CONVOY ELECTRON PRODUCTION BY HEAVY IONS IN SOLIDS*

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The term "convoy" electron refers to those electrons ejected in fast ion-atom and ion-solid collisions closely matched in vector velocity to that of the incident heavy particles responsible for their ejection. Similarities and differences among electrons ejected into such states through binary electron capture to continuum and electron loss to continuum processes in single ion-atom encounters are compared and contrasted to more complex ejection processes occurring in solid targets. Puzzles posed by the apparent strong projectile Z dependence but weak emergent ion charge dependence of the yield in the case of solid targets are reviewed. Very recent progress in resolving these puzzles has been made by recent observations that the apparent mean free path for electron scattering out of the forward direction within the target is observed to be an order of magnitude greater than that for free electrons of equal velocity provided the projectile charge is high.

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1. Introduction

To introduce the subject of convoy electron production in solids, we quote from a recent review (1). A sharp cusp in the velocity spectrum of electrons, ejected in ion-atom and ion-solid collisions, is observed when the ejected electron velocity $\vec{v}_e$ matches that of the emergent ion $\vec{v}_p$ in both speed and direction. In ion-atom collisions, the electrons originate from capture to low-lying, projectile-centered continuum states (ECC) for fast bare or nearly bare projectiles, and from loss to those low-lying continuum states (ELC) when loosely bound projectile electrons are available. Most investigators now agree that ECC cusps are strongly skewed (1) toward lower velocities, and exhibit full widths half maxima roughly proportional to v_p (neglecting target-shell effects, which are sometimes strong.) A close examination of recent ELC data shows that ELC cusps for heavy ions are instead nearly symmetric, with widths nearly independent of v_e in the velocity range 6 - 18 a.u., a result only recently predicted by theory (2). "Convoy" electron cusps produced in heavy ion-solid collisions at MeV/u energies exhibit approximately velocity independent widths very similar to ELC cusp widths. While the shape of the convoy peaks is approximately independent of projectile Z, velocity, and of target material, it is found that the yields in polycrystalline targets exhibit a strong dependence on projectile Z and velocity. While attempts have been made to link convoy electron production to binary ECC or ELC processes, sometimes at the last layer, or alternatively to a solid state

wake-riding model, our measured dependences of cusp shape and yield on projectile charge state and energy are inconsistent with the predictions of such theories (1).

When coincidence with emergent ion charge state q_e is required, ECC cusps can be sorted as to whether 0,1,2 ... additional bound-state captures occurred during the same collision which generated the continuum electron.(3) Similarly, ELC cusps can be sorted as to how many additional electrons were lost. The shapes observed are relatively independent of whether or not additional capture or loss events occurred. The yields (production cross sections) tend to mimic the beam velocity, projectile Z, and projectile charge q dependence of corresponding single- and multiple-electron bound state capture and loss cross sections.

For convoy electron production in solids, as detailed below cusp shapes are again found to be approximately independent of q_e . More remarkably, for polycrystalline and randomly oriented monocrystalline targets, the yields are found to be nearly independent of q_e , i.e., to mirror the unweighted statistical fraction of emergent ions of each charge state even though there is an appreciable projectile Z dependence, and until recently reason to believe that the observed convoys originate in many cases at a depth well within one mean-free path for charge changing of the exit surface. For well-channeled ions, however, the convoy yield is strongly suppressed, pointing to the necessity of close approach to an atomic string in the bulk as a necessary precursor of convoy production.

2. Convoy electron production: Shapes and yields from singles and coincidence measurements.

Our experiments with p, O, Si, and Ni projectiles in the velocity range 6 - 14 a.u. on C, Al, and Au polycrystalline targets all yielded convoy electron cusps of similar shape with the same FWHM of $\Gamma = 0.25 \pm 0.02$ a.u., where the half angle of collection $\Theta_0 = 1.8^\circ$, and the energy resolution of our analyzer was 1.4%. One shortcoming of these convoy experiments is that the electron-velocity distribution was 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 convoy electron-velocity distribution and yield would clarify the situation. A typical experimental result is shown in Fig. 1 for 12-MeV C^{2+} ions incident on a $40\text{-}\mu\text{g}/\text{cm}^2$ Al target. A total, or singles, spectrum is shown together with electron-velocity spectra observed in coincidence with emergent C^{6+} ions (~25% of the total), C^{4+} ions (~20% of the total), and C^{3+} ions (~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.

Inspection of Fig. 1 lead to the conclusion that no significant

differences in the velocity distribution of the convoy electrons were evident as the final charge state of the projectile was 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/u)], or the target material (aluminum, carbon, and gold) did not alter the conclusion that to a good approximation the shape of the convoy electron-velocity distribution from solids is independent of the final projectile charge state. This statement also applies to crystalline targets.

When we examined the yield of convoy electrons in coincidence with a particular final charge state of the projectile, we found the curious result that the convoy electron yield, per coincidently registered projectile, is approximately independent of the final, projectile charge state. That is, the fraction of electrons coincident with a particular final charge state just mirrors the fraction of projectiles having that particular charge state. If the atomic number and energy of the incident projectile or the target material was changed, we found that each coincident convoy electron yield varies in the same proportions as the noncoincident convoy electron yield.

These results strongly mitigated against the surface production of convoy electrons and suggested the bulk of the material as

the origin of these electrons. To further explore this possibility, we utilized O^{6+} , O^{7+} , and O^{8+} beams traversing a Au single-crystal target and employed channeling techniques, where it is known that the incident ion charge state often remains intact (frozen) throughout the entire crystal passage (e.g., ~80% of 2.5-MeV/u O^{8+} ions traversing the $\langle 110 \rangle$ direction will remain 8 + throughout a 300- $\mu\text{g}/\text{cm}^2$ gold monocrystal). We found that the shapes of the velocity distributions are independent of whether a channeling or random direction of a single-crystal target or a polycrystalline target is chosen and do not depend on q or q_e , the incident and emergent charge states. However, in the channeling data, the yields which were found to be independent of the final projectile charge state for polycrystalline targets depend strongly on incident charge q 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. (See Ref. 1 for additional details.) It was 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 the distance of approach to lattice sites is likely to be closest. The yield entries in Table I can be surprisingly well reproduced by postulating two classes of channeled ions: those that approach close enough to a lattice site (effective distance $\sim 0.65\text{ } \text{\AA}$) to produce a convoy electron with the high probability (equal to the random or polycrystalline rate); and those which remain at larger separa-

tions, producing convoys with negligibly low probability. Two immediate tests of this extreme assumption were 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 -460 eV and are therefore far more efficient at contributing to capture according to the Bohr $v_e \sim v_p$ 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.

The experimental evidence (1) allowed 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), which is dominated by electron capture to excited states ($\geq 90\%$ of the time). The excited states most copiously populated have $n = 2$ and 3, with populations of higher states possible at the $\geq 10\%$ 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 suggested that convoy production is initiated by capture(s) followed immediately by electron loss to the continuum. If as in the data of ref. 1 the mean-free path for such dual events is ≥ 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, although it is possible that secondary elastic scattering would cause some unknown degree of repopulation.

In Ref. 1 we argued that since the escape depth of ~ 1 keV free electrons is $\sim 10 - 20$ Å, the net production of several convoy electrons per emergent ion is depleted by electron scattering to $\sim 10^{-2} - 10^{-4}$ observable electrons per ion. We shall see, there is now reason to modify this argument. A curious and unexplained enigma remained unresolved by our otherwise very successful model. Three facts needed to be reconciled. 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 in C was ~ 200 Å, so that any ion traversing the final 20 Å of target had little likelihood of changing charge. The fundamental question posed was as follows: How can the correlation between emergent-ion charge-state 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 ($Z \sim 2.7$), yet is approximately independent of the emergent-ion charge (Z screened by zero to two tightly bound K electrons).

Additional puzzles are posed by data taken using light projec-

tiles. For hydrogen and helium projectiles, the yield of convoy electrons mimic the background electron yield (4), but do not track projectile charge state evolution. Other investigators (5,6) measuring the energy distribution of convoy electrons using mostly hydrogen and helium projectiles note two components in the observed electron velocity distribution (a sharp feature atop an underlying helmet-shaped hump offset from the center of the hump). This two-component nature of the cusp in ejected electron energy (longitudinal velocity) has not been observed in work with heavy projectiles. A second cusp component of different origin (4), arising from field ionization in the electrostatic spectrometer normally used by us and others, has been shown to be an artifact of the particular apparatus used in some studies (7,4).

Meanwhile, the production of Rydberg states of projectiles emerging from solid targets has been investigated by several authors. For heavy projectiles, it is found (8) that Rydberg states of high l are plentiful, contrary to the predictions of a single atom pickup model (e.g. calculated in the first Born approximation). However, no significant initial nonstatistical sublevel population has yet been observed (7), in contrast to proton data which show strong indication of alignment in the excitation (4).

A partial but incomplete resolution of the puzzles concerning the n and l -state distribution of foil-excited Rydberg states

was achieved in work by Betz et al. (8) and by Yamazaki and Oda (9). These authors have emphasized the possible importance of the sudden change in projectile shielding as the projectile exits; the ion sees a potential step $\sim Ze^2/\omega_p v$ where ω_p is the bulk plasmon frequency of the target, and Z and v describe the projectile atomic number and velocity, respectively. Betz et al. trace the origin of such high Rydberg-state production to the electrons transferred from the continuum to high n - and l -states at this potential step; Yamazaki and Oda invoke the same mechanism to explain the generation of what they term intrinsic convoy electron production (the spike on the helmet) in terms of free electron transfer to the sharp feature from the underlying secondary electrons.

