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Laird D. Schearer
Missouri University of Science and Technology

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Optical pumping in a flowing helium afterglow with additive metal impurity atoms*

L. D. Schearer
Physics Department, University of Missouri-Rolla, Rolla, Missouri 65401
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It is demonstrated that in a Penning collision of optically oriented metastable helium atoms with an impurity metal atom such as barium, both the longitudinal and transverse components of spin angular momentum are conserved. This results in a polarization of the ion which may be detected by optical emission in the case of excited ion levels or absorption for the ground states. A series of experiments is described which demonstrates the spin dependence of the Penning-collision process. The magnetic resonance of excited-state ions may be observed directly; from the resonance-linewidth radiative lifetimes may be measured.

I. INTRODUCTION

Utilizing an optically pumped helium afterglow I have examined the spin dependence of Penning ionization and charge-transfer collisions between the helium-afterglow constituents (primarily ions and metastable atoms) and impurity atoms and molecules.

Reactions of an electronically excited species with other atoms and molecules are important processes in stellar and planetary atmospheres. The large abundance of helium in these atmospheres makes any collision process involving it of intrinsic interest. For example, helium metastable atoms play an important role in the physics of the atmospheres of stars and planets. Additionally, thermal-energy collisions are important in many other phenomena such as gas discharges, shock waves, photolysis, radiation chemistry, and plasmas in general.

One of the important tools in the laboratory measurements of reaction rates for collisions of this type is the fast-flowing helium afterglow. The use of this technique has enjoyed much success in the measurements of thermal-energy ion-neutral reaction rates, associative detachment reactions, and deexcitation rates for metastable helium atoms by a variety of atomic and molecular gases.[1]

For the most part the use of the flowing-helium afterglow has been limited to interactions between the excited helium atoms and ions and the more common atomic and molecular gases, particularly atmospheric constituents. Relatively little work has been done with the flowing afterglow in which the additive impurities are metal vapors. Although reactions of this type are of considerable interest to atmospheric scientists, the development of cw laser systems in which the inversion mechanism has been ascribed to thermal-energy heavy-particle reactions further emphasizes the lack of information concerning them. Add to this the involvement of electronically excited helium in gas lasers,[2] optically pumped magnetometers,[3] polarized gas targets and beams,[4] and one has compelling reasons to investigate collision processes of this type.

Further, very little work has been reported which is concerned specifically with the spin dependence of Penning-ionization processes although it was long ago recognized that spin was in an important parameter. Only recently have spin-dependent collision processes been utilized as a mechanism for transferring a polarization from one species to another via inelastic collisions in which significant amounts of energy exchange take place.[5]

Of particular interest are collisions involving the helium metastable ($2\,^3S_1$) state and impurity atoms and molecules. With the exception of neon, the energy of the helium metastable atom (19.8 eV) is sufficient to ionize any impurity atom or molecule. This ionization process was first investigated by Penning in the mid-thirties and the process now bears his name, namely, Penning ionization.

In the sections following we describe the use of the fast-flowing helium afterglow to investigate the optical excitation of ions resulting from Penning ionization and charge-transfer reactions:

$$\mathrm{He}(2\,^3S_1) + X \rightarrow \mathrm{He}(1\,^1S_0) + (X^+)^* + e + \text{kinetic energy} \tag{1}$$

and

$$\mathrm{He}^+(^2S_{1/2}) + X \rightarrow \mathrm{He}(^1S) + (X^+)^*, \tag{2}$$

where $(X^+)^*$ denotes the ionized excited state of the atom or molecule X.

If reaction (1) is to proceed with substantial cross section at thermal energies, the internal energy of the helium metastable atom must be

greater than the energy associated with $(X^+)^*$, the excess energy being carried off as kinetic energy of the electron. The energy dependence for charge-transfer reactions (2) is more stringent. For the processes studied here the cross section is largest for transitions in which $(X^+)^*$ lies between 0.1 and 0.4 eV below He^+.[6]

The above conditions are necessary but not sufficient. The Wigner-spin rule in addition requires that total spin must be conserved. In the metastable states such as the $^3S_1$ level of helium, electron exchange with the impurity atom of molecule X is required for deexcitation of the helium to the ground $^1S_0$ state. It is this feature of Penning ionization that we exploit to obtain ensembles of polarized excited- and ground-state ions and electrons.

Examination of the polarization of the optical emission as a function of the spin polarization of the helium metastable atoms and ions establishes the strong spin dependence of the reaction. This strong spin dependence of the collision is utilized to produce polarized electrons and ions which are then available for the purposes of radio-frequency spectroscopy and the study of spin-dependent collisional processes.

In Sec. II, the characteristics of a fast-flowing helium afterglow are described which are of importance to the study of Penning and charge-transfer reactions. The optical pumping process in helium is then described—the technique utilized to obtain an initial polarization of the helium-afterglow products. Following this is a description of the effect of additive impurities which appear as a transfer of the longitudinal component of magnetization from the helium metastable atoms and ions to the impurity ions. A series of experiments are described which demonstrate the effectiveness of the polarization transfer and in which magnetic resonances of excited ions are observed.

Examination of the optical polarization of the emission from excited Penning ions as a function of the spin polarization of the metastable helium atoms establishes that both the longitudinal and transverse components of spin are well-conserved in the collision. This strong spin dependence of the collision is utilized to produce ensembles of polarized electrons and ions in excited and ground levels. The polarized electrons and ions are then available for a wide variety of experimental procedures.

This technique effectively extends optical-pumping experiments in ions to a much wider range of states and species than is generally available. This is a region that is generally limited in access due to the difficulty of obtaining pumping-light sources and the lack of excitation methods which yield a sufficiently high density of ions in the states to be pumped.[7] The use of Penning ionization and polarization transfer from metastable helium atoms overcomes both these difficulties.

II. THE FAST-FLOWING HELIUM AFTERGLOW

A. Configuration

Reactions of the type described by (1) and (2) can, of course, be studied in an active discharge. However, in this case inelastic collisions between hot electrons and the gaseous constituents may contribute to the populations of the final states of the ions thus obscuring the particular collisional processes of interest. The use of time-resolved spectroscopy in which the discharge is briefly excited and the observations made at varying time intervals after cessation of the pulse effects a temporal separation of the active discharge and events which occur in the afterglow. Experiments of this type have been used to advantage to differentiate between Penning processes (1) and charge-transfer processes (2) and to obtain cross sections for them. The disadvantage of this method is that measurements must be made against the background of a rapidly decaying plasma.

The fast-flowing afterglow can be used to obtain a spatial separation of the active discharge and the afterglow while at the same time providing steady-state conditions under which the appropriate observations can be made.

The fast-flowing afterglow utilized in this report is a modification of one constructed and described earlier by Schmeltekopf *et al.*[1] The flow tube consists principally of a 1-m-long Pyrex tube, 7.5 cm in diameter. One end of the tube is connected to a high-speed, high-capacity mechanical forepump backed by a Roots blower. The pumping speed at the entrance to the pump is 540 ft^3/min. The other end of the tube is terminated in a converging-diverging nozzle. Helium tank gas of standard purity (99.995%) is metered through a flow valve to the nozzle and is expanded into the flow tube. A microwave cavity of the Evenson type is used to excite a discharge in the throat of the nozzle. The gas velocity in the flow tube varies between 10^3 and 10^4 cm sec^{-1} and the total pressure ranges between 0.1 and 1 Torr.

As the gas moves downstream the short-lived products of the active discharge decay, the hot electrons are thermalized, and singlet metastables are converted to triplets through exchange collisions with slow electrons. The principal constituents remaining downstream are thermal electrons, atomic-helium ions, helium triplet metastable atoms, and neutral helium. Under typical operating conditions we estimate the densities of

the various constituents to be, for He($^1S_0$), 10^{16} cm^{-3}; He^+ and e, 10^{10} cm^{-3}; He($2\,^3S_1$), 5×10^{10} cm^{-3}, approximately 20 cm downstream from the excitation region. At this point provision is made to inject the impurity species.

