Coincidence Experiments Concerning Convoy-Electron Production by 1–8.5 MeV/u Highly Ionized Projectiles Traversing Polycrystalline Solids and Axial Channels in Gold

S. B. Elston, I. A. Sellin, M. Breinig, S. Huldt, L. Liljeby, and R. S. Thoe
University of Tennessee, Knoxville, Tennessee 37916, and Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830

and

S. Datz and S. Overbury
Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830

and

R. Laubert
East Carolina University, Greenville, North Carolina 27834
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Convoy electrons produced by 1–8.5 MeV/u O^{q+} ions traversing polycrystalline C, Al, and Au targets and ⟨110⟩ and ⟨100⟩ axial channels in Au are detected in coincidence with emergent ions of charge state q_e. The ***yield*** integrated over electron velocity for *channeled* ions depends strongly on q_e, but the ***shape*** is found to be independent of q_e, v, and target material. For polycrystalline and randomly oriented monocrystalline targets, the yield is found to be ***independent*** of emergent-ion charge state q_e. The data suggest that convoy-electron production is a bulk process.

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A sharp cusp in the spectrum of electrons, ejected in ion-solid and ion-atom collisions, is observed when the electron velocity $\vec{v}_e$ matches that of the emergent ion, $\vec{v}$, in both magnitude and direction.[1-3] In ion-atom collisions,[4] the electrons originate from capture to the continuum (ECC) for fast bare or nearly bare projectiles, and from loss to the continuum (ELC) when loosely bound projectile electrons are available. The ECC cusps are strongly skewed toward *lower* velocities and exhibit a full width at half maximum roughly proportional to v. A close examination of our recent ELC data show that ELC cusps are nearly *symmetric*, with widths independent of v in the velocity range 6–18 a.u., a result unpredicted by recent theory.

In contrast, the cusps characterizing "convoy"-electron production in ion-solid collisions are skewed toward *high* electron velocities, but exhibit velocity-independent widths that we find to be very similar to ELC widths. While the shape of the convoy peaks is independent of projectile Z_1 and of target material, the yields in polycrystalline targets exhibit a strong dependence on projectile Z and velocity.[3] Numerous attempts have been made to link convoy-electron production to binary ECC or ELC processes, usually at the exit surface.[3-5] As an alternative, a solid-state wake-riding model has been proposed.[6] Measured dependences[3] of shape and yield on projectile charge state and energy are inconsistent with the predictions of either theory.[6,7]

To aid in unraveling this puzzling array of similarities and differences, we have recently initiated coincidence experiments to investigate the dependence of shape and yield of the convoy-electron spectrum on the charge q_e of the associated emergent ion, for ions traversing polycrystalline targets as well as for well-channeled ions traversing a gold single crystal. For the best channeled ions, the fact that their charge often does not change during their entire passage[8] through the crystal makes such channeled ions useful probes. In our experiments, ion beams of ~ 1 nA intensity and various incident charge states were obtained from the Oak Ridge National Laboratory tandem accelerator ($Z_1 = 6$, 8; $E_1 = 1$–2.5 MeV/u), from the Brookhaven National Laboratory tandem accelerator ($Z_1 = 8$, 14; $E_1 = 1$–4 MeV/u), and from the Lawrence Berkeley Laboratory SuperHILAC ($Z_1 = 18$, $E_1 = 6$–8 MeV/u). Most electron spectra were acquired with electrostatic analyzers operated at an energy resolution $\Delta E/E$ of (1–1.4)% [full width at half maximum (FWHM)], which was set by source size and by a suitable aperture at the exit focus. The solid targets

