Beam Charge Dependence of Ion-Surface Scattering

B. Hird, P. Gauthier, J. Bulicz, and R. A. Armstrong
Ottawa-Carleton Institute for Physics, Ottawa University Campus, Ottawa, Ontario, Canada K1N 6N5
(Received 1 August 1991)

Evidence is presented that ions retain no information about the incident charge state after they have been scattered from a clean surface. Measurements of oxygen-silicon surface scattering, under conditions which favor binary collisions with individual surface atoms, were found to give the same ratio of negative to positive scattered ions under identical scattering conditions for both an incident O^- ion beam and an incident O^+ ion beam.

PACS numbers: 79.20.Nc

Ion-surface scattering at a few hundred to a few keV energy has been extensively used for surface characterization under circumstances where most of the detected ions have undergone a single elastic collision with an atom at the surface. The charge may change during the scattering, and extensive experimental and theoretical work has been done on the scattered charge fractions when positive ions are scattered from clean surfaces. The theory predicts quite well the observed dependence of the charge fractions on the energy states of the electron in the ion and in the surface. Thus the charge-changing information contributes to the general understanding of the model for the ion-surface atom collision process. For recent reviews see Los and Geerlings [1] and Brako and Newns [2].

We report a direct experimental test of the loss of memory of the initial charge state during ion-surface scattering. Identical measurements were repeated except that first a positive-ion beam and then a negative-ion beam of the same element at the same energy were used. If the memory of this initial charge state is lost then all the outgoing properties of the scattering process should be identical. A particularly sensitive test is the ratio of negative to positive charges on the scattered ions.

A property of the scattering models is that the collision process is effectively divided into three parts [3]: (a) the initial interaction before the ion reaches the surface; (b) a close encounter, or hard collision, in which the ion trajectory changes direction; and (c) an interaction between the surface and the projectile as it leaves. Experimentally the intermediate charge states of the particle are not directly accessible.

Since the early work by Hagstrum [4], who examined the initial neutralization processes in detail, it has been generally assumed that the first two parts of the interaction are not important in determining the final charge state. More recent formulations of the theory [5] give very small order-of-magnitude estimates for the survival probability of the initial charge state through the first two parts of the collision. For example, the observed neutral atoms in the scattering of a sodium atom beam from a tungsten surface are interpreted theoretically as ionized atoms which have been reneutralized on the way out [6,7].

In spite of being in good agreement with the model, none of the experiments distinguishes between neutralization on the way in and neutralization on the way out. Perhaps the best test of the memory-loss assumption of the model was made by Bronckers and de Wit [8] who compared the angular distributions of Ne^+ and O^- (from fragmented incident H_2O^+ ions) scattered from Cu(110). They found that both Ne^+ ions and O^- ions show strong shadowing and blocking cone effects from first-layer atoms consistent with a close encounter which is independent of the ingoing charge state (Molière scattering). They concluded that the charge state of the ion immediately before the collision is unimportant, and that the final charge state is determined during the outgoing part of the trajectory. A difficulty in interpretating the Bronckers and de Wit data is the low yield of Ne^+ scattering, a consequence of the large rare-gas ionization potential, compared to the negative-ion yield of oxygen.

The only previously reported measurements which compare otherwise identical positive- and negative-ion beam-surface scattering are those of van Wunnik, Geerlings, Granneman, and Los [9] for hydrogen scattering from tungsten covered with a monolayer of cesium. They initially found some differences in the negative hydrogen fractions in proton scattering from this surface which they explained by incomplete memory loss for proton-cesium scattering and complete memory loss for proton-tungsten scattering. In an attempt to confirm this postulate they then compared the negative-ion yields using positive and negative hydrogen beams under conditions which optimized the proportion of ions scattered from the cesium monolayer. In contradiction to their postulate they found no difference, within the accuracy of their measurements, between the negative-ion yields for the differently charged beams.

The experimental conditions reported here were chosen so that most of the scattered ions were the result of scattering by a single surface atom. A clean surface was chosen to provide repeatability, and an amorphized surface was chosen to minimize surface structure channeling and blocking effects, even though these are unlikely to be charge dependent [8].

Positive and negative oxygen-ion beams of energies between 6.82 and 19.9 keV were scattered from a clean sil-

icon surface at an incident grazing angle of 5° (perpendicular energy component 52 to 144 eV). The ion beams were produced in an rf-type ion source, accelerated, mass selected by a 30° magnetic deflection, and then passed through a 1-m-long differential section at 1×10^{-8} Torr into a chamber which was kept at about 2.5×10^{-10} Torr during the measurements. The entrance and exit slits of the differential section, as well as restricting gas flow, determined the geometry of the beam, so that it had a width of 0.25 mm, and a 2-mrad angular spread at the silicon target.

