Target Orientation Dependence of the Backscattered Intensities and Charge Fractions Observed for Large Angle Quasi-Binary Collisions of Ar^{q+} Ions with Au(110)

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Abstract

Using a time-of-flight technique we have investigated the flux intensity variations for 5 keV Ar^{q+} ions (q=2,7,9,11) backscattered from Au(110) by 120° in quasi-binary collisions. A strong dependence on target orientation is found for the neutral flux, while for the charged components only a weak dependence is seen. An analysis of the observed dependences based on trajectory simulations clearly shows site-specific neutralization differences between the various possible binary collisions and a role of reionization processes in formation of multiply charged scattered ions.

1 Introduction

The large angle scattering technique has long been used in surface elemental and structural analysis and is only recently seeing increased interest as a tool for the study of multiply charged ion (MCI)-surface interactions [1, 2]. While this technique is more challenging than the grazing incidence approach in terms of achieving the very low energy scattering conditions needed for studying, e.g., above-surface projectile neutralization, it offers the advantage of opening to study interactions restricted to one, or at most two collisions along well defined scattering trajectories. The present contribution is to demonstrate the potential of the technique for MCI-surface charge exchange studies and to develop and supplement the results and ideas concerning path-dependent neutralization of multiply charged Ar ions incident on Au(110) already briefly reported elsewhere [3].

2 Research Approach

2.1 Experimental Technique

The measurements were carried out at the ORNL Multicharged Ion Research Facility with previously described apparatus[2, 4] that implements an ultra-high vacuum (10^{-10} mbar), floating scattering chamber equipped with a time-of-flight (TOF) analyzer. This analyzer features a floatable drift tube, permitting simultaneous measurements of energy distributions and charge fractions of projectiles scattered from the single crystal target into 120°. The target was attached to a sample mount with two rotational degrees of freedom and was prepared by cycles of sputter cleaning under grazing incidence with 2 keV Ar^+ ions and successive annealing at about 450° C. The chopped primary beams of argon MCI were decelerated from (10 x q) keV to a final energy close to 5 keV before impacting the Au(110) surface.

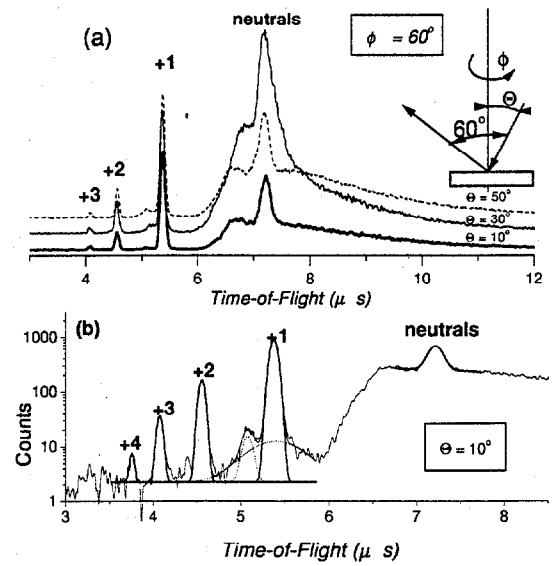

Figure 1: (a) Scattered projectile TOF spectra for 5.7 keV Ar^{9+} ions incident on Au(110) for three different incidence angles; all spectra are in linear scale and normalized to the same integrated target current; (b) An example of the non-linear fitting used to determine the "BC" fluxes (see text for details).

Fig. 1(a) shows typical scattered projectile TOF spectra for Ar^{9+} incident ions, and illustrates the separation of the various scattered charge states achieved by use of the floatable TOF drift tube and the variations of the scattered flux intensities with incidence angle. The spectra exhibit rather sharp peaks at energies very close to those calculated for elastic binary collisions (BC) between the incident projectile and individual target atoms. In the case of the neutrals the "BC" peak sits on top of a broad structure arising from multiple collision events. The exact shape of this structure is not known *a priori* and is the primary source of uncertainty in the non-linear fitting procedure performed to determine the areas (i.e. experimental fluxes) of the "BC" peaks. Usually the peaks were fitted by Gaussians (thick lines in Fig. 1(b)) while the multiple scattering background was approximated by a combination of a constant offset and additional Gaussians (dashed lines) in the case of the singly charged scattered ions or a linear slope (not shown) in the case of the scattered neutrals. As can be seen, for neutrals both the shape and intensity of the multiple collision background, as well as the intensities of "BC" peaks are a strong function of incidence angle. Although not illustrated in the figure, the spectral shapes and intensities also depend

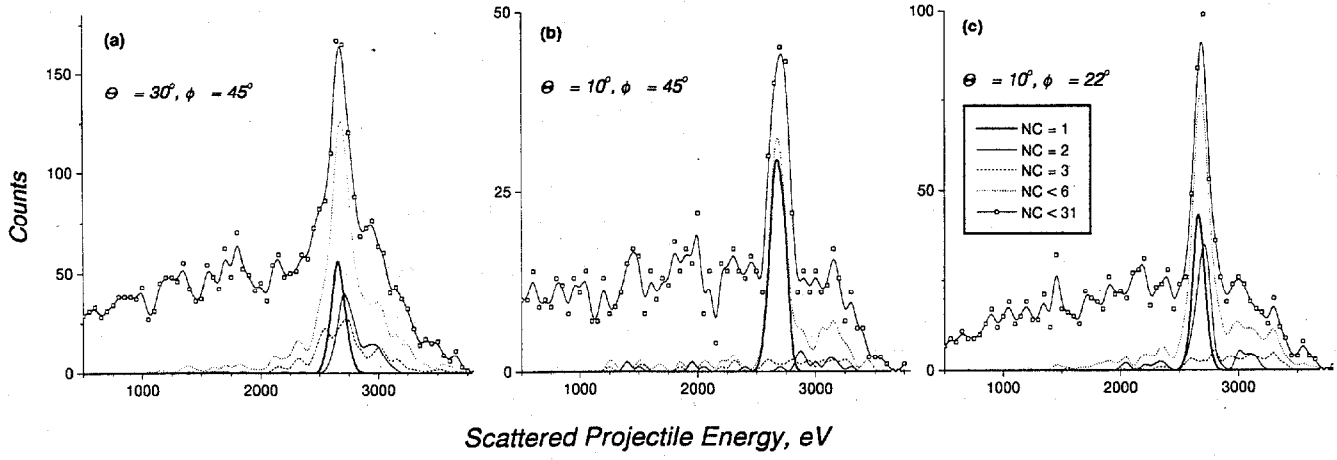

Figure 2: Simulated energy loss spectra for three different target orientations (lines with symbols). The spectral contributions from low multiplicity collision events are separately indicated; NC in the figure denotes the number of collisions a particular spectral feature arises from.

strongly on the target azimuth orientation. The non-neutral peaks have practically no multiple collision "background" (although high energy "knees" arising from low multiplicity collision events are clearly visible on the left side of singly charged ion peaks) indicating very low survival probabilities of non-neutral scattered projectiles in multiple collisions. In addition it is noted that the non-neutral peaks manifest much weaker orientation dependent variations.

2.2 Computer Trajectory Simulations

To understand incidence angle and target azimuthal orientation dependencies of the scattered fluxes, projectile trajectory simulations using the MARLOWE (version 14c) code [5] were carried out. MARLOWE treats the interaction between the projectile and the surface in an *elastic* binary collision approximation. An exponential-sum screened Coulomb interaction potential was chosen with default parameters from [6], and the generally accepted missing-row model was used for the reconstructed Au(110) surface. The primary beam energy distribution was simulated in a series of MARLOWE tasks with different initial particle kinetic energies. Due to computing time constraints the angular acceptance was increased by a factor of 4 over the experimental acceptance angle of 2°. Almost 4×10^6 trajectories were generated for each target orientation investigated, of which $\sim$ (0.03 - 0.08)% were scattered into the detector cone. For each of the latter trajectories, the number of collisions, scattering angles, etc. were saved, permitting its subsequent reconstruction.

