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ELECTRON CAPTURE AND LOSS TO CONTINUUM STATES IN GASES AND SOLIDS*

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I. Introduction

Among the many processes that occur when fast particles interact with target atoms there is the interesting case where the projectile captures target electrons, not necessarily into well-defined bound states, but into slightly unbound continuum states [1]. The net result is that one or more target electrons leave the target with speeds and directions that are nearly coincident with that of the bombarding projectile, but are not bound to the projectile. Present theories inadequately describe single and multiple capture processes to either bound or unbound states in the medium-velocity range.

Cognizance of the continuum electron capture phenomenon is important in experiments measuring pure bound state capture cross sections as well. Recent theory and experiment indicate that in some cases this channel can account for more than 50% of the total ionization cross section [2], and in others carried out in our laboratory, one or more bound state capture events are found to accompany continuum capture $\approx 20\%$ of the time. Hence experiments that separate bound state capture from continuum capture must be compared to theories that explicitly include the simultaneous continuum capture contribution. For solid targets the situation is even more complicated. Until very recently it has not even been known whether the origin of the forward continuum electrons observed in ion-solid collisions is associated with the surface or the bulk of the material.

The characteristic feature of electrons captured into the continuum (ECC) is that they possess a velocity, in magnitude *and* direction, close to that of the projectile ion. Hence the projectile ion emerges from the interaction region, which can be a gaseous or solid target, accompanied by electrons which move with the same velocity. Measurements by Crooks and Rudd [3] of the electron velocity distribution resulting from proton bombardment of He revealed a cusp-shaped peak in the forward ejected electron velocity distribution with the peak velocity equaling the ion velocity. Independently, Harrison and Lucas [4] observed a similar peak in the velocity distribution of electrons accompanying protons emerging from solid carbon targets. These authors have continued such work with other collaborators in a series of investigations [4], [5], [6], [7] for various H, He projectiles, mostly in thin solid and occasionally in gaseous targets, at projectile energies up to 1.2 MeV/u. Extensive work for both solid and gaseous targets and H, He projectiles has also been undertaken by Menendez, Duncan and co-workers [8], and for solid targets by Meckbach et al [9]. The latter authors have written an excellent summary article, whose title "Do present 'charge transfer to the continuum' theories correctly describe the production of $v_e = v_i$ electrons

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in ion beam-foil collisions?" emphasizes several controversial disagreements between the various data and theoretical predictions. Chiu, McGowan, and Mitchell [10] have extended the work of Meckbach et al. to gaseous targets, and find support for the electron capture to continuum (ECC) description in gaseous targets, in opposition to deficiencies they find in such a description for solid targets. However, Steckelmacher et al. [7] have criticized the interpretation given the data of Meckbach et al. on instrumental resolution and background treatment grounds, and argue for the validity of the ECC model for the solid-target case as well, and against the rival, solid-state "wake-riding" description [9], [11]. The latter model considers the possibility that electrons may be trapped in an oscillatory electron density fluctuation polarization potential extending behind and moving with the projectile, finally being liberated at the exit surface.

An excellent review of the theoretical aspects of the ECC process in ion-atom collisions has recently been provided by Shakeshaft and Spruch [12]. A number of experimental findings complementary to these discussed here have been covered by Meckbach et al. [9].

II. Experimental Procedure

A key feature of our experimental procedure is the easy interchange of short gaseous and thin solid targets at the same physical position, with all apparatus aperture sizes, dimensions, positions, and other experimental details unaltered. It has therefore been possible to cancel most systematic apparatus effects in comparing gaseous and solid target results. By using single ion-atom collision techniques, by using bare and few-electron ions of appreciably higher charge than heretofore, by extending the velocity range of measurement appreciably above that of earlier experiments, and by studying charge-state variation over an appreciably wider range than used previously, we have been able to test experimentally features of continuum electron-capture and -loss theories which have been inaccessible in previous experiments. These somewhat orthogonal tests complement rather than duplicate earlier experimental tests, but, like them, raise new questions about still other conspicuous disagreements.

Schematic diagrams of suitable apparatus are displayed in Figs. 1 and 2. Projectile ions from the Oak Ridge National Laboratory Tandem, Brookhaven National Laboratory Tandem, Lawrence Berkeley Laboratory Super-HILAC, or other accelerator traverse a thin gaseous or interchangeable solid target over a velocity range corresponding to 0.7-8.5 MeV/u. Beams are typically collimated to 1/3 mm diameter and $\pm 0.025^\circ$ angular spread, and then traverse a 4 mm thick target cell terminated by ~ 2 mm apertures (or a foil target centered at the same location). In the case of Fig. 1, the beam and accompanying electrons then enter along the central ray of a 180° spherical-sector analyzer of mean radius 3.8 cm, whose $\Delta E/E$ of 1.4% full width at half maximum is set by a 0.71 mm analyzer exit aperture, and the source dimensions at the center of the cell. Typical gas pressures of ~ 15 mTorr are established by a standard feedback-controlled capacitance-manometer system. Single-collision conditions and negligible charge changing are established by extremely linear plots of integrated cusp cross sections versus pressure for the various incident ions and energies. The plot intercepts typically coincide precisely with very small residual cusp cross sections measured at zero pressure. Spurious signals from electron loss by contaminant beams can be ruled out, as deliberate changes in the accelerator vacuum by a decade in pressure produced no noticeable effect on pressure linearity. The final collimation

Fig. 1 Schematic diagram of electrostatic version of the experimental apparatus, showing coincidence arrangement used.

