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# Alignment of ions in Penning collisions

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The authors have observed the alignment of the $5p\,^2P_{3/2}$ state of strontium ions produced in Penning collisions between an unpolarized beam of helium metastable atoms and a strontium vapor target. The alignment is shown by a linear polarization of the optical emission from the excited ion. For a 66-meV beam of helium metastable atoms a 3.5% linear polarization of the emission relative to the beam axis was measured. It is shown how the alignment may be used to determine the probabilities for populating the various final quasimolecular states of the ion-atom pair. The alignment of the Penning ions is an important new parameter in the description of these reactions.

## I. INTRODUCTION

The process of Penning ionization has been the subject of many experimental and theoretical investigations. Typically in this process the excitation energy of a metastable atom is transferred to a target atom with the resultant ionization of this target atom. Although most studies have dealt with the total cross section for this process, coherent final states for the process have also been recently observed. In one case, Schearer has demonstrated in an afterglow experiment that the ions are formed in a coherent state if the metastable atoms are in a coherent state.[1] This has been shown to be a direct consequence of the applicablity of the Wigner spin rule.[2] In a second case, using a beam of metastable helium atoms Hotop and Niehaus have observed that the electrons ejected in the Penning ionization of argon are in a coherent state.[3] Subsequently Ebding and Niehaus have shown this to be a general characteristic of the Penning ionization process.[4] In these experiments the beam serves to define a preferred direction for the system. In this article we report for the first time the observation of a coherent state for the ions formed in Penning ionization utilizing a beam of unpolarized metastable atoms. In our particular experiment the coherent state is that of the strontium ions formed in the excited $5p\,^2P_{3/2}$ state by Penning ionization by metastable He($2s\,^3S_1$) atoms. The emission accompanying transitions from this excited state to the ion ground state is observed to have a positive linear polarization relative to the beam axis. This is a direct consequence of the alignment of the excited ions formed in Penning ionization. In Sec. II, the alignment of the Sr$^+$($5p\,^2P_{3/2}$) ion is shown to be related to the differential scattering cross section of the collision pair and the relative populations of the final quasimolecular states of the Penning process. The observation of an ion alignment thus provides unique information about the process and represents an important new tool in the study of Penning ionization. Section III describes the experiment and the results we obtain.

## II. THEORY

The theoretical framework for the Penning ionization process has been presented by Nakamura[5] and Miller.[6] *Ab initio* calculations have been performed for the Penning ionization of hydrogen atoms by metastable helium atoms.[7] Semiempirical calculations have also been made for the ionization of argon by helium metastables.[8] The analysis here is based on some principal features of the reaction which can be deduced from the above theory. In particular, the validity of the semiclassical description of the relative motion of the atom is assumed. In addition, the result that the ionization occurs in the region of closest approach of the atoms is used.

The electron angular momentum about the internuclear axis at the ionization is conserved. This suggests that a convenient description of the process would reference the state of the ejected electron to this axis.

A calculation similar to those performed for collisional depolarization of excited states of atoms by helium[9,10] indicates that the strontium-ion final state depends only on the system in the region of closest approach and not on the direction of the outgoing helium atom. This reflects the result that collisional depolarization of excited states by helium is characteristic of a hard-core rather than a long-range Van der Waals interaction.[9] The internuclear axis when ionization occurs is therefore the preferred reference axis for the ions. Taking the beam axis as the $z$ direction, the reaction axis is taken to be directed from the target atoms to the metastable atom at ionization. This is specified as the $\theta_r, \phi_r$ direction.

The discussion is simplified by using two properties which appear to be valid for the Penning process. The first is that the reaction occurs in the region of the repulsive potential.[8] For each scattering angle there is therefore an unique reaction axis. The second is that the cross section is independent of the electron spin.[1,2] This property is partially verified by our observation that the $5p\,^2P_{1/2}$ and $5p\,^2P_{3/2}$ states of $\mathrm{Sr}^+$ are statistically populated in the experiment reported here. It suffices therefore to neglect the electron spin in the course of the calculation and at the end couple the appropriate spin-density matrix (in the present experiment that of a random spin) to the orbital angular momentum of the ion to obtain the appropriate multiplet. The procedure is similar to that used to calculate the coherent ion state formed in Penning ionization by oriented metastable helium atoms.[2] Finally, in the absence of spin dependence, the process will exhibit cylindrical symmetry about the beam axis.