Fig. 2 shows selected results of the performed computer simulations. The simulations reproduce quite well the flux variations with target orientation seen experimentally and were used as the basis for analyzing the collision events contributing to the "BC" peaks. These events can be divided into three types: pure binary (Fig. 2b), "in-plane" (or close to "in-plane"), quasi-binary (Fig. 2c), and "out-of-plane" ("zig-zag" type motion) low multiplicity collision events which form rather broad structures in the vicinity of the "BC" peaks (Fig. 2a). In the present context, by "quasi-binary" is meant that class of double collisions, usually a combination of large angle ("hard") and small angle ("soft") scattering events which have nearly identical energy losses as the true "BC" events, and by "collisions" are meant those projectile-target atom encounters which satisfy the MARLOWE default criterion of having the impact parameter less than a maximum value ($\sim$1.6Å for Au), determined from the target lattice dimensions. In contrast to the first two collision types, the third considerably stretches the base of the "BC" peak, making stripping of the "BC" flux in the measured spectra rather difficult. In order to avoid such complications we avoided measurements at incidence angles in the vicinity of 30° (relative to the surface normal), and instead worked at either 10° or 50°. The simulations show that, at these two incidence angles, the "BC" peaks are built up only from first two types of events, irrespective of target azimuth orientation. Higher multiplicity events (i.e. number of collisions > 2) do not contribute to the "BC" peak, and form instead the smooth pedestal upon which the peak sits.

3 Selected results and discussions

Fig. 3 shows the target azimuth dependences of the total scattered "BC" fluxes (i.e. summed over all scattered charge states) for four different Ar charge states incident on gold at 10°. Two main features characterize the measured dependences. (1) The scattered fluxes look virtually identical for all incident charge states, i.e. have identical azimuth dependences within the experimental errors. (2) The measured total fluxes are in excellent agreement with backscattering yields based on projectile trajectory simulations (using neutral scattering potential - a fact which is remarkable in its own right!). This agreement provides the basis for using the computer simulated trajectories to identify the events making up the "BC" peaks for our geometry (Fig. 4). Such analysis, previously reported elsewhere[3], shows that single collisions in only the top three target layers contribute to the peaks. They are denoted **SI**, **SII**, and **SIII**, respectively. Target atoms not directly at the surface/vacuum interface were found to have negligible contribution, presumably resulting from shadowing effects due to the near normal incidence conditions employed. The "hard" "soft"

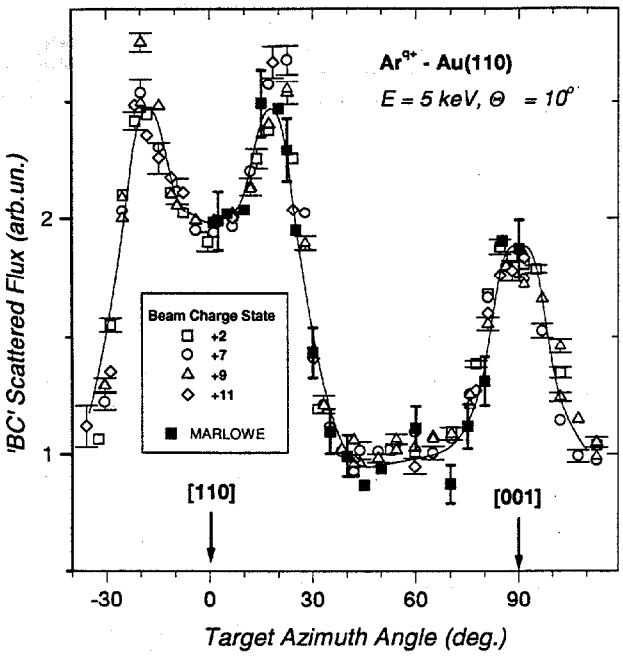

Figure 3: Azimuth dependences of the total (i.e., summed over all scattered charge states) "binary collision" fluxes for four 5 keV Ar beams in different charge states incident on Au(110) at 10°(open symbols). The simulation results are shown as the line with solid squares.

