Measured Multipole Moments Of Continuum Electron Transfer Angular Distributions

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Abstract

The velocity space distribution of electrons emitted near the forward direction from collisions involving fast, highly stripped oxygen ions with gaseous and solid targets is presented and described in terms of multipole moments of the ejected charge distribution, which permits direct comparison with recent theory. The measurements are produced by employing position-sensitive electron detection to combine emission angle definition with conventional electrostatic spectrometry. Agreement obtained between theory and distributions observed for binary continuum electron loss processes coupled with a similar multipole content observed with solid targets suggests a model of convoy electron production dominated by electron loss from the projectile within the bulk of the target. Further, the connection between multipoles of the projectile electron emission distribution in single collisions and the state of excitation of that projectile excited states may provide the basis for a probe of the state of ions traversing bulk solid matter.

I. Introduction

The spectrum of electrons emitted near zero degrees in ion-atom and ion-solid collisions is dominated by a strong 'cusp'-shaped peak corresponding to electrons nearly matched in vector velocity to that of the projectile ion [1]. Advances in theory and experimental technique permit simplified and unified descriptions of the cusp spectrum and suggest new means to study excitation states of ions traversing condensed matter. The signature of cusp electrons produced by electron capture to the continuum of the projectile ion (ECC) is now seen as the result of a strong dipole moment in the emitted charge distribution, explainable in second Born approximation [2]. Projectile ionization processes, or electron loss to the continuum
(ELC), produce a transversely emitted charge distribution characterized by
even-order multipole moments (monopole, quadrupole, hexadecapole, etc.) and
maximum multipolarity \( k = 2n \) determined by the principal quantum number \( n \) of
the contributing projectile orbital \([3,4]\). The so-called 'convoy' cusp
produced by swift charged particles passing through solid materials has been
observed to possess the transverse signature of the projectile loss mechanism
and to become enriched in higher-order multipoles with increasing projectile
speed \([5]\). This latter observation can be interpreted as reflecting steady-
state excitation of high \( n- \) and \( l- \)states during passage through the bulk
material, followed by electron loss processes which populate the cusp region
of the spectrum. If this interpretation is correct, detailed measurements of
the convoy electron cusp can, under appropriate circumstances, provide a
probe of the state of excitation of charged particles penetrating condensed
matter.

The history underlying this current state of understanding of cusp
formation is too lengthy to detail here; several reviews of relevant theore-
tical and experimental work, which pertain to the domain of high velocity,
high-Z projectiles, have appeared \([1]\). The existence of the cusp is seen as
merely a consequence of the Coulomb final state interaction, the details of
the shape of the cusp, however, reflect the entire history of the collision:
anisotropies arise in the doubly differential cross section (DDCS) which are
collision mechanism-dependent. It is important to appreciate that detailed
comparison between measurements and most existing theory often requires
collision velocities large compared to the characteristic orbital velocities
of any electrons that can contribute to the cusp, usually so that the
first and at most second-order Born approximations can be made with some
confidence. Joachim Burgdörfer \([3]\) has developed a density matrix descrip-
tion of the ELC process which exploits smooth continuation of projectile
excitation across the ionization limit to show that a set of dynamical
multipoles originally introduced to describe bound-state coherences are
suited for the description of continuum-state coherences as well. Conse-
quently, the anisotropies in the DDCS for ELC can be expressed as expectation
values of the dynamical multipoles. Burgdörfer has shown this method to be
extensible to other electron transfer to continuum (ETC) processes and to
collisionally excited Rydberg manifolds [6]. A unified approach thus
emerges in which the same fundamental parameters describe the populations of
cusp and Rydberg final states.

