Energy- and angle-dependent trends in the trapping probability of O\(^+\) incident on Cu(001)

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Abstract

We have measured the trends in the trapping probability of 5–600 eV O\(^+\) ions incident on Cu(001) at 45° and along the sample normal. The trapping probabilities, measured using Auger Electron Spectroscopy (AES), depend strongly on the incident ion energy, but not on the incident angle. Significant differences are observed between these trends and those previously measured for Na\(^+\) incident on Cu(001) over a similar range of energies and angles, particularly for the 45° angle of incidence. We conclude that these observations reflect differences in the ion-surface scattering potentials. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

Near-surface trapping processes in hyperthermal energy ion–surface collisions are central to thin film growth and surface processing techniques that employ hyperthermal energy particles (e.g., sputter, pulsed laser, and ion-assisted growth techniques, and reactive ion etching) [1]. It is known, for example, that growth modes and film properties can be modified by the use of energetic particles [2]. These techniques often involve collisions between surface atoms and various incident ion species over a range of incident energies and angles. The use of energetic beams with control of the incident species, energy and scattering geometry can greatly assist in understanding the fundamental trapping mechanisms involved.

When hyperthermal energy ions are incident on a surface, they may scatter from the surface, become trapped above the top layer of surface atoms or become implanted below the surface atoms. The outcome depends on the incident ion energy and scattering geometry, as well as on the incident ion species, surface species and surface structure. These factors also determine the scattering potential, which in turn plays an important role in determining the trapping probabilities. For instance, an ion has a certain probability of becoming trapped above the top-layer of surface atoms if it loses enough energy during inelastic
collisions with surface atoms that it cannot escape from the attractive potential well, which is typically a couple of eV deep. We refer to this as the surface trapping probability, $P_S$, which distinguishes it from the combined probability of trapping either on the surface or subsurface, which we refer to as the trapping probability, $P_T$. Simple energy-transfer arguments predict the following trends in $P_S$ and $P_T$. At low energies, a few eV, $P_S$ is high since ions incident with such low energies easily lose sufficient energy during the ion-surface collision that they cannot escape the attractive potential well. As the incident ion energy increases, but is still below the energy required to penetrate the surface, the scattered ions have more energy to escape the attractive well, and $P_S$ decreases. As the incident ion energy continues to increase, $P_T$ is expected to rise due to implantation.

We have measured $P_T$, using Auger electron spectroscopy (AES), for 5–600 eV O$^+$ ions incident on Cu(001), along the ⟨100⟩ azimuth, at normal and 45° incident angles. The main goal of this study was to understand the importance of incident energy and scattering geometry in determining $P_T$. We have found that the measured trapping probabilities depend very sensitively on the incident ion energy, but not on the scattering geometry. Although there have been previous investigations of O trapping on metal surfaces [3,4], none have looked at the dependence of trapping probabilities on the incident angle.

In this paper we will contrast our measurements to those obtained for Na$^+$ incident on Cu(001) [5,6]. By comparing our results to those obtained for the better understood Na–Cu system, we can identify factors that are important in determining the trends in the trapping probability. Furthermore, since Na and O are close in mass, we might expect that the trapping probabilities, based on the simple energy transfer arguments presented above, should follow similar trends. These simple arguments, however, do not correctly predict the trapping probabilities for Na ions incident at 45°. In fact, trajectory analysis of the Na–Cu system has revealed that the trajectory types that lead to surface trapping can depend very sensitively on the apparent surface corrugation seen by the incident ions. We will conclude by summarizing the results of a preliminary calculation that indicates that the apparent surface corrugation seen by the incident O ions is larger than that seen by the incident Na ions, suggesting an explanation for the differences observed between the O and Na trapping probabilities at 45° incidence.