Fig. 2 Schematic diagram of magnetic sector spectrometer. Ion beam enters spectrometer (1) and exits to Faraday cup (or CEM) (2). Electrons follow curved path (3) through pole pieces (4). Mirror pieces (5) focus field edges for improved focusing. Shield pieces (6) minimize magnetic fields along electron path, house gas cell (7) defined by aperture (8), angle-defining aperture (9), and exit aperture (10). Channel electron multiplier (CEM) detector has negatively biased cone (11).

guarantees that the beam never encounters any aperture or surface in the gas cell or spectrometer regions, as verified by very low spurious signals obtained at zero pressure. Magnetic fields are reduced to $< 3 \times 10^{-6}$ T over the spectrometer volume by three orthogonal external coils. Changes in coil current of $\approx 20\%$ produce negligible changes in cusp shape and integrated production cross sections.

Upon emerging from the target, the ion beam passes through an opening in the larger radius sector of the electron energy analyzer and is subsequently focused by a magnetic quadrupole doublet, dispersed horizontally according to its charge state by a perpendicular magnetic field, and finally collected by a channel electron multiplier located approximately 6 m from the target. This arrangement allowed us to determine the charge state of the emergent ions and permitted beam normalization. Hence, multi-channel scaling techniques could be employed in collecting the data; standard coincidence electronics and techniques were also used. The start signal for a time-to-amplitude converter (TAC) was generated by the detection of an energy-analyzed electron, while the stop signal was generated by the detection of a charge-analyzed ion. The resultant TAC distribution has a width ~ 6 ns at full width at half maximum, and indicates that a coincidence event between the detected electron and a charge-analyzed particle has occurred. In a similar manner the number of accidental events is counted. The total particle flux was adjusted so that the true-to-accidental ratio is typically 100 to 1 but is always constrained to exceed 5 to 1 by beam flux adjustment. This experimental arrangement allowed us to measure the number and energy distribution of all the emitted electrons (i.e., the "singles" spectrum), the number and energy distribution of electrons (corrected for accidental events) in coincidence with a particular charge state of the emergent ion (coincidence spectrum), the total number of ions of a certain charge state, and the total number of ions.

Figure 2 shows a magnetic variant appropriate to use of projectiles above the Coulomb barrier, where problematic nuclear radiation backgrounds from entrance collimations can be suppressed through use of "virtual" entrance slits produced by non-magnetic collimators located *within* the analyzer. Using such experimental arrangements, we have employed bare, one-, and multi-electron projectile ions on He, Ne, and Ar targets (under single collision conditions); thin self-supporting polycrystalline C, Al, Ag and Au targets; and monocrystalline Au targets oriented for axial channeling ($<110>$ and $<100>$) and random directions. Since there are several distinct aspects to the results obtained, we discuss them separately.

A. Electron Capture to Continuum in Gases by Bare Projectiles

When the incident projectile is bare and the target is a dilute gas, we find that the cusp-shaped ECC velocity distribution is skewed toward low velocities. It is a matter of current controversy whether the asymmetry discussed in the continuum electron velocity distribution is produced by contributions from the second Born amplitude (which are predicted to be asymmetric) added to those arising from the first Born term (which are symmetric). Hence experiments in this velocity domain offer unique tests for the validity of many approximations employed. The full width at half maximum (FWHM), Γ , of the velocity distribution can be represented approximately by the first Born theoretical result $\Gamma \approx (3/2) v_i \theta_0$ (θ_0 is half the analyzer polar acceptance angle and v_i the ion velocity). Regarding yields for Ne and Ar targets, we find that the cross section has a projectile charge dependence of Z_1^n ($n \sim 2.3$) and energy E^{-m} ($m \sim 2.75$) dependence, dependences more closely resembling

those found for bound state capture at comparable projectile velocities than the n and m dependences theoretically predicted for ECC at asymptotically high velocities using a hydrogenic target approximation.

