Collective Propensity of Orientation for Multielectron Ions in Collisions

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The orientation parameter, $\langle J_y\rangle/J(J+1)$, of the $^2F_{7/2}$ state of Ar^+, formed in the two-electron process $He^+ + Ar \rightarrow He(1s^2) + Ar^+(3p^4 4p, {}^2F_{7/2})$ has been measured at various scattering angles. The measured value of $\langle J_y\rangle$, the component of the total angular momentum perpendicular to the collision plane, is interpreted as resulting from a collective circulation of the inactive $3p^4$ core electrons as well as the active $4p$ electron. We find that over the range of scattering angles studied, this collective-electron circulation is in the direction of the rotating internuclear axis, the direction given by the propensity rule, suggesting that this rule is more general than heretofore proposed.

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A property of excited states of ions and atoms formed in collisions with directed beams of electrons, ions, atoms, and molecules is that the electron distributions of states are usually nonspherical when the orbital angular momentum quantum number is greater than zero. They can also circulate with a net angular momentum perpendicular to the collision plane of any given collision [1].

Experimental measurements of the electron charge distribution and the circulation of the probability flux density of electronic states, formed in collisions, have provided, at the most fundamental level, unprecedented detail about the dynamical characteristics of the state of the atom and have often provided ultimate tests of detailed collision models and calculations [1]. Even in the absence of detailed calculations, new levels of insights into collision dynamics often result from such measurements [2].

The electron circulation is of special interest because of a propensity rule for colliding diatom systems, which states that the electron probability flux density of an excited atom or ion tends to circulate in the direction of the rotating internuclear axis [1,3]. Stated another way, the angular momentum of the colliding nuclear system tends to get transferred into the angular momentum of the circulating electrons of the excited atoms or ions.

The propensity rule is important because it is one of those few broad principles that connect and organize seemingly complicated and diverse diatom, inelastic collisions; it can be thought of as an approximate selection rule. Because of this importance, the propensity rule has been the focus of several recent theoretical and experimental investigations in diatom collisions, in an attempt to extract the underlying physics that connects different systems [4-6].

The experimental studies of the propensity rule to date, and the supporting theories, have been for either a pure one-electron system or, in the case of a multielectron system, a single active electron outside a filled shell that can be treated as a quasi-one-electron system. Special effort has even been made to find and study systems that have one active electron around an isotropic core [5,6].

We present here experimental results that indicate the propensity rule may be even more universal than suspected, in that a new electronic circulation can occur in more than the active, excited electron. In the case when an atom or ion has unfilled inner shells, a collective propensity of the electron circulation may also occur in the unexcited electrons. Specifically, we find in the case of the simultaneous removal by charge transfer and excitation of outer-shell electrons of Ar in 1 keV collisions with He^+, in the reaction

$$He^+ + Ar(3p^6) \rightarrow He(1s^2) + Ar^+(3p^4 4p, {}^2F_{7/2}) ,$$

that even the electrons in the partially filled-inactive $3p^4$ core, as well as the active $4p$ electron, circulate according to the propensity rule. Their net orbital angular momentum approaches the theoretical maximum value for some laboratory scattering angles. This suggests a common interaction of the $3p$ electrons during the collision, resulting in their collective circulation, but an excitation of only one of them. Such collective effects have not been studied in any detail and can only be observed using the techniques of alignment and orientation presented here.

The collective electron circulation is related to the mean value of the total angular momentum $\langle J_y\rangle$ perpendicular to the collision plane, a plane defined by the incident beam and the outgoing neutral $He(1s^2)$. Since argon has a nuclear spin of zero, only electronic spin and orbital angular momentum contribute to $\langle J_y\rangle$. Incorporating the formalism of Fano and Macek [7], we describe $\langle J_y\rangle$ by using their definition of the orientation parameter, O^c_{1-}, which for atomic states of definite J is

$$O^c_{1-} = \frac{\langle J_y\rangle}{J(J+1)} = \frac{\sum_{M_J} \mathrm{Tr}\rho(M_J) M_J}{J(J+1)} ,$$

where $\rho(M_J)$ is the density matrix of the excited state. We use the coordinate system of Fano and Macek where the initial beam direction defines the z axis. We also use the Fano-Macek normalization of $J(J+1)$.