In an attempt to sort out these problems, we recently measured the yields (10) of convoy electrons associated with 15.2 MeV/amu Ni^{24+} and Ni^{26+} ions transversing C and Al. By observing how the yield saturates with increasing thickness, we obtained effective mean free paths (MFPs) for the scattering of convoy electrons out of the collector half angle (1.54°). Figure 2 shows a typical plot. The MFPs thus derived are about 10 times those expected for free electrons of the same velocity! -- e.g. at an equivalent electron energy of 8.3 keV we find a convoy electron MFP of $\sim 2000 \pm 200$ Å in carbon, as opposed to the free electron value of ~ 10 nm.

In our recent experiments Ni^{24+} and Ni^{26+} ions of 15.2 MeV/u energy were directed through polycrystalline C and Al foils of

thicknesses 3-500 $\mu\text{g}/\text{cm}^2$. The methods used have been described fully by Latz et al. (6) and need not be repeated here. The unique beam used was made available on short notice by the staff of the GSI accelerator at Darmstadt.

The observed convoy electron yields (in arbitrary units) are plotted vs. target thickness in Fig. 2. Additional details may be found in Ref. 10. Our experiments track the rise in convoy production from effectively very thin targets to as thick as desired. Thus a yield growth curve is measured from which a crude estimate of the MFP can be obtained by fitting to $1-\exp(-x/\lambda)$, where x is the target thickness, and λ the effective MFP for convoy production. While such a simple estimate is not expected to be accurate, even at the level of a factor of two, the size of the effect observed is so large that there can be no doubt as to its significance. The approximate independence of the earlier coincidence data now becomes easier to understand, since the yield could be influenced by the charge state history of the projectile quite deep in the foil.

We have examined an interesting suggestion (11) that the effect might be due to parasitic dependence on electron loss from the projectile as it penetrates the foil, as the L- and M-shell electrons approach charge state equilibrium in the foil thickness and projectile velocity range in question. However, the electron loss cross section from Ni^{26+} (2 L-shell electrons) is almost an order of magnitude larger (12) than for Ni^{26+} (only

K-shell electrons), while our measured yields are comparable.

A model which quantitatively explains the order of magnitude increase in MFP has recently been proposed by Bottcher et al. (10). The elements of the model are as follows. An electron is visualized as moving with the velocity originally equal to that of the ion beam p_0 . Collisions with target atoms create a small transverse component $p_{\perp}$ which does a random walk combined with drift in the ionic field as electron and projectile proceed through the foil.

Electrons are detected when their net scattering angle is within the detector half-angle θ_0 . If the average scattering angle at each collision is θ_s , and the total number of scatterings (about one per atomic layer of the foil) is N_s , the net angle $N_s^{1/2}\theta_s$, whence $\theta_s \sim 0.05-0.02$ deg for 10^7-10^8 scattering layers. The observed $\theta_s = \theta_s^0 - \theta_c$, where θ_s is the deflection in one target collision, and θ_c is the "Coulomb focusing angle" through which the electron is pulled back between scattering events by the electric field F of the projectile. If the interlayer distance is a and the projectile velocity is v_p , the electron changes its transverse velocity by Fa/v_p between layers, so that $\theta_c \sim Fa/v_p^2$ (13). The inverse MFP for scattering through angles $> \theta_{\min}$ is proportional to

$$\int_{\theta_{\min}}^{\theta_{\max}} \sigma(\theta) \sin\theta d\theta \quad \text{and thus to } \theta_{\min}^{-2} \quad \text{if}$$

we assume scattering ($\sigma \sim \theta^{-4}$ at small angles) and $\theta_{\max} \gg \theta_{\min}$. The MFP is thus enhanced by a factor $\zeta = (\theta_S^0/\theta_S)^2 = [1 + (\theta_C/\theta_S)]^2$. The mean force F entering this formula is difficult to specify without detailed calculations, but in the region of charge states q and ionic speeds v such that $v < q$ we can take Bohr's estimate that $F \gtrsim$ binding force of a valence electron (~ 0.5 a.u. in C, Al) for pickup to occur. Thus $\theta_C \sim 0.1$ deg and $\zeta \sim 9-36$ in the present experiments. If our formula is applied to O^{8+} at ~ 2 MeV/amu (the energies used in Ref. 1) a similar enhancement is found, suggesting a MFP $\gtrsim 200$ Å. comparable to the MFP for ion charge changing. Thus, the correlation with q_e should be reduced, assisting the interpretation of the coincidence data. In the region $v \gg q$, F is much smaller and $\zeta \sim 1$, in harmony with Ref. (4) which finds a normal MFP for protons at $v = 5$ a.u.