In the framework of the method advanced by Burgdörfer, the DDCS for ETC
processes is expanded in the zero-velocity limit as

\[
\frac{d\sigma}{d\hat{v}} = \left(\frac{a}{v}\right) \sum_{k=0}^{\infty} P_k(\cos\theta) \beta_k
\]

where \( \hat{v} \) is the electron emission velocity in the projectile rest frame (PRF),
\( P_k \) are the Legendre polynomials, and \( \beta_k \) are the asymmetry parameters deriv-
able from the theory. Contact can also be made with the double-series
expansion of the DDCS introduced by Meckbach, Garibotti, and co-workers [7]
and employed in previous work in our laboratory [8] by expanding the \( \beta_k \) to
account for finite-velocity corrections to the cross section:

\[
\frac{d\sigma}{d\hat{v}} = \left(\frac{a}{v}\right) \sum_{k,j=0}^{\infty} B_{kj} v^j P_k(\cos\theta).
\]

II. Method

Much experimental data on ETC processes to date have been singly
differential (in electron energy or longitudinal velocity component), even
though in most cases a small range of collection angles have been employed.
As a result, much of the collision-dependent information contained in the
above DDCS expansions is lost in apparatus-dependent averaging over emission
angles interior to the spectrometer collection cone. We employ an apparatus
developed in our laboratory which subdivides a forward-oriented collection
cone of about 5 degrees half-angle into differential angular elements of
about one-third degree full angle. These elements are sized so that the
effective angular resolution in the PRF corresponds to a differential slice
in transverse electron emission velocity $v_t$ of a size comparable to that of
the slice in longitudinal emission velocity $v_\parallel$ determined by the electron
spectrometer employed. The apparatus collects data simultaneously from all
elements within this forward cone by means of position-sensitive detection
techniques and thereby permits efficient data acquisition, eliminates
mechanical scanning linkages, and automatically determines the zero-degree
direction.

The major elements of the electron spectrometric apparatus are diagram-
med in Figure 1. The target region, which is a ~0.5 cm thick cell for
gaseous targets and a self-supporting foil for solid target measurements, is
viewed by a spherical sector electrostatic spectrometer having a mean
deflection radius of 5.5 cm and a deflection angle of 160 deg. The primary
ion beam exits the spectrometer through a hole in the outer sector. Apart
from small aberrations and in the absence of extraneous fields, the focusing
properties of the spectrometer reestablish, with unit magnification at the
exit aperture, the in-beam angular distribution of electrons having energies
within the spectrometer pass-band. Collision region emission angles thus
correspond one-to-one with arrival angles at the exit aperture. A drift
region 15 cm in length following the exit aperture permits the preserved
emission angles to develop into transverse position coordinates at the
location of a position-sensitive detector (PSD).

The PSD consists of tandem microchannel plate electron multipliers,
followed by a circular-arc-terminated resistive anode [9]. Four charge pulse outputs at the corners of the anode are independently amplified and then decoded using a simple ratiometric method to recover the primary event position and thus the emission angles or, more fundamentally, the transverse velocity components as a digital quantity that, in combination with the spectrometer pass energy, determines the emission coordinates of the detected electron. By scanning the analyzer deflection field, the entire three-dimensional \( \dot{V} \)-distribution of the ETC cusp can be obtained (in velocity space the projectile and laboratory frame distributions are related through the simple translation \( \dot{V}_{\text{PRF}} = \dot{V}_{\text{LF}} - \dot{V}_p \), where \( \dot{V}_p \) is the projectile velocity). While the intrinsic position resolution of the PSD alone is 100 to 150 \( \mu \text{m} \), the corresponding angular resolution of \( \sim 0.05 \text{ deg} \) is broadened by the size of the 1 mm diameter spectrometer exit aperture (1.5 mm for ECC data) to \( \sim 0.35 \text{ deg} \) (0.5 deg). The spectrometer energy resolution is 0.9% FWHM. The three-dimensional (differential in emission energy and polar and azimuthal angles) distributions produced by the apparatus are assessed for multipole content by means of a fitting procedure based on the multipole expansions discussed earlier. The analysis is a straightforward extension of that used for earlier singly differential data published by our laboratory [8].