B. Afterglow diagnostics

A variety of techniques are utilized to determine the properties of this flowing afterglow. For example, one can examine the afterglow emission spectroscopically. In a pure helium afterglow at pressures below 1 Torr we find that only atomic lines from helium are present. This afterglow emission is presumably due to the recombination of He ions and electrons. Molecular emission was not observed at pressures below 1 Torr.

The purity of the afterglow is determined also from the spectroscopic properties. The most persistent impurity is nitrogen which results from small leaks in the system. Nitrogen is easily detected since the Penning ionization of N_2 results in the production of N_2^+ in the $B^2\Sigma$ state. Subsequent radiative decay at 3914 Å is easily monitored. For the experiments reported here the nitrogen contamination was such that the emission of radiation at 3914 Å was at least one and generally two orders of magnitude smaller than emission at 3889 Å which results from the recombination of helium ions. The use of research-grade helium obtained from Linde Air Products precludes the existence of other unwanted impurities.

To obtain the flow velocity the microwave discharge is pulsed and the time delay until the pulse reaches downstream is measured. If we plot the time delay as a function of distance downstream, we obtain the flow velocity. The flow velocity increases with increasing background pressure. At approximately 0.2 Torr the flow velocity is about 10^4 cm sec^{-1}. Above 10 Torr the flow is supersonic as indicated by the presence of diamond-shaped shock waves in the afterglow emission. Figure 1 shows the time delay for the arrival of the afterflow emission at various points downstream. The spreading out of the pulse at increasing distances from the excitation region is a result of diffusion. The pressure in the system is obtained by inserting a calibrated Pirani gauge into the afterglow. The pressure measurements in the system are unreliable and are used primarily to reproduce conditions from day to day.

The helium triplet metastable density is monitored by the absorption of resonance radiation at 1.08 ($2\,^3S$-$2\,^3P$) or 3889 Å ($2\,^3S$-$3\,^3P$) provided by a helium lamp. From the path length and fractional absorption an estimate of the metastable density is obtained. By varying the microwave power to the excitation cavity and its location, densities up to 10^{11} cm^{-3} can be obtained. At this density the metastable population appears to saturate; increasing the μ-wave power does not result in higher metastable densities although the afterglow emission intensity continues to increase. Again this measurement yields only estimates of the metastable density. We were unable to detect absorption at 2 μm ($2\,^1S$-$2\,^1P$) due to the presence

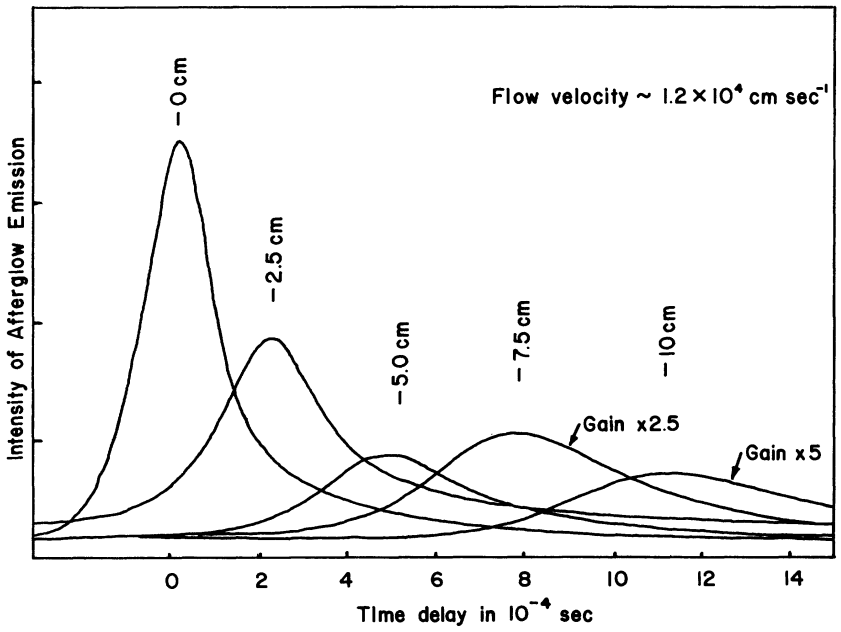

FIG. 1. Time of arrival of afterglow emission at varying distances into the reaction zone. The point shown as 0 cm is 25 cm downstream from the discharge pulse. A plot of the time delay vs distance downstream yields the flow velocity.

of singlet metastables. We estimate the singlet metastable density to be at least one and probably two orders of magnitude lower than the triplet density.

The use of additive impurities is also a sensitive test for the relative densities of afterglow constituents. Neon is excited in a collision of the second kind by both the singlet metastable and the triplet metastable. Fluorescence at 6328 Å is indicative of the presence of the singlet-helium metastable atom while emission at 1.15 μm is due to excitation by the helium triplet. No emission which originates from the excited levels of Ne which are populated by collisions with singlet-helium metastable atoms is observed. This provides additional confirmation that the singlet metastable atoms created in the discharge region are rapidly converted to $He(2\,^3S_1)$ atoms by spin-exchange collisions with the thermalized electrons.

With the addition of Zn-metal atoms one can obtain a relative measure of the ratio of He^+ to $He(2\,^3S_1)$. The cross sections for Penning ionization of Zn by helium-triplet metastable atoms and charge transfer with He^+ have been measured[6]:

$$He(2\,^3S_1) + Zn(^1S_0) \rightarrow He(1\,^1S_0) + Zn^+(4\,^2D_{3/2}) + e\,, \tag{3}$$

followed by

$$Zn^+(4\,^2D_{3/2}) \rightarrow Zn^+(4\,^2P_{1/2}) + h\nu(5894\,\text{Å})\,. \tag{4}$$

For charge transfer:

$$He^+(1\,^2S_{1/2}) + Zn(^1S_0) \rightarrow He(1\,^1S_0) + Zn^+(5\,^2D_{5/2})\,, \tag{5}$$

followed by

$$Zn^+(5\,^2D_{5/2}) \rightarrow Zn^+(5\,^2P_{3/2}) + h\nu(6102\,\text{Å})\,. \tag{6}$$

The cross sections for the two processes are comparable; consequently one needs only to measure the relative intensities of the 5894- and 6102-Å emission to obtain a relative measure of the $He(2\,^3S_1)/He^+(1\,^2S_{1/2})$ densities. That the emission at 5894 and 6102 Å is due to different processes can easily be shown. In a *pulsed* helium afterglow the emissions decay at different rates with the 5894-Å emission following the decay of the $He(2\,^3S_1)$ density. In the *flowing* helium afterglow a microwave antenna can be used to heat slightly the electrons. The increased electron temperature increases the loss rate of the electrons and ions due to ambipolar diffusion and reduces their density. In this case the emission at 6120 Å is reduced while the 5894-Å emission arising from Penning collisions with the neutral helium metastable atoms remains unchanged. We find that in the afterglow region the intensities are comparable (within a factor of 5), and conclude that the ion density is within a factor of 5 of the helium $(2\,^3S_1)$ density. Since the afterglow is a neutral plasma, the electron density is the same as the ion density. In a later section we provide experimental evidence which indicates that under our normal operating conditions a large fraction (~30%) of the ions and electrons in the afterglow region result from the collision of two helium metastable atoms in a Penning-ionizing collision.

C. Spin dependence of Penning ionization

Penning-ionizing collisions generally proceed with a large cross section if the Wigner-spin rule is satisfied. In many cases the 19.8 eV of internal energy for the $He(2\,^3S)$ atom is sufficient to both ionize the impurity and leave the ion in an excited state. For example, as a result of a Penning collision with Cd there are six possible states of the ion that are energetically accessible. The total quenching cross section of $He(2\,^3S_1)$ by Cd has been measured by Padovani and Schearer[9] as 4.5×10^{-15} cm^2. Cermak[10] has measured the relative cross sections for excitation of these Cd^+ levels.