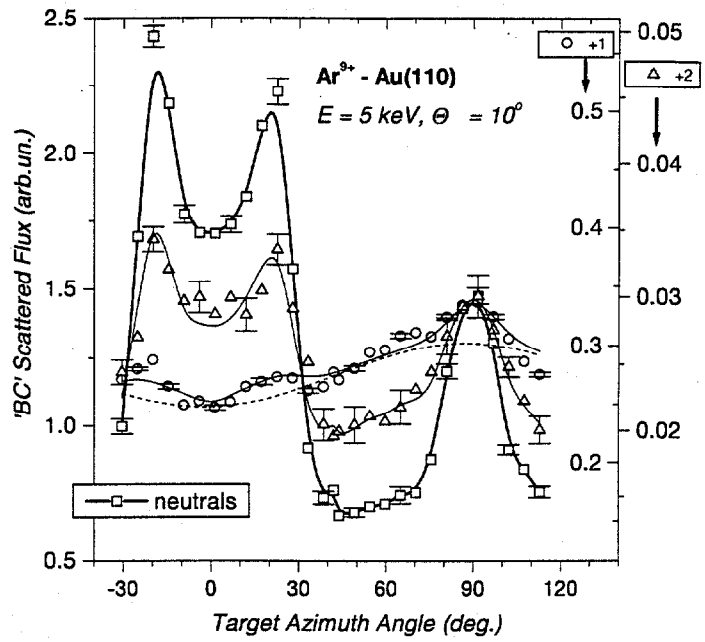

Figure 5: Azimuth dependences of scattered neutral and charged "binary collision" fluxes for 5 keV Ar^{9+} ions incident on Au(110) at 10°. The thick line is drawn through the neutral data points to guide an eye. The thin lines are fits discussed in the text.

quasi-binary collision events originate exclusively from the second and third target layers (denoted **DII** and **DIII**) and make contributions comparable to the **S** types at some azimuthal positions. It was shown that each collision type was characterized by a distinct and unique dependence on target azimuth. Further analysis based on differences in azimuthal dependences for neutral and charged scattered components (already noted above) led to the conclusion that during low energy large-angle projectile backscattering, only single binary collisions in the topmost target layer (**SI** type) resulted in incomplete neutralization, even for an incident projectile charge as high as 11+. Evidence was also found that reionization of neutrals at the second collision for quasi-binary events having the first "hard" collision in the second layer (**DII** type) also contributed to the charged scattered flux intensity.

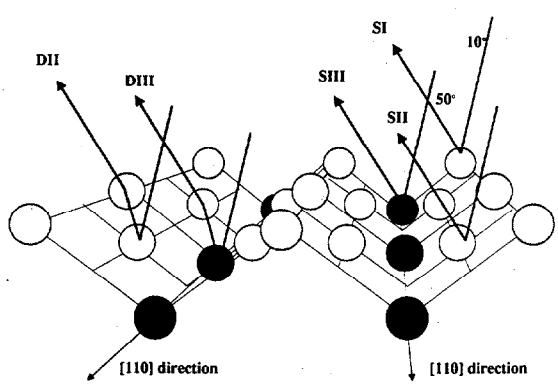

Figure 4: Schematic representation of the (2x1) reconstructed Au(110) surface and the 5 different collision types (see text for details).

Additional new information on the MCI-surface charge exchange interaction can be gained from more detailed considerations of the target orientation dependences for neutral and charged fluxes presented on Fig. 5 and Fig. 6. We first focus on the azimuthal dependence for the singly charged scattered projectiles presented in Fig. 5. The considerably more "flat" character (compared to the neutral scattered component) is a signature of the just mentioned binary collision scattering from the topmost target layer which is, as expected, independent of target azimuthal orientation. The 1+ component bears evidence of reionization in quasi-binary collisions in the presence of the main maximum at 90° and the two local maxima at approximately ±20°, which characterize **DII** type events [3]. But aside from these two features, the dependence exhibits a clearly visible *sinusoidal* type behavior (with amplitude of 10% from the mean value - much bigger than experimental error bars) which can not be ascribed to any of the remaining collision events. The thin line in Fig. 5 drawn through the singly charged data points represents a weighted combination of **SI** and **DII** curves added to the *sinusoidal* variation: $0.57\times\mathbf{SI}\times(1 + 0.1\times\sin\phi) + 0.04\times\mathbf{DII}$. It is worth noting that this sinusoidal feature is common to all the investigated beams incident on the target at 10°, but is not as prominent in our data taken at at normal incidence (possibly due to the relatively larger error bars (±8%) that the latter set of measurements had). It seems reasonable to assume that this is a manifestation of a collective charge exchange interaction of the target atoms with the projectile which the MARLOWE simulation does not take into account.