III. Results and Discussion

Figure 2 displays a selection of results of ETC cusp velocity distribution measurements, corresponding least-squares fits to the multipole expansion described earlier, and theoretical predictions [3,6] in summary form. The data shown are contours of equal intensity of electron emission in the emission-energy and polar-emission-angle plane for single collisions of \( v_p = 10.1, 14.3, \) and \( 16.2 \text{ au} \) \( \text{He}^+ \) ions with \( \text{He} \) and \( \text{Ar} \) targets. The axes are scaled
so that equal intervals in either direction represent approximately equal intervals in longitudinal projectile frame emission velocity \(v_L\) and transverse projectile frame emission velocity \(v_T\). As expected from earlier singly differential work [10], the distributions are dominated by ELC from the loosely bound \(n = 2\) levels by an order of magnitude over ECC and ELC from \(n = 1\). The immediate appearance of these data are of strongly transverse emission, as predicted by Burgdörfer's calculation [3, 6], and in striking contrast to the strong dipole character of the ECC distribution obtained from a \(v_p = 15.4\) au collision in Ne, shown in part f of Figure 2. Also shown is the theoretical angular distribution calculated by Burgdörfer, convoluted with the spectrometer acceptance function.

Excellent agreement is obtained for fitted values of the multipole strengths \(\beta_k\) of the argon data at the higher two velocities; somewhat poorer agreement at 10 au is not of concern because the first Born approximation is suspected to be less accurate at lower \(v_p\) [6]. A lack of agreement between helium target data and theory is of much greater concern. The simpler structure of this target and the use of more accurate scattering functions than were available for argon would at first suggest a more accurate description for helium. An experimental source of a difference between these targets is unlikely because the data sets were alternately acquired for He and Ar targets by merely switching between target gas bottles on a time scale of minutes, while the accelerator operators were instructed to leave all beam conditions (i.e. focusing and steering) unchanged. Further, although pixel-by-pixel 'gas-dump' background subtractions (see section 5) contribute more heavily to statistical and systematic errors with helium targets, the ELC to background ratio at the cusp peak was \(\approx 11:1\) in the worse case.

Lacking a known, sufficiently large source of experimental error that
would selectively affect the helium data, we speculate that doubly inelastic collisions between the active projectile electron and target electrons may be responsible for the difference. Such processes are taken into account in the theory within the framework of a closure approximation and this contribution is fractionally more important for He because of its much smaller nuclear charge. Previous investigations [11], however, have indicated only minor errors in asymmetry parameters are induced by the closure approximation. In addition, the closure approximation cannot account for a difference in radial distribution. It is also possible, of course, that an as yet unidentified excitation or ionization process is occurring either in addition to, or in combination with simple electron loss to the forward peak.

Our recent measurements [5] of the multipole moments of the solid target convoy electron distribution, while limited in scope, are intriguing in that they suggest the feasibility of applying knowledge gained from ELC studies such as those in the previous section and extensions of theoretical studies like those of Burgdörfer [3,6] to probe the processes leading to convoy emission from condensed matter targets. They reveal multipole content of order well beyond the quadrupole and hexadecapole moments obtained for ELC from \( n = 2 \) orbitals and offer an opportunity to perform unique, new measurements sensitive to excitations of swift projectiles that occur while immersed in bulk material. Figure 3 presents contoured emission distributions we have observed for convoy electron production together with corresponding data for ELC from equal velocity \( 0^5+ \) in argon. The targets employed (15 \( \mu \)g/cm\(^2\)) were substantially below equilibrium thickness. The resemblance between the convoy cusps and those for ELC is striking - both are strongly transverse - and the convoy data displays no evidence of the strong dipolarity which is the hallmark of ECC (see Figure 2f). We interpret this feature of the data to
signify a prominent role for ELC in the convoy production process.

The displayed ELC and convoy distributions differ in one important respect, however: attempts to fit multipole moments to the convoy distributions in the same manner as was done with the ELC data produced poor results until multipoles of order up to \( k = 10 \) were included in the fitting procedure. The resulting fitted values of \( \beta_k \) are given in Table I for the three impact velocities studied. As can be seen from a close examination of the \( \beta_k \) values, the enhanced multipolarity of convoy emission skews toward higher multipoles with increasing projectile velocity. Recalling that the maximum predicted multipolarity \( P_k \) of ELC from a given \( n \) level is \( k = 2n \), the data then suggests convoy production which behaves as loss from highly excited states formed by capture or capture plus excitation within the bulk material of the target. Also shown in the table are values of \( B_k \equiv \beta_k(\text{convoy})/\beta_k(\text{ELC}) \) for \( k = 2,4 \), which reflects the extent to which high \( k \) values for the convoy distributions rise beyond those for ELC at larger projectile velocities. The observation of high \( n,l \) excitation within the target agrees with recent reports made by Betz, et al. [12] that such high states, produced within the bulk, are required to explain long-lived cascade tails of foil-excited \( \text{Si}^{15+} \) Ly \( \alpha \).

