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Target orientation dependence of the charge fractions observed for low-energy multicharged ions (Ne^{q+} and F^{q+}) backscattered from RbI(100)

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Abstract

Using a time-of-flight (TOF) technique we have investigated projectile neutralization in the backscattering from RbI(100) of Ne and F multicharged ions in the keV energy range. The energy distributions and charge fractions were measured as a function of the polar incidence angle and the target azimuthal orientation. We found significant variations in the neutralization probability for the different projectiles suggesting that level matching effects play an important role. The observed scattered charge state distributions show significant dependences on both incidence and azimuth angle. These variations are attributed not only to the presence or absence of encounters with nearest neighbors, but also to the presence of more atoms lying along particular directional strings. Interpretation of the results was facilitated by carrying out detailed trajectory simulations. Sample data for few-keV F^{7+} and Ne^{8+} incident projectiles are presented to illustrate the underlying concepts.
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1. Introduction

In this article we give a brief overview of recent studies of projectile neutralization during large-angle backscattering interactions with an insulator surface, RbI(100), carried out at the ORNL Multicharged Ion Research Facility. In contrast to measurements of projectile neutralization during grazing surface interactions [1], which involve a large number of lattice sites, our backscattering studies have as one of their goals improved understanding of projectile neutralization in the limit of one or two surface-atom collisions. As shown in published results [2,3] for Au(110), such collisions can be identified by energy loss analysis of the scattered projectiles to select binary and quasi-binary collisions. It was further shown that information about projectile neutralization at several different distinguishable surface lattice sites could be obtained from analysis of the target azimuth dependencies observed for the various scattered charge states. Using classical trajectory Monte Carlo simulations, projectile neutralization during true binary and quasi-binary double collisions with the different surface lattice sites could be determined [2].

Extending our projectile neutralization measurements to alkali halide targets, we have tried to determine the effect of the significantly reduced electron density characterizing such targets, and to assess the possibility of resolving the individual target constituents by energy loss analysis. Initial measurements [4] with CsI(100) could not resolve energy losses associated with scattering from the halogen and alkali sites due to the small mass difference of the target atoms. However, subsequent measurements [5] with a RbI(100) target showed clear resolution of halogen and alkali site scattering. This finding opened the door to a site-specific study of neutralization in quasi-binary double collisions, where the identity of the "hard" collision scattering site is

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determined by energy loss analysis, and the identity of the "soft" collision partner is selected by suitable azimuth orientation of the target relative to the projectile scattering plane. A <100> direction selects an alkali–halogen or halogen–alkali collision sequence, and a <110> direction selects a halogen–halogen or alkali–alkali sequence. Aspects of site-specific projectile neutralization are briefly discussed and illustrated below.

2. Experiment

The measurements were performed at the ORNL Multicharged Ion Research Facility (MIRF) [6] using an ultra high vacuum (10^{-10} mbar) apparatus previously described [7]. A chopped primary ion beam is decelerated from (10×q) keV to final energies in the 4–5 keV range before impinging on a single crystal RbI(100) surface. The sample is mounted on a manipulator that allows variations of the ion incident direction with respect to the surface normal (θ) or to a main crystallographic axis (ϕ) (see Fig. 1). Ions and atoms scattered from the sample are detected by a multichannel plate (MCP) located at a scattering angle of 120° from the incident beam direction. The time-of-flight (TOF) analyzer, which has a floatable drift tube, allows simultaneous measurement of energy distributions and charge fractions for the scattered projectiles [7]. An important feature of this approach is the ability to analyze all scattered charge states, including neutrals, essential for complete determination of scattered charge fractions.

In order to reduce the induced ion bombardment damage before each measurement, the surface was prepared using cycles of grazing bombardment with 2 keV Ar^+ ions and annealing at 450 °C. This method effectively removes impurities, and yields very smooth ordered surfaces. To prevent macroscopic charging, the RbI target was maintained at a temperature of 250 °C during the measurements.

3. RbI(100) Crystal

RbI has a diatomic cubic structure (rock salt) of alternating atoms ABAB on each side of the unit cell (e.g., a <100> direction, with 0.366 nm atomic spacing). In a <110> direction, there are alternating parallel atomic rows AAAA and BBBB (0.517 nm atomic spacing). This means that by changing the azimuth target orientation we can change the adjacent atomic specie that can participate in the quasi-binary collision. A <100> direction selects quasi-binary collisions of the type: Rb–I and I–Rb, while a <110> direction, quasi-binary collisions of the type: Rb–Rb and I–I. The salient features of the crystal structure and the scattering geometry are indicated schematically in Fig. 1.

4. Trajectory simulations

To understand target orientation dependences of the scattered fluxes, and the types of collisions comprising them, projectile trajectory simulations using the MARLOWE code (version 15b) [8] were carried out. MARLOWE treats the interaction between the projectile and target atoms in an elastic binary collision approximation (BCA). We chose an exponential-sum screened Coulomb interaction potential to represent the projectile and target species. The scattered beam energy distribution was simulated in a series of MARLOWE tasks with different projectiles and incidence conditions. For all interactions resulting in backscattering from the surface, the number of

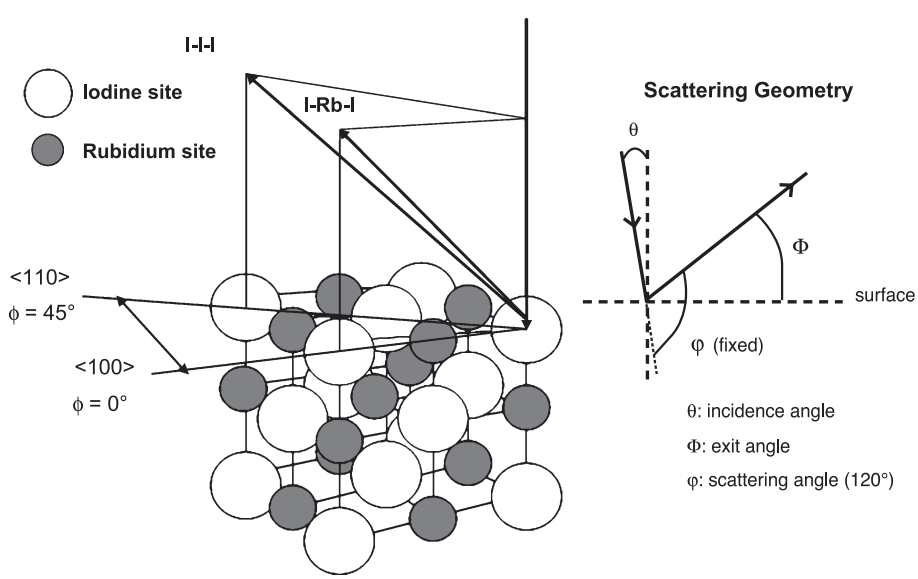

Fig. 1. Schematic diagram of the RbI(100) target, showing the different atomic strings along a <100> and <110> direction, and the scattering geometry.

collisions, scattering angles, etc., were saved; this permitted subsequent reconstruction of all such trajectories. To improve counting statistics the simulated angular acceptance was increased by a factor of 4 over the experimental acceptance angle of 2°. Of the 10^8 trajectories generated for each target orientation investigated, about (0.03–0.08)% were scattered into the detector acceptance cone.

The simulations were used as the basis for analyzing the collision events contributing to the "binary collision" (BC) peaks seen experimentally. Results will be described in the next section.

5. Results and discussion

In Fig. 2 we show the TOF spectra for 4.3 keV F^{7+} normally incident on the RbI(100) surface along the two principal directions: <100> (ϕ=0°) and <110> (ϕ=45°). These spectra are fairly typical for multicharged ion projectiles when a floatable drift tube is used (biased at −3 kV in this case). The spectra show that different recoil charge states are well separated.

We can also resolve F projectiles backscattered in a binary collision from either Rb or I atoms, because the two surface constituents have significantly different masses (site-specificity for each final charge state). We see that binary collision peaks associated with backscattering from Rb and I sites are clearly resolved for final charge states as high as 3+. The small peak observed at TOF=0 is used to calibrate the flight time scale. This peak is produced by soft X-ray events registered on the TOF detector resulting from projectile impact on the surface. The scattered neutral binary collision peaks sit on top of a broad background structure which arises from multiple collisions whose trajectories extend deeper into the RbI bulk. This multiple collision background is not a significant feature for charged recoil ions. This indicates that charged recoil ions do not survive in multiple collision cascades. We also note that the intensities of the scattered charge states shown in Fig. 2 are dependent on the target azimuthal orientation.