To determine O^c_{1-} uniquely, for a given scattering angle of the $He(1s^2)$, one must measure the normalized Stokes parameter P_1,P_2,P_3 for the polarized radiation of the $^2F_{7/2}$ to $^2D_{5/2}$ transition (461 nm) emitted perpendicular to the collision plane, as well as the linear polariza-

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tion P_4 for radiation emitted in the collision plane, along the Y axis of Fig. 1. These quantities are defined as

$$P_1=\frac{I_\parallel-I_\perp}{I_\parallel+I_\perp},\quad P_2=\frac{I(45°)-I(-45°)}{I(45°)+I(-45°)},$$

$$P_3=\frac{I(\mathrm{RHC})-I(\mathrm{LHC})}{I(\mathrm{RHC})+I(\mathrm{LHC})},\quad P_4=\frac{I'_\parallel-I'_\perp}{I'_\parallel+I'_\perp},$$

where $I_\parallel$, $I_\perp$, and $I(\pm 45°)$ are the intensities, respectively, of the radiation polarized parallel, perpendicular, and at ±45° to the initial beam direction. The parameter P_3 measures the degree of circular polarization, where RHC (LHC) refers to the classical right- (left-) handed circular polarization or negative helicity.

Fano and Macek have provided a convenient formalism for describing the emission intensity of linear and circularly polarized light from atomic systems in terms of the expectation values of various combinations of Cartesian components of total angular momentum [7]. In addition to O^c_{1-}, already defined, there are three alignment parameters, A^c_0, A^c_{1+}, and A^c_{2+}, that describe the excited state in terms of expectation values of the Cartesian components of total angular momentum, as well as the orientation parameter. From the general expressions for various polarization components of the intensity provided by the Fano-Macek formalism, one can determine four relationships relating P_1 through P_4 to the orientation parameter and the three alignment parameters. These relationships are readily found to be

$$O^c_{1-}\equiv\frac{\langle J_y\rangle}{J(J+1)}=\frac{2P_3[P_4+1]}{h^{(1)}[P_1P_4-P_1-P_4-3]},$$

$$A^c_0\equiv\frac{\langle 3J_z^2-\mathbf{J}^2\rangle}{J(J+1)}=\frac{-2[P_1(2P_4+1)+P_4]}{h^{(2)}[P_1P_4-P_1-P_4-3]},$$

$$A^c_{1+}\equiv\frac{\langle J_xJ_z+J_zJ_x\rangle}{J(J+1)}=\frac{-2P_2[P_4+1]}{h^{(2)}[P_1P_4-P_1-P_4-3]},$$

$$A^c_{2+}\equiv\frac{\langle J_x^2-J_y^2\rangle}{J(J+1)}=\frac{+2(P_1-P_4)}{h^{(2)}[P_1P_4-P_1-P_4-3]},$$

where $h^{(1)}$ and $h^{(2)}$ depend only upon the initial and final J in the $^2F_{7/2}$ to $^2D_{512}$ transition. For this transition, $h^{(1)}=4.5$ and $h^{(2)}=-0.75$ [7].

The Stokes parameters and P_4 were measured by detecting the characteristic radiation in coincidence with the scattered He($1s^2$) at specific scattering angles, using the apparatus schematically shown in Fig. 1. He$^+$ was formed in a uniplasmatron source, a beam was extracted, mass analyzed, and collimated before interacting with a target of Ar formed in a gas jet from a small hypodermic needle. Scattered neutral helium atoms were detected by microchannel plates mounted in a chevron configuration. Individual copper anodes behind the microchannel plates that collected the amplified electron charge defined the various scattering angles. A wedge-shaped mask over the microchannel plates limited the azimuthal scattering angle to ±10° about the X' and Y' axis. The primary He$^+$ ion beam was bent into a Faraday cup by a set of deflection plates that are not shown in Fig. 1. Great care was taken in the calibration of the transmission properties of the linear polarizer and the determination of the phase shift and fast axis of the near-quarter-wave plate [8,9].