A simple estimation supports this hypothesis. Since, for the (2x1) Au(110) surface reconstruction, the surface electron density of states closely follows the corrugation of the surface structure [7] we can suppose that the interaction between projectile and the surface can be characterized by sum of terms proportional to $(1/R_i)^n$, where R_i is a distance between projectile and i^{th} atom in the surface, and n is a parameter. If we consider now a single scattering from a target atom on the ridge leading to a scattered 1+ ion (the dominant collision type for this scattered charge state), the total "charge exchange" between

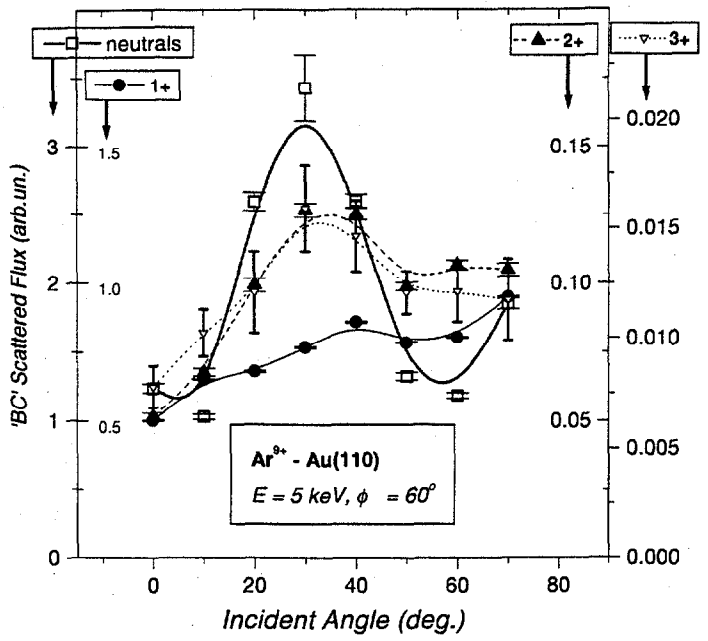

Figure 6: Incident angle dependences of scattered neutral and charged "binary collision" fluxes for Ar^{9+} ions incident on Au(110) at 60° target azimuthal orientation.

the projectile and the surface can be estimated by an integral from the sum over some interval (R_1, R_2) along the projectile trajectory. Considering R_1 and R_2 as parameters and taking into account only nearest neighbors of the target atom (two in the ridge and four in the second row) we found that "charge exchange" had a *cosine* dependence on azimuth angle with the maximum in the direction of the missing rows, in qualitative agreement with the experimental data, which show a minimum in the 1+ intensity along the [110] missing row direction).

Several other conclusions can be drawn from this exercise. First of all, to achieve noticeable variations with azimuth by the above functional form, the distances R_1 and R_2 have to be on the order of the target lattice dimension and n has to be > 4 (while interactions of longer range, i.e. smaller n, or at distances large or small compared to the lattice dimension will show a much weaker dependence, if any, on azimuth orientation). This observation supports a suggestion previously made in [3], namely that large distance electron capture on the receding trajectory plays an important role for projectile neutralization. It is interesting that this "charge exchange memory" survives a very close encounter with the target atom during the scattering into 120°. Detailed theoretical work is obviously needed to explore more quantitatively all the above mentioned features.

Finally, we turn to the azimuth dependence for the doubly charged scattered ions also presented in Fig. 5. Surprisingly, the latter dependence is not as "flat" as that of the singly charged scattered ions. It can in fact be reproduced very well by a weighted combination of neutral and singly charged scattered ion intensities with weights 0.009 and 0.05 respectively (see the second thin line in Fig. 5). As illustrated in Fig. 6, which shows flux dependences of neutral, 1+, 2+ and 3+ scattered projectiles as function on incidence angle with fixed azimuth, the 2+ scattered ions also show a "more like neutrals" dependence on incidence angle than the 1+ scattered ions. This means that the majority of doubly charged scattered ions do not represent survival from neutralization in the scattering, but rather formation due to different kinds of reionization processes occuring most likely on the outgoing part of projectile trajectories. As can be seen in Fig. 6, this is also true for triply charged ions (the azimuth dependence of triply charged ions was not shown in Fig. 5, because of the relatively larger statistical errors for this low intensity scattered charge state).

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