Kinetic excitation of solids induced by energetic particle bombardment: Influence of impact angle

Christian Heuser\textsuperscript{a,}\textsuperscript{*}, Mario Marpe\textsuperscript{a}, Detlef Diesing\textsuperscript{b}, Andreas Wucher\textsuperscript{a}

\textsuperscript{a}Fachbereich Physik, Universität Duisburg-Essen, 47048 Duisburg, Germany
\textsuperscript{b}Fachbereich Chemie, Universität Duisburg-Essen, 45117 Essen, Germany

\textbf{A B S T R A C T}

The kinetic excitation of a solid surface under bombardment with energetic particles is studied via internal electron emission in a metal–insulator–metal junction. In particular, the dependence of the measured tunneling yield on the projectile impact angle is studied. The resulting impact angle distribution is compared with predictions of the total excitation energy profile calculated using the SRIM 2006 Monte Carlo program package. While the calculated profiles fail to explain the experimental data, it is shown that a simple calculation of impact angle dependent projectile backscattering qualitatively reproduces the observed trends.

© 2008 Elsevier B.V. All rights reserved.

\textbf{1. Introduction}

When an energetic particle impinges on a surface, its kinetic energy is dissipated via elastic collisions with target atoms (nuclear stopping) and via electronic excitation processes (electronic stopping). Nuclear stopping generates fast, non-thermal particle kinetics which are usually described by a collision cascade or a collisional spike and may lead to the ejection of surface material (“sputtering”). Electronic stopping, on the other hand, gives rise to a fast local and temporal heating of the electronic sub-system of the solid, which is mediated by an electronic friction experienced by all moving particles and, in addition, by electron promotion processes occurring in close atomic collisions. The resulting “kinetic” excitation manifests, for instance, in the “external” emission of electrons from the bombarded surface [1] and is also responsible for the fact that part of the sputtered atoms leave the surface in excited or ionized states [2,3].

Theoretical estimates [4] predict the kinetic excitation process to be dominated by low-energy excitations, where the generated “hot” electrons do not have enough energy to overcome the surface barrier and be emitted into the vacuum. Detection of such carriers is, however, possible by means of a buried tunnel junction, where they can lead to “internal” emission currents [5]. This strategy is applied here to detect hot electrons and holes produced by the impact of 10 keV Ar\textsuperscript{+} ions onto a metallic surface. As a complement to our earlier work [6–8] investigating the dependence of the bombardment induced internal emission yield on parameters like kinetic energy or charge state of the projectiles, particular emphasis is put here on the influence of the impact angle, since it is expected that this dependence reveals valuable information about the depth distribution of the generated excitation.

\textbf{2. Experiment}

The buried tunnel junction is realized in form of a metal–insulator–metal (MIM) film structure. The top metal electrode of this device (polycrystalline silver) represents the actual target material, the surface which is bombarded by the energetic primary ion beam. Electrons and holes excited in the selvage of the surface via kinetic processes following the projectile impact can migrate to the metal–oxide interface, overcome the tunnel barrier represented by the oxide film (∼3 nm amorphous AlO\textsubscript{x}) and be detected as an internal emission current in the underlying metal substrate electrode (polycrystalline aluminum). Projectile ions are generated by a commercial ion source delivering a beam of positively charged argon ions with energies reaching from 5 to 15 keV, which was operated in a pulsed mode in order to clearly discern bombardment induced effects and keep the total ion fluence low. The bombardment induced internal emission current was measured and...
divided by the primary ion current in order to determine the internal emission yield as a function of the projectile impact angle. The primary ion current was measured using a faraday cup with a diameter of about 1 mm to minimize effects of electron emission.

The design and production of the MIM devices as well as the procedures to measure the tunneling current have been described in great detail earlier [5,8]. Briefly, the substrate (Al) and top (Ag) metal electrodes are formed as 5 mm wide stripes which are oriented perpendicular to each other, thus limiting the electrically active junction to the overlap area of 5 × 5 mm². The relatively large active area was chosen to adapt the variation of the irradiated spot upon changing the ion impact angle to oblique incidence. The diameter of the beamspot was about 100 μm. To ensure that projectiles strike the active area the signal was optimized by varying the beam position until a stable signal was observable. Both electrodes are separated by an amorphous AlO Xu layer of about 3 nm thickness. The thickness of the on top Ag layer is a crucial parameter in this type of experiments. On one hand, it must be chosen larger than the mean range of the projectile ions (≤10 nm depending on impact angle [9]) in order to prevent projectile induced damage of the oxide film. On the other hand, it must be comparable with (or smaller than) the effective electron mean free path (9–15 nm [8]) to prevent significant loss of the generated hot charge carriers during their passage towards the junction. A nominal thickness of 20 nm was therefore chosen as a good compromise between these contradicting requirements. The surface roughness of the resulting Ag layer is typically about 4 nm. Moreover, the I–V characteristics of the MIM devices were recorded frequently before, during and after the measurements in order to ensure that the electrochemical properties of the junction did not change during the experiments. All experiments were performed at zero bias voltage between two metal electrodes.

3. Results and discussion

Changing the angle of incidence results in a modification of the depth distribution of energy deposited in the collision dynamics induced by the projectile impact. As a consequence, the kinetic excitation dynamics will change as well, and hot charge carriers are created at different depths below the surface. In a MIM experiment, this is important since the hot carriers need to be transported to the junction depth in order to contribute to the internal emission current. Measuring the tunneling yield as a function of projectile impact angle therefore reveals important information about both the depth distribution of the excitation processes and the transport properties of the top metal film. In order to unravel both effects, the excitation distribution is modeled by Monte Carlo simulation using the SRIM 2006 software package [10].

The dependence of the measured tunneling yield on the impact angle of the Ar⁺ projectile ions is shown in Fig. 1. The data were acquired for a MIM device with a top Ag layer of 20 nm thickness bombarded with Ar⁺ ions of two different impact energies of 5 and 7 keV and averaged over a number of different experimental runs, with the indicated error bars representing the typical standard deviation. One observation is immediately evident: The observed impact angle dependence does not seem to critically depend on the kinetic impact energy of the projectiles, indicating that the effect is generated by geometrical effects rather than differences in penetration depth of the projectiles. In fact, for impact angles up to 40° with respect to the surface normal, the tunneling yield is essentially independent of the impact geometry. This finding is remarkable, since the penetration depth decreases by about 30% in this interval. Under more oblique incidence conditions, on the other hand, the tunneling yield starts to strongly decrease with increasing impact angle.

In principle changing the impact angle might change the depth distribution of excitation energy generated by the projectile impact. If hot electrons are created closer to the surface, they must travel a longer distance to reach the buried junction, thus resulting in an increased probability of losing part of their energy via inelastic scattering. The latter, in turn, will lead to a reduced probability to overcome the tunneling barrier. Therefore, grazing incidence might produce a shallower excitation distribution, thus leading to the observed decrease of the tunneling yield. Note, however, that this effect can only play a dominating role if the average excitation depth is comparable with (or larger than) the (effective) electron mean free path.

On the other hand, projectiles will be backscattered from the surface with larger probability if the impact angle is increased. As a consequence, more kinetic energy might be taken away by backscattered projectiles, and the total amount of excitation energy deposited into the solid might decrease upon the transition from normal to oblique incidence, again qualitatively explaining the trend observed in Fig. 1.

In order to qualitatively discuss these possibilities, we employ a simple statistical estimate of the inelastic energy loss experienced by the projectile and all moving recoil atoms in the course of the collision cascade initiated by the projectile impact. Calculating the total energy transferred into the electronic system using the statistical Monte Carlo code SRIM 2006, we find depth distributions of the deposited “ionization” energy which are depicted in Fig. 2. It is obvious that these distributions change with impact geometry in such a way that the excitation energy is deposited closer to the surface under oblique incidence conditions. While this is strongly evident in the excitation induced by the projectile, the effect becomes much less pronounced for the distribution induced by the recoil atoms. Integrating the distributions over depth, we find total excitation energies depicted in Fig. 3. The data were normalized to the projectile impact energy and therefore denote that fraction of the original kinetic energy which is converted into electronic excitation in the course of the entire collision cascade. Two observations are evident. First, the major part of the deposited excitation energy appears to be generated by recoils rather than by the projectile itself. In that respect, the depth distribution variations observed in Fig. 2 might not have as dramatic consequences as it appears at first sight. Second, the total deposited excitation energy exhibits a qualitatively similar trend as the measured tun-

![Fig. 1. Impact angle dependence of measured tunneling yield of an Ag/AlOx/Al MIM device with a 20 nm top Ag film thickness bombarded with Ar⁺ of the indicated impact energy. The data were averaged over several measurement cycles with the error bars indicating the standard deviation.](image-url)
 tunneling yield as depicted in Fig. 1. