Path-Dependent Neutralization of Multiply Charged Ar Ions Incident on Au(110)

V. A. Morozov and F. W. Meyer
Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6372
(Received 8 June 2000)

Using a time-of-flight technique we have investigated the backscattering of 5 keV Ar^{2+} and Ar^{11+} projectiles from Au(110). A strong dependence on target azimuth is found for the neutral flux resulting from (quasi)binary projectile collisions with target atoms, while for the charged components only a weak dependence is seen. A deconvolution of the observed dependences based on trajectory simulations clearly shows site-specific neutralization differences between the various possible binary collisions occurring with surface atoms. Such differences must be properly accounted for in order to permit meaningful comparison with theory.

DOI: 10.1103/PhysRevLett.86.736 PACS numbers: 79.20.Rf, 34.50.Dy, 61.18.Bn, 79.20.Ap

The interaction of slow multiply charged ions (MCI) with a metal surface triggers a complicated cascade of inelastic charge exchange processes, which has been the subject of numerous studies [1]. MCI neutralization occurring during such interactions is presently thought to take place in three distinct phases: along the approach trajectory, during the close interaction with the surface, and on the way out.

For neutralization occurring on the approach trajectory, a classical over-the-barrier model [2] has been developed, which has proved to be very successful in treating many interesting phenomena of the MCI-surface interaction such as "hollow" atom formation and energy gain by image charge acceleration [3]. In this model, electron capture occurs at large above-surface distances into high lying projectile Rydberg states that are in resonance with the target Fermi level, and that subsequently relax by a complex autoionization cascade.

At the projectile approach velocities considered here, there is insufficient time for the hollow atom to relax prior to close interaction with the surface. Most of the captured electrons therefore remain in their initial loosely bound Rydberg levels, and are "peeled" from the projectile as these levels are shifted above the Fermi level in the exponentially increasing electron density of the electron selvedge seen by the approaching projectile. During the close interaction with the surface that follows, additional neutralization processes become active. These processes are very efficient in populating projectile inner shells directly (e.g., side-feeding processes [4]), and are thus much faster than the neutralization/relaxation processes occurring on the way in. The "freezing in" of the final projectile charge state occurs as the projectile recedes from the surface [5]. This phase affects mainly the relative intensities of scattered neutrals and 1+ ions, but can also include projectile Auger relaxation effects in the case of highly charged incident ions.

Of these three phases, the second presents by far the most challenges in terms of theoretical treatment. While the object of a number of recent investigations [6], many of the quantitative details of the processes active in this region remain poorly understood. One impediment to progress on the theoretical side is the absence of quantitative experimental data on MCI projectile neutralization for well-defined trajectories involving single binary collisions with surface atoms. Only recently have grazing MCI-surface interaction experiments been developed that, for example, permit coincidence studies of electron emission along selected classes of projectile trajectories [7] and for selected energy losses [8]. In this Letter we report the first such trajectory resolved data for highly charged ions in the case of large angle scattering (LAS). The LAS geometry has long been used in surface elemental and structural analysis [9] and is only now seeing increased interest as a tool for the study of inelastic MCI-surface interactions [10,11].

Our data show that there can be significant dependence of the final scattered projectile charge state on the precise trajectory taken. While path-dependent neutralization has been documented in the past [12,13] for singly charged incident ions, it has to date not received proper attention in the case of MCI-surface interactions. Remarkably, we find that, for both low and high charge state incident projectiles, the degree of neutralization varies with the specific site on the surface with which the "hard" LAS collision occurs, from essentially complete neutralization for interactions with the sidewalls or troughs of the reconstructed Au(110) surface [14], to significant ion survival for interaction with the ridges.

The measurements were carried out at the ORNL Multicharged Ion Research Facility with an apparatus that implements an ultrahigh vacuum (10^{-10} mbar) floating scattering chamber and time-of-flight (TOF) analyzer with floatable drift tube [11], permitting simultaneous measurements of energy distributions and charge fractions [15] 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 $(10 \times q)$ keV to a final energy close to 5 keV and

 0031-9007/01/86(4)/736(4)$15.00

were incident on the Au(110) surface at 10° relative to the surface normal.