The conservation of total spin angular momentum expressed by Wigner-spin rule implies that a component of spin angular momentum is conserved about *some* axis. Let us assume for the present that the axis along which spin is conserved is defined by an external magnetic field. For a specific reaction of the type described by Eq. (1),

$$He(2\,^3S_1\uparrow\uparrow) + Zn(^1S_0\downarrow\uparrow) \rightarrow He(^1S_0\downarrow\uparrow) + Zn^+(4\,^2D_{5/2}\uparrow) + e(\uparrow), \tag{7}$$

where the arrows represent the orientations of the electron spin along the external magnetic field immediately before the collision for the left-hand side and immediately after the reaction on the right-hand side. $He(2\,^3S_1\uparrow\uparrow)$, then describes a helium metastable atom in the $m_j=+1$ Zeeman sublevel of the $2\,^3S_1$ metastable state.

Immediately after the collision the electron spin associated with the ion is oriented along the external-field direction but then couples to the orbital angular momentum. After a time on the order of the period of the fine-structure separation the ion is in an eigenstate. The probability that a particular Zeeman level is occupied is then just the squared amplitude of the term containing $m_s=+1/2$. For example, if we write the wave functions of the $^2D_{5/2}$ level in the m_s, m_L representation:

$$
\begin{aligned}
&|J,m_J\rangle \rightarrow |L,m_L,m_s\rangle ,\\
&|5/2,5/2\rangle \rightarrow |2,2,1/2\rangle ,\\
&|5/2,3/2\rangle \rightarrow \sqrt{4/5}\,|2,1,1/2\rangle + \sqrt{1/5}\,|2,2,-1/2\rangle ,\\
&|5/2,1/2\rangle \rightarrow \sqrt{3/5}\,|2,0,1/2\rangle + \sqrt{2/5}\,|2,1,-1/2\rangle ,\\
&|5/2,-1/2\rangle \rightarrow \sqrt{2/5}\,|2,-1,1/2\rangle + \sqrt{3/5}\,|2,0,-1/2\rangle ,\\
&|5/2,-3/2\rangle \rightarrow \sqrt{1/5}\,|2,-2,1/2\rangle + \sqrt{4/5}\,|2,-1,-1/2\rangle ,\\
&|5/2,-5/2\rangle \rightarrow \qquad\qquad |2,-2,-1/2\rangle ,
\end{aligned}
\tag{8}
$$

the relative excitation rates are in the ratio of 5:4:3:2:1:0 if the helium metastable atom is in the $m_j = +1$ level.

From the $5\,^2D_{5/2}$ level the ion decays to the $5\,^2P_{3/2}$ with the emission of 7479-Å radiation. Using the transition probabilities for electric dipole emission along the field direction shown in Table I we can write:

$$
\begin{aligned}
&I(\sigma^+) \propto N_{5/2}(10) + N_{3/2}(6) + N_{1/2}(3) + N_{-1/2}(1) ,\\
&I(\sigma^-) \propto N_{1/2}(1) + N_{-1/2}(3) + N_{-3/2}(6) + N_{-5/2}(10) ,
\end{aligned}
\tag{9}
$$

where σ^+ emission arises from a transition in which $\Delta m_j = -1$. If we then take $N_{5/2} : N_{3/2} : N_{1/2} : N_{-1/2} : N_{-3/2} : N_{-5/2}$ as 5:4:3:2:1:0, we find

$$
\frac{I(\sigma^+) - I(\sigma^-)}{I(\sigma^+) + I(\sigma^-)} = 0.70 . \tag{10}
$$

Defining the J polarization as

$$
P(J) = \frac{1}{J} \sum_{m_J} \frac{N(m_J)}{N} \langle m_J | J_z | m_J \rangle , \tag{11}
$$

and the spin polarization as

$$
P(S) = \frac{1}{S} \sum_{m_J} \frac{N(m_J)}{N} \langle m_J | S_z | m_J \rangle , \tag{12}
$$

we obtain $P(J) = P(S) = 0.467$. Thus, beginning with $P(S) = 1.00$, for the system of $\mathrm{He}(2\,^3S_1, m_J = +1) + \mathrm{Zn}(^1S_0)$ initially, we find that as a result of the $\vec{S}\cdot\vec{L}$ coupling in the excited state of the ion, the final ion polarization is 0.467 and the optical polarization is 0.70. We note also that the $\vec{S}\cdot\vec{L}$ coupling in the excited ion is required if the optical polarization is to be nonzero. This, of course, is a consequence of the selection rule $\Delta m_s = 0$ in electric dipole transitions.

If one defines the polarization of the $\mathrm{He}(2\,^3S_1)$ system as

$$
P = [N(m_s = +1) - N(m_s = -1)]/N , \tag{13}
$$

and an alignment of the $\mathrm{He}(2\,^3S_1)$ system as

$$
A = [N(m_s = +1) + N(m_s = -1) - 2N(m_s = 0)]/N , \tag{14}
$$

it is easily shown that there is no alignment transfer to the ions if there are no selection rules on Δm_L in the collision process. It follows then that no effects of the spin dependence of the collision process will be seen if the observation direction is orthogonal to the external magnetic field.

D. Optical pumping in the helium afterglow

If we are to observe the effects described in Sec. II C, we must first prepare the $\mathrm{He}(2\,^3S_1)$ atoms in the $m_J = +1$. Fortunately, the technique of optical pumping provides a method whereby helium metastable atoms in this state can be obtained. With few exceptions optical pumping of the $\mathrm{He}(2\,^3S_1)$ atoms in the afterglow follows that of optical pumping in the active discharge of a closed vessel. For a detailed discussion of optical pumping in helium the reader is referred to the work of Colegrove and Franken,[11] and Schearer.[12]

In the usual optical-pumping experiment circularly polarized resonance radiation from a helium lamp is directed along the external magnetic field direction and absorbed by the ensemble of metastable helium atoms. In the process of absorption and reemission a portion of the angular momentum

TABLE I. Relative transition probabilities: $^2D_{5/2}$-$^2P_{3/2}$. θ is angle between direction of observation and the external magnetic field.

		$^2P_{3/2}$			
	m_J \ $m_{J'}$	$\frac{3}{2}$	$\frac{1}{2}$	$-\frac{1}{2}$	$-\frac{3}{2}$
$^2D_{5/2}$	$\frac{5}{2}$	$5(1+\cos^2\theta)$	0	0	0
	$\frac{3}{2}$	$4\sin^2\theta$	$3(1+\cos^2\theta)$	0	0
	$\frac{1}{2}$	$\frac{1}{2}(1+\cos^2\theta)$	$6\sin^2\theta$	$\frac{3}{2}(1+\cos^2\theta)$	0
	$-\frac{1}{2}$	0	$\frac{3}{2}(1+\cos^2\theta)$	$6\sin^2\theta$	$\frac{1}{2}(1+\cos^2\theta)$
	$-\frac{3}{2}$	0	0	$3(1+\cos^2\theta)$	$4\sin^2\theta$
	$-\frac{5}{2}$	0	0	0	$5(1+\cos^2\theta)$

in the light beam is transferred to the helium metastable system. By recourse to the usual optical-pumping-rate equations one can easily show that under the conditions described above that *both* an alignment defined by Eq. (14) and a polarization defined by Eq. (13) are produced in the helium metastable system.

The production of an alignment or polarization in the sample is accompanied by a change in the transparency of the absorbing ensemble of atoms; consequently, by monitoring the transmitted resonance radiation one can observe the progress of the optical-pumping process. The fractional change in the absorption of the resonance radiation is a measure of the degree of alignment (or polarization) produced. Unfortunately, in the case of helium there is no unique relationship between the observable (fractional change in absorption) and alignment or polarization.

In the experiments to be described the sources of the resonance radiation are helium-gas filled, electrode-less capillary lamps excited with about 20 W of rf power between 20 and 50 MHz. One or more lamps are focused on the afterglow and directed along the external field. The circular polarizers are obtained from the Polaroid Corporation as their type HR linear polarizer and a phase retardation plate of a quarter wave at 1.08 μ. Silicon photovoltaic cells with peak sensitivity at 1.06 μ are obtained from RCA and used to monitor the transmitted resonance radiation.