While we have not yet been able to establish the relative importance of multiple continuum capture, we do find it possible to examine quantitatively the importance of simultaneous bound- and continuum-state capture measured in coincidence, relative to the processes of single-electron bound-state capture and of continuum-state capture alone. For example, for bare O^{8+} ions with velocities ~ 9 au undergoing single collisions in Ar gaseous targets, we find that for every 100 events observed in which a continuum capture occurs with *no* additional simultaneous bound-state capture, additional simultaneous capture of one, two, and three electrons into bound states is observed in about thirty, about ten, and about one collision events, respectively. Since our results directly demonstrate that often one or more closely associated additional bound-state captures occur whenever a continuum-capture event is observed, it is natural to raise the question, in what fraction of the events do one or more continuum-state captures occur when a bound-state capture occurs? Though this question is not directly answered in our recent experiments because of counting-rate limitations in the ion-counting coincidence channel, we *can* answer this question indirectly by comparing the total cross section for coincident continuum and bound-electron capture with total cross sections measured by other authors for what has always tacitly been assumed by them to represent bound-state captures alone. We find that we can consistently account for a significant fraction of what have traditionally been termed one-, two-, and three-electron bound-state-capture cross sections in terms of two-, three-, and four-electron capture events instead, where an unobserved electron has been ignored. The immediate consequence is that such comparisons as are made with bound-state capture theories are thus being made with the wrong theories. Comparisons must instead be made with theories which explicitly include the correlated continuum-capture contributions.

Figure 3 presents representative ECC coincidence spectra for bare O^{8+} ions in Ar, with the coincident final ion charge labeling each curve. From comparative yields, ratios of 1:2:3:4 electron events (ECC + zero, one, two, or three bound-state captures) are extracted, where in each case the start signal was registration of a continuum-electron capture. While in principle the apparatus does not distinguish between single and multiple ECC leading to an electron start signal, multiple ECC is thought very likely to cause large scatter of the continuum electrons in phase space well away from the condition $\vec{v}_e \simeq \vec{v}_i$ as a result of their mutual Coulomb repulsion, a hypothesis consistent with an unsuccessful search we made to detect them.

In Fig. 4, data concerning the projectile Z , v_i , and scattered-ion charge-state (q_e) dependence of the cross section for zero, one, two, and three bound-state captures for O^{8+} and C^{6+} ions in Ar gas are presented along with lines to guide the eye. A number of interesting comparisons with the single and multiple bound-state-capture data (ECC tacitly ignored) obtained by MacDonald and Martin [13] for the same collision system at overlapping velocities can be drawn. In earlier, non-coincidence experiments, we had already noted many qualitative similarities concerning velocity and Z dependences for such total capture data [13], [14] with our ECC data. Several such dependences are mimicked.

Interesting properties of the data presented in Fig. 4 include the following:

Fig. 3 Spectrum of continuum-capture electrons measured in coincidence with $b = 0, 1, 2, 3$ additional bound-state captures (scattered ion charge $q' = 8, 7, 6, 5$) for ~ 9 au (30 MeV) O^{8+} in Ar.

(1) Given an ECC event for 20 MeV O^{8+} on Ar, cross sections for additional coincident captures of $b = 0, 1, 2, 3$ bound-state electrons scale approximately as $2.1:1:0.38:0.11$, or $\sim (1/3)^b$. This can be compared with corresponding ratios at 20 MeV read from [13] of $\sigma_{87}:\sigma_{86}:\sigma_{85} \sim 1:0.25:0.036$. The corresponding scaling for ECC + bound-capture events for C^{6+} in Ar at 19 MeV scales as $\sim (1/4)^b$.

(2) In the range from 20 to 30 MeV, the trends with beam energy of ECC coincident with $b = 0, 1, 2$ are qualitatively similar to those observed in [13], and scale approximately as $E^{-1.8}:E^{-2.0}:E^{-2.8}$.

(3) If the O^{8+} ECC data on Ar at 20 and 30 MeV are interpolated to the same energy per nucleon as our 19-MeV C^{6+} data, the projectile Z scaling for ECC with $b = 0$ is $\sim Z^{1.7}$, whereas that for ECC with $b = 1$ is $\sim Z^{2.9}$. This observation is pertinent with respect to interpretation of the $\sim Z^{2.3}$ scaling we had noted earlier [15] in non-coincident ECC data for C^{6+} , O^{8+} , and Si^{14+} on Ar at similar velocities. Now one sees that the exponent represents a statistical average over $b = 0, 1, 2, 3, \dots$ processes coincident with ECC.

Fig. 4 Differential cross section for production of continuum-capture electrons in coincidence with $b = 0, 1, 2, 3$ additional bound-state electrons for O^{8+} in Ar at 20 and 30 MeV, and for C^{6+} in Ar at 19 MeV, averaged over a cone of half angle $\Theta_0 = 1.6^\circ$ centered on 0° and integrated over velocity intervals $v_e = v_i (1 \pm \alpha)$, $\alpha = 0.04$.

