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## Coherence Transfer in a Penning Ionizing Collision

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Modulation of the intensity of light emitted by  $\text{Sr}^+(^2P_{3/2})$  ions at the resonance frequency of metastable helium atoms has been observed, demonstrating that coherence has been transferred in the Penning collision. The coherent excitation of the Penning ions suggests that the techniques of "coherence spectroscopy" such as level-crossing and Hanle experiments can be extended to excited states of Penning ions which are currently inaccessible by means of optical excitation.

This Letter reports the first observation of the coherent excitation of ions in a Penning ionizing collision between metastable helium atoms and strontium atoms. The coherence appears as a modulation of the  $\text{Sr}^+$  emission at  $4077 \text{ \AA}$  ( $5^2P_{3/2}$ - $5^2S_{1/2}$ ) at the Zeeman resonance frequency of the helium metastable atoms and is evidence of the coherence between eigenstates of the  $\text{Sr}^+(^2P_{3/2})$  ions. The coherence is characteristic of the helium metastable atom and must have been transferred to the  $\text{Sr}^+$  in the Penning collision producing the ion. In the experiment to be described we show that coherence is transferred in a Penning reaction which in turn (1) implies that the transverse component of spin is conserved in the collision, (2) demonstrates the essential nature of the electron exchange postulated for Penning reactions,<sup>1,2</sup> and (3) shows that coherence spectroscopy may be successfully applied to excited states of ions which result from Penning collisions.

The experiments of Colegrove, Schearer, and Walters,<sup>3</sup> Partridge and Series,<sup>4</sup> and Ruff and Carver<sup>5</sup> showed that coherence between the eigenstates of an atom can be transferred from one atom to another in a collision. In the first of these, coherence between the eigenstates of the ground  $\text{He}^3$  atoms is transferred to the metastable atom by spin-exchange collisions. Modula-

tion at the nuclear-magnetic-resonance frequency of the ground state is detected by observing the absorption of resonance radiation by the metastable atoms. In the second experiment modulation at the hydrogen resonance frequency is observed in a transverse beam of sodium light. Observation of modulation in both experiments was taken as evidence of coherence transfer in spin-exchange collisions.

Gough<sup>6</sup> considered the question "whether there is any possibility of transfer of coherence in a collision of the second kind" and leaves the answer open. He had observed the Hanle effect in the cadmium fluorescence when a mixture of cadmium and mercury vapors is illuminated with polarized mercury light. Subsequently, Series<sup>7</sup> interpreted these observations as an experimental demonstration of coherence transfer in collisions of the second kind. More recently Elbel, Niewitecka, and Krause<sup>8</sup> have observed coherence transfer between the  $^2P_{1/2}$  and  $^2P_{3/2}$  states in sodium resulting from collision with helium atoms. In neither of these cases, however, was a modulation of the fluorescence observed.

The experimental apparatus is schematically represented in Fig. 1. It closely resembles the arrangement used earlier to observe the transfer of the longitudinal component of magnetization from metastable helium to a variety of ions.<sup>9</sup>

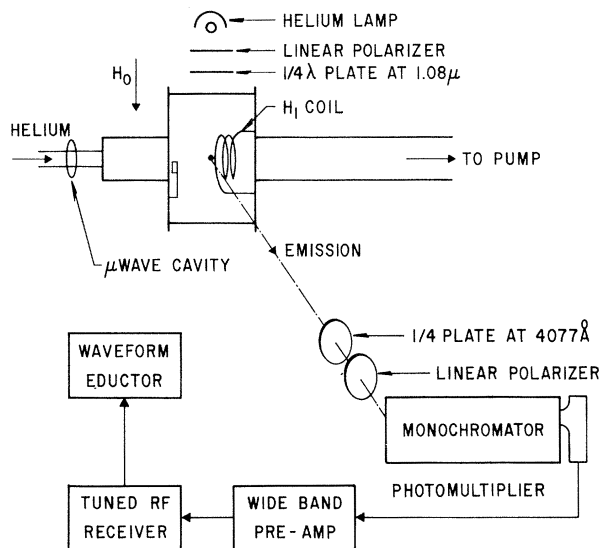
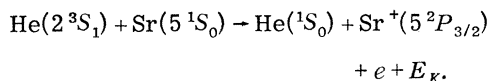


FIG. 1. Schematic representation of experimental apparatus.

The metastable helium atoms are optically pumped in the usual manner in the afterglow region of a flowing-helium discharge. The flowing-gas apparatus is located in a static magnetic field defined by a 52-cm diam Helmholtz-coil pair. The field is perpendicular to the flow and parallel to the pumping light beam. A weak radio-frequency magnetic field at the resonance frequency of the helium metastable atoms induces a coherent superposition of the eigenstates of the metastable system (a coherent precession of the transverse component of magnetization). The time-varying eigenstates can be observed by passing a second circularly polarized beam of resonance radiation from a helium lamp through the optical pumping region orthogonal to the field direction. This "crossed" light beam is then intensity modulated at the resonance frequency as first shown by Dehmelt<sup>10</sup> and Bell and Bloom<sup>11</sup> in the related Na optical-pumping experiments.

