorescence signal obtained when a narrow-line (<200 MHz). Nd-doped lanthanum hexaluminate (LNA) laser[4,5] is scanned through the helium transitions. When using D_1 light to pump the sample, care must be exercised to avoid any excitation of the D_2 transition. This is difficult at the higher laser powers required for the limiting polarization since the wings of the D_2 transition can be excited by D_1 light. It has thus been necessary to tune the laser slightly to the "blue" side of the D_1 transition in order to obtain high polarizations.

This presents two difficulties. The laser frequency tends to drift over time so that it must be stabilized; tuning to the side of a transition introduces additional complications. Also, one is limited to laser systems in which the laser emission bandwidth is much less than the 1.7-GHz Doppler width. An alternative possibility is to pump with the D_0 component, which is well isolated, and sacrifice the possibility of obtaining polarizations approaching 100%. It is the purpose of this paper to describe a simple modification to the conventional optical pumping technique which permits the use of D_0 pumping light while simultaneously offering the possibility of obtaining a 100% spin polarization of the metastable helium atoms.

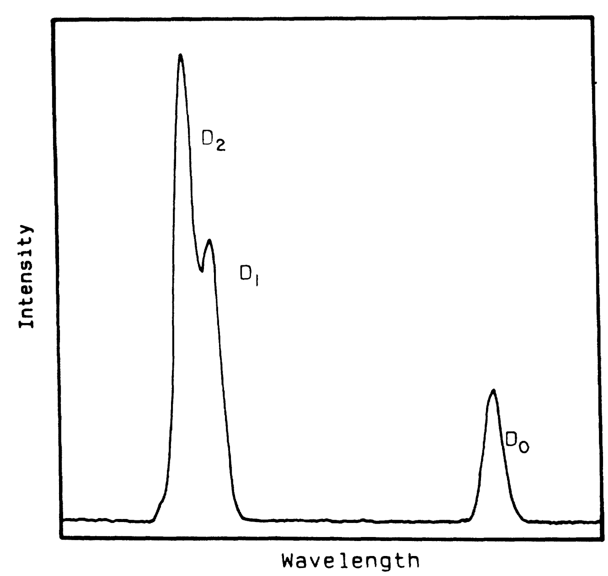

FIG. 1. Fluorescence spectrum obtained from a ^{4}He discharge cell as a narrow-band LNA laser is tuned through the metastable resonance transitions at 1083 nm. The three peaks correspond to the D_0, D_1, and D_2 lines. The fluorescent intensities are approximately in the ratio of their statistical weights 1:3:5.

RATE EQUATION ANALYSIS OF D_0 PUMPING

Rate equations for the populations of the Zeeman levels of the metastable $2\,^3S_1$ can be simply written for D_0 pumping with circularly polarized light. Atoms are removed from the metastable $m_j=-1$ at a rate determined by the intensity of the pump light. Atoms excited to the $2\,^3P_0, m_j=0$ level return with equal probabilities to the metastable Zeeman levels. The necessary rate equations for the metastable levels are then

$$\frac{dn_{-1}}{dt}=-\frac{n_{-1}}{\tau_p}+\frac{n_{-1}}{3\tau_p}+\left[\frac{n}{3}-n_{-1}\right]\frac{1}{\tau_r}\,,$$

$$\frac{dn_0}{dt}=\frac{n_{-1}}{3\tau_p}+\left[\frac{n}{3}-n_0\right]\frac{1}{\tau_r}\,,$$

where τ_p is the characteristic pumping time and τ_r is the characteristic relaxation time. $n_{-1,0,+1}$ are the densities of the respective m_j levels. The reciprocal of the characteristic pumping time is proportional to the intensity of the pump beam. Solving the above for the level populations we obtain

$$P=\frac{n_{+1}-n_{-1}}{n}=\frac{\alpha}{2\alpha+3}\,,$$

where P is the electron spin polarization and α is τ_r/τ_p.