The experimentally measured fractional change in absorption as the metastable atoms are pumped is approximately 0.005–0.01; the optically induced polarization is, however, substantially larger—on the order of 0.3. The source of the apparent discrepancy is in the composition of the resonance radiation. The pumping light consists of three components near 1.08 μ due to transitions from the $2\,^3P_0$, $^3P_1$, and $^3P_2$ levels labeled for convenience as the D_0, D_1, and D_2 component. The transitions are within 1 Å of each other and cannot efficiently be isolated. Again, by recourse to the rate equations one can easily show that while all three components tend to pump the atoms in the same direction, the intensity of the transmitted D_2 component decreases as the sample is pumped while the intensity of the D_0 and D_1 components increase. Thus, there are cancellations in the observed optical signals when one averages over the three components. This can be clearly seen from Fig. 2 where we have scanned the change in the absorption through the three resonance components. This effect renders unsuitable the measurement of the fractional absorption to indicate the degree of polarization of the sample. If one monitors only the D_0 component, the measured fractional absorption is more nearly a representation of the polarization present. In this case one can show

$$(\Delta I/I)_{D_0} = 3[(N-)/N - 1], \tag{15}$$

where the term on the left represents the change in fractional absorption, $N-$ is the population of the $m_J = -1$ level, and N is the total metastable population.

Byerly[13] has shown that in the active discharge the lifetime of the helium metastable atom in a given magnetic sublevel is limited by collisions of the helium metastable atoms with the hot electrons of the discharge. In the flowing afterglow the

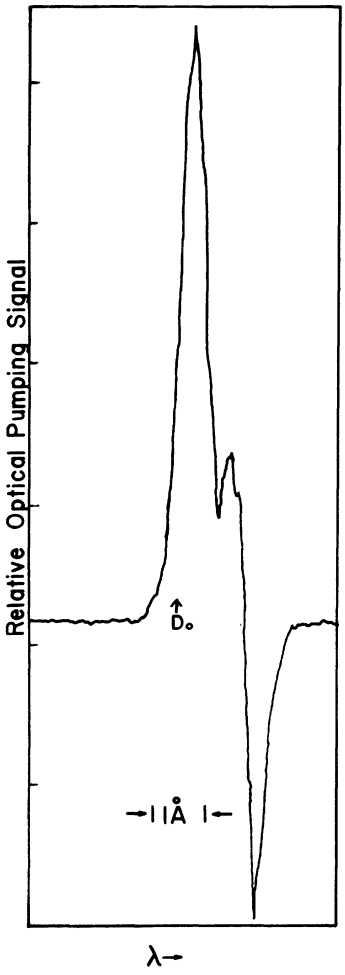

FIG. 2. Change in absorption due to optical pumping as a function of wavelength of the pumping light. The absorption decreases as the He metastable atoms are pumped for the D_0 and D_1 components but increases for the D_2 component. The use of a broad-band detector results in a reduced signal. The fractional change in the intensity of the D_0 component is about 0.3. The bulk of the absorption is due to the D_2 and D_3 components.

electrons have thermalized. In this case the principal source of relaxation appears to be spin-exchange collisions between the thermal electrons and the metastable helium atoms. The principle difference between the two cases is that in the active discharge the relaxation process is one in which angular momentum is lost from the system while in the afterglow the angular momentum is stored in the system although the spin-exchange collisions redistribute it. Thus, we find that the optically induced polarization in the afterglow system is substantially larger than that obtained in an active helium discharge. Further support for this observation is to be found in optical-pumping experiments in the helium-three isotope. In this case it had been found that as the discharge level is reduced the attainable polarization increases substantially. Polarizations of the nuclear-spin system in He^3 exceeding 50% have been reported.[14]

The helium (2^3S_1) metastable magnetic resonance at $g=2$ is easily observed by monitoring the intensity of the transmitted resonance radiation. The usual practice is to apply a fixed rf magnetic field perpendicular to the external field H. When the equation

$$h\nu = g\mu_B H, \tag{16}$$

where μ_B is the Bohr magneton and ν is the frequency, is satisfied, transitions induced by the oscillating magnetic field alter the polarization produced by the pumping beam and hence, the transmitted radiation.

The magnetic resonance curve of the helium metastable atoms is then obtained by scanning the external magnetic field through the resonance condition. Under the conditions described earlier, the signals are large and easily observed on an oscilloscope. The width of this resonance line is determined by the spin-state lifetime in a gradient-free region and in the limit of small rf-field intensity. In practice the resonance lines in the laboratory are broadened by field gradients and produce a linewidth of about 10 mG. Since the constants in Eq. (16) are known for the helium metastable atom, one has a simple, convenient, and accurate method of calibrating the external magnetic field intensity.

III. EXPERIMENTAL OBSERVATIONS

A. Polarization transfer to ions

Figure 3 is a schematic representation of the apparatus. Obvious modifications to the basic system which are utilized for the different observations are indicated in subsequent sections. Basically, the helium metastable atoms are prepared in the appropriate state by optical pumping. The optical pumping is accomplished in the field-free region approximately 30 cm downstream from the excitation source. In this region the impurity specie are introduced. Subsequently, the Penning-ionizing collision occurs and the optical emission from the excited impurity ion is observed along the external field direction. The emission is isolated with a grating monochromator and its polarization is analyzed for a difference between the σ^+ and σ^- components. The polarization analysis is accomplished by passing the optical emission through a linear polarizer and a rotating quarter-wave plate. The signal is synchronously detected as twice the rotation frequency by an appropriate photomultiplier. This procedure detects directly the quantity

$$I(\sigma^+) - I(\sigma^-).$$

The presence of this signal on the ion emission demonstrates that the excited ion is indeed polarized. If we then apply a resonance magnetic field to the helium metastable atoms at $g=2$ and observe a change in the quantity defined by Eq. (11), we can assert that the excited ion is polarized and that its polarization was acquired as a result of spin conservation in the Penning collision with the optically oriented helium metastable atoms. Figure 4 is the signal obtained for $Zn^+(4^2D_{3/2}$-$4^2P_{3/2})$ emission at 5894-Å as the external magnetic field is scanned slowly through the helium metastable resonance condition. The helium metastable polarization for the conditions in which Fig. 4 was obtained, is about 22% as determined from

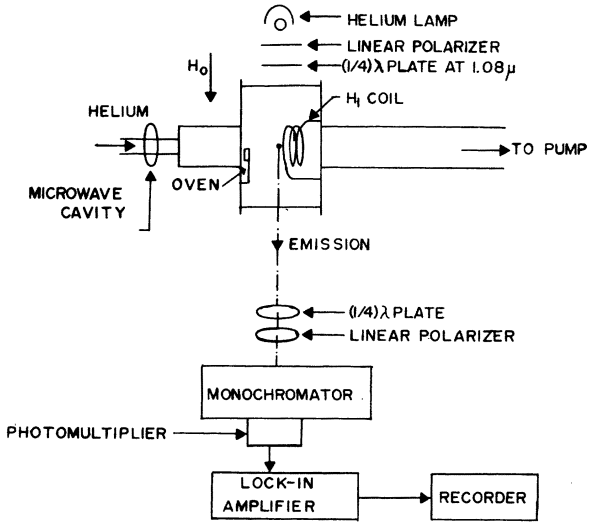

FIG. 3. Schematic representation of apparatus. The expanded region shown is used to inhibit condensation of metal atoms on glass surfaces.

a measurement of the fractional change in the absorbed D_0 component of the helium-pumping radiation. The measured optical polarization of the Zn^+ emission shown is 16%. On the basis of this observation and Eq. (10) we conclude that spin angular momentum is well-conserved about the field direction in the Penning collision. Similar results are obtained for a wide variety of metal vapors. The atoms, transitions, and wavelengths, for which substantial polarization transfer have been observed are shown in Table II.