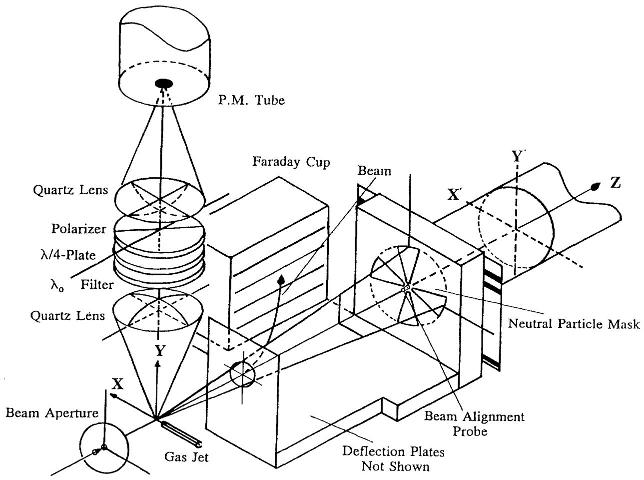

FIG. 1. Schematic of apparatus for measuring polarized radiation in coincidence with He($1s^2$).

Our results of the orientation parameter measurements for the $^2F_{7/2}$ state of Ar$^+$ are shown in Fig. 2 for positive scattering angles in the first quadrant of the X-Z plane. Results for negative angle scattering give identical results within the quoted errors, except the results are of opposite sign. The error bars in the scattering angle direction represent the finite width of the detector anodes behind the microchannel plates. The error in O^c_{1-} represents 1 standard deviation and takes into account counting statistics, uncertainties in the polarizer angle and transmission properties of the polarizer, as well as uncertainties in the value of the retardation of the near-quarter-wave plate and the fast-axis direction of the quarter-wave plate. We note that the expectation value of the angular momentum, $\langle J_y\rangle$, is negative for positive scattering angles, with the value of O^c_{1-} reaching -0.164 ± 0.003 for the smallest measured angle, 0.94°.

Included in Fig. 2 are the approximate impact parameters that are covered in our scattering range. These have been determined using tabulated Hartree-Fock charge densities to determine central scattering potentials. We also note that these impact parameters are just beyond the maximum of the radial wave function of the $3p$ orbitals of $^2F_{7/2}$ of Ar$^+$ [10]. These radial functions are shown in Fig. 3.

In order to discuss the contributions of the $3p^4$ core and the active $4p$ electron to O^c_{1-}, we first consider the coupling scheme that best describes the excited states of Ar$^+$ [11]. This scheme is one where the excited $4p$ electron of interest is coupled to the $3p^4$ core by L-S cou-

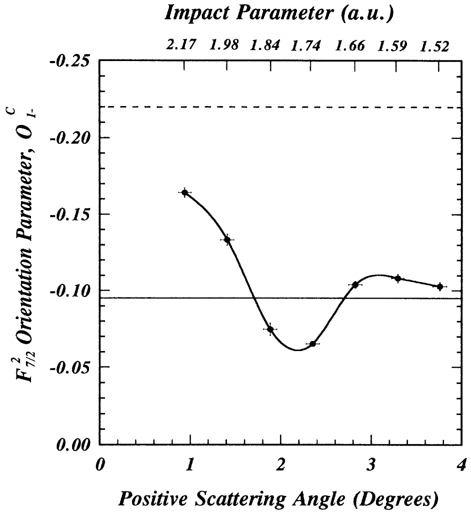

FIG. 2. Orientation parameter O^c_{1-} of $Ar^+(^2F_{7/2})$ formed in collisions with He^+, as functions of laboratory scattering angle and impact parameter. Dashed line represents maximum possible orientation. Solid line represents maximum possible contribution of $4p$ electron to orientation.