With the above conditions the process can be characterized by a scattering amplitude $f_\mu^{(l)}(\epsilon, \theta_s)$. This is the amplitude for the Penning ionization to occur with the following results. First, the helium atom is scattered into the angle $\theta_s$ relative to its incident direction. Second, the ejected electron has energy $\epsilon$, total-angular-momentum quantum number $l$, and angular momentum about the reaction axis $\mu\hbar$. And third, the strontium ion is in a $5p\,^2P$ state and has an electron orbital angular momentum of $-\mu\hbar$ about the reaction axis. This constraint on the electron angular momentum of the ion is the result of the conservation of angular momentum about the reaction axis, the decoupling of spin and orbital electron angular momentum, and the $\Sigma$ character of the ground-state strontium-$\mathrm{He}(2s\,^3S_1)$ system. The reaction axis lies in the scattering plane and makes an angle $\theta_r$, uniquely determined by $\theta_s$, with the beam axis and $\phi_r = \phi_s$, the azimuthal scattering angle.

Since the final state of the ion is a $^2P$ state the projection of the angular momentum on the reaction axis can only be zero, corresponding to a $^2\Sigma$ state, or $\pm\hbar$, corresponding to the $^2\Pi$ states. It is important to note that although it can be argued that the amplitude corresponding to the $\Sigma$ state is dominant,[11] it is not possible to *a priori* show that $\Pi$ amplitudes are negligible.

In terms of these amplitudes the differential cross section for Penning ionization with the helium atom scattered in the direction $\theta_s$ is, with the standard sum over the unobserved final states,

$$\frac{d\sigma(\theta_s)}{d\Omega_s} = \sum_{\mu=-1}^{+1} \sum_{l=|\mu|}^{\infty} \int |f_\mu^{(l)}(\epsilon, \theta_s)|^2 d\epsilon \,. \tag{1}$$

The lack of interference between amplitudes is the result of a unique reaction axis for a given scattering direction. In order to obtain the density matrix describing the ions it is necessary to use a common basis set for all reaction axes. The eigenstates of the angular momentum projected onto the beam axis is the preferred set. The beam defines the $z$ direction. These states are related to the corresponding states on the reaction axis at the angle $\theta_r, \theta_s$ by the rotation matrices[12]

$$\mathfrak{D}_{m\mu}^{(1)}(-\phi_s, \theta_r, 0) = d_{m\mu}^{(1)}(\theta_r) e^{-im\phi_s} \,, \tag{2}$$

with $m\hbar$ and $\mu\hbar$ the components of the electron orbital angular momentum on the $z$ axis and reaction axis, respectively. In terms of the eigenstates of the $z$ component of electron orbital angular momentum, i.e., still neglecting the electron spin, the density matrix for the ions is

$$\langle m|\rho|m'\rangle = \sigma_T^{-1} \sum_{\mu=-1}^{+1} \sum_{l=|\mu|}^{\infty} \int \mathfrak{D}_{m\mu}^{(1)}(-\phi_s, \theta_r, 0)\, |f_{-\mu}^{(l)}(\epsilon, \theta_s)|^2 \mathfrak{D}_{m'\mu}^{(1)}(-\phi_s, \theta_r, 0)^* \, d\Omega_s \, d\epsilon \,, \tag{3}$$

where $\sigma_T$ is the total Penning cross section resulting in a $5p\,^2P$ ion. The unique reaction axis for each scattering direction together with the sum over the states of the unobserved ejected electron again removes any interference between amplitudes.

The integration over $\phi_s$ can be carried out and yields the diagonal density matrix

$$\langle m|\rho|m'\rangle = \delta_{mm'} 2\pi\sigma_T^{-1} \sum_{\mu=-1}^{+1} \sum_{l=|\mu|}^{\infty} \int |f_{-\mu}^{(l)}(\epsilon, \theta_s)|^2 [d_{m\mu}^{(1)}(\theta_r)]^2 \sin\theta_s \, d\theta_s \, d\epsilon \,. \tag{4}$$

Using the obvious symmetry $|f_{-1}^{(l)}(\epsilon, \theta_s)| = |f_{+1}^{(l)}(\epsilon, \theta_s)|$ the density matrix is found to depend on three parameters. The first, the probability $p_0$ of forming the ion in a $\Sigma$ state relative to the reaction axis, is given by

$$p_0 = 2\pi\sigma_T^{-1} \sum_{l=0}^{\infty} \int |f_0^{(l)}(\epsilon, \theta_s)|^2 \sin\theta_s \, d\theta_s \, d\epsilon \,. \tag{5}$$