Our final picture is thus of a net flux into quasi-bound states due to the attractive field of the ion; on exit these states have a moderate-to-high probability of ionizing in the potential step and producing a cusp electron yield corresponding to long MFPs. We thus succeed in unifying and extending the ideas of Betz (8) and Yamazaki and Oda (9) concerning the association of electrons and projectile within the foil and the role of the potential step on exit.

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References

- (1) M. Breinig, S.B. Elston, S. Huldt, L. Liljeby, C.R. Vane, S.D. Berry, G.A. Glass, M. Schauer, I.A. Sellin, G.D. Alton, S. Datz, R. Overbury, R. Laubert and M. Suter, Phys. Rev. A25 (1982) 3015; and references therein.
- (2) J. Burgdörfer, M. Breinig, S.B. Elston, and I.A. Sellin, Phys. Rev. A28 (1983) 3277; and references therein.

- (3) C.R. Vane, I.A. Sellin, S.B. Elston, M. Suter, R.S. Thoe, G.D. Alton, S.D. Berry, and G.A. Glass, Phys Rev. Lett. 43 (1979) 1388.
- (4) E.P. Kanter, D. Schneider, Z. Vager, D.S. Gemmell, B.J. Zabranski, G. Yuan-zhuang, P. Arcuni, PM. Koch, D.R. Mariani and W. Van de Water, Phys Rev. A 29 (1984) 583; Z. Vager, et al., Phys. Rev. Lett. 48 (1982) 592; Y. Yamazaki and N. Oda, Phys. Rev. Lett. 52 (1984) 29.
- (5) A. Gladieux and A. Chateau-Thierry, Phys. Rev. Lett. 47 (1981) 786.
- (6) R. Latz, R. Astner, H.J. Frischkorn, P. Koschar, J. Pfenning, J. Schrader and K.O. Groeneveld, Nucl. Inst. Meth. 194 (1982) 315.
- (7) P. Engar, M. Breinig, R. DeSerio, I.A. Sellin, C.E. Gonzalez-Lepera, S.D. Berry, M. Bloemer and T. Underwood, Conference Abstracts, XIII International Conference on Atomic Physics, #41, Seattle, Washington (July, 1984), and references therein.
- (8) H.D. Betz, et al., Phys. Rev. Lett. 50 (1983) 34.
- (9) Y. Yamazaki and N. Oda, Phys. Rev. Lett. 52 (1984) 29.
- (10) I.A. Sellin, S.D. Berry, M. Breinig, C. Bottcher, R. Latz, M. Burkhard, A. Folger, H.J. Frischkorn, K.O. Groeneveld, D. Hofmann, & P. Koschar, to be published in Physics of Forward Electron Production in Ion-Atom and Ion-Solid Collisions, K.O. Groeneveld, W. Meckbach and I.A. Sellin, eds. (Springer-Verlag, Heidelberg, 1984).
- (11) H.-D. Betz, private communication.

- (12) H. Gould, D. Greiner, P. Lindstrom, T.J.M. Symons and H. Crawford, Phys. Rev. Lett. 52 (1984) 180. Also private communication with H. Gould.
- (13) C. Bottcher, J. Phys. B. 11 (1978), 3887.

Figure Captions

Fig. 1. The longitudinal velocity distribution of convoy electrons emerging from a solid when 12-MeV C^{2+} is incident on a $40\text{-}\mu\text{g}/\text{cm}^2$ Al target. The "singles" or noncoincident 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.

Fig. 2. Convoy electron yield vs. target thickness. The open circles and squares refer to Ni^{24+} ions at 15.2 MeV/u in aluminum and carbon foils and the enclosed circles to Ni^{26+} in aluminum.

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 in	q_e out	8+	7+	6+
8+	Y<110>	21 (68)	39 (28)	82 (4)
	Y<100>	37 (59)	58 (35)	79 (6)
	Y<Rand>	100 (26)	100 (59)	100 (15)
7+	Y<110>	29 (42)	24 (51)	58 (7)
	Y<100>	37 (52)	47 (42)	71 (6)
	Y<Rand>	100 (25)	100 (60)	100 (15)
6+	Y<110>	37 (31)	29 (42)	21 (27)
	Y<100>	39 (49)	45 (42)	47 (9)
	Y<Rand>	100 (27)	100 (57)	100 (16)

INTENSITY

5.80 6.20 6.60 7.00 5.80 6.20 6.60 7.00

VELOCITY (A.U.)

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