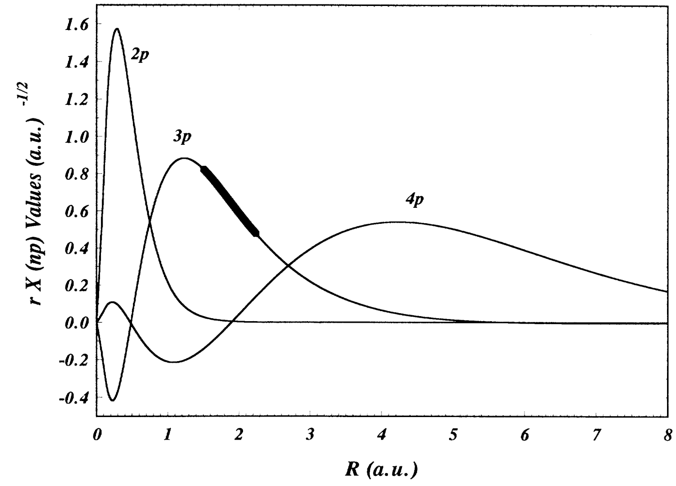

FIG. 3. Radial wave functions $rX(np)$ for p orbitals of Ar^+. The enhanced line on $3p$ function indicates region of present measurements.

pling. The $3p^4$ core electrons also couple among themselves by L-S coupling to produce either a ^3P, ^1D, or ^1S core state. This coupling scheme appears to hold very well for states with an active $4p$ electron. The $^2F_{7/2}$ state is then described by an active $4p$ electron coupled to a ^1D core by L-S coupling. In general the expectation value $\langle J_y \rangle$ of the $^2F_{7/2}$ state has contributions from both the active $4p$ electron and the inactive $3p^4(^1D)$ core electrons.

The maximum negative value of O^c_{1-} for the $^2F_{7/2}$ state occurs when $\langle J_y \rangle = M_J = -\frac{7}{2}$, giving a maximum value of $O^c_{1-} = -0.22$. For this case the pure state function $\psi(\frac{7}{2}, -\frac{7}{2})$ can be written as the product

$$\psi(\tfrac{7}{2}, -\tfrac{7}{2}) = \psi_c(M_L = -2)\psi_a(m_l = -1)\chi_a(-\tfrac{1}{2}),$$

where ψ_c is the wave function for the core and ψ_a and χ_a are the orbital and spin functions of the active $4p$ electron. For this extreme case, the maximum total orientation is the sum of the orientation of the $3p^4(^1D)$ core electrons and the orientation of the active $4p$ electron. In particular, the $4p$ electron gives a maximum possible contribution to the total orientation, namely, $(-\frac{3}{2})/J(J+1) = -0.095$, which is 43% of the theoretical maximum, -0.22. As a point of reference we show this value of -0.095 in Fig. 2.

Our measured value of -0.164 for the $^2F_{7/2}$ orientation, at this angle, is clearly not described by the pure state function product given above, although the state is clearly dominated by large negative values of M_J. However, the value of -0.095 does provide an upper limit for the contribution of the active $4p$ electron for any possible state. Since this upper limit of -0.095 is considerably less than the measured value (see Fig. 2) and is significantly outside the error of the measured value, we must conclude that significant orientation of the $3p^4(^1D)$ core is present at the small scattering angles.

In addition, the sense of the net orientation, or circulation of the inactive $3p^4$ electrons, is in the same direction as the active $4p$ electron; both circulate in the direction given by the propensity rule, that being the same direction as the internuclear axis rotation. As stated earlier, this suggests a common interaction of the $3p$ electrons during the collision. Our results indicate that excitation is not a requirement for the transfer of angular momentum.

Both the core state configuration and active electron are dominated by single electron states of $m_l = -1$. Thus, the $^2F_{7/2}$ state of Ar^+ for 1 keV collisions exhibits a net collective circulation of the $3p^4$ and $4p$ electrons at our smallest scattering angles; the contribution of the $3p^4$ electrons to the net circulation is roughly the same as the active $4p$ electron.

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