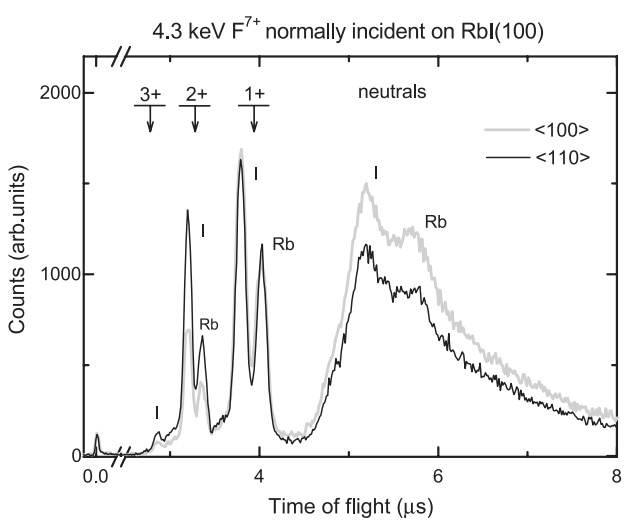

Fig. 2. Raw TOF spectra for 4.3 keV F^{7+} normally incident on the RbI(100) surface, along <100> and <110> directions.

In Fig. 3 we compare recoil energy spectra obtained for different projectiles: 4.3 keV F^{7+} and 4.9 keV Ne^{8+}. The various scattered charge state peaks shown were corrected for collection and detection efficiency effects [7] during the TOF to energy transformation. The corrections can be significant and must be properly accounted for to obtain an accurate determination of scattered charge fractions.

The spectra in Fig. 3 were acquired along a <110> direction where the hard–soft collisions are of the I–I or Rb–Rb type. We note significant differences between the scattered neutral and 1+ fractions in each spectrum. In contrast to the F projectile spectrum, the Ne spectrum shows almost no discernible binary-collision neutral formation for these conditions. Surprisingly, the neutral yield is much higher for fluorine, even though the exit angle is less grazing. This result suggests that the level matching effects seem to be important.

In Fig. 2 we showed that the intensities of the scattered charge states are dependent on the target azimuthal orientation. This effect was presented in more detail in a previous paper (see Ref. [9]) where we showed the azimuthal variations of the scattered charge fractions for normally incident Ne^{8+} projectiles resulting from quasi-binary collision backscattering from Rb and I lattice sites. Interestingly, both Rb- and I-backscattered projectile charge states showed the same trend with azimuth orientation, i.e., smoothly decreasing degrees of neutralization in going from the <100> to the <110> azimuthal direction.

More dramatic variations have been observed when changing the incidence angle. Variations in 4.3 keV F^{7+} energy spectra for two different angles: θ=−9° and θ=15°, along a <100> direction are shown in Fig. 4. We note that there is a significant dependence of both neutrals and non-neutral charge state fractions with angle. Incident angles of θ=−9° and θ=15° correspond to exit passing distances to the next neighbor of 0.25 and 0.1 nm, respectively. The closer the exit trajectories pass neighboring atom on the surface, the higher the neutralization probability. In fact, for the 0.1 nm case, there are almost no 3+ and 2+ charge states, and we can perfectly distinguish the two peaks corresponding to F^0.

