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ION POLARIZATION VIA PENNING COLLISIONS WITH OPTICALLY PUMPED METASTABLE HELIUM

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Transfer of spin polarization from optically pumped metastable helium atoms to the ions of Cd, Sr, and Zn following Penning collisions has been demonstrated indicating that a component of spin angular momentum is conserved. The ion polarization, detected optically, exceeds 10% at ion concentrations on the order of 10^{10}-10^{11} cm^{-3}.

The electron spin polarization of ions utilizing an optical pumping process generally requires novel techniques which are frequently limited in their application. For example, Baker et al.[1] stripped an electron from He^3 ground-state atoms which were polarized following electron exchange collisions with optically oriented metastable helium atoms to generate a polarized ion beam. Major and Dehmelt[2] in an elegant experiment utilized a spin-dependent selective destruction process between optically oriented Cs and He^+ to produce polarized $(He^3)^+$. Ackermann, zu Putlitz, and Weber[3] polarized Sr^+ by the direct interaction of resonance radiation after removing the ions from the region in which they were created, and recently, Mitchell and Fortsan[4] induced a Rb^+ nuclear polarization following resonant charge-transfer collisions with optically pumped Rb atoms.

The purpose of this Letter is to describe a new technique involving ionizing collisions between optically pumped metastable helium atoms and free atoms which provides a high concentration of highly polarized ions. This technique avoids some of the limitations of the earlier methods utilizing spin-dependent collisions and is applicable to a wide variety of atoms.

According to the Wigner spin rule[5] total spin should be conserved in Penning collisions

$$X^* + Y \rightarrow X + Y^+ + e. \qquad (1)$$

In general ionization occurs when the excited atom X^* posesses energy greater than the ionization potential of atom Y. We wish to further assert that a component of spin angular momentum is conserved if the collision duration is sufficiently short. We have exploited reactions of the type given by (1) to obtain polarized ions of Zn, Cd, and Sr which have undergone Penning collisions with optically pumped metastable helium atoms. The ion polarization is detected by monitoring the polarization of the light emitted by the excited ion following radiative decay. The fractional change in the polarization of the radiated light exceeds 0.10 indicating an ion spin polarization of this order.

The experimental apparatus used in this polarization transfer is a flowing-gas system of the type described by Schmeltekopf and Broida.[6] The essential elements are shown in Fig. 1. Helium gas is excited with a microwave generator and the excited gas is allowed to flow downstream. The primary constituents remaining in the flow by the time the gas reaches the reaction

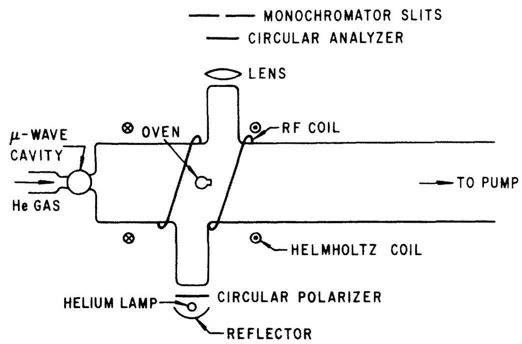

FIG. 1. Schematic of flowing-gas system. Entrance and exit windows are extended from the tube to avoid condensation of the metal. The flow velocity is on the order of 10^3-10^4 cm sec^{-1}. The metastable density in the reaction region is about 10^{11} cm^{-3}.

region are ground-state helium atoms, triplet metastable atoms, ions, and thermal electrons. A small oven containing small amounts of Cd, Zn, or Sr metal is placed in the reaction region. As the metal is vaporized the free metal atoms undergo ionizing collisions with the metastable helium atoms. In the case of Cd the particular reactions we have observed are

$$He(2^3S_1) + Cd(^1S_0) \rightarrow He(^1S_0) + Cd^+(^2D_{5/2}) + e \qquad (2)$$

followed by radiative decay of the ion:

$$Cd^+(^2D_{5/2}) \rightarrow Cd^+(^2P_{3/2}) + h\nu(4416\ \text{Å}),$$
$$Cd^+(^2P_{3/2}) \rightarrow Cd^+(^2S_{1/2}) + h\nu(2145\ \text{Å}). \qquad (3)$$

The radiation from the reaction region is focused on the slits of a $\frac{1}{2}$-m Jarrell-Ash spectrometer and detected with an S-5 photomultiplier at the exit slits. If the $He(2^3S_1)$ atoms are now prepared in a particular Zeeman sublevel and we assume that a component of spin angular momentum is conserved, we can write Eq. (1) as

$$(^3S_1\uparrow\uparrow) + (^1S_0\uparrow\downarrow) \rightarrow (^1S_0\uparrow\downarrow) + (^2D_{5/2}\uparrow) + e\uparrow, \qquad (2)$$

where the arrows represent the spin state of the electrons. If we assume that the ion is formed with equal probability in states of different m_l and that a component of spin angular momentum is conserved, we find that the m_J sublevels of the $^2D_{5/2}$ state are populated in the ratios 0:1:2:3 :4:5. Subsequent radiative decay of the ion to the $^2P_{3/2}$ state gives rise to a polarization of the 4416-Å light which can be detected.