Figure 1 shows scattered particle TOF spectra for Ar^{2+} and Ar^{11+} incident ions, and illustrates the separation of the scattered charge states achieved by use of the floatable drift tube. The spectra exhibit sharp peaks at positions expected for *elastic* binary collisions (BC) between projectile and target atoms. In the case of the neutrals the "BC" peak sits on top of a broad structure arising from multiple collision events.

Figure 2 shows the target azimuth dependences of the BC fluxes for both incident charge states. Four features of the measured dependences are noted. (1) The neutral scattered fluxes manifest strong variations with target azimuth (a factor of 3 and 5 for incident 2+ and 11+, respectively). (2) The charged scattered fluxes show much weaker variations (on the order of 25% and 40%, respectively). (3) The total scattered fluxes (i.e., summed over all scattered charge states) have identical azimuth dependences within the experimental errors. (4) The measured total fluxes are in remarkable agreement with LAS yields based on projectile trajectory simulations using the MARLOWE (version 14c) code [16].

MARLOWE treats the interaction between the projectile and the surface in an *elastic* binary collision approximation. In what follows, "collision" means a projectile-target atom encounter which satisfies the MARLOWE default criterion of having the impact parameter b less than a maximum value ($b_{max} \sim 1.6$ Å for Au) determined from the target lattice dimensions. An exponential-sum screened Coulomb interaction potential was chosen with default parameters from [17], and the generally accepted missing-row model was used for the reconstructed Au(110) surface. Because of 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 azimuth orientation investigated, of which ~0.04% were scattered into the detector cone. For each such trajectory, the number of collisions, scattering angles, etc., were saved to permit its subsequent reconstruction.

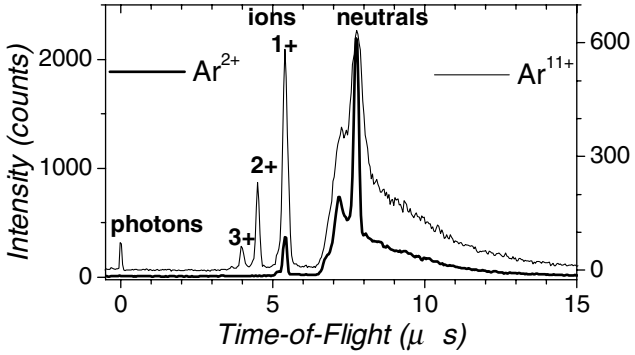

FIG. 1. Time-of-flight spectra for 5 keV Ar^{2+} and Ar^{11+} ions incident on Au(110) at 10° and 2° off the [001] direction, and scattered by 120°.

The simulation reveals that the "BC" peak is in fact built up from two types of events — pure single (SC) and "quasibinary" double (DC) collisions [18]. "Quasibinary" means that class of double collisions, usually a combination of large angle ("hard") and small angle ("soft") scattering events, resulting in final energies encompassed in the observed main energy loss peak [19]. Higher multiplicity events (i.e., number of collisions >2) do not contribute to the "BC" peak, and form instead the pedestal upon which the peak sits.

An analysis of the target layers, in which the events making up the "BC" peak occur for our geometry, shows that single collisions in only the top three target layers contribute. They are denoted SI, SII, and SIII, respectively.

