The development of projectile excitation during ion-solid collisions observed through convoy electron emission

Stuart B. Elston

Department of Physics and Astronomy,
University of Tennessee, Knoxville, TN 37996

and

Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA

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Abstract

Preliminary results and interpretation are presented for a recent experiment designed to explore the evolution of electronic excitation of a projectile ion during flight through a solid target by monitoring the development of high-order multipole moments of the convoy electron angular distribution as a function of target thickness.

1. Introduction

A number of studies over recent years has drawn increasing attention to the importance of understanding the detailed nature of and role played by electronic excited states of projectile ions traversing solid media, and the mechanisms by which this excitation develops. This problem is a long-standing one, dating back at least to the early paper of Bohr and Lindhard.¹ Experiments observing the production of photons by projectiles with velocities corresponding to the order of one to a few MeV/u have suggested the existence of states of high angular momentum in bulk solid media, analogous to Rydberg states of isolated atoms.² Complementary studies of convoy electron production (from oxygen ions) reveal multipole moments of the emission angular distribution up to order at least 10, consistent with the population of states describable in a hydrogenic basis by principal quantum numbers up to at least 5.³ In this latter case ($n = 5$), one considers electronic orbitals comparable in size to the typical interatomic target spacing, where dynamic screening of the projectile charge should be significant and the use of such a simple basis as a hydrogenic one should be highly suspect. By means of contrast, however, earlier work aimed at describing the approach of an ion to charge-state equilibrium,⁴ which pointed out the need to consider excitation of the projectile, found satisfactory agreement with the exit charge distribution by including only a single, low-lying excited state. While this list is by no means complete, it is seen that information is and has been accumulating which is observable by a variety of techniques whose combined sensitivity spans the full range of states of the projectile.

It is widely recognized that the complex sequence of collisional excitation, ionization, and capture events that occur during the passage of a typical projectile through a solid present a formidable challenge to theory.⁵ Central to this challenge is a need to understand the nature of both bound and continuum electronic states of a Coulomb projectile whose trajectory is immersed in the 'sea' of electrons and ion cores of an amorphous or crystalline solid, which sea is strongly disturbed by the projectile's passage. Given even a reasonable approximation to the electronic projectile states, one must then attempt to model the evolution of an initial state during successive collisions which may often be severe enough to make perturbation methods useless. Of particular interest to the analysis of results presented in this paper is that it has lately become clear that one cannot assume that an electron 'pumped up' via the 'density effect'¹ beyond an approximate screening distance becomes forever ionized, but instead must be followed as long as it is transported within the nearby phase space of the projectile Coulomb center.⁶

This work presents preliminary results and interpretation of a recent experiment designed to probe the evolution of excited states of projectile ions while traversing thin

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amorphous solid targets, as revealed by the development of high-order multipole moments describing the angular distribution of convoy electron emission. The experiment employed zero-, one-, and three-electron Ar projectiles, to permit simple assumptions to be made about initial projectile states. The relatively large projectile velocity (37 a.u.) and the use of thin, amorphous carbon targets were chosen to reduce the above-mentioned transport effects.

II. Method

The basic apparatus used in the present experiment, and the methods used to analyze the angular distributions observed, have been described in some detail previously.⁸ Briefly, beams of 36 MeV/u Ar^{q+}, with $q = 15, 17$, and 18 provided at the LISE facility of the Grand Accélérateur National d'Ions Lourds (GANIL) were directed onto self-supporting carbon foil targets with nominal thicknesses (quoted by the manufacturer⁹) in the range from 0.5 to 21 $\mu\text{g}/\text{cm}^2$. The projectile beam diameter was typically 1.0 mm and the beam emittance was limited to about 1π mm-mrad. Electrons emitted within a forward cone of about 4° were analyzed by an electrostatic spherical sector electron spectrometer equipped with a position-sensitive detector (PSD) to provide simultaneous resolution of energy ($\delta E/E \approx 1\%$) and emission angles ($\delta\theta \approx 0.5^\circ$). The PSD output, decoded by the ratiometric charge-division method, along with the spectrometer pass energy completely determine, within limits imposed by the resolution, the velocity components of the detected electron. Thus by scanning the spectrometer deflection field we map the entire $\vec{v}$ -distribution over a region of interest, in this case around the projectile vector velocity. Multipole moments of the angular distribution were extracted from the emission distributions by using a non-linear least-squares fitting routine as described below. Auxiliary measurement of the exit charge state fractions for 37 a.u. Ar¹⁵⁺ incident on both thin ($\leq 15\mu\text{g}/\text{cm}^2$) and thick ($\geq 100\mu\text{g}/\text{cm}^2$) carbon foils were performed on-line but with separate targets floated from the same stock as those used for the emission distributions. Target thicknesses were calibrated in-situ before and after the main experiment using electron energy loss measurements obtained with an electron gun and the emission spectrometer.

III. Results and Discussion

Two typical observed emission distributions are displayed in Figure 1 as contours of equal emission intensity. Those shown are for Ar¹⁷⁺ with projectile velocity $v_p = 37$ a.u. incident on carbon foil targets of nominally 1 and 8 $\mu\text{g}/\text{cm}^2$ thicknesses. Considerable evolution of the emission distribution, presumably reflecting projectile excitation as a result of multiple collisions, is evident.