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were placed at the entrance focus of the analyzer. Incident ions were usually collimated to 0.06 ± 0.02 deg, so that, for example, ≳ 97% of all incident 2.5-MeV/u oxygen ions were within typical channeling acceptance angles. Apertures set the analyzer acceptance angle θ_0 to ~1.5–1.7 deg. A hole in the outer plate of the analyzer permitted transmission of the emergent ion beam, which was subsequently focused by a quadrupole doublet, dispersed horizontally ~5 deg by a magnet, and finally collected by a second CEM, some 6 m downstream from the targets. Magnetic fields in the electron analysis region were reduced to ≲ 10^{-6} T by external coils. Pressures of ~ 10^{-7} Torr were maintained in the beam lines to reduce ion-charge–changing collisions. Data normalization was possible with use of either a 1-m-long Faraday cup located just upstream of the ion CEM, or with the CEM itself. Electrons in the singles spectrum could be allocated among ions with final charge q_e, with use of a time-to-amplitude converter (TAC). Energy-analyzed electrons generated start signals, while stop signals were generated by q_e-analyzed ions. In the TAC spectrum (~6 ns FWHM), ratios of reals to accidentals were often ≳ 100 and were always constrained to exceed 5 by incident–beam-flux adjustment. The data thus consisted of the number and velocity distribution of all electrons collected (the "singles" spectrum), the number and velocity distribution of electrons observed in coincidence with q_e (corrected for accidental events and for a measured ion collection/detection efficiency of 85%), the total number of emergent ions of each preselected q_e, and the total number of projectile ions.

The results of the spectrum-shape analysis are being prepared for publication elsewhere. We note that, in high-statistics data runs of duration ≳ 10 h, we were unable to detect any appreciable change in the shape regardless of the choice of Z_1, Z_2, v, or q_e, or whether a particular channeling or random direction was chosen.

For the solid targets used (30 μg/cm^2 C, 50 μg/cm^2 Al, 100 μg/cm^2 Au), as well as for the Au single crystal (~300 μg/cm^2) oriented in a random (nonchanneling) direction, we find that *the convoy-electron yield per projectile ion is independent of emergent projectile charge state, and simply mirrors the statistical fraction of the corresponding projectile-charge-state distribution.* Since in both the ion-atom and the ion-solid collision case there is a rather strong projectile Z dependence, this observation is a surprise, and argues strongly against a surface-layer origin for convoy electrons.

For 2.4-MeV/u oxygen beams traversing the Au single crystal, Table I displays the convoy-electron yield $Y(\langle ijk\rangle)$ per emergent ion for a particular initial and final charge state. The measured fraction $[\mathrm{CF}(q,q_e)]$ of emergent ions with this final charge state is also displayed in parentheses. $Y(\langle ijk\rangle)$ and Y(rand) refer to particular channeling and random directions, respectively. For the random direction the yield is found to be independent of the final projectile-charge state, q_e. Hence the fraction of convoy electrons coincident with a particular q_e simply mirrors the *unweighted fraction* of projectiles having that q_e. It is also evident that convoy-electron production for well-channeled ions is much suppressed, with the greatest suppression arising in the most open channel ($\langle 110\rangle$) and for the most compact projectiles (O^{8+}, O^{7+}).

In a channel, a large fraction, F_A, of ions is confined to collisions with Au conduction electrons ($5d^{10}6s^1$), for which the capture and loss cross sections are sharply reduced[8]; the remaining, smaller fraction, $F_B = 1 - F_A$, experiences collisions with more strongly bound electrons. We expect ions in group B to approach charge-state equilibrium rapidly, and the corresponding convoy-electron yield Y_B to be well approximated by Y(rand).