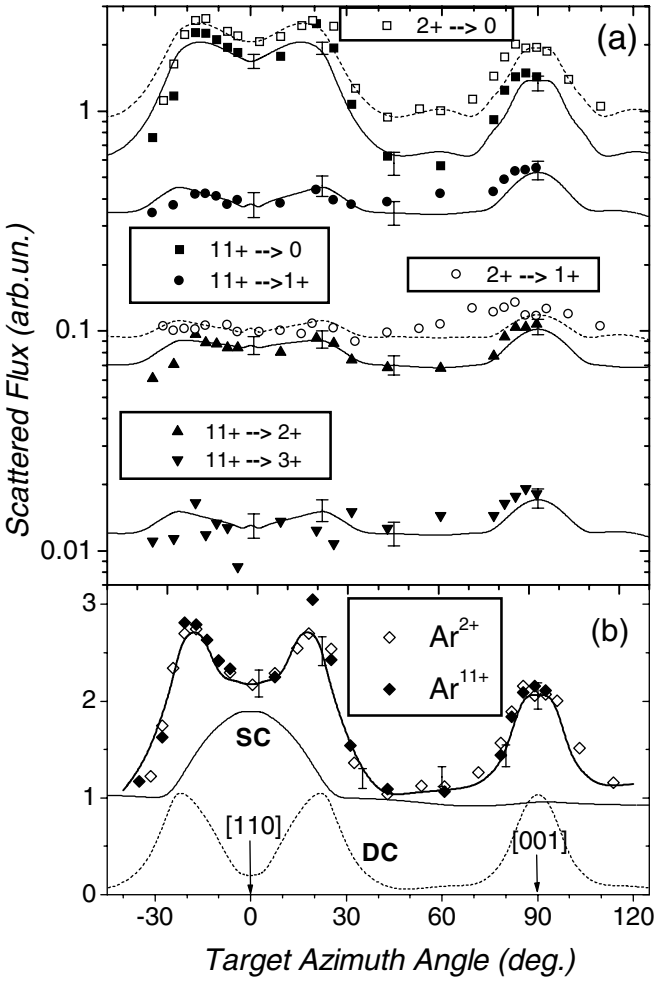

FIG. 2. (a) Azimuth dependences of scattered neutral and charged "binary collision" fluxes for Ar^{2+} (open symbols) and Ar^{11+} (solid symbols) ions incident on Au(110) at 10°. Lines are fits to the data (see text for details). Experimental errors bars are smaller than the size of the symbols, except for Ar^{3+} scattered ions, where they are of the order of the typical simulation error bars shown. (b) Azimuth dependences of the total (see text) "binary collision" fluxes for both incident ions. Also shown are the simulated azimuth dependences for single (SC = SI + SII + SIII) and quasibinary double (DC = DII + DIII) collisions with typical error bars. Their sum is shown as the thick line.

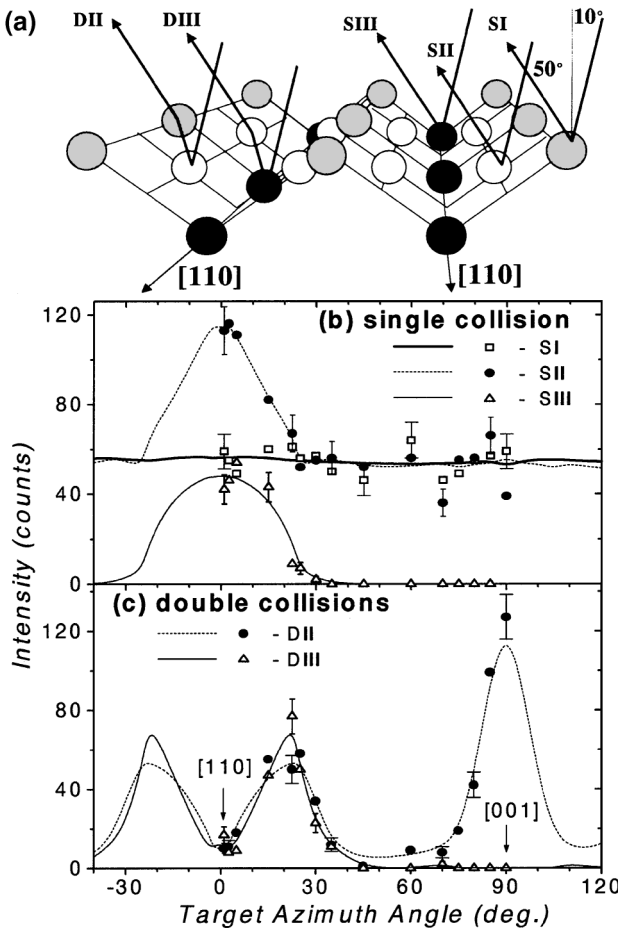

FIG. 3. (a) Schematic representation of the (2×1) reconstructed Au(110) surface and the five different collision types. Simulated azimuth dependences for single (b) and quasibinary double (c) collision fluxes. The lines are smoothed curves through the simulated data points, extended over the full measured range.

Target atoms not directly at the surface/vacuum interface make a negligible contribution to the "BC" peak, presumably resulting from shadowing effects [9] due to our near normal incidence conditions. The hard-soft quasibinary collisions originate exclusively from the second and third target layers and will be denoted DII and DIII, the Roman numeral designating the layer in which the hard collision occurs. Figure 3 shows these five collision types as well as their individual target azimuth dependences. A striking feature of the simulation results is that each collision type is characterized by a distinct and unique dependence on target azimuth.