B. Magnetic resonance of excited ions

With the results of the Sec. III A we have shown that the excited ions are polarized and that the polarization can be detected optically. If the g factor for the excited ion is different from the helium $2\,^3S_1$ atom ($g=2$), it should be possible to observe the effects of a resonance magnetic field applied to the ion system. Figure 5 is the demodulated output of the photomultiplier as the external magnetic field is slowly scanned. The resonance at $H=7.6$ G ($g=2$) is due to the helium metastable atoms; the resonance $H=15.2$ G ($g=1$) is due to double quantum transitions in the helium metastable system; at $H=12.5$ G is the $g=1.2$ resonance due to the even isotopes of $Cd^+(5\,^2D_{5/2})$; the resonance at $H=10.7$ G is due to the $F=2$ component of the odd isotopes of $Cd^+(5\,^2D_{5/2}g_F=1.4)$ and $I=1/2$ which occurs with an abundance of 25% in a natural cadmium sample. The $F=3$ ($g_F=1$) resonance is masked by the double-quantum resonance signal of the helium metastable system.

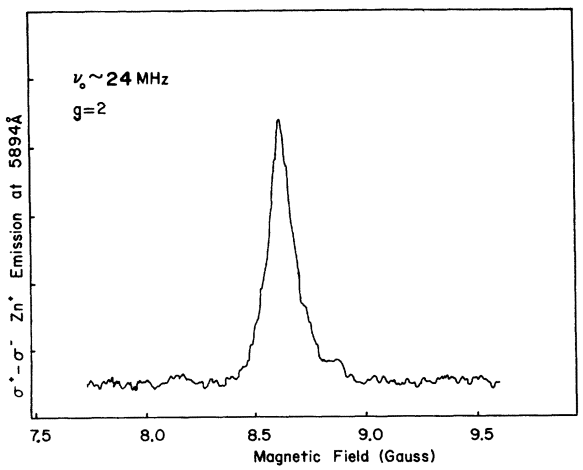

FIG. 4. Change in $\sigma^+ - \sigma^-$ emission at 5894 Å as a function of applied magnetic field in the presence of an oscillating magnetic field at 24 MHz. The resonance occurs at the $g=2$ resonance of the $2\,^3S_1$ helium atoms. The vertical scale is decreasing intensity polarization.

The full width at half-height of the resonance signal is related to the lifetime of the ion in a given magnetic sublevel by the relationship

$$\Delta\nu = 1/\pi\tau\,, \tag{17}$$

where $\Delta\nu$ is the width in frequency units and τ is the lifetime. Lifetime limiting processes include the effects of the resonance magnetic field, collisional mixing, collision-induced decay, and radiative decay. By measuring the linewidth with progressively smaller resonance field intensities and extrapolating to zero, one eliminates broadening due to this source. Since collisional effects are pressure dependent, the linewidth is measured as a function of the afterglow total pressure. In the limit of zero pressure the linewidth is a measure of the radiative lifetime.

In Table III are the measured radiative lifetimes for $Zn^+(4\,^2D_{5/2})$, $Zn^+(4\,^2D_{3/2})$, and $Cd^+(5\,^2D_{5/2})$ obtained in the limit of zero field and pressure broadening. It should be noted that this technique is applicable to a wide variety of ions, and levels since the afterglow configuration is especially amenable to the insertion of small coils and/or

TABLE II. Ions in which polarization transfer is observed.

Ion	Wavelength (Å)	Transition
Cd^+	4416	$5\,^2D_{5/2}$-$5\,^2P_{3/2}$
	3250	$5\,^2D_{3/2}$-$5\,^2P_{1/2}$
Zn^+	7480	$4\,^2D_{5/2}$-$4\,^2P_{3/2}$
	5894	$4\,^3D_{3/2}$-$4\,^2P_{1/2}$
	6102[a]	$5\,^2D_{5/2}$-$5\,^2P_{3/2}$
Ca^+	3934	$4\,^2P_{3/2}$-$4\,^2S_{1/2}$
	3968	$4\,^2P_{1/2}$-$4\,^2S_{1/2}$
Sr^+	4078	$5\,^2P_{3/2}$-$5\,^2S_{1/2}$
	4216	$5\,^2P_{1/2}$-$5\,^2S_{1/2}$
Ba^+	4554	$6\,^2P_{3/2}$-$6\,^2S_{1/2}$
	4934	$6\,^2P_{1/2}$-$6\,^2S_{1/2}$
	6142	$6\,^2P_{3/2}$-$5\,^2D_{5/2}$
Mg^+	4481	$4\,^2F_{7/2}$-$3\,^2D_{5/2}$
Pb^+	5606	$7\,^2P_{3/2}$-$7\,^2S_{1/2}$
Yb^+	3694	$6\,^2P_{3/2}$-$6\,^2S_{1/2}$
N_2^+	3914	$B\,^2\Sigma$-$X\,^2\Sigma$
Eu^+	4522	$z\,^9P_3$-$a\,^7S_3^0$
	4435	$z\,^9P_4$-$a\,^7S_3^0$
	4205	$z\,^9P_3$-$a\,^9S_4^0$
	4129	$z\,^9P_4$-$a\,^9S_4^0$

[a] Populated by charge transfer with He^+.

cavities (to obtain large resonance fields in small volumes) and ovens for the various impurity species. Within the experimental uncertainty the linewidths were independent of pressure over the range 0.2–0.6 Torr. If the entire uncertainty in the linewidth measurement is attributed to collisional mixing over this pressure range, an upper limit on the magnitude of the collisional mixing cross section can be calculated. We find $\sigma(\text{mix}) \leq 2 \times 10^{-17}$ cm^2 for the $Cd^+(5\,^2D_{5/2})$ level and $\sigma(\text{mix}) \leq 4 \times 10^{-17}$ cm^2 for the $Zn^+(4\,^2D_{5/2})$ levels. The small depolarization cross section due to collisions of the excited ions with helium ($1\,^1S_0$) is a consequence of the electron configuration for these states which is of the form $nd^9 \times (n+1)S^2$. The orbital angular momentum arises from a "d" hole and the outer two electrons are in S orbitals. The outer electrons are evidently effective in shielding the core electrons from perturbations due to collisions. Similar arguments have been used to explain the small depolarization cross section observed for $Ne(3\,^3P_2)$-$He(1\,^1S_0)$ collisions.[15]

C. Ground-ion resonances

Polarization of the excited ions is detected by observation of the emitted light when the ion decays radiatively. The ground state of the ion is also populated either directly as a result of the Penning process or as a result of cascade from higher-lying levels which have been populated by Penning collisions. In either case the ground ion is polarized. The ground-ion lifetimes are relatively long, being limited by ambipolar diffusion and recombination, and, thus accumulate in densities large enough to be observed by optical absorption.

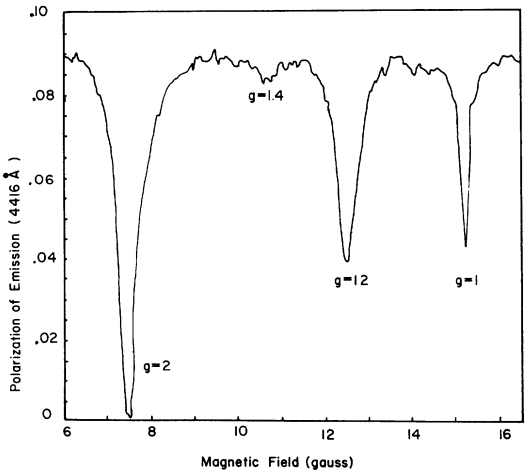

FIG. 5. Magnetic resonance signals from Cd^+.

TABLE III. Lifetime measurements.

Ion	Level	Lifetime (this expt) (10^{-6} sec)	Lifetime (other expt) (10^{-6} sec)
Cd^+	$5\,^2D_{5/2}$	0.773 ± 0.027	0.783 ± 0.011[a] 0.83 ± 0.07[b]
Zn^+	$4\,^2D_{5/2}$	1.61 ± 0.11	...
Zn^+	$4\,^2D_{3/2}$	2.22 ± 0.11	4.65 ± 0.02[b]

[a] M. Barrat and J. P. Barrat, Compt. Rend. 257, 1463 (1963).