For ions traversing the most open channel ($\langle 110\rangle$) near channel center, where the low electron density suppresses both capture and loss, and low electron momentum further suppresses capture, we expect the lowest convoy-electron

TABLE I. Percentage convoy-electron yield per emergent ion, for O^{8+} incident at 2.4 MeV/u on Au in the $\langle 110\rangle$, $\langle 100\rangle$, and random directions. The yield is normalized to the measured random yield of ~3.8×10^{-4} e/ion. The number in parentheses is the percentage of emergent ions in state q_e.

q_{in}	q_e^{out}	8+	7+	6+
8+	$Y(\langle 110\rangle)$	21 (68)	39 (28)	82 (4)
	$Y(\langle 100\rangle)$	37 (59)	58 (35)	79 (6)
	Y(rand)	100 (26)	100 (59)	100 (15)
7+	$Y(\langle 110\rangle)$	29 (42)	24 (51)	58 (7)
	$Y(\langle 100\rangle)$	37 (52)	47 (42)	71 (6)
	Y(rand)	100 (25)	100 (60)	100 (15)
6+	$Y(\langle 110\rangle)$	37 (31)	29 (42)	21 (27)
	$Y(\langle 100\rangle)$	39 (49)	45 (42)	47 (9)
	Y(rand)	100 (27)	100 (57)	100 (16)

yields. Assuming, therefore, that for group-A ions the convoy-electron yield $Y_A(8^+, \text{in}; 6^+, \text{out})$ is very small, we may infer the fractions $F_B(q_e)$ of class-B ions that emerge with charge q_e from measured yields and charge fractions for each channel. Knowing the F_B's, we calculate $Y_A(q, q_e)$ for all q and q_e for a consistency check and confirm that all the Y_A's are small compared to Y_B. The fraction of ions belonging to group B is given by $F_B = \sum_{q_e} F_B(q_e)$ and $F_A = 1 - F_B$. We find that $F_A \cong 0.78$ for the $\langle 110 \rangle$ channel and $F_A \cong 0.68$ for the $\langle 100 \rangle$ channel. With use of these F_A values and crystallographic values for the geometric area of each channel, the effective area available to group-B ions and unavailable to group-A ions is 1.3 Å^2 out of 5.83 Å^2 for $\langle 110 \rangle$, and *also* 1.3 Å^2 out of 4.14 Å^2 for $\langle 100 \rangle$. A unique value $r_{\text{crit}} = 0.65$ Å, corresponding to $\pi r^2_{\text{crit}} = 1.3$ Å^2, can be deduced. Thus, *only ions that have impact parameters < 0.65 Å (group B) produce convoy electrons, with an efficiency approximating the random value.*

We find it possible to construct a simple model which reproduces most of the observed values. Convoy-electron production is initiated in close collisions throughout the bulk while the final ion-charge state is determined at the exit surface. A close relationship between ELC and convoy-electron production is suggested by the fact that the widths of the convoy-electron and ELC cusps are equal within uncertainty and are independent of projectile nuclear charge, projectile velocity, and target.[3] We suggest that convoy-electron production is initiated by single- or multiple-electron capture events having cross sections $\sim 10^{-17}$ cm^2,[9] predominantly to excited states $\gtrsim 90\%$ of the time,[10] followed immediately by ELC. Subsequent electron scattering (elastic and inelastic) leads mainly to scattering into a wide range of angles, effectively extinguishing the convoy-electron population. (Some unknown degree of repopulation is possible by secondary elastic scattering.) The net production of several convoy electrons per emergent ion is depleted by electron scattering to $\sim 10^{-3}$–10^{-2} observable electrons per ion. In Au atoms the $6s$ and $5d$ electrons have kinetic energies < 10 eV. But the $5p$ and $4f$ electrons—which have binding energies of $\sim$250–460 eV and are therefore far more efficient at contributing to capture according to the Bohr $v_e \sim v$ matching criterion—have mean radii ranging from 0.60 to 0.28 Å. Therefore, the "magic" distance of 0.65 Å can be assigned a plausible physical interpretation.

Our experiment is the first to explore convoy-electron production under channeling conditions. Our interpretation of the data is that *ions must approach an atomic string within the well-defined distance of ~ 0.65 Å to generate convoy electrons*, independent of the particular channel chosen. This value is very suggestive of electron capture in the bulk, because of the availability of more strongly bound electrons ($5p$ and $4f$) at radii less than or equal to this value (but unavailable at greater radii).