To assess the relative contributions of the different collision classes, we fit the measured fluxes of the different scattered charged states by weighted linear combinations of the five identified collision types (Table I). Justification for this approach is the excellent agreement [see Fig. 2(b)] between the azimuth variations of the total experimental "BC" fluxes and the calculated LAS intensities (i.e., the sum of SI, SII, SIII, DII, and DIII). The agreement between the simulation results, based on a neutral-neutral scattering potential on the one hand, and the experiment, using MCI projectiles on the other, is remarkable in its own right, and will be commented on more fully elsewhere [20]. Within the uncertainty of the present simulation ($\pm 10\%$), we find that, for both incident charge states, only the SI and DII classes are required to fit the azimuth dependences of the observed charged scattered fluxes, while all five types of collisions are required to fit the observed neutral fluxes. The fitting results are summarized graphically in Fig. 2(a) and show good agreement with the experimental data.

TABLE I. Weights of the different collision classes shown in Fig. 3 used to fit the experimental data in Fig. 2(a). Weights for classes SII, SIII, and DIII are 1 and 0 for the neutral and charged scattered ion fluxes, respectively.

Outgoing charge state	Incoming Ar^{2+}		Incoming Ar^{11+}	
	SI	DII	SI	DII
0	0.80	0.98	0.10	0.78
+1	0.20	0.02	0.75	0.20
+2	⋯	⋯	0.125	0.02
+3	⋯	⋯	0.025	0.005

The fact that our simulations of the scattered ion azimuth dependences do not require contributions from the SII trajectory class, while DII trajectories are definitely needed, is interesting and not trivial. The simulations show that, for both trajectory classes, the hard collisions are practically identical (scattering angles 120° and ~118°, respectively). The only difference is in the presence of a soft second collision in the latter class, with scattering angles up to 20°, leading to the "zigzag"-type trajectories that have already been noted elsewhere [13]. We ascribe the ion fraction for this trajectory class to reionization at the second collision. This issue together with additional evidence pointing to this conclusion will be discussed in greater detail elsewhere [20].

On the basis of the information displayed in Table I, it appears that, during low energy projectile LAS, only binary collisions in the topmost target layer result in incomplete neutralization, even for an incident projectile charge as high as 11+. The fact that complete neutralization occurs in single collision encounters in already the second and third layers is remarkable in that, for the (2×1) Au(110) surface reconstruction, the surface corrugation extends to the third layer, i.e., second and third layer target atoms lie exposed at the solid vacuum interface. Since the electron density of states, while exponentially decreasing with distance above the surface, still follows the corrugation of the surface structure at distances as large as 5 Å above the surface [21], it does not appear to offer enhanced opportunity for projectile neutralization on the way in for those trajectories impacting the corrugation troughs. A more likely scenario may involve large distance (i.e., $b > b_{\max}$) electron capture from lattice atoms. For SII and SIII collisions leading to the 120° scattering investigated here, the scattered projectile will always have the opportunity for such an encounter in the first and the first

and second layers, respectively. It will also spend more time close to the corrugated electron selvedge than for SI collisions. Thus, while such encounters will not satisfy MARLOWE criterion of a collision, they may well provide opportunity for the quasiresonant charge transfer from target atoms or overbarrier transitions from the Fermi electron sea required to complete the projectile neutralization. Work is obviously needed to explore theoretically this proposed scenario.

A central conclusion of this paper is that an analysis of experimental charge fraction data such as that presented above (or the equivalent thereof) is required before meaningful comparison with dynamical neutralization models of binary collisions becomes possible. Without such analysis, there is no guarantee that the observed scattered charge states do not originate from different collision types, leading to potentially significant errors. For example, in the case of the Ar^{11+} incident ions, the mean charge (often used as a base for comparison to neutralization models [22]) for the SI collisions calculated from Table I equals 1.1. This value is at least a factor of 2 larger than the mean charge, deduced directly from the relative intensities of the charge dispersed "BC" peaks of Fig. 1, which varies from 0.23 to 0.56 depending on the target azimuth. More subtly, even in the azimuth range at about 60°, where the contribution from quasibinary double collisions is negligible [see Fig. 2(b)], this approach underestimates the correct mean charge for the SI class. This is because the total "BC" flux in this range consists of two equally probable [see Fig. 3(a)] components, SI and SII, only the first of which contributes, according to our fit results, to the scattered ion flux.