[b] E. Geneux and B. Wanders-Vincenz, Helv. Phys. Acta 33, 185 (1960).

If a source of ion resonance radiation is circularly polarized and passed through the reaction region of the afterglow along the external field direction, the relative populations of the Zeeman sublevels of the ground ion can be probed. The sources of the ion resonance radiation are commercial hollow cathode lamps operated at 25 mA. Changes in the polarization of the ions appear as changes in the transmission of the circularly polarized probing light. Polarization of the ground ions of Ca^+, Sr^+, and Ba^+ have been detected in this manner. Figure 6 shows the transmission of ion resonance radiation from a Sr^+ hollow cathode lamp at 4077 Å ($^2P_{3/2}$-$^2S_{1/2}$) and 4215 Å ($^2P_{1/2}$-$^2S_{1/2}$) as the external field is scanned through the $g=2$ resonance condition. As expected, the D_1 and D_2 components have opposite polarity. The ion density is such that 28% of the resonance radiation is absorbed over a path length of approximately 7 cm.

The metal vapors used in these experiments were those of natural abundance, and the largest

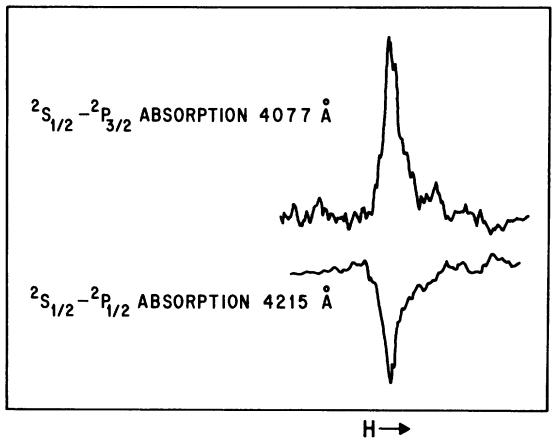

FIG. 6. Transmission of strontium-ion resonance radiation at 4077 and 4215 Å is a function of helium metastable polarization. The resonances are at $g=2$.

percentage of the metals were of the even isotope for which $g=2$. Since this is the same g factor as the helium metastable, no information about ion lifetimes or accurate measurement of g factors are available from the resonance curves of Fig. 6. In the case of Ba, 17.81% is of odd isotope with $I=3/2$. A resonance signal at $g=0.5$ should be observable with a signal/noise of 0.04 of that shown in Fig. 6. Improvements in the existing apparatus, longer signal integration times, and brighter sources of resonance radiation should permit the direct observations of the odd isotope resonances of these ground ions, thus extending optical-pumping techniques to a wide variety of paramagnetic ions in their ground state.

D. Penning-electron polarization

Having established that spin angular momentum is well-conserved in the collision process, Eq. (7) indicates that the spin of the Penning electron should also be oriented along the externally applied magnetic field. Thus, following the Penning collisions in the reaction region of the flowing afterglow, we would expect to have an ensemble of polarized electrons, and the problem now becomes one of suitably detecting them. For this we exploit the spin-exchange process between the free electrons and alkali-metal atoms due to Dehmelt.[16] A second oven containing Na metal is inserted into the afterglow. The original Na atoms are, of course, unpolarized, but subsequent to a spin-exchange collision with an electron, they acquire the polarization originally present in the free electrons. The presence of polarized Na atoms in the ground $^2S_{1/2}$ state is detected by passing circularly polarized light from a Na lamp along the field direction through the Na vapor. The transmission of the resonance radiation is then a measure of the polarization of the Na atoms. Figure 7 is a scan of the magnetic field through the $g=2$ resonance of the helium metastable atoms and the $g=0.5$ resonance of the Na atoms. A grating monochromator is used to isolate the D_1 and D_2 components of the Na resonance radiation. The probing light is sufficiently weak that it induces only a small polarization itself. In the absence of the helium optical pumping, the signal at $g=2$ disappears completely and the $g=0.5$ signal reduces to the curve labeled "*b*". The signals are reduced by more than an order of magnitude in the absence of a source of Penning electrons. For the curves shown in Fig. 7, Ar was used as the source of Penning electrons. Similar results were obtained with Cd, H_2, and N_2.

There is a possibility that the source of electron polarization is via spin-exchange collisions between the Penning electrons and the helium metastable atoms; certainly the exchange cross sections are sufficiently large. To eliminate this possibility the impurity density was maintained at a sufficiently high level that the helium metastable density in the vicinity of the Na oven was almost totally depleted by Penning quenching collisions. This, coupled with a flow velocity near 10^4 cm sec^{-1}, renders it unlikely that an electron created in the Penning collision would "find" a helium metastable atom and subsequently "find" a Na atom all in the millisecond or so required for the flowing gas to move between the Penning reaction zone and the Na probing light.

Walters *et al.* have extracted a beam of polarized electrons from a flowing-helium afterglow to which an impurity species was added.[17] The electron beam polarization is measured by Mott scattering from a gold foil after acceleration to 120 keV. They obtained polarizations in excess of 30%. Presumably the process described in the preceding paragraphs is responsible for the large observed polarizations.

E. He-He collisions

The collision between two helium metastable atoms results in the formation of He^+, an electron, and a deexcited helium atom in a Penning col-

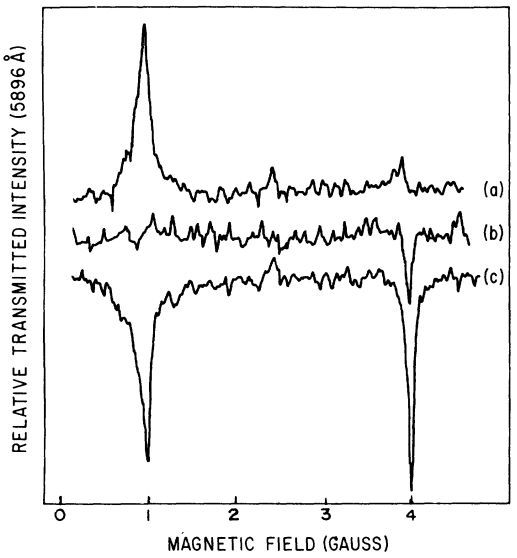

FIG. 7. Transmission of Na resonance light at 5896 Å. In curve (c) the helium pumping light and the sodium probing radiation have the same polarization. In curve (a) the polarizations have the opposite sense. Curve (b) is obtained when the helium metastable system is unpolarized. The resonance near 1 G is due to the helium metastable atoms and/or free electrons ($g=2$). The resonance near 4 G is due to the $^2S_{1/2}$ Na atoms.

lision. The cross section is large (10^{-14} cm^2) and has been measured earlier by several workers.[18] A requirement that the cross section be large is that the Wigner-spin rule must be satisfied. We have already seen that this rule implies that a component of spin angular momenta must be conserved along the external field direction. Now consider the case when a helium metastable atom in the $m_J = +1$ level collides with another helium metastable atom. There are three possibilities:

$$\begin{aligned} &\mathrm{He}^M(\uparrow\uparrow) + \mathrm{He}^M(\uparrow\downarrow) \rightarrow \mathrm{He}(^1S_0\uparrow\downarrow) + \mathrm{He}^+(\uparrow) + e(\uparrow), \\ &\mathrm{He}^M(\uparrow\uparrow) + \mathrm{He}^M(\downarrow\downarrow) \rightarrow \mathrm{He}(^1S_0\uparrow\downarrow) + \mathrm{He}^+(\downarrow) + e(\uparrow) \\ &\qquad\qquad \text{or } \rightarrow \mathrm{He}(^1S_0\uparrow\downarrow) + \mathrm{He}^+(\uparrow) + e(\downarrow), \\ &\mathrm{He}^M(\uparrow\uparrow) + \mathrm{He}^M(\uparrow\uparrow) \not\rightarrow \end{aligned} \tag{18}$$

If the colliding helium metastable atoms are both in the $m_J = +1$ magnetic sublevel, the Wigner-spin rule cannot be satisfied and the reaction must be zero.