Extraction of multipole moments from the emission distributions was done to quantify the observed evolution and to make contact with theories of the doubly-differential cross-section through the multipole expansion

$$\frac{d\sigma}{d\vec{v}} = \frac{\sigma_0}{v} \sum_{k=0}^{\infty} \beta_k P_k(\cos\theta) \quad (\beta_0 \equiv 0),$$

where $\vec{v} = \vec{v}_e - \vec{v}_p$ is the projectile-frame emission velocity, the β_k are the multipole strengths, the P_k are Legendre polynomials, and σ_0 is the total emission cross-section. Non-linear least-squares fitting is used to compare this expansion, transformed into the laboratory frame with first-order relativistic corrections and convoluted with the spectrometer resolution function, to the observed distributions; the β_k are determined as the fitting process converges.

Figure 1. Contours of equal convoy electron emission intensity at intervals of 10% of the peak value for 37 a.u. velocity one-electron argon ions incident on 1 and 8 $\mu\text{g}/\text{cm}^2$ amorphous carbon targets. The horizontal and vertical axes indicate components of emission velocity parallel (v_p) and transverse (v_t) to the incident projectile directions.

Two particularly interesting multipole moments, β_1 and β_2 , are shown as a function of target thickness and incident projectile charge state in Figure 2. We note that in addition to the moments shown in Figure 2, the observations support significant odd order moments up to order 5 and even moments up to order (at least) 10. Efforts are presently underway to model in some detail the multiple collision processes leading to the excitation implied by these multipoles; the present paper will be limited to a discussion of how useful quantitative results can in principle be obtained from multipole strength evolution data like that of the figure.

From the auxiliary exit-charge-state distribution measurements, one can determine total cross-sections for electron capture and loss for each of the relevant projectile charge states, in much the same manner as has been done by Rozet¹⁰ for similar collision systems. The corresponding mean-free-paths (mfp's) for K- and L-shell ionization and K-shell excitation for 36 MeV/u Ar ions in carbon are consistent with theoretical predictions of Gillespie¹¹ and are of the order of 30000, 10000, and 32000 a.u., respectively. With respect to these and similar processes, then, the present experiment can be considered to be under single collision conditions, since the targets employed have thicknesses in the range 100 to 2000 a.u. However, the excitation cross-sections for shells L- and higher can be expected to be larger, producing smaller mfp's, so that given the population of an L-shell level by excitation or capture, further excitation can be expected to occur rapidly. Of equal importance, inelastic free-electron mean-free-paths for equal-velocity electrons (i.e., ≈ 37 a.u.) are typically 600 a.u.¹² Consequently the thicker targets in this experiment should exhibit the effect of multiple collisional excitation and convoy electron transport. The multiple collision aspect should be particularly evident in the case of incident Ar^{15+} , which carries a loosely-bound L-electron into the target, perhaps even at intermediate thicknesses.

Figure 2. The dependence of dipole (β_1) and quadrupole (β_2) moments extracted from the emission angular distributions on target thickness; the solid lines are drawn to guide the eye. Error bars are based on statistical uncertainties associated with the fitting process and on uncertainty in foil thickness.

Considering that the three incident charge states shown in Figure 2 represent three relatively unique initial projectile states (no electron, one 1s electron, and 3 electrons of which one is the more-loosely-bound 2s and is likely to dominate loss and excitation processes), one can take the zero-target-thickness limit of the set of β_k for each given incident charge state to be characteristic of ELC from the appropriate shell or of ECC in the case of the bare projectile. This is possible because the observed thin-target yield of convoy electrons from one-electron Ar^{17+} , i.e. ECC to a charge-17 center plus K-shell ELC, exceeds that from bare Ar^{18+} (ECC) by a factor of 50, so that at worst the one-electron data represents K-shell ELC contaminated by 2% ECC. Further, the Ar^{15+} (three electron) convoy yield is a factor of ≈ 20 greater yet, suggesting that the distribution of β_k values for incident Ar^{15+} is dominated by ELC from the L-shell. The specific β_k values obtained in this way from the zero-thickness limit are in quite reasonable agreement with previous experimental data^{3,13} and with theory from several sources¹⁴.

The effect of plural- or multiple-collision processes on convoy electron emission is to 'mix' the three characteristic β_k distributions introduced above, and to add other distributions characteristic of higher excited projectile states, in an evolving linear superposition. This mixing occurs with targets of increasing thickness, and the coefficients of this superposition of β_k distributions are related to the cross-sections which connect

the various states. For example, the thin-target Ar^{18+} bare incident ion data display the strong, negative dipole moment which is typical of the ECC process at high collision velocities and which is observed in singly-differential as a skewing toward emission energies below the central, 'cusp' energy. This large, negative β_1 value gives way at larger thicknesses to a small dipole moment and an increasingly strong negative quadrupole moment (β_2), which is characteristic of the ELC process as can be seen in the thin-target Ar^{17+} distribution. We interpret this particular behavior as evidence of so-called capture-loss⁷ in which an electron is captured into a bound orbital and then, in a subsequent collision, is lost to the continuum in an electron capture-loss to the continuum process (ECLC), and which, to our knowledge, has never previously been resolvable from ECC. The measurements and analysis technique outlined here should provide a quantitative measure of the relative size of these two mechanisms.

Other information of interest, such as the K- to L-shell excitation cross-section which underlies the initial growth of a β_4 moment and the rate of approach to projectile excitation equilibrium, is potentially available, pending analysis, in the data of Figure 2 and corresponding plots of other measurable multipole strengths; space, however, does not permit an exhaustive discussion here.

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