Conversion of the $\sim 20\%$, $\sim 5\%$, and $\sim 1\%$ likelihoods of $b = 1, 2$, and 3 bound-state captures accompanying 9 au O^{8+} -Ar ECC events into cross sections leads to the following results:

(1) Assuming a cusp shape of the form given by Dettmann and co-workers [4], and integrating the ECC cusp over intervals of $v_e = v_i (1 \pm \alpha)$, $\alpha = 0.5$, and over a cone of half-angle $\Theta_0 = 30^\circ$ leads to cross section estimates of $\sim 9.3 \times 10^{-18}$, $\sim 3.0 \times 10^{-18}$, $\sim 9.9 \times 10^{-19}$, and $\sim 8.9 \times 10^{-20}$ cm² for $b = 0, 1, 2, 3$, respectively. Here use is made of a spectrometer solid angle and efficiency calibration based on absolute cross sections measured by Stolterfoht, Schneider, and Ziem [16] for L-Auger production by H^+ on Ar and on absolute cross sections for continuum capture measured by Rødbro and Andersen [17] and by Cranage and Lucas [5].

(2) Alternative determinations of the ratio of integrated ECC yields to the projectile stripping yields we have measured for O^{8+} on Ar at the same

energy, followed by normalization to the total-electron-loss cross sections obtained from [13], leads to independent coincident $b = 0, 1, 2, 3$ cross section estimates of $\sim 3.0 \times 10^{-18}$, $\sim 2.6 \times 10^{-18}$, $\sim 8.5 \times 10^{-19}$, and $\sim 7.7 \times 10^{-20}$ cm², respectively. Comparing these estimated cross sections with those of [13] leads to the conclusion that coincident ECC accompanies bound-state-capture events roughly 20% of the time for $b = 1, 2$, or 3 . Hence we conclude that many measurements tacitly assumed to correspond to pure bound-state-capture processes - and indeed generally a large number of other capture cross-section measurements [13], [14] tacitly assumed to correspond to pure bound-state-capture processes - are actually compound, because of the neglect of an important coincident-electron-capture channel!

A word of caution about the dependence of the FWHM of ECC distributions on such dynamical variables as v_i , Θ_0 is in order. While the first Born result of Dettmann et al. [4] does yield $\Gamma \propto (3/2)v_i \Theta_0$, in approximate agreement with our experimental work and that of others for ECC in gases, in our results there is a strong left-right asymmetry in the dependence observed on v_i . While we find that Γ in most cases scales roughly as v , only the total FWHM does so. If divided into left-right (L, R) HWHM's corresponding to $v_e \leq v_i$, $v_e \geq v_i$, Γ_R turns out always to be of the order expected from consideration of analyzer resolution (typically $\approx 1\%$), while Γ_L accounts for the bulk of the growth of Γ with v . Furthermore, there are significant shell effects. For Ne, Γ_L is seen to rise significantly just as the beam velocity is varied through the range $v_i \approx v_K$, where v_K represents the K-shell electron velocity. Similar K-shell effects, though less well resolved, are seen in Ar. Ar possesses three shells whose contributions overlap. Theory has yet to fold in the internal velocity distribution of ECC electrons in explicit quantitative calculations, as the approximation $v_i \gg Z_{\text{TARG}} v_B$ (v_B is the Bohr velocity) is normally made. The onset of the rise in Γ_L occurs just where the Bohr velocity-matching criterion for electron capture would predict.

B. Electron Loss to Continuum in Gases (ELC)

Drepper and Briggs [18] have put forward a binary collision theory concerning electron loss by fast projectile ions (ELC), in which the laboratory-frame distribution of the large forward ejected-electron peak displays a cusp in both energy and angle, centered in velocity near $\vec{v}_e = \vec{v}_i$. The theoretical cusp shape closely resembles that predicted in ECC theories [4], [19], [20] for bare ions.

Such cusps can be studied if multi-electron projectiles are used on gaseous targets. A typical result is shown for various O^{q+} , Si^{q+} ions incident on an Ar gaseous target in Figs. 5 and 6. We note a nearly symmetric cusp which is considerably (a factor of ~ 2) narrower than a typical ECC cusp. Superimposed on the cusp are very definite structures which are identified as projectile Auger lines resulting from the transition $(1s^2 2p n l) \rightarrow (1s^2 2s) + ke^-$, with $n = 6, 7, 8, \dots$. Transitions for $n \leq 5$ in oxygen are energetically forbidden, as then the 2s-2p energy difference is insufficient to liberate the Rydberg electron.

The wide range of charge states used for Si^{q+} on Ar ($q = 5-14$) gives rise to a monotonically decreasing ELC integrated cusp yield with q , producing a factor of ~ 10 drop in going from $q = 5$ to $q = 11$. These yields have been found to be weakly dependent on v_i , verifying a prediction of Briggs and Drepper that in the present velocity range, such electron-loss cross sections may tend to velocity independence. Significant departures (e.g., a factor

Fig. 5 Normalized spectra for electron capture and loss to continuum states in the forward direction for 2.5 MeV/A O^{q+} and Si^{q+} on Ar. Zero-count base lines are shown for each spectrum. Statistical errors are about 2% at the peaks.