At helium metastable densities approaching 10^{11} cm^{-3} ionizing collisions between metastable pairs is a significant process affecting the properties of the afterglow. Under appropriate conditions it can be the dominant source of electrons and ions. If this reaction is a significant source of electrons in the afterglow, it should be possible to modify the charge density by placing a large fraction of the helium metastables in the $m_J = +1$ level thereby reducing the metastable reaction rate.

To detect the charge density in the afterglow, a pair of copper plates 14×32 mm were inserted into the flow region and connected to a recording microammeter. The current flow between the plates is then monitored as a function of the helium metastable polarization. Figure 8 shows the current flow as the external field is scanned through the helium metastable resonance at $g = 2$. At resonance the metastable polarization is substantially reduced, and the current flow is *increased*. This observation verifies directly the requirement that the Wigner spin be satisfied if the reaction is to proceed with a large cross section.

There have been several earlier measurements of the He-He reaction rate. In these cases the reaction rates have been measured by observing the total metastable density in an unpolarized sample. If the three Zeeman levels are equally populated (as is generally the case in an electrical discharge) $\frac{2}{9}$ of the metastable-metastable collisions are between atoms both of which are in either the $m_J = +1$ or $m_J = -1$ levels. If the total density is observed, the measured cross section is a weighted average of $\frac{7}{9}$ with a large cross section and $\frac{2}{9}$ with a small (or zero) cross section. Thus, the observed cross sections are smaller than the actual cross sections for metastable quenching.

If we assume that the electrical discharge populates each of the three metastable Zeeman levels at equal rates, it can be easily shown that an alignment of the helium metastable atoms will be produced because of the unequal loss rates for the three levels via He^M-He^M collisions. To test this possibility, we examined the polarization of the 1.08-μm ($2\,^3P$-$2\,^3S_1$) radiation emitted by an electrical discharge in pure helium gas at 0.2-Torr pressure. We find that the 1.08-μm light emitted perpendicular to the external field direction is polarized [i.e., $I(\pi) \neq I(\sigma)$] and that the polarization can be quenched by the application of a resonance magnetic field to the helium metastable system which equalizes the Zeeman-level populations.

F. Polarized helium ions: Charge transfer

For a partially polarized metastable helium ensemble, Eq. (18) predicts that the helium ions should acquire a substantial polarization. The He^+ polarization cannot be detected directly. However, assuming that a charge-transfer collision con-

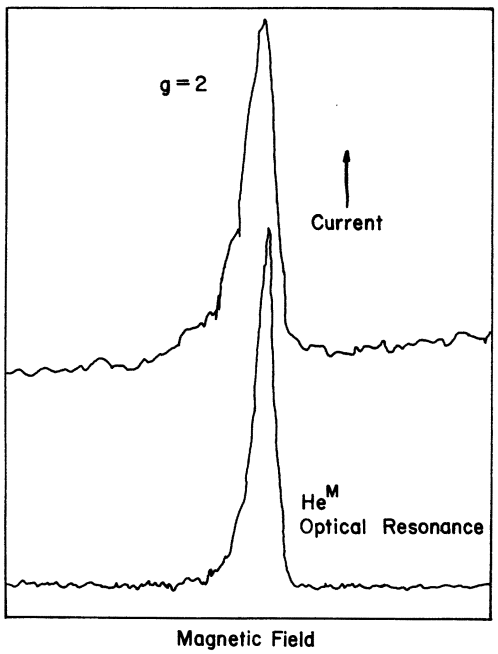

FIG. 8. Resonances shown occur at $g = 2$. The lower curve is the conventional helium metastable optical resonance. The upper curve is obtained by measuring the charge flow between copper electrodes placed in the afterglow region.

serves spin, one can utilize this process to transfer the polarization to a second species which can then be observed optically. A charge-transfer process between the helium ion and Zn yields $Zn^+(5\,^2D_{5/2})$ which subsequently decays radiatively to the $5\,^2P_{3/2}$ level with emission at 6102 Å. The cross section is 2.08×10^{-15} cm^2 for this charge-transfer collision.[8] The polarization of the emitted light at 6102 Å follows the polarization of the helium metastable system. The ion polarization in this case must have been derived from the charge-transfer collision with the He^+ since the $Zn^+(5\,^2D_{5/2})$ is not energetically accessible by Penning ionization of Zn with the helium metastable. We therefore conclude that the helium ion is polarized and that charge transfer is a spin conserving process.

G. He^M–molecular collisions

The addition of molecular impurities to a flowing-helium afterglow also results in a Penning ionization although the optical spectrum is complicated by other channels of reactions such as dissociative excitation. Others have exploited this type of experiment frequently labeled chemiluminescence. Our interest in this section is whether polarization can be transferred from the helium metastable system to molecular ions in a Penning reaction.

In the Penning collision of He^M with molecular nitrogen the $B\,^2\Sigma$ state of the ion is populated; it subsequently decays to the $X\,^2\Sigma$ ground state with emission in a number of bands. The $v'=0\rightarrow v=0$ decay is centered near 3914 Å, and with a $1/2M$ Jarrell-Ash spectrometer the P and R branches may be separated and the individual rotational lines observed. If we now monitor the polarization of the emission as before, we find that the emission is polarized when the helium metastable system is optically oriented. Figure 9 is a plot of the observed polarization as the wavelength is scanned through the 3914-Å band. The polarity of the signal changes as we go from the P branch in which the rotational quantum number, K, increases by 1 and the R branch for which $\Delta k=-1$. The area under the two portions of the curve is approximately the same so that a broad-band detector cannot be used to detect the polarization.

The observed polarization is sufficiently large that it should be possible to observe magnetic resonance transitions between the magnetic Zeeman sublevels of the excited ion to obtain g factors and lifetimes of the individual rotational levels. It may also be possible to observe transitions between the spin doubled components of individual rotational levels.

In Fig. 10 we display the optical spectrum of silane (SiH_4) from 2000–2500 Å which has been excited in the flowing-helium afterglow. Of particular interest are the observed Si I and Si II lines. At longer wavelengths (not shown) one observes both SiH and SiH^+ bands. We have not yet determined the mechanism by which these states are formed, but we include the spectra here to show the further value of flowing-helium afterglows. We have not yet examined the polarization

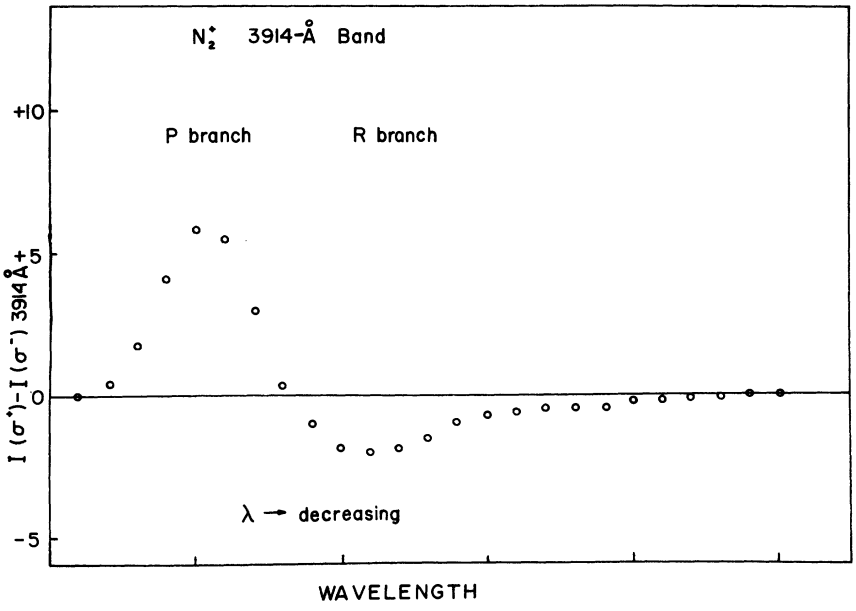

FIG. 9. Polarization of emission from N_2^+ as a function of wavelength for O-O vibrational band at 3914 Å.

of the emission to determine whether spin is conserved in these more complex collision processes.