The $He(2^3S_1)$ atoms are optically pumped in the conventional manner in the region just upstream from the oven containing the impurity. The external magnetic field of about 2 G is applied perpendicular to the flow direction. The radiation emitted by the excited ion along the field direction is examined at a point several centimeters downstream from the oven. The following observations are made as the external magnetic field is scanned through the metastable helium resonance condition at $g_J = 2$. (1) The total intensity of the emitted radiation at 4416 Å is independent of the spin polarization of the helium. (2) If the emitted ion radiation is circularly analyzed, i.e., only the σ^+ radiation observed, the intensity of 4416-Å light decreases when the helium spin polarization is saturated by an rf magnetic field. Figure 2 is a recording of the helium metastable resonance obtained when observing the σ^+ 4416-Å radiation. (3) The phase of the above signal changes when either (a) the σ^- radiation is observed at 4416 Å or (b) the spin polarization of the helium metastable atoms is reversed. (4) The measured quantity

$$[I(\sigma^+) - I(\sigma^-)]/[I(\sigma^+) + I(\sigma^-)]$$

is on the order of 0.10 indicating a relatively large polarization of the ion and an efficient polarization transfer.

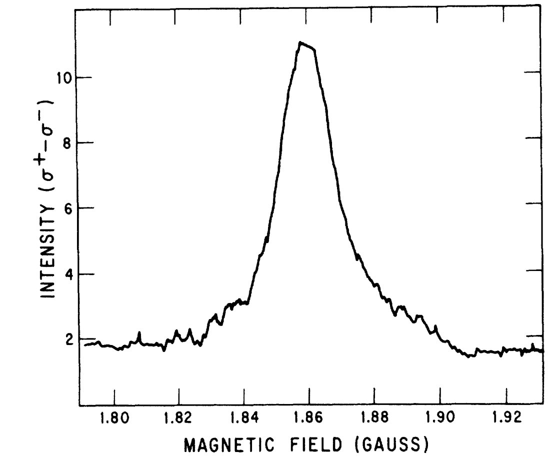

FIG. 2. Recording of helium metastable resonance obtained while monitoring polarization changes in the 4416-Å radiation from the Cd ion. The resonance rf field was amplitude modulated at 400 Hz. The signal was synchronously detected with a time constant of 3 sec.

The above experiment was repeated with Zn and Sr. In the case of Zn^+ the $^2D_{5/2}$ atom radiates to the $^2P_{3/2}$ level with emission at 7480 Å. In Sr^+ the transition $^2P_{3/2} \rightarrow {}^2S_{1/2}$ is monitored at 4077 Å. Similar results are obtained for each of these cases.

In summary, we have demonstrated that a component of spin angular momentum is well conserved following a Penning reaction. The polarization of the ion should not be disturbed by radiative processes[7,8]; consequently, a large polarization of the ion ground-state atoms can be obtained. This technique should be applicable to a wide variety of ions and provide a useful source of beams of highly polarized ions. We finally wish to point out that the electron that comes off in the reaction should also be polarized. The method of producing a polarized electron beam introduced by Walters and his colleagues[9] could be improved by several orders of magnitude by the addition of an impurity such as Cd to the optical-pumping cell.

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EXISTENCE OF THERMODYNAMICS FOR REAL MATTER WITH COULOMB FORCES

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It is shown that a system made up of nuclei and electrons, the constituents of ordinary matter, has a well-defined statistical-mechanically computed free energy per unit volume in the thermodynamic (bulk) limit. This proves that statistical mechanics, as developed by Gibbs, really leads to a proper thermodynamics for macroscopic systems.

In this note we wish to report the solution to a classic problem lying at the foundations of statistical mechanics.

Ever since the daring hypothesis of Gibbs and others that the equilibrium properties of matter could be completely described in terms of a phase-space average, or partition function, $Z = \mathrm{Tr}e^{-\beta H}$, it was realized that there were grave difficulties in justifying this assumption in terms of basic microscopic dynamics and that such delicate matters as the ergodic conjecture stood in the way. These questions have still not been satisfactorily resolved, but more recently still another problem about Z began to receive attention: Assuming the validity of the partition function, is it true that the resulting properties of matter will be extensive and otherwise the same as those postulated in the science of thermodynamics? In particular, does the thermodynamic, or bulk, limit exist for the free energy derived from the partition function, and if so, does it have the appropriate convexity, i.e., stability properties?

To be precise, if N_j are an unbounded, increasing sequence of particle numbers, and Ω_j a sequence of reasonable domains (or boxes) of volume V_j such that $N_j/V_j \to \text{constant} = \rho$, does the free energy per unit volume

$$f_j = -kT(V_j)^{-1}\ln Z(\beta, N_j, \Omega_j) \tag{1}$$

approach a limit [called $f(\beta,\rho)$] as $j\to\infty$, and is this limit independent of the particular sequence and shape of the domains? If so, is f convex in the density ρ and concave in the temperature β^{-1}? Convexity is the same as thermodynamic stability (non-negative compressibility and specific heat).

Various authors have evolved a technique for proving the above,[1,2] but always with one severe drawback. It had to be assumed that the interparticle potentials were short range (in a manner to