A small oven containing strontium metal intercepts a portion of the flow. As the strontium atoms enter the reaction region they undergo Penning collisions with the 19.78-eV helium  $^3S_1$  atoms. A particular reaction is given by



The  $\text{Sr}^+$  excited ion decays radiatively to the ground ion level  $^2S_{1/2}$  with emission at  $4077 \text{ \AA}$ . The emission perpendicular to the magnetic field is collimated, passed through a circular analyzer, isolated with a 0.25-m Bausch and Lomb

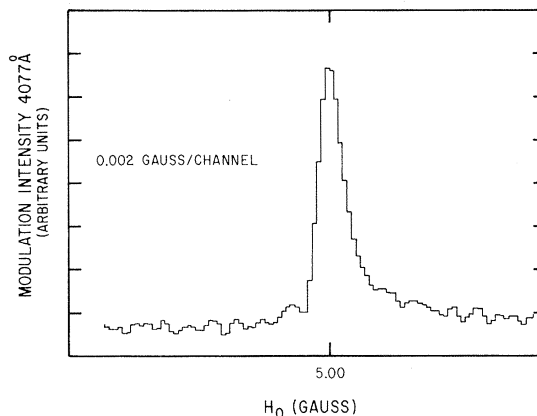


FIG. 2. Helium metastable resonance ( $g=2$ ) observed as a modulation of the  $\text{Sr}^+$  emission at  $4077 \text{ \AA}$ . The curve shown is the demodulated output of the receiver tuned to the helium metastable resonance frequency at 14 MHz. The magnetic field  $H_0$  is successively scanned through the resonance condition. The asymmetry in the resonance curve is a result of leakage of the 14-MHz field which mixes with the modulated light signal in the photomultiplier. The phase of the 14-MHz signal changes on either side of resonance. By increasing the leakage the resonance dispersion curve can be obtained.

grating monochromator, and detected with an S-13 photomultiplier. The photomultiplier output is terminated in a  $93\text{-}\Omega$  resistance to match the input impedance of a wide-band preamplifier. The output of the amplifier is fed to the antenna terminals of a Hallicrafters SX-130 communications receiver which is tuned to the rf magnetic field frequency. As the external magnetic field is scanned through the metastable helium resonance condition, a voltage appears at the detector output of the receiver. This voltage is applied to a PAR waveform eductor which is triggered synchronously with the start of the magnetic-field scan. The averaged output following 2000 scans is shown in Fig. 2. For the curve shown, the resonance frequency of the helium metastable atoms was 14 MHz ( $g=2$ ). Similar resonances were obtained for resonance frequencies between 1.5 and 21 MHz.

The duration of the Penning collision is short compared to any other magnetic interaction; consequently, the electron spins in the newly formed ion are uncorrelated with the orbital angular momentum. After a time corresponding to the fine-structure separation of the ion, the electron spin is coupled to the orbital angular momentum. However, since the orientation of the electron spin survives the collision, the total angular momentum of the ion is driven at the precessional

frequency of the metastable helium atoms. This frequency is very different from the resonance frequency of the  $\text{Sr}^+(^2P_{3/2})$  ion. The coherence initially present in the metastable helium system is transferred to the ion and persists until the phase difference between the precession frequencies of the metastable helium atoms and the  $\text{Sr}^+(^2P_{3/2})$  ions becomes appreciable. If the excited ion decays radiatively before this phase difference becomes large, the emission viewed perpendicular to the field direction through a circular analyzer will be amplitude modulated at the helium metastable resonance frequency. Coherence transfer will be observed, then, provided  $(\omega_0 - \omega_i)\tau \ll 1$  and will not be observable if  $(\omega_0 - \omega_i)\tau \gg 1$ , where  $\omega_0$  and  $\omega_i$  are the resonance frequencies of the metastable helium atoms and excited ions, respectively, in radians per second, and  $\tau$  is the radiative lifetime.

Partridge and Series<sup>4</sup> have developed a theory for coherence transfer in atomic collision with specific reference to the  $\text{He}^3$  optical-pumping system. The qualitative assertions of the preceding paragraph are in agreement with their conclusions, and only slight modification of their theory of coherence transfer is necessary to accommodate the observations reported here for Penning ionization.

The excited-state lifetime of  $\text{Sr}^+(5^2P_{3/2})$  has been measured to be  $\tau = 6.53$  nsec,<sup>12</sup> and  $(\omega_0 - \omega_i)\tau = 1$  at 26 G. Consequently, over the metastable resonance-frequency range covered in this experiment (1.5–21 MHz), the coherence-transfer condition was well satisfied. Limitations in the experimental equipment available prevented us from testing the condition in the region where  $(\omega_0 - \omega_i)\tau \gg 1$ . However, the radiative lifetime of  $\text{Cd}^+(5^2D_{5/2})$  is 773 nsec.<sup>13</sup> This ion emits at 4416 Å and was readily substituted for the strontium in the flowing-gas apparatus. In this case  $(\omega_0 - \omega_i)\tau > 1$  for fields exceeding 0.18 G, and modulation of the emitted radiation was not observed in spite of the fact that a large longitudinal component of polarization is transferred to the  $\text{Cd}^+(5^2D_{5/2})$  ion.<sup>9</sup>

Observations of the intensity of the light modulation as a function of field strength can be uti-

lized as a type of coherence spectroscopy to obtain the quantity  $\omega\tau$  for a variety of excited states of Penning ions, many levels of which are inaccessible by the more conventional methods. Level crossing experiments<sup>14</sup> and the related Hanle effect have generally been limited to systems which directly interact with light, the light providing a coherent excitation of the levels to be studied. This requires that these levels be optically connected to a ground or metastable level with radiative transitions in accessible spectral regions so that available light sources can be used for excitation. Level crossing, like modulation phenomena, can be described in terms of coherence between eigenstates. Since we have now demonstrated that coherence may be transferred from the helium metastable system to the ions of a second species created in a Penning collision, it should be possible to extend level-crossing observations to the excited states of Penning ions which are inaccessible by optical excitation.

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