2. Experimental techniques

These experiments were performed in an ultra high vacuum (UHV) system that has been described in detail elsewhere [7–10]. Only the features relevant to the present work are summarized here. The UHV chamber is equipped with a beamline for producing well-collimated, mono-energetic, low and hyperthermal energy ion beams in the energy range from 5 eV to 10 keV. The O$^+$ ions were extracted from a Colutron ion source [11] using a source gas mixture of 15% O$_2$–85% Ne, enriched with pure O$_2$ when necessary to increase the O$^+$ ion yield. The Cu(001) single crystal was prepared by standard sputter and anneal cycles. Surface cleanliness and long range order were monitored using AES and low energy electron diffraction (LEED), respectively. The base pressure was $3 \times 10^{-11}$ Torr and operating pressures were below $2 \times 10^{-10}$ Torr.

The beam was tuned into the 1 mm entrance aperture of a Faraday cup mounted on a manipulator directly below the Cu(001) sample. Typical currents in the Faraday cup were ~0.5 nA. A cross-sectional profile of the incident beam was measured for each incident energy by sampling the current in the Faraday cup at many positions over a uniform grid. Typical beams were roughly Gaussian in shape, with 1 mm half widths.

A trapping measurement was performed by exposing the sample to the incident O beam for a well defined time, $t$. The oxygen coverage on the sample, $\Theta(r,t)$, was measured using AES, and had a spatial distribution that reflected the shape of the incident ion beam and the incident angle. We assume that we are in the low coverage limit where $\Theta(r,t)$ is linear with the ion beam dose and trapping probability. As will be seen below, this
assumption is justified by our data. The coverage is then given by
\[
\Theta(r, t) = P_T(E, \theta_i) j_n(r)t, \tag{1}
\]
where \(E\) and \(\theta_i\) are the incident beam energy and angle (measured relative to the surface normal), respectively, \(r\) is the position on the sample measured from the center of the coverage on the sample, \(P_T(E, \theta_i)\) is the trapping probability, and \(j_n(r) = j \cdot \hat{n}\) is the normal component of the incident ion current density. The dose of O on the sample is given by \(j_n(r)t\). The current density was obtained from the measured beam current using a numerical deconvolution procedure with Wiener optimized filtering to remove the effect of the finite size of the Faraday cup aperture [12,13].

To simplify the experimental procedure and minimize the amount of time the sample was exposed to the incident Auger electron beam, \(P_T\) was only evaluated at the center of the O spot profile on the sample, at \(r = 0\). From Eq. (1),
\[
P_T(E, \theta_i) \frac{\Theta(0, t)}{j_n(0)t} = \frac{\Theta(0, t)}{j(0)\cos(\theta_i)} t, \tag{2}
\]
where \(j(0)\) is the current density at the center of the incident ion beam. The center of the oxygen coverage on sample was found by performing vertical and horizontal Auger line scans across the sample, with the Auger tuned to the O (503 eV) peak position. Once the center of the oxygen profile on the sample had been located, the coverage at this location was determined by scanning the Auger, in differential mode, across the O (503 eV) and Cu (920 eV) peak positions and measuring the ratio, \(R_{O/Cu}\), of the peak-to-peak heights of the Auger O (503 eV) and Cu (920 eV) signals. Once \(R_{O/Cu}\) had been evaluated at the center of the spot profile on the sample, a full Auger scan from 85 to 920 eV was performed to verify that there was no contamination from other sources, paying particular attention to the intensity at the C (272 eV) Auger peak position. The full Auger scan was also used to obtain the correction to \(R_{O/Cu}\) from the background of secondary electrons [14]. A second full Auger scan was performed far from the center of the O spot profile to ensure that there was no measurable level of trapping from background O.