To change from a positive- to a negative-ion beam, all the electric and magnetic fields throughout the beam transport system were reversed except in the ion source itself. The negative ions were then produced from positive ions inside the exit canal of the ion source by electron capture collisions with the residual gas there. Somewhat different operating conditions were required to optimize the negative-ion beam to a maximum intensity of a few percent of the available positive-ion beam. Beam currents which were used on the target were typically 10^{-11} A.

A cylindrical electrostatic analyzer, with a deflection angle of $\pi/\sqrt{2}$, focused the ions scattered from the target onto a slit, behind which was placed a channel electron multiplier which counted individual ions. The collection angle of ions from the target was 2° and the experimental energy resolution, as measured with the direct beam, was 3%. The analyzer was mounted on a track inside the UHV chamber to allow measurements of the scattered ions at different angles. For these data an angle of 25° to the surface was used.

All the scattered-ion count rates were normalized to the beam intensity by using a fixed second channel electron multiplier which counted ions that had been backscattered from the target. The cones of both counters were biased sufficiently negative to reject all secondary electrons. At these ion energies the counting efficiency of channel electron multipliers is close to 100%, for all the charge states of the particles scattered from the target, so that a direct comparison of the negative and positive beam intensities was obtained.

The target was cleaned by 2-keV argon-ion sputtering ($2\ \mu A/cm^2$ for 15 min). It was then heated sufficiently to outgas the surface argon, but not to anneal the crystal structure. A reflection high-energy electron diffraction pattern showed no diffraction spots. Previously it had been verified that, when similar sputtering was followed by annealing to about 1000°C for 10 min, a clear Si(100)-(2×1) pattern was obtained. There was a complete absence of any detectable peak in the energy spectrum of the scattered ions at the energy corresponding to the scattering from carbon when measured at angles where this peak did not overlap other peaks.

Figure 1 shows energy spectra with 6.82 keV and with 16 keV beam energies. The scattered energy spectra of the negative ions differed from the positive ions in that

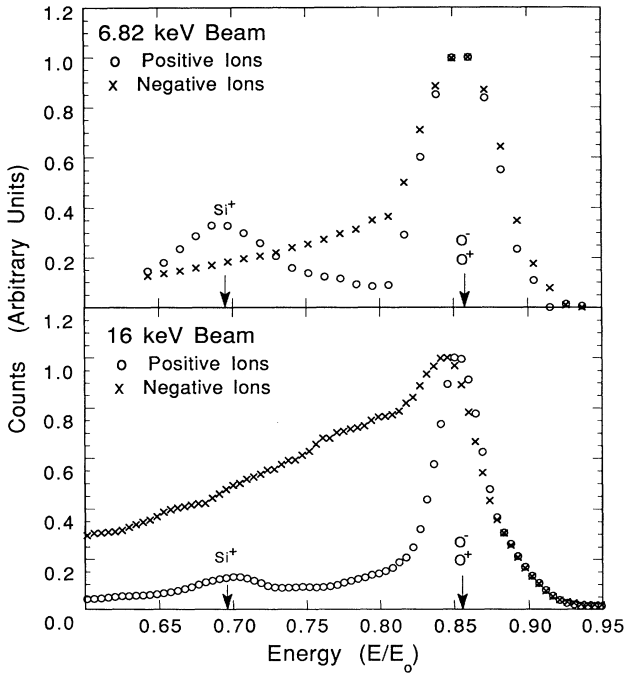

FIG. 1. The energy spectra of ions scattered at 25° to the silicon surface for 5° incident ion beams. The scattered energies are given as the ratio to the incident beam energy. The arrows show the predictions for an elastic collision with a single silicon atom on the surface.

there was a continuum below the surface peak, which increased with increasing beam energy. This confirms the observation of Bronckers and de Wit [8] that multiple subsurface scattering which involves energy loss by inelastic processes is more evident in the scattered negative ions than in the scattered positive ions. Another difference was that the positive-ion spectra showed a surface recoil peak in which an oxygen ion was scattered downwards into the surface and the recoiling silicon ion was emitted from the surface. The absence of this recoil peak in the negative-ion spectra suggests that Si^- ions are not formed when silicon atoms leave the surface. All the peaks in the spectra appeared at the calculated energies, confirming that they were mainly due to single-atom surface scattering. The exception was the negative-ion peak which was displaced slightly at high beam energies because it stood on the edge of the continuum of subsurface scattered ions. The shapes of the spectra taken with positive beams and with negative beams were indistinguishable from each other, and from the spectrum of ions which had been scattering from a single-crystal Si(100) surface at azimuthal angles away from a channeling direction.