It is seen that at high pumping light intensities, the limiting polarization is 50% if right circularly polarized D_0 light alone is used. If one now adds linearly polarized light perpendicular to the field direction, which induces transitions between the $m_j=0$ Zeeman levels, a new set of rate equations is obtained:

$$\frac{dn_{-1}}{dt}=-\frac{n_{-1}}{\tau_p}+\frac{n_{-1}}{3\tau_p}+\frac{n_0}{3\tau_p^*}+\left[\frac{n}{3}-n_{-1}\right]\frac{1}{\tau_r}\,,$$

$$\frac{dn_0}{dt}=-\frac{n_0}{\tau^*}+\frac{n_{-1}}{3\tau_p}+\frac{n_0}{3\tau_p^*}+\left[\frac{n}{3}-n_0\right]\frac{1}{\tau_r}\,,$$

with the steady-state solution

$$P=\frac{n_{+1}-n_{-1}}{n}=\frac{\alpha(\alpha^*+1)}{(\alpha^*+1)(\alpha+2)+(\alpha+1)}\,,$$

where τ_p^* is the characteristic pumping time of the π polarized light, with $\alpha^*=\tau_r/\tau_p^*$.

Now the steady-state solution yields a metastable-state polarization which saturates at 100% when both pumping beams are intense. These results are easily visualized by examining the left-hand side of the level diagram of Fig. 2. With right circularly polarized D_0 light alone, only the $m_j=-1$ level population is modified; the relative populations of the $m_j=0$ and $+1$ levels remain unchanged. Linearly polarized π light, however, depletes the $m_j=0$ level population, leading to an enhanced polarization of the electron spins.

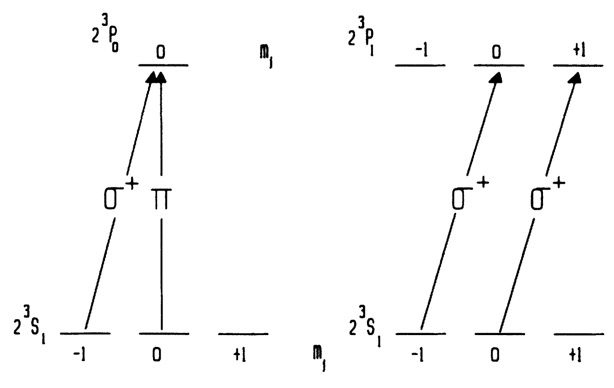

FIG. 2. Abbreviated energy-level diagram. The left-hand side of the figure are the levels involved in the optical pumping process with D_0 light. The slanted line represents absorption by the circularly polarized component and the vertical line, absorption of the linearly polarized light. On the right-hand side of the diagram are the levels involved with the probe-beam absorption. The probe beam is right circularly polarized and is tuned to the D_1 transition. Absorption by the probe beam is a direct measure of the populations of the $m_j=0$ and -1 levels.

EXPERIMENTAL OBSERVATIONS

Figure 3 is a schematic representation of the apparatus. The laser used to pump the metastable ^{4}He ensemble is a recently developed arc-lamp-pumped LNA laser.[6,7] This laser provide up to 3 W on the helium resonance transition at 1082.908 nm. The beam is divided by a beam splitter. The right circularly polarized light with intensities to 1 W is directed into the helium discharge cell parallel to an externally applied magnetic field of several gauss. A second beam from the laser is directed into the cell perpendicular to the field direction. The linear polarization of this laser beam is parallel to the externally applied field direction in order to induce $\Delta m_j=0$ transitions as described above. The intensities of either beam could be varied by introducing appropriate attenuators.

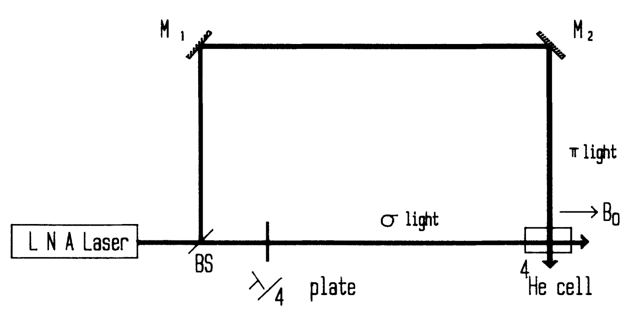

FIG. 3. Schematic representation of the apparatus. BS is a beam splitter. Approximately 30% of the power is split off from the laser beam and with mirrors M_1 and M_2 directed into the discharge cell transverse to the applied field direction B_0. The field intensity is about 3 G. The LNA laser output is linearly polarized; the quarter wave plate shown circularly polarizes the beam. The laser intensity incident on the discharge cell is changed by inserting neutral density filters into the beam path.