To convert \(R_{O/Cu}\) to an absolute coverage \(\Theta(0, t)\), in monolayers (ML), it was necessary to calibrate the Auger measurement. This was done by exploiting the fact that upon exposure of the sample to thermal O\(_2\) the saturation coverage, \(\Theta_{sat}\), is 0.5 ML, corresponding to a \((2 \times 2\sqrt{2})R45\) superstructure observable using LEED [15]. \(\Theta_{sat}\) corresponds to a measured peak-to-peak ratio of \(R_{O/Cu} = 0.19\). The coverage at the center of the spot profile after each beam deposition is then given by
\[
\Theta(0, t) = \left(\frac{\Theta_{sat}}{R_{sat}}\right) R_{O/Cu}(0, t). \tag{3}
\]

From Eq. (2), the trapping probability becomes
\[
P_T(E, \theta_i) = \left(\frac{\Theta_{sat}}{R_{sat}}\right) \left(\frac{R_{O/Cu}(0, t)}{j_n(0)t}\right). \tag{4}
\]

We evaluated \(P_T\) for \(\theta_i = 0^\circ\) and \(\theta_i = 45^\circ\) for which \(P_T\) is given by
\[
P_T(E, 0) = \left(\frac{\Theta_{sat}}{R_{sat}}\right) \left(\frac{R_{O/Cu}(0, t)}{j(0)t}\right),
\]
\[
P_T(E, 45) = \left(\frac{\Theta_{sat}}{R_{sat}}\right) \left(\frac{R_{O/Cu}(0, t)}{j(0)t\cos(45^\circ)}\right). \tag{5}
\]

For these two incident angles, and for a range of incident ion energies between 5 and 600 eV, \(R_{O/Cu}(0, t)\) was measured for five different beam doses. A linear least-squares fit determined \(R_{O/Cu}/(j_n(0)t)\), from which we evaluated \(P_T\) using Eq. (5). Fig. 1 shows \(R_{O/Cu}\) plotted as a function of the dose for \(\sim 14\) eV O\(^+\) incident on Cu(001), for \(\theta_i = 0^\circ\) and \(\theta_i = 45^\circ\). It can be seen that these data lie on straight lines, justifying our assumption that we are in the low coverage regime where \(\Theta(r, t)\) is linear with the dose and trapping probability.

3. Results

Fig. 2 shows \(P_T\) as a function of incident ion energy, for \(\theta_i = 0^\circ\) and \(45^\circ\). It can be seen that, within the sensitivity of our measurement, there is little difference between \(P_T\) at these two angles. However, there is a significant dependence of \(P_T\) on the incident ion beam energy. \(P_T\) decreases by almost a factor of two between 5 and 50 eV (see
Fig. 2 inset), while above 50 eV there is a general upward trend.

The slow increase above 50 eV can be explained by considering the origin of the measured Auger peak-to-peak ratio, $R_{O/Cu}$. Since typical probing depths of AES are 10–30 Å [16], $R_{O/Cu}$ is sensitive to the presence of O over this entire depth (i.e., buried or implanted) and is not a measure of the amount of O trapped on the surface alone. Thus the increase in subsurface O with increasing energy leads to an increase in $R_{O/Cu}$. A further complication in the interpretation of the data at energies where subsurface trapping is possible arises from the depth-dependent sensitivity of the Auger signal. The Auger signal is more sensitive to surface than to subsurface O since the Auger electrons emitted from the subsurface O are attenuated as they escape the sample [17]. Thus, the conversion of $R_{O/Cu}$ to absolute coverage is not quantitative once subsurface penetration has started. We assume that the increase in $P_T$ above 50–60 eV is due to subsurface O, and that although subsurface penetration occurs below this energy, it does not become significant until the incident ion energy has increased to ~50 eV [18].

4. Discussion

It is possible to draw some general conclusions about the O trapping probabilities based on our understanding of the Na–Cu system. In the following discussion we will contrast our measured O trapping probabilities to those previously measured for the model Na–Cu system [5,6]. We will start this discussion by describing the Na–Cu results.