The other two are the averages of $\cos^2\theta_r$ for those ions formed in the $\Sigma$ state and in the $\Pi$ states relative to the reaction axis; denote them by $\beta_\mu$ with $\mu = 0$ for the $\Sigma$ state and $\mu = \pm 1$ for the $\Pi$ states. These are given by

$$\beta_\mu = \frac{\int \sin\theta_s \cos^2\theta_r \sum_{l=|\mu|}^{\infty} |f_\mu^{(l)}(\epsilon, \theta_s)|^2 d\theta_s d\epsilon}{\int \sin\theta_s' \sum_{l'=|\mu|}^{\infty} |f_\mu^{(l')}(\epsilon', \theta_s')|^2 d\theta_s' d\epsilon'} . \qquad (6)$$

Assuming that the spin state of the $Sr^+(5p\,^2P)$ ion is random the electron orbital angular momentum of the ion is coupled, in the *LS* coupling scheme, to a random spin. The resulting $^2P_{1/2,3/2}$ system is described by a density matrix obtained by combining the matrix given for the orbital angular momentum with that corresponding to a random spin.[2] The experiment reported here observes only the subdensity matrices corresponding separately to the $^2P_{1/2}$ and $^2P_{3/2}$ ions. Only the $^2P_{3/2}$ ions can exhibit alignment[13] and they are described by a density matrix which is diagonal for those electron states which are eigenstates of angular momentum projected on the beam axis. In terms of these states the nonzero matrix elements are

$$\langle \tfrac{3}{2} | \rho | \tfrac{3}{2} \rangle = \langle -\tfrac{3}{2} | \rho | -\tfrac{3}{2} \rangle = \tfrac{1}{8}(2+\Delta) \qquad (7a)$$

and

$$\langle \tfrac{1}{2} | \rho | \tfrac{1}{2} \rangle = \langle -\tfrac{1}{2} | \rho | -\tfrac{1}{2} \rangle = \tfrac{1}{8}(2-\Delta) , \qquad (7b)$$

where

$$\Delta = P_1(3\beta_1 - 1) - p_0(3\beta_0 - 1) \qquad (7c)$$

and

$$p_1 = \tfrac{1}{2}(1 - p_0) . \qquad (7d)$$

$p_1$ and $p_0$ are the probabilities of forming a $\Pi$ or $\Sigma$ state on the reaction axis.

The linear polarization of the emission from this $^2P_{3/2}$ state in the decay to the $^2S_{1/2}$ ground state of the ion is[13]

$$P = -3\Delta/(8-\Delta) . \qquad (8)$$

The polarization is therefore simply related to the relative probabilities for the excited ion to be created in the $\Pi$ or $\Sigma$ state along with the average square of the cosine of the angle for the interatomic axis for which the reaction occurs.

## III. EXPERIMENT AND DISCUSSION

The beam source of helium metastable atoms utilized in these experiments is a modification of a previously reported design in which an electric discharge is maintained in a gas undergoing nozzle expansion.[14] Under the appropriate discharge conditions helium metastable densities exceeding $10^{14}$ atoms/sr sec are obtained with a most probable beam energy of 66 meV and a width of the velocity distribution of 45%. Although the ratio of singlet to triplet metastable densities was not measured, it is likely to be small as in other discharge sources.[15]

The metastable beam entered a 97*l* reaction volume after passing through a differential pumping wall. At the center of the reaction volume a resistively heated boron nitride crucible containing strontium metal provided a strontium vapor density of the order of $10^{14}/cm^3$. The interaction volume was approximately 1 $cm^3$ and the metastable beam current was approximately $10^{11}$ metastable atoms/sec.

The optical detection system observed the interaction volume at right angles to the beam axis through a window in the vacuum wall. The optical emission from excited target ions was focused by two quartz lenses through a half-wave plate rotating at angular frequency $\omega$ and a fixed linear polarizer. The light transmitted by the polarizer was filtered with a interference filter or monochromator and detected by an *S*-19 phototube. When the light incident on the detection system had a net linear polarization, then a time-varying signal was detected by the phototube with the dependence given by $\cos^2(2\omega t)$. The amplitude of this signal is directly proportional to the magnitude of the linear polarization. The system was calibrated in terms of the magnitude and sign of the polarization by introducing another fixed linear polarizer in front of the half-wave plate whose transmission properties were known. In order to check the level of instrumental polarization in the detection system, a magnetic field solenoid was placed in a direction parallel to the observation direction and near the interaction volume. By establishing a large magnetic field transverse to the beam direction, all coherence produced in the excited ion states is destroyed; hence, the resulting emission is completely depolarized.