The electron-spin polarization of the metastable atoms was observed in either or both of two ways. In the first and easiest method we used the spin-dependent optogalvanic effect to determine the ensemble polarization.[8] This method relies on a change in the cell impedance for detection. The cell impedance may be shown to depend directly on the orientation of the metastable spins. The second, perhaps more reliable albeit more complicated method, utilizes a weak probe beam to monitor the population of the $m_j=-1$ and 0 levels by optical absorption. Each method and the results obtained are described in the sections following.

OPTOGALVANIC DETECTION OF POLARIZATION

The spin-dependent optogalvanic effect was described earlier.[8] Only its essential elements are repeated here. In the weak electrical discharge of the type used for helium optical pumping, a significant source of the free electrons in the discharge cell comes from metastable-metastable collisions,[9,10] the free electrons determining the plasma conductivity. The discharge impedance can then be modified if the cross section $He^m+He^m \rightarrow He^+ + He + e$ for the production of free electrons in the discharge can be modified. Since this collision reaction must satisfy spin conservation rules, it is easily shown that if both metastable atoms are in the same $m_j=+1$ or -1 state, spin cannot be conserved and the cross section goes to zero. Thus the cell conductivity is a measure of the ensemble polarization.

In Fig. 4 we show the results obtained. The optogalvanic signal obtained when using circularly polarized D_0 light to pump the metastable atoms saturates when the pump power of our LNA laser reaches about 80 mW as shown by the open triangles. If now a small amount of π polarized, D_0 light is directed along an axis perpendicular to the defining field direction, the polarization dramatically increases as illustrated by the solid triangles of Fig. 4. The polarization now saturates at twice the signal previously observed. No further increase in the optogalvanic signal is observed with an increase of either the circularly or linearly polarized components.

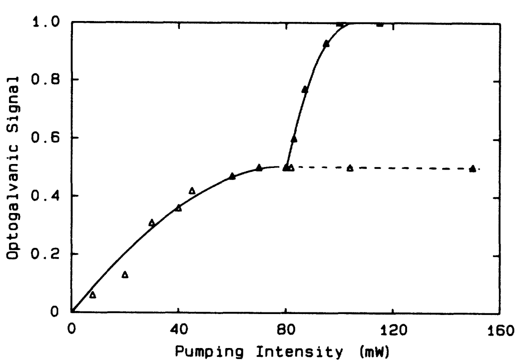

FIG. 4. Spin-dependent, optogalvanic signals. These signals are obtained by monitoring the rf-discharge current, which changes as the cell impedance is modified. In a weak helium discharge metastable-metastable collisions are a major source of the discharge sustaining electrons. The cross section for these collisions is spin dependent and is thus a measure of the metastable orientation. The open triangles show the optogalvanic signal when the circularly polarized component alone is present. The signal saturates at about 90 mW. When the linearly polarized component is added, the signal dramatically increases, doubling with the addition of less than 30 mW and saturating at a new value. The presumption is that the new saturation value corresponds to 100% polarization of the metastable spin system.

OPTICAL ABSORPTION MEASUREMENTS

The relative populations of the $m_j=-1$ and 0 levels can be observed directly in optical absorption if a weak probe beam of right circularly light tuned to the D_1 ($2\,^3S_1-2\,^3P_1$) transition at 1083.025 nm is incident on the discharge cell along the direction of the externally applied magnetic field. As is seen from the right-hand side of Fig. 2, this component of light interrogates directly the $m_j=-1$ and 0 populations.

The electron-spin polarization can be related to the change in the optical absorption of the D_1 light, $P=\Delta I/I$, where ΔI is the change in absorbed light as the ensemble is pumped and I is the total absorbed light when the sample is unpolarized. In Fig. 5 the results obtained with the weak probe beam are displayed. The right-hand side of the figure shows the absorption of the probe laser beam when the strong laser is circularly polarized and tuned to the D_0 transition. The optical signal indicates a polarization of 50%. The LNA laser power is about 250 mW. The beam diameter at the entrance to the discharge cell is about 3.0 cm. The weak probe is obtained from a diode-pumped LNA laser.[5] Its bandwidth is less than 40 MHz with a measured intensity of less than 100 μW. The beam is also expanded to a diameter of 1.5 cm.