Fig. 6 Energy spectrum of electrons emitted in the forward direction in collisions of 25 MeV O^{4+} on Ar. The spectrum shows a cusp-shaped peak produced mainly by projectile electron loss, centered at an energy corresponding to the projectile velocity. In both wings of this cusp a series of lines is visible, which originate from projectile autoionizing states of the following kind: $(1s^2 2p \ n\ell) \rightarrow (1s^2 2s) + ke^-$. For O^{4+} , $n \leq 5$ is energetically forbidden.

of > 2 over the range 2.5-3.9 MeV/A) from this weak velocity dependence set in only for $q = > 11$, just in the range where the ELC cross sections are dropping below the more strongly velocity-dependent ECC cross sections.

Coincidence experiments identifying the final projectile charge states allow allocation of the rich Auger structure observed among excited projectile states, which can thus be tagged according to parent ion charge state. The interpretation of these structures is presented elsewhere [21].

Generally speaking, there is much to be learned about basic collision dynamics from the coincidentally observed Auger spectra. For example, in experiments involving 18 MeV C^{4+} ions incident on Ar, Elston et al. [22] find it possible to intercompare absolute cross sections for single C K excitation, single C K ionization (sometimes accompanied by an excitation of a second electron), and double C K ionization (sometimes accompanied by an excitation of a third electron) from a unified set of spectra, all obtained under single-collision conditions.

Moreover, kinematics greatly aids the performance of such coincidence experiments. Owing to the high projectile velocity, the emitted electrons are folded into a forward cone, thereby aiding collection. Near zero deg, there is *zero* Doppler spread to first order, thereby reducing the need for very high resolution to overcome line blending problems. At the relatively high ion energies employed, the coincidence registration efficiency (including detection and collection efficiencies in the ion channel as well as electronic dead time and discriminator losses) can be made to exceed 85 to 90%. This high registration efficiency arises because the ion detection efficiency is high, and scattering angles are small. Coincidence data of useful statistical significance can be obtained with total ion currents in the 10^{-12} to 10^{-14} A range within a few hours.

As in the case of ECC, a word of caution about the FWHM of ELC distributions is in order. Though the ELC distributions are much more symmetric than the corresponding ECC ones, the predicted growth [18] in width $\propto v_i$ is *not* observed. Instead, for C^{9+} , O^{9+} , Si^{9+} , and Ar^{9+} traversing He, Ar at energies in the range 1-8.5 MeV/u, in numerous instances we find Γ to be only weakly dependent on v_i .

C. Convoy Electron Production in Solid Targets

"Convoy" electrons is the generic name [11], [23] that has been given to electrons that accompany ions as they emerge from solid targets. As remarked earlier, the ease of interchange of gaseous and solid targets in our apparatus while leaving all other geometrical and kinematic conditions fixed has permitted us to compare gaseous and solid target results under conditions where most systematic apparatus effects cancel to first order. A key parameter for comparison of theory and experiment is the full width at half maximum Γ_θ of the longitudinal velocity distribution of the electrons emerging in the forward direction. In the ECC theory of Dettmann et al. [4], Γ_θ can be expressed as $\Gamma_\theta = (3/2)v_i \theta_0$, where v_i is the projectile velocity in au and θ_0 is half of the acceptance angle, in radians, of the electron velocity analyzer. For ECC in gaseous targets, experimental results are only roughly consistent with this prediction (see the conclusion of Section A). For solid targets, some authors [9], [10], [24] observe Γ 's that are essentially independent of the velocity of the projectile. These results are at odds with the ECC prediction and the assumption that the "last target layer" is the source of the continuum

electrons. The properties of the distribution of continuum electrons coming from wake-riding (WR) states are predicted to be significantly different from those for ECC electrons. The predicted FWHM's of the longitudinal velocity distribution for wake-riding electrons are shown in Fig. 7; they depend on the target material and the atomic number of the projectile and are a decreasing function of the projectile energy. Results of our corresponding measurements are also shown in Fig. 7.

Results of our experiments with protons, oxygen, silicon, and nickel projectiles on polycrystalline C, Al, Ag, and Au targets all give a (somewhat geometry-dependent) FWHM of $\Gamma = 0.25$ au, but do *not* show the dependences on the variables (e.g., Z , v_i) discussed in present theories. The experimental

Fig. 7 The full width at half maximum of the longitudinal electron velocity distribution, Γ_{ℓ} , in au, for continuum electrons as a function of the electron velocity v_e , in au. The incident projectile energy, in MeV/u, appears at the top of the figure. The dashed line is the ECC prediction. The solid lines are the predictions of the WR theory for an Al target with the indicated projectiles. The lowest solid curve marked Ag is the prediction of WR theory for protons incident on Ag. The experimental data for solids correspond to $\Gamma_{\ell} \approx 0.25$ au and are represented by the heavy solid line. The open points represent the gaseous target (Ne and Ar) results for O^{8+} and Si^{14+} .

results are clearly at variance with present theoretical considerations, although distinct differences in the cusp electron distribution are evident for solid compared to gaseous targets.