For helium metastable atoms incident on strontium, a nonzero polarization was observed for the $5p\,^2P_{3/2}$-$5s\,^2S_{1/2}$ emission of the strontium ion. The magnitude of the optical polarization was 3.5% with an uncertainty of 0.5%, and the polarization was parallel to the beam axis. The measured 0.5% uncertainty was determined primarily by the signal-to-noise ratio of the averaged data. There was no uncertainty in the sign of the polarization because it is determined simply by a visual comparison of the phase of the actual signal and the calibration signal.

The residual magnetic field in the interaction region was less than 0.5 G. Since the radiative lifetime of the strontium ion level we are observing is about 6 nsec, the residual field contributes a negligible error to the polarization measurement.[16] In addition the averages over the beam velocities and the thermal velocity of the target have a negligible effect compared to the uncertainty in the measurements.

The importance of cascade contributions to the intensity of the observed ion line was determined by taking an optical emission spectrum of the excited strontium. At these collision energies the $5p\,^2P_{3/2}$-$5s\,^2S_{1/2}$ transition at 407.8 nm has the largest emission cross section. Any emission whose lower state was the $5p\,^2P_{3/2}$ ion state characteristically had an intensity smaller than that of the $5p\,^2P_{3/2}$-$5s\,^2S_{1/2}$ intensity by a factor of 10 or more. Thus, the neglect of cascade contribution is justified for these initial measurements.

It follows from the discussion in Sec. II that given the differential scattering cross section, such as exists for the He($2\,^3S_1$)-Ar system,[15] the alignment measurement reported here would determine by Eq. 7(c) the relative population of the quasi molecular states which occur in this Penning reaction. Knowledge of this state population for a given Penning collision pair will eliminate the need for an *a priori* assumption of the population that has often been necessary to interpret other Penning studies.[17] The alignment of the ion formed in Penning ionization reactions is therefore an important new parameter in the description of these reactions.

Finally, we note that the observation of ion alignment is not restricted to the He($2s\,^3S_1$)-Sr system. Initial measurements in the same apparatus with Ca and Ba targets have also shown a nonzero alignment in the $^2P_{3/2}$ ion level. Tentatively, the Ca polarization is near +5.5% and the Ba polarization is under +1%. Thus, it appears that the Penning ions will, in general, exhibit a nonzero alignment at these collision energies.

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[1] L. D. Schearer and L. A. Riseberg, Phys. Rev. Lett. 26, 599 (1971).

[2] W. F. Parks and L. D. Schearer, Phys. Rev. Lett. 29, 531 (1972).

[3] H. Hotop and A. Niehaus, Chem. Phys. Lett. 8, 497 (1971).

[4] T. Ebding and A. Niehaus, Z. Phys. 270, 43 (1974).

[5] H. Nakamura, J. Phys. Soc. Jpn. 26, 1473 (1969).

[6] W. H. Miller, J. Chem. Phys. 52, 3563 (1970).

[7] W. H. Miller, C. A. Slocomb, and H. F. Schaefer, J. Chem. Phys. 56, 1347 (1972).

[8] A. P. Hickman and H. Morgner, J. Phys. B 9, 1765 (1977).

[9] A. I. Okunevich and V. I. Perel. Sov. Phys. JETP 31, 356 (1970).

[10] A. Omont, J. Phys. Radium 26, 26 (1965).

[11] L. A. Riseberg, W. F. Parks, and L. D. Schearer, Phys. Rev. 8, 1963 (1973).

[12] A. R. Edmonds, *Angular Momentum in Quantum Mechanics*, 2nd ed., (Princeton University, Princeton, 1957), Chap. 4, p. 55.

[13] U. Fano and J. H. Macek, Rev. Mod. Phys. 45, 553 (1973).

[14] D. W. Fahey, W. F. Parks, and L. D. Schearer, Rev. Sci. Instrum. 49, 503 (1978).

[15] B. Brutschy, H. Haberland, and K. Schmidt, J. Phys. B 9, 2693 (1976).

[16] F. H. K. Rambow and L. D. Schearer, Phys. Rev. 14, 1735 (1976).

[17] D. A. Micha and H. Nakamura, Phys. Rev. A 11, 1988 (1975).