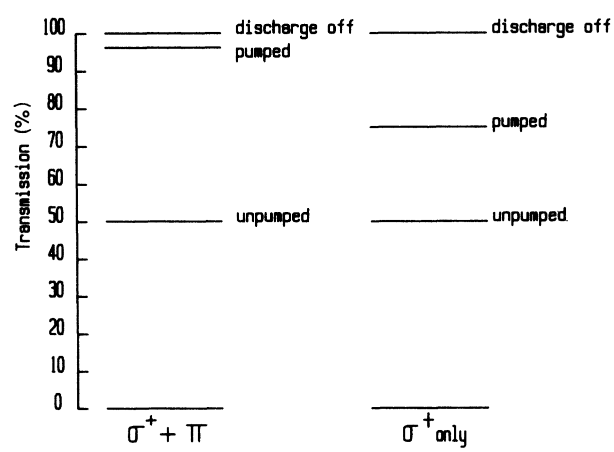

FIG. 5. Optical absorption of a probe laser light beam at the D_1 transition by the helium discharge cell. With the pump laser tuned to the D_0 transition and circularly polarized the transmission increases from 50% to 75% as the metastable atoms are oriented by the optical pumping process (right-hand side of diagram). When the linearly polarized component is added the transmission increases to more than 96% indicating an almost total depletion of the $m_j=-1$ and 0 levels and a near 100% polarization of the electron spins.

If the second linearly polarized component at D_0 is now directed into the cell as shown in Fig. 3, the probe beam absorption drops to less than 4%. The transverse beam power is 100 mW with a diameter of 3.0 cm at the cell entrance. The cell is basically transparent to the D_1 probe beam, indicating that all the atoms have been pumped out of the $m_j=-1$ and 0 absorbing levels and into the $m_j=+1$ level. This corresponds to a metastable polarization of nearly 100%.

APPLICATIONS

Walters and his colleagues have demonstrated a versatile, efficient source of polarized electrons which utilizes the optical pumping process in a flowing helium afterglow.[11] With the results reported here, the pumping efficiency is enhanced and improved polarizations and/or beam currents can be expected.

This modification enables one to use a laser whose bandwidth is comparable to or slightly broader than the Doppler width since the D_0 transition is well separated from the $D_{1,2}$ components. Such a laser is the recently reported arc-lamp-pumped LNA laser.[6,7] This is a very reliable, moderately inexpensive laser that is capable of several watts of output power within the Doppler width of the helium absorption lines. It is an excellent replacement for the costly and inefficient Ar^+-ion-pumped LNA laser[4] or the low-power diode-pumped LNA laser.[5]

CONCLUSIONS

We have demonstrated a simple, but elegant modification to the conventional optical pumping process which yields a ^{4}He metastable spin polarization of nearly 100%. The modification permits the use of an inexpensive, arc-lamp-pumped LNA laser, which is capable of providing several watts of tunable laser emission at the helium transitions. The comparatively large power available from the laser implies that large numbers of metastable atoms can be polarized to nearly 100%. With the Penning ionization process, which is used to efficiently remove one of the polarized electrons from the polarized helium metastable atom, it should be possible to obtain a higher electron polarization at a greater beam current than is currently available.

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[1] F. D. Colegrove and P. A. Franken, Phys. Rev. **119**, 680 (1960).

[2] L. D. Schearer, in *Advances in Quantum Electronics,* edited by J. R. Singer (Columbia, University Press, New York 1961), pp. 239–251.

[3] L. D. Schearer, Phys. Rev. **160**, 76 (1967).

[4] L. D. Schearer, M. Leduc, D. Vivien, A-M. Lejus, and J. Thery, IEEE J. Quantum Electron. **QE-22**, 713 (1986).

[5] J. Hamel, A. Cassimi, H. Abu-Safia, M. Leduc, and L. D. Schearer, Opt. Commun. **63**, 114 (1987).

[6] C. G. Aminoff, C. Larat, M. Leduc, and F. Laloe, Rev. Phys. Appl. **24**, 827 (1989).

[7] L. D. Schearer and P. Tin, J. Appl. Phys. **68**, 943 (1990).

[8] L. D. Schearer and P. Tin, Opt. Soc. Am. B **6**, 1771 (1989).

[9] J. C. Hill, L. L. Hatfield, N. D. Stockwell, and G. K. Walters, Phys. Rev. A **5**, 189 (1972).

[10] L. D. Schearer, Phys. Lett. **33A**, 325 (1970).

[11] L. G. Gray, K. W. Giberson, Chu-Cheng, R. S. Keiffer, F. B. Dunning, and G. K. Walters, Rev. Sci. Instrum. **54**, 271 (1983). See also G. H. Rutherford, J. M. Ratliff, J. G. Lynn, F. B. Dunning, and G. K. Walters, Rev. Sci. Instrum. **61**, 1460 (1990).