We are indebted to S. H. Overbury for setting up the MARLOWE simulations and to D. M. Zehner for the loan of the Au crystal. This research was sponsored in part by the Office of Fusion Energy Sciences, and by the Office of Basic Energy Sciences of the U.S. Department of Energy under Contract No. DE-AC05-00OR22725 with UT-Batelle, LLC. V. A. M. was appointed through the ORNL Postdoctoral Research Associates Program administered jointly by Oak Ridge Associated Universities and ORNL.

[1] A. Arnau *et al.,* Surf. Sci. Rep. **27**, 113 (1997), and references therein.
[2] J. Burgdörfer *et al.,* Phys. Rev. A **44**, 567 (1991).
[3] H. P. Winter and F. Aumayr, J. Phys. B **32**, R39 (1999), and references therein.
[4] L. Folkerts and R. Morgenstern, Europhys. Lett. **13**, 377 (1990).
[5] See, e.g., H. Winter, Comments At. Mol. Phys. **26**, 287 (1991).
[6] J. Thomaschewski *et al.,* Phys. Rev. A **57**, 3665 (1998); J. Burgdörfer *et al.,* Nucl. Instrum. Methods Phys. Res., Sect. B **98**, 415 (1995).
[7] C. Lemell *et al.,* Phys. Rev. Lett. **81**, 1965 (1998).
[8] P. Roncin *et al.,* Phys. Rev. Lett. **83**, 864 (1999).
[9] H. Niehus *et al.,* Surf. Sci. Rep. **17**, 213 (1993); J. W. Rabalais, Surf. Sci. **299/300**, 219 (1994); M. Aono and R. Souda, Jpn. J. Appl. Phys. **24**, 1249 (1985); S. H. Overbury, in *The Handbook of Surface Imaging and Visualization,* edited by A. T. Hubbard (CRC Press, New York, 1995), pp. 375–383.
[10] W. Huang *et al.,* Phys. Scripta **T80**, 228 (1999); R. Schuch *et al.,* Nucl. Instrum. Methods Phys. Res., Sect. B **157**, 309 (1999).
[11] F. W. Meyer and V. A. Morozov, Nucl. Instrum. Methods Phys. Res., Sect. B **157**, 297 (1999).
[12] E. Taglauer *et al.,* Phys. Rev. Lett. **45**, 740 (1980); R. M. Charatan *et al.,* Surf. Sci. **302**, 205 (1994).
[13] C. A. Keller *et al.,* Phys. Rev. Lett. **75**, 1654 (1995).
[14] W. Moritz and D. Wolf, Surf. Sci. **88**, L29 (1979); R. Koch *et al.,* Appl. Phys. A **55**, 417 (1992).
[15] V. A. Morozov and F. W. Meyer, Rev. Sci. Instrum. **70**, 4515 (1999).
[16] M. T. Robinson, Radiat. Eff. Defects Solids **130**, 3 (1994); Phys. Rev. B **40**, 10 717 (1989).
[17] J. F. Ziegler, J. P. Biersack, and U. Littmark, *The Stopping and Range of Ions in Solids* (Pergamon, New York, 1985), Vol. 1, p. 48.
[18] V. A. Morozov and F. W. Meyer, Phys. Scr. (to be published) (HCI 2000 Conference Proceedings).
[19] For example, for scattering along the [001] direction, the first and second (in brackets) moments of the angular and impact parameter distributions for quasibinary events are 117.8° (2.5°) and 0.133 Å (0.007 Å) for the first collision and 6.8° (5.5°) and 1.14 Å (0.24 Å) for the second.
[20] V. A. Morozov and F. W. Meyer (to be published). It is not yet clear whether this implies that complete neutralization has occurred prior to reaching the turning point, or whether in LAS, where there is significant interpenetration of the respective electron clouds of the two collision partners, the overall scattering is not particularly sensitive to the charge state of the partially stripped MCI.
[21] J. Tersoff and D. R. Hamann, Phys. Rev. Lett. **50**, 1998 (1983).
[22] W. Huang *et al.,* Phys. Rev. A **56**, 3777 (1997); W. Huang *et al.,* Nucl. Instrum. Methods Phys. Res., Sect. B **135**, 336 (1998).