The relative intensities of the surface scattered ions were estimated from the peak areas. In the negative-ion spectrum there was some difficulty in separating out the inelastic continuum which extended under the surface scattered peak. Inelastic processes are less likely to retain

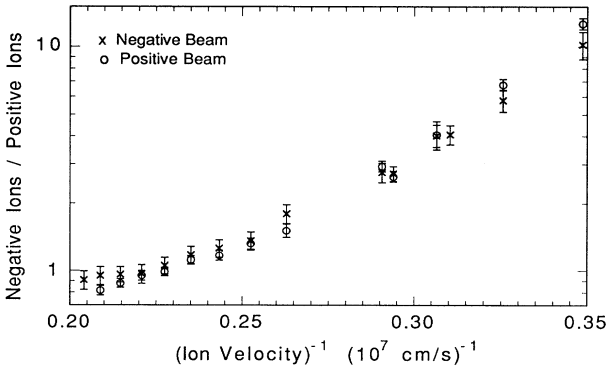

FIG. 2. The ratio of negative ions to positive ions which are emitted at 25° to the surface when ions are incident on the surface at 5°. The velocity range corresponds to ion-beam energies between 19.9 and 6.82 keV.

information about the initial charge because they involve multiple collisions. The relative intensities of the surface scattering were therefore estimated by fitting the maxima and the higher sides of the surface peaks.

The ratio of the intensity of the surface scattered positive ions to negative ions is shown in Fig. 2 as a function of the inverse of the ion velocity, which is proportional to the interaction time of the ion with the surface. Each point has an estimated accuracy between 5% and 10%, mainly from counting statistics. The range of beam energies was limited at the low end by the weak positive-ion intensity, and at the high end by the increased difficulty of estimating the negative surface scattered peak above the inelastic continuum. Including all the data, the average of the ratio of the results with a negative beam to those with a positive beam at the same energy comes to 1.021 ± 0.03. Within statistical error this is unity, so that the whole body of data provides strong evidence that all memory of the initial charge state is lost for keV oxygen scattering from silicon.

Many processes determine the ratio of positive to negative scattered ions, but nearly all of them act equally on the positive and negative incident beam. If memory loss is complete, it is possible to understand our observed variation of the ratio with ion velocity by considering charge changing only during the outgoing part of the trajectory. Hagstrum [4] assumed an exponential form $R(s) = A\exp(-as)$ for the neutralization transition probability. Here A and a are constants, and s is the perpendicular distance from the surface. This relation simulates the decreasing interaction as the ion recedes from the surface. The probability that an ion, after a close encounter, survives to emerge from the surface without a change of charge is then given by

$$P(v_0) = \exp\left[\frac{A}{av_0}[\exp(-as) - 1]\right],$$

where v_0 is the perpendicular component of the outgoing velocity [3]. Assuming that the positive and negative charge neutralization rates are independent of each other, then the charge ratio outside the surface becomes

$$\frac{\text{negative ions}}{\text{positive ions}} = \exp\left[\frac{1}{v_0}\left[\frac{A_+}{a_+} - \frac{A_-}{a_-}\right]\right].$$

The observed ratio should therefore increase or decrease exponentially with $(v_0)^{-1}$, depending on the sign of the coefficient. Figure 2 shows that the data follow roughly an exponential increase. The neutralization rate is therefore higher for positive ions than for negative ions, as might be expected from the lack of subsurface positive-ion scattering. The data at our higher inverse velocities give a value for the exponential coefficient $(A_+/a_+ - A_-/a_-)$ of 4×10^8 cm/s. The departure from a simple exponential at our lowest inverse velocities may be due to the increased penetration of the ions below the surface, with a consequent decrease in the relative number of positive ions which emerge.

[1] J. Los and J. J. C. Geerlings, Phys. Rep. **190**, 133 (1990).
[2] R. Brako and D. M. Newns, Rep. Prog. Phys. **52**, 655 (1989).
[3] P. Hochmann, M. A. Miller, and J. W. Rabalais, J. Phys. Chem. **89**, 2751 (1985); J. W. Rabalais, J-N. Chen, R. Kumar, and M. Narayana, J. Chem. Phys. **83**, 6489 (1985).
[4] H. D. Hagstrum, Phys. Rev. **96**, 336 (1954); **122**, 83 (1961); **150**, 495 (1966).
[5] P. W. Anderson, Phys. Rev. **124**, 41 (1961); D. M. Newns, Phys. Rev. **178**, 1123 (1969).
[6] R. Brako and D. M. Newns, Surf. Sci. **108**, 253 (1981).
[7] E. G. Overbosch, B. Rasser, A. D. Tenner, and J. Los, Surf. Sci. **92**, 310 (1980).
[8] R. P. N. Bronckers and A. G. J. de Wit, Surf. Sci. **104**, 384 (1981); **112**, 111 (1981).
[9] J. N. M. van Wunnik, J. J. C. Geerlings, E. H. A. Granneman, and J. Los, Surf. Sci. **131**, 17 (1983).