The power and speed with which our method can determine high-order multipole content of ejected electron distributions and the inferred connection with high \( n,l \) excitation make several follow-up measurements of interest. Perhaps most obvious is a measurement of the target thickness dependence of the multipole content of convoy emission; addition of coincident detection of emergent ion charge state would permit measurement and comparison, in the same apparatus and under the same conditions, of mean free paths for convoy electron production and extinction and for projectile charge-
changing. While it is expected that elastic and inelastic electron scattering processes which occur after convoy production and prior to or during exit from the foil surface (including the effect of the exit potential 'step') must be taken into account in any detailed examination of convoy multipole distributions, the strong Coulomb 'focusing' [13] provided by the nearby projectile ion may make these effects smaller that they would appear to free electrons of the same speed, at least for the highly charged ions studied here. Further, there are indications that the effect of the static exit surface potential step is at least partially compensated for by a dynamical screening effect [14]. We are therefore cautiously optimistic that the present results will provide a basis for a detailed probe of the state of excitation and ionization of ions traversing bulk condensed matter.

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Figure Captions

Figure 1.
Schematic diagram of the spectrometric apparatus, equipped with position-sensitive detection to achieve resolution of emission angles.

Figure 2.
Contour plots for ELC by $^5$O$^+$ in Ar and He at $v_p = 14.3$ au, and ECC by $^8$O$^+$ in Ne at 15.4 au. Contours shown represent multiples of 12.5% of the peak height. Horizontal scale represents lab frame electron energy; vertical scale represents polar electron ejection angle. Scaling is chosen so that isotropic emission would produce essentially circular contours.
(a) Measured distribution for Ar target. (b) Corresponding best fit to (a), normalized to peak height of data. (c) Theoretical distribution (Ref. 11) after convolution with spectrometer acceptance, also normalized to peak of (a). (d) Measured distribution for He target. (e) Corresponding He target fit, normalized to peak of (d). (f) Measured ECC distribution for 15.4 au $^8$O$^+$ in Ne.

Figure 3.
Contour plots of emission distributions for convoy electrons produced in thin carbon targets compared with those for ELC in argon. Contour levels shown represent intervals of 20% of each peak height. The horizontal dimension of each plot represents longitudinal velocity components; the vertical dimension is transverse to the projectile direction. Equal velocity scales are shown in each direction, with the bar indicating an interval of 0.2 au. Isotropic angular distributions would have circular contours.
TABLE I. Comparison of $B_k$ values for convoy distributions with those for ELC. Also shown are the even order coefficient ratios $B_k$ defined in the text.

| $v_p$ (au) | Target | $B_2$  | $B_4$  | $B_6$  | $B_8$  | $B_{10}$ |
|------------|--------|--------|--------|--------|--------|----------|
| 16.2       | Carbon | -0.78  | 0.25   | -0.32  | 0.19   | -0.16    |
|            | $A_{ra}$ | -0.68  | 0.13   |        |        |          |
|            | $A_{rb}$ | -0.70  | 0.18   |        |        |          |
|            | $B_k$   | 1.1    | 1.9    |        |        |          |
| 14.3       | Carbon | -0.82  | 0.29   | -0.26  | 0.07   | -0.03    |
|            | $A_{ra}$ | -0.62  | 0.12   |        |        |          |
|            | $A_{rb}$ | -0.67  | 0.16   |        |        |          |
|            | $B_k$   | 1.3    | 2.4    |        |        |          |
| 10.1       | Carbon | -0.48  | 0.11   | -0.18  | 0.09   | -0.05    |
|            | $A_{ra}$ | -0.26  | 0.05   |        |        |          |
|            | $A_{rb}$ | -0.56  | 0.10   |        |        |          |
|            | $B_k$   | 1.8    | 2.2    |        |        |          |

*aExperimental values from Reference 6.

*bTheoretical values from References 9 and 11.
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