Goodstein et al. [5,6] measured trends in $P_3$ for 5–600 eV Na$^+$ incident on Cu(001) along the (100) azimuth, at $\theta_i = 8^\circ$ and $\theta_i = 45^\circ$. A novel technique was used that relied on charge transfer processes specific to the Na–Cu system, and that is very sensitive to Na trapped on the surface. It was found that for $\theta_i = 45^\circ$, $P_3$ was highly nonmonotonic between 5 and 75 eV, dropping to zero at 20 eV before rising again to a local maximum at 75 eV. Above 75 eV, $P_3$ decreased monotonically. In contrast, the O trapping probability decreases
monotonically between 5 and \( \sim 50 \text{ eV} \) (Fig. 2) and, most notably, there is no minimum at 20 eV. Similar results have been obtained below 50–60 eV, using AES, for \( \text{O}^+ \) incident on \( \text{Ni}(111) \) at \( \theta_i = 0^\circ \) [3].

Trapping mechanisms for the Na–Cu system have been studied in great detail with a classical trajectory simulation code [19], using an accurate Na–Cu scattering potential which consists of an attractive image potential and a sum of repulsive Hartree–Fock pair potentials [20]. Fig. 3 shows the results of the simulated trapping probabilities for Na with energies from 5 to 300 eV. Although absolute coverages were not measured, excellent qualitative agreement was obtained between the trends in the data and the simulated trends in \( P_S \). Systematic trajectory analysis for the Na–Cu system at 45° [5,6] demonstrated that the nonmonotonic trend in \( P_S \) could be explained in terms of the trajectory types allowed by the apparent surface corrugation seen by the incident ions, which changes as a function of the incident ion energy and incident angle. Fig. 4 shows examples of low-corrugation and high-corrugation trajectories that lead to surface trapping. For the Na–Cu system at the 45° scattering geometry it was found that there is a clear transition at 20 eV between the low- and high-corrugation trajectory types, resulting in a minimum in the surface trapping. The dramatic difference we have observed in the measured trapping probabilities at \( \theta_i = 45^\circ \) for O and Na indicates that the apparent surface corrugation seen by the incident O ions must differ significantly from that seen by the incident Na ions. Therefore, we expect the energy-dependence of the low- and high-corrugation trajectories to be different for the two systems.

We are currently in the process of calculating an O–Cu interaction potential for use in our classical trajectory simulation [18]. Our preliminary calculations show that the \([\text{O–Cu}]\) pair potential is significantly less repulsive than the \([\text{Na–Cu}]\) pair potential, resulting in a higher apparent surface corrugation for incident O ions compared to the incident Na ions. Furthermore, the results of our classical trajectory simulation, using this
preliminary potential, qualitatively reproduce our O trapping data [18]. This suggests that, in general, trapping experiments may provide a good test of the interaction potential. However, a more detailed microscopic understanding of the O trapping trajectories must await the completion of this calculation as well as a thorough analysis of the trajectory types that occur in the O–Cu system. Finally, the role of charge transfer in determining trends in the trapping probabilities, as well as the sensitivity of the trapping to the attractive well used in our simulations, must also be investigated.

5. Summary

We have presented the energy- and angle-dependent trends in the trapping probabilities of 5–600 eV O$^+$ ions incident on Cu(001) at $\theta_i = 0^\circ$ and 45$^\circ$. These trends depend strongly on the incident ion energy, but not on the incident angle. The trapping probability decreases with increasing energy up to $\sim$50 eV. Above this energy the trapping probability rises with increasing energy, which we attribute to subsurface implantation. Using Na–Cu as a model system, we know that the energy-dependent apparent surface corrugation seen by the incident ions, which is determined by the ion-surface scattering potential, plays an important role in determining the trajectory types that lead to surface trapping. In order to study the O trapping probabilities in more detail, it is necessary to obtain an accurate repulsive O–Cu pair potential for use in the classical trajectory simulation [18]. Measurements of trapping probabilities may provide a sensitive test of scattering potentials at these energies.

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References