To obtain additional insights about the various target orientation dependences of the scattered fluxes, and the types of collisions involved, we performed MARLOWE projectile trajectory simulations. In particular, we focused on the collision events contributing to the binary collision peaks (BC) seen experimentally. Simulations reveal that the BC peak is in fact built up from two kinds of events—single and "quasi-binary" collisions. The quasi-binary

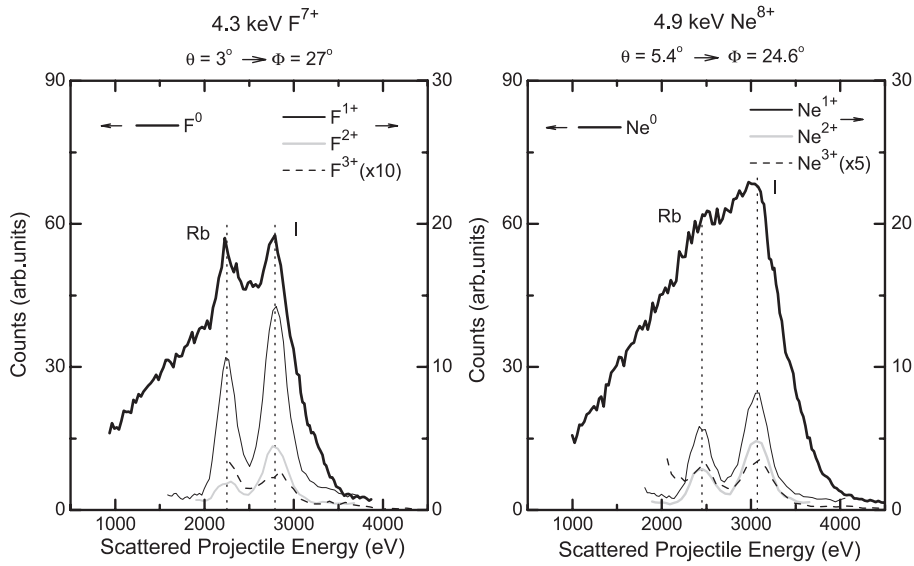

Fig. 3. Energy spectra of scattered neutral and positively charged ions for two different incident projectiles: 4.3 keV F^{7+} and 4.9 keV Ne^{8+}; obtained from TOF spectra acquired along a <110> direction. Vertical dotted lines indicate the calculated position for the binary peaks.

backscattering collisions occur predominantly from only the surface layer, thus blocking and shadowing effects on the exit trajectory after hard collisions in deeper layers do not play a role in determining possible incidence variations of the projectile quasi-binary collision backscattered flux. These quasi-binary surface events are "hard–soft" or "soft–hard" collision sequences that have an energy loss very close to, and experimentally unresolvable from, that of a single "hard" collision. Moreover, the quasi-binary surface collision cascades can even involve two, three or four collisions, usually with combination of one large angle ("hard") event and one or more small angle ("soft") scattering event.

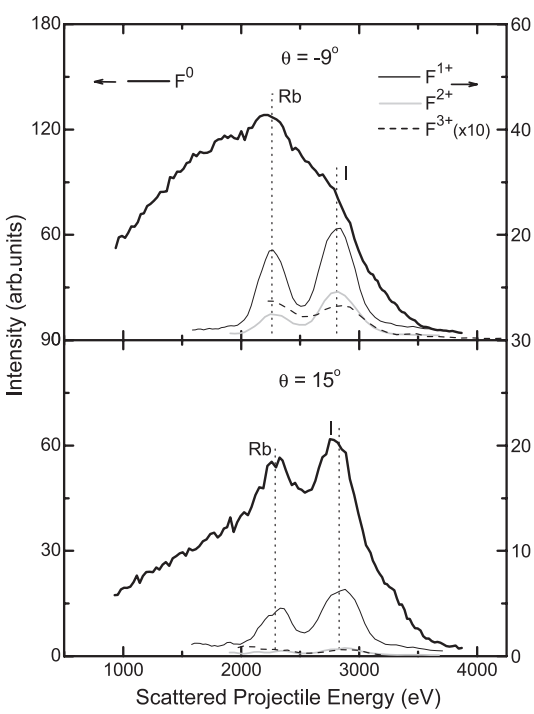

Fig. 4. Energy spectra of scattered neutral and positively charged ions for incident 4.3 keV F^{7+}, obtained from TOF spectra acquired along a <100> direction for two different incidence angles: $\theta=-9^\circ$ and $\theta=15^\circ$. Vertical dotted lines indicate the calculated position for the binary peaks.

The fact that quasi-binary events are particularly prominent for incidence angles having a more grazing exit trajectory is illustrated in Fig. 5a. In fact, for $\theta=15^\circ$, double, triple and quadruple collisions are the only types that occur, as shown in Fig. 5b. High multiplicity events (i.e., number of collisions>10) do not contribute to the BC peak, and form instead the pedestal upon which the peak sits. The most prominent features are the two peaks at the expected energy for binary collisions with the Rb or I atoms (see Fig. 5a). The ratio of the Rb and I binary collision peaks (areas) corresponds closely to the calculated ratio of the relevant differential scattering cross sections of these two target atoms for 120° backscattering (i.e., about 70%). The relative contributions of true binary and quasi-binary double (and at higher incidence angle, triple) collisions to the BC peak vary strongly with the incidence angle, and underlie the measured incidence variations of scattered charge fractions that were shown in Fig. 4. We reemphasize that binary and quasi-binary collisions contributing to the observed backscattering peaks occur almost exclusively at the surface-vacuum interface, even away from normal incidence (see Fig. 5b).

A study of incident ion neutralization involving quasi-binary collisions would be incomplete without also addressing effects associated with azimuthal orientation. Azimuthal change from a <100> to a <110> direction not only changes the passing distance to the nearest neighbor, but also the identity of this atom. In fact, the type of quasi-binary collisions transitions from I–Rb or Rb–I to I–I or Rb–Rb

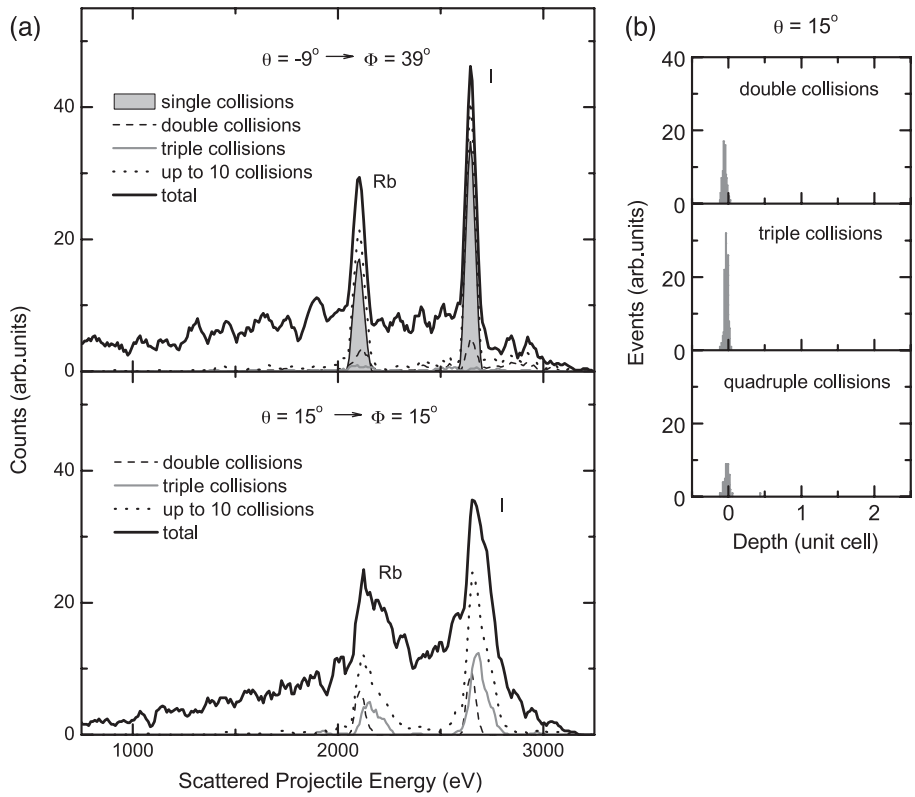

Fig. 5. MARLOWE simulation results for 4.3 keV F projectiles incident on RbI(100) that duplicate incidence conditions of the experimental spectra shown in Fig. 4, i.e., along a <100> direction, for two different incidence angles: $\theta=-9^\circ$ and $\theta=15^\circ$. (a) Energy distributions of scattered projectiles and (b) layers where the different collision cascades are generated for the representative case of $\theta=15^\circ$. Note that even for multiplicity as high as 4, collisions occur predominantly on the topmost layer.