H. Transfer of the transverse component of spin angular momentum

All the preceding experiments were concerned with the conservation of the longitudinal component of spin angular momentum. In the optically pumped helium metastable system the transverse component of the magnetic moment averages to zero because of the random phases of the precessing magnetic moments. It is, however, possible to induce a coherent precession of the transverse magnetization as shown by Dehmelt.[19] This is accomplished by the application of a weak resonance magnetic to the helium metastable system which couples the individual Zeeman levels. The coherent precession of the transverse component appears as a modulation of the transmission of the 1.08-μ resonance radiation at the resonance frequency and is detected by passing a second resonance light beam through the optically pumped ensemble of helium metastable atoms perpendicular to the external field direction. We now wish to inquire if the transverse component of spin angular momentum is conserved in the Penning-collision process.

Immediately after the Penning ionization has taken place the electron spins in the newly formed ion are uncorrelated with any orbital angular momentum. The electron spin then couples to the orbital angular momentum in a time which is a measure of the strength of the $\vec{L}\cdot\vec{S}$ coupling. The total angular momentum, J, then precesses about the external field. If the excited ion decays radiatively in a time short compared to the precession period, the total angular momentum of the ion will be driven at the precessional frequency of the helium metastable system. When this emission is observed along a direction perpendicular to the external field through a circular polarization analyzer, the intensity is amplitude modulated at the helium metastable resonance frequency. As the difference between the precessional frequencies of the helium metastable atom and the excited ion increases relative to the excited-ion decay rate, a phase difference develops between the precessing helium metastable atom and the observed ion emission. The phase difference is given by[20]

$$\phi = \tan^{-1}[(g-2)\mu_B H]\tau\,, \tag{19}$$

where $g\mu_B H$ is the precession frequency of the total angular momentum of the excited ion in the field H and τ is the radiative lifetime. As the phase difference becomes appreciable, the amplitude modulation of the emission decreases.

The lifetime of $Sr^+(5\,^2P_{3/2})$ is 6.53 nsec[21]; consequently, in magnetic fields smaller than 20 G, the

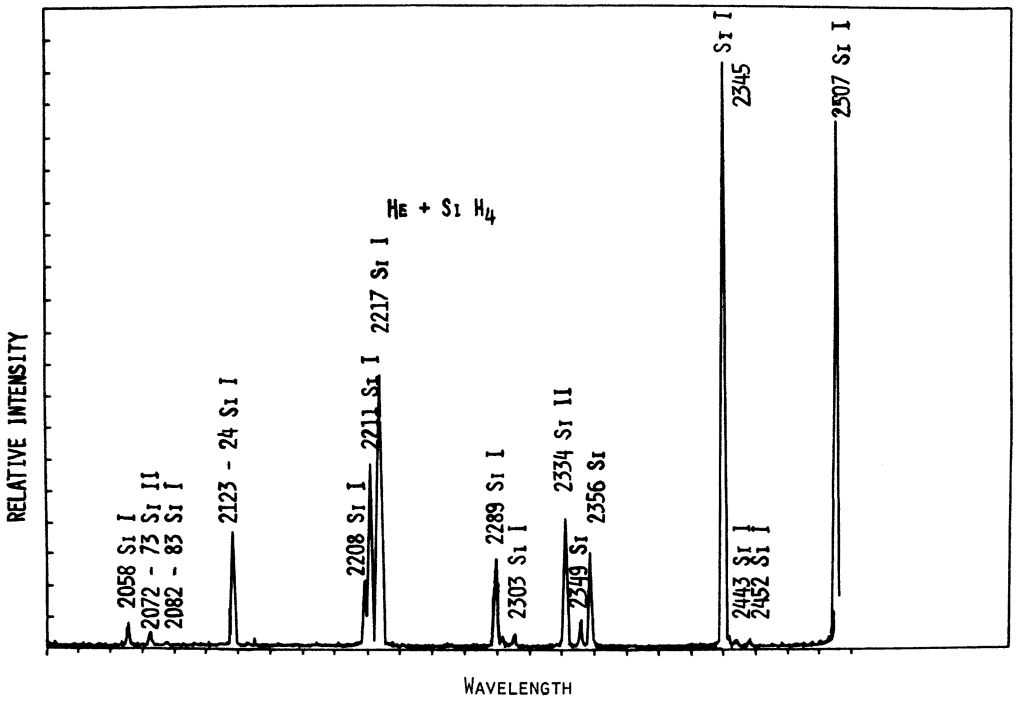

FIG. 10. Afterglow spectra of SiH_4 excited by collisions with helium metastable atoms and ions.

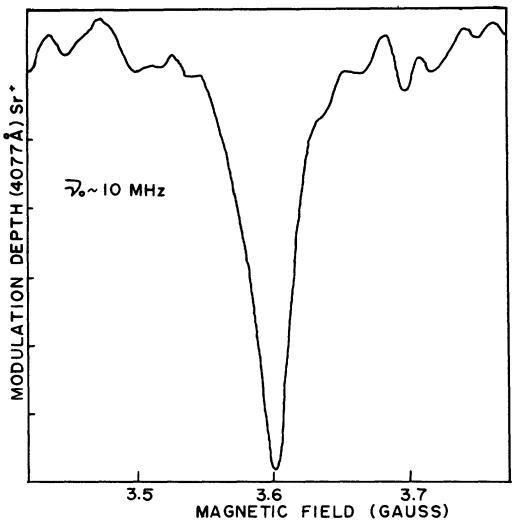

FIG. 11. Modulation amplitude of Sr^+ emission at 4077 Å. The 10-MHz modulation occurs at the $g=2$ resonance of the helium metastable atoms.

phase shift is small. We have observed the modulation of the emission at 4077 Å in $Sr^+(5\,^2P_{3/2}$-$5\,^2S_{1/2})$ which has been excited by Penning ionization with metastable helium atoms which are driven coherently at $g=2$ by a weak resonance magnetic field. In the flowing-helium afterglow the metastable atoms are optically oriented by a beam of resonance radiation directed along the external field direction. A weak resonance magnetic field at $g=2$ orthogonal to the pump beam couples the Zeeman levels of the metastable atoms and causes a coherent precession of the transverse component of the magnetization. By passing a second circularly polarized resonance light beam from a helium lamp through the afterglow perpendicular to both the pumping beam and the resonance magnetic field, the coherent precession of the metastable spin system is observed as a modulation of the transmitted helium resonance radiation.

The addition of Sr vapor into the afterglow region results in a Penning ionization of the Sr and excitation of the $Sr^+(5\,^2P_{3/2})$ level. The Sr^+ 4077-Å light emitted along a direction perpendicular to both the external magnetic field and the resonance magnetic field is observed through a circular analyzer. The emission is amplitude modulated at $g=2$ as shown in Fig. 11. We thus conclude that the transverse component of spin angular momentum is also conserved in the Penning process.

Observations of the intensity of the modulated light as a function of external field strength can be utilized to measure the quantity $\omega\tau$ for a variety of excited ions produced by Penning ionization. Either the phase shift ϕ in Eq. (19) can be determined or one can plot the amplitude of the modulation in phase with the helium metastable precession as a function of the external field. The amplitude of the in-phase component follows the relationship[20]

$$I \propto 1/[1+(\Delta\omega\tau)^2], \tag{20}$$

where $\omega\tau$ is the difference between the precession frequencies of the helium metastable atom and the excited ion in an external magnetic field.

IV. CONCLUSION

The fast-flowing optically pumped helium afterglow has shown itself to be a valuable tool for the observation of spin-dependent, inelastic collision processes. Its applications range from simple optical spectroscopy of excited ions to magnetic resonance spectroscopy of excited- and ground-state ions from which ion radiative lifetimes and g factors may be obtained. The technique can also yield beams of polarized electrons and polarized ions. More specifically it extends the range of optical-pumping experiments to ionic species.

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