A typical velocity spectrum is shown in Fig. 8, for 108 MeV Si incident on Ar (gaseous) and Au (solid) targets. For solid targets the velocity distribution is narrower and exhibits a skew toward the high velocity side.

One shortcoming of the above convoy experiments (of about two years ago) is that the electron velocity distribution is averaged over all of the final projectile charge states. Guided by parallel experimental experience in our ECC studies, it became evident that a coincidence experiment specifying the relationship between the final charge state of the projectile and the electron velocity distribution and yield would clarify the situation. A typical experimental result is shown in Fig. 9, for 12 MeV C^{2+} ions incident on a $40 \mu\text{g}/\text{cm}^2$ Al target. A total, or singles, spectrum is shown together with

Fig. 8 The differential production cross section $d\sigma/dv_e$ as a function of the electron velocity, v_e , of continuum electrons emerging near 0° with respect to the ion beam, for 108 MeV Si^{13+} in Ar gas at 30 mTorr and for Si^{9+} on a $100 \mu\text{g}/\text{cm}^2$ Au foil. The electron energy scale appears at the top of the figure. The mean emergent charge from the solid target is also 13.

Fig. 9 The longitudinal velocity distribution of convoy electrons emerging from a solid when 12 MeV $^{12}\text{C}^{2+}$ is incident on a 40 $\mu\text{g}/\text{cm}^2$ Al target. The "singles" or non-coincident distribution is shown in the lower left-hand corner. The electron velocity distributions in coincidence with the final charge state of C^{6+} , C^{4+} , and C^{3+} are shown in clockwise arrangement starting in the upper left-hand corner. The vertical scale (intensity) is arbitrarily normalized for each spectrum.

electron velocity spectra observed in coincidence with emergent C^{6+} ions ($\sim 25\%$ of the total), C^{4+} ions ($\sim 20\%$ of the total), and C^{3+} ions ($\sim 1.2\%$ of the total). The C^{5+} distribution accounted for 45% of the total and hence need not be shown. Lower final charge states were observed, but with intensities of less than 0.5% of the total. Hence reliable electron velocity distributions (i.e., distributions corresponding to a true-to-accidental ratio greater than 5) could not be obtained. Each particular coincident distribution required $\sim$ one to three hours of collection time. The sum of the electron spectra obtained in coincidence account for $\sim 92\%$ of the total number of electrons observed, indicating a satisfactory ion collection geometry and a high detection efficiency for the electron multiplier used as a particle detector.

Inspection of Fig. 9 leads to the conclusion that no evident differences in the velocity distribution of the convoy electrons are observed as the final

charge state of the projectile is changed. In fact, the FWHM is the same for all the final charge states of the projectile. Changing the incident energy of the projectile (from 12 MeV to 33 MeV carbon), the charge state of the projectile, the atomic number of the projectile (carbon, oxygen, and silicon [all at 1 MeV/amu]) or the target material (aluminum, carbon, and gold) does not alter the conclusion that to a good approximation the final convoy electron velocity distribution from solids is independent of the final projectile charge state. This statement also applies to crystalline targets. Recent theoretical considerations of Day [23] indicate a diffraction structure in the electron velocity distribution and a Z_1 dependence of the FWHM of approximately $Z_1^{1/2}$. No diffraction structure is observed in Fig. 9; however, it is unclear whether the present electron analyzer would be able to resolve the proposed structure.

The yield of convoy electrons may be estimated from an appropriate normalization and integration of the longitudinal velocity distribution. The measuring instrument normally has a solid angle of acceptance centered on zero deg, $\Delta\Omega$, and an energy resolution $\Delta E/E$ which fold the actual transverse and longitudinal velocity distribution characteristics. Hence if the width and shape of the longitudinal and transverse velocity distributions were known accurately, we could obtain the total electron yield from a measurement of the yield in the longitudinal direction. They are currently known only approximately; a corresponding Ansatz is needed to permit total yield estimation.

We represent the velocity distribution in the longitudinal ($||$) and transverse ($\perp$) directions by cusps of exponential shape, of half widths α and β , respectively. The total yield, in terms of the yield measured along the longitudinal direction, Y_R , is:

$$Y = Y_R \left\{ \frac{1}{1 - (1 + Cv_i) \exp. (-Cv_i)} \right\}, \quad (1)$$

where v_i is the projectile velocity, $C = \frac{2\Theta_0 \ln 2}{\Gamma_t}$, and Θ_0 is the half-angle of the forward acceptance cone.

For solid targets we find $\Gamma_\lambda = 0.25$ au, and infer $\Gamma_t = 1.8 \Gamma_\lambda = 0.45$ au, independent of the projectile velocity. (Here we have used the relationship $\Gamma_t \approx 1.8 \Gamma_\lambda$, measured by Meckbach et al. in lower-energy ion-solid collision studies.)