collisions. To determine if the change in the second partner of the collision has any influence on the neutralization probability we also studied incidence angles where the passing distance of the receding trajectory is identical for both azimuth directions. Results are shown in Fig. 6. The spectra correspond to 4.3 keV F^{7+} projectiles incident at an angle $\theta=-9^\circ$ in a <100> direction, and $\theta=3^\circ$ in a <110> direction. These incident angles correspond to a passing distance of ~0.25 nm for the second target atom. Vertical dotted lines in Fig. 6 indicate the calculated position for the peaks corresponding to fluorine ions backscattered on either, Rb or I. We see that there is a significant difference in the scattered neutral yield along the two azimuthal directions. Thus we see (at least for F) that equivalent passing distances do not result in equivalent neutralization along <100> and <110> directions.

Our MARLOWE trajectory simulations again indicate that, for both these cases (i.e., $\theta=-9^\circ$ along <100> direction and $\theta=3^\circ$ along the <110>), the single and quasi-binary collision backscattering occurs predominantly from the topmost layer and that contributions from single and quasi-binary collisions to the total energy spectrum are almost identical. Consequently, azimuthal differences in the neutralization are associated with the collision pair: Rb–I and I–Rb along <100> vs. I–I and Rb–Rb along <110>.

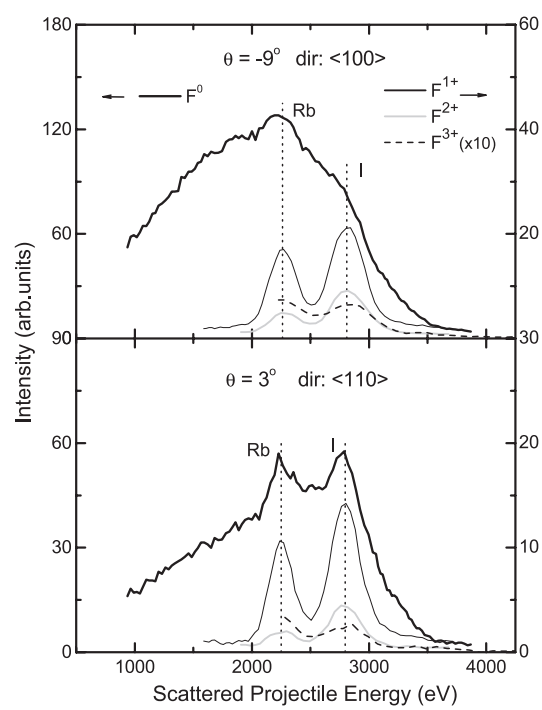

Fig. 6. Energy spectra of scattered neutral and positively charged ions for incident 4.3 keV F^{7+}, obtained from TOF spectra acquired under selected incidence conditions. The receding trajectory passing distance to the nearest neighbor is 0.25 nm along the two directions, <100> and <110>. Vertical dotted lines indicate the calculated position for the binary peaks.

6. Summary and conclusions

Investigations of projectile neutralization in backscattering from RbI(100) for few-keV Ne^{8+} and F^{7+} incident ions indicate:

(i) Significant variations in the scattered neutrals and 1+ charge states for the different projectiles. Ne spectra show no discernible binary collision neutral formation for projectile incidence close to normal incidence. This suggests the importance of level matching effects even for the complex multi-electron transfer processes involved in the large-angle backscattering binary collisions studied here.

(ii) Significant variations in the scattered charge states (principally neutrals) with incidence angle and target azimuth orientation. These variations are attributed not only to the presence or absence of encounters with nearest neighbors, but also more distant atoms lying along the particular directional strings.

(iii) For the same passing distance to the second partner of a quasi-binary collision sequence, the neutralization probability is not the same in <100> and <110> directions. Instead, differences are probably associated with different quasi-binary collision sequences, i.e., Rb–I and I–Rb along <100> vs. I–I and Rb–Rb along <110>.

(iv) Even away from normal incidence, the MARLOWE simulations indicate that subsurface layer contributions to the binary collision flux are not significant. Thus, proximity effects can be studied without the added complexity of competing or distorting effects that could arise for collisions in deeper target layers.

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