The skewness of the electron velocity distributions from solids toward electron velocities $v_e > v$ can be qualitatively and quantitatively explained by considering the velocity dependence, $\sim v_e^{1.6 \pm 0.1}$, of the mean free path of free electrons in bulk solids.[11] We may correct the observed cusp shape by a velocity-dependent factor reflecting the exponential attenuation due to electron scattering within the bulk. The result of this procedure is shown in Fig. 1 for the typical case of 16-MeV O^{3+} traversing C. The resultant symmetric, peak-normalized curve is closely similar to experimental ELC cusps from C^{q+}, O^{q+} traversing Ne and Ar (apart from low-energy Auger lines which appear in the cusp wings).[4]

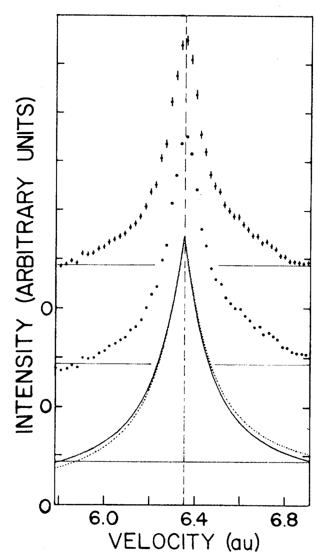

FIG. 1. Spectrum of convoy electrons emergent near 0 deg from 16-MeV O^{3+} ions traversing a 30-μg/cm^2 C foil. The upper data points are obtained from the raw spectrum (lower points), through a correction factor $v_e^{-1.6}$, to account for the electron–escape-depth velocity dependence estimated from Ref. 11. The lower curves represent respective fitted cusp shapes, which better display the degree of symmetrization produced.

Since no fitting procedure beyond overall peak normalization is used, the ability to symmetrize quantitatively the skewed peak is viewed as support for the bulk production of convoy electrons, a small fraction of which escapes through the surface.

A curious and unexplained enigma remains unresolved by our otherwise very successful model. Three facts need to be reconciled. The free-electron–scattering data suggest that all of the *observed* convoy electrons—though they are produced throughout the bulk—originate within the final ~20 Å of passage through the target (otherwise they scatter out). Yet the mean free path for projectile-ion charge changing under our conditions is ~200 Å, so that any ion traversing the final 20 Å of target has little likelihood of changing charge. The fundamental question posed is as follows: How can the correlation between emergent-ion charge and convoy-electron yield be broken in a distance of ~20 Å? Unless the correlation *is* broken, it is very difficult to understand why the convoy-electron yield is strongly dependent on projectile nuclear charge ($Z_1^{\sim 2.7}$), yet is independent of the emergent-ion charge (Z_1 screened by zero to two tightly bound K electrons).

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Ion Dip Spectroscopy: A New Technique of Multiphoton Ionization Spectroscopy Applied to I_2

Donald E. Cooper, Charles M. Klimcak, and John E. Wessel
The Aerospace Corporation, Los Angeles, California 90009
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A new method of high-resolution multiphoton spectroscopy based on competition between ionization and stimulated-emission channels is described. It combines features of multiphoton photoionization with those of optical-optical double resonance. Results obtained for I_2 demonstrate the method and identify intermediate states involved in multiphoton photoionization. The method offers the potential for extremely sensitive sub-Doppler spectroscopy of complex molecules.

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In this Letter, we describe and demonstrate a new two-wavelength method of multiphoton photoionization spectroscopy[1] (MPI) termed ion dip spectroscopy. Results are presented for application to multiphoton spectroscopy of the iodine molecule.[2,3] Ion dip spectroscopy combines the extreme sensitivity of resonance-enhanced two-photon photoionization[4] with the high resolution of techniques such as coherent anti-Stokes Raman scattering (CARS), stimulated–Raman-gain spec-

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