For $\Theta_0 = 1$ deg, C takes the value 5.38×10^{-2} . (For solid targets we measure a higher fraction of the total electrons as the projectile velocity is increased than for ECC, where the corresponding cusps are wider.)

For completeness, the yield Y_R in the longitudinal direction can be expressed as:

$$Y_R = 1.6 \times 10^{-7} \frac{\bar{q} y}{Q} \frac{\Delta V}{k} \frac{E_i}{\Delta E} \frac{1}{E_i} \frac{1}{\Theta_0^2}, \quad (2)$$

where $\bar{q}$ is the mean charge of the collected ions, Q the charge in pC collected in the Faraday cup, E_i the energy of the convoy electrons in eV, $\Delta E_i/E_i$ the energy resolution of the analyzer, k the analyzer constant in V/eV, and ΔV

the change in volts of the analyzer voltage for each step. For typical analyzer parameters $\Delta E_i/E_i = 1.4 \times 10^{-2}$, $k = 0.416$ V/eV and $\Theta_0 = 1.8^\circ$, (2) reduces to:

$$Y_R = 8.5 \times 10^{-6} \frac{\bar{q} \Delta V y}{Q E_i} \frac{\text{electrons}}{\text{particle} \cdot \text{deg}^2}. \quad (3)$$

Here y is the total number of electrons collected within the interval $(v_i - \beta)$ to $(v_i + \beta)$, where β was chosen as twice the full width at half maximum of the longitudinal electron energy distribution, Γ_ℓ . Numerically, $\beta = 0.5$ au, since Γ_ℓ is independent of the target material and of the charge, velocity, and atomic number of the projectile. The yield of convoy electrons, as given by (2) or (3), is independent of the incident charge state of the projectile and of the target thickness (the target thickness was changed by a factor of two), but does change for different target materials as well as with the energy and atomic number of the incident projectiles.

The data analyzed accordingly are shown in Fig. 10 for the various projectile-target combinations used as a function of the incident particle energy. In the interest of clarity, only the oxygen projectile data is shown for all of the targets. For the H^+ , Si^{9+} , and Ni^{9+} particles, only the C and Au target data are shown. The near linearity and parallelism of the similar target curves for each projectile shown in Fig. 10 suggest a common velocity dependence for the various projectiles, and an approximate velocity-independent Z_1 dependence. Indeed, we find that we can summarize all of our results by the empirical equation

$$Y = 1 \times 10^{-4} C(Z_2) Z_1^m E_1^{-n}, \quad (4)$$

where Z_1 is the atomic number of the incident particle of energy E_1 in MeV/amu; $m = 2.75 \pm 0.2$; $n = 2.25 \pm 0.1$; and $C(Z_2)$ is a constant depending on the target material: $C(\text{Au}) = 1.65$, $C(\text{Ag}) = 1.25$, $C(\text{Al}) = C(\text{C}) = 1.0$. All the values have estimated uncertainties ± 0.15 .

To our knowledge, the only other measurement of the yield of convoy electrons from solids is by Dettmann et al. [4] for 225 keV protons on carbon. Eq. (4) predicts a result within 30% of their measurements. Meckbach et al. [9] report relative yield measurements for 200-500 keV protons on carbon and find an energy dependence of $n \approx 3.1$, which is in reasonable agreement with our results.

D. Convoy Production Measured in Coincidence with Emergent Ion Charge

When we examine the yield of convoy electrons in coincidence with a particular final charge state of the projectile, we find the curious result [24] that the convoy electron yield, per coincidently registered projectile, is independent of the final projectile charge state. That is, the fraction of electrons coincident with a particular final charge state is just mirrored by the fraction of projectiles having that particular charge state. If the atomic number and energy of the incident projectile or the target material is changed, we find that each coincident electron yield varies in the same proportions as the non-coincident convoy electron yield.

Fig. 10 The yield of convoy electrons prepared according to (1) and (4) for different target materials and different incident projectiles. The dashed lines are to guide the eye.

These results strongly mitigate against the surface production of convoy electrons and suggest the bulk of the material as the origin of these electrons. To further explore this possibility, we utilized 06^+ , 07^+ , and 08^+ beams traversing a Au single-crystal target and employed channeling techniques, where it is known that incident ion charges often remain intact (frozen) throughout their entire passage (e.g., $\sim 80\%$ of 2.5 MeV/u 08^+ ions traversing the $\langle 110 \rangle$ direction will remain 8^+ throughout a 300 $\mu\text{g}/\text{cm}^2$ gold crystal). As mentioned previously, the shapes of the velocity distribution for channeled ions obtained in coincidence with the final projectile charge state are indistinguishable from those obtained with polycrystalline targets or from non-coincident (singles) distributions. However, in the channeling data, the yields which were found to be independent of the final projectile charge state for polycrystalline targets exhibit instead a strong diminution, dependent on incident as well as exit charge state. Table I presents in matrix form the yields per emergent ion for various incident, q , and exit, q_e , charge state pairs, together with the corresponding measured charge state fractions and coincident fraction values. It is immediately evident that convoy production for well-channeled ions is much suppressed, with the greatest suppression arising in the most open channel $\langle 110 \rangle$. It is also evident that the highest yields occur when distances of approach to lattice sites is likely to be closest. The yield entries in Table I can be surprisingly well

Table I 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 $\sim 3.8 \times 10^{-4}$ electrons/ion. The number in parentheses is the fraction (%) of emergent ions in state q_e .

q_e out q in		$8+$		$7+$		$6+$	
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)

reproduced by postulating two classes of channeled ions, those that approach close enough to a lattice site (effective distance ~ 0.65 Å) to produce a convoy with high probability (equal to the random or polycrystalline rate) and those which remain at larger separations, producing convoys with negligibly low probability. Two immediate tests of this extreme assumption are well verified. First, the number 0.65 Å explains both the $\langle 110 \rangle$ and the $\langle 100 \rangle$ yield figures. Second, it happens that for Au atoms the 6s and 5d electrons have kinetic energies < 10 eV. But the 5p and 4f electrons - which have binding energies of ~ 250 to 460 eV and are therefore far more efficient at contributing to capture according to the Bohr $v_e \sim v_i$ 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 most plausible physical interpretation.

E. Model for Convoy Electron Production in Solids

The experimental evidence to date allows us to construct the following model for convoy electron production in solids. Convoy production is initiated in a close collision, most probably a single or multiple-electron capture event ($\sim 10^{-17}$ cm² capture cross section [13]), which, according to the work of

Brown et al. [25], is dominated by electron capture to excited states ($\sim 90\%$ of the time). The excited states most copiously populated [25] have $n = 2$ and 3, with populations of higher states possible at the $\sim 10\%$ level. Because binary encounter theory predicts that e^- loss cross sections scale approximately as n^2 , and because of the peak width and symmetry considerations noted above, we suggest that convoy production is initiated by capture(s) followed *immediately* by electron loss to the continuum. If the mean free path for such dual event convoy production were, say, ~ 250 Å, then several convoys per projectile would be produced in a solid a few thousand Å thick. Subsequent electron-electron scattering (elastic and inelastic) leads mainly to scattering into a wide range of angles, effectively extinguishing the convoy population, though it is possible that secondary elastic scattering would cause some unknown degree of repopulation. In any event, since the escape depth of ~ 1 keV electrons is ~ 10 -20 Å, the net production of several convoy electrons per emergent ion is depleted by electron scattering to $\sim 10^{-2}$ to 10^{-4} observable electrons per ion.

The skew of the electron velocity distributions toward electron velocities $v_e > v_i$ can be qualitatively and quantitatively explained by considering the velocity dependence of the electron scattering cross section in solids [26]. We correct the observed cusp shape by a velocity-dependent factor ($\sim v^{-1.6}$) reflecting the exponential attenuation resulting from electron scattering within the bulk. The result of this procedure is shown in Fig. 11 for 16 MeV O^{3+} traversing carbon. The resultant symmetric, peak-normalized curve is similar to experimental electron loss cusps from C^{9+} and O^{9+} traversing Ne and Ar as shown in Figs. 5 and 6 (except for the additional low-energy Auger lines which appear in the cusp wings). Since no fitting procedure beyond overall peak normalization is used, the ability to quantitatively symmetrize the skewed peak is viewed as support for the bulk production of convoy electrons, a small fraction of which escape through the surface.

Despite the many appealing features of the model we have described, there remains an enigma. Since binary electron capture and loss processes are both Z - and q -dependent, it seems reasonable to assume a similar dependence for convoy production. Since 10^{-17} cm² charge changing cross sections appropriate to the ion velocities used [13] imply ~ 200 Å mean free paths for capture and loss at solid target densities, it is then difficult to account for the complete destruction of correlation between convoy production and emergent ion charge, if the convoys observed indeed originate in the final ~ 10 Å of target passage. What destroys the correlation so effectively? Is it possible that the mean free path for free electron scattering is very small compared to that for convoy electron scattering owing to the strong drag provided by the projectile ion? Is it possible that there is rapid capture and loss of excited electrons with mean free paths $\ll 10$ Å which re-equilibrate the charge in the last layers? Is it possible that shielding of projectile K-shell electrons by the solid medium is so complete that convoy production is q -independent? What other models do as well at accounting for the shape and channeled ion yield data as the model proposed while avoiding this enigma? It seems that clear, substantial clues to a complete understanding of convoy production are provided by the present data. There remain, however, perplexing problems in understanding the unexpected lack of correlation of shape and yield with q_e .

Fig. 11 Spectrum of convoy electrons emergent near 0 deg from 16 MeV O^{3+} ions traversing a $30 \mu\text{g}/\text{cm}^2$ 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). The lower curves represent respective fitted cusp shapes, which better display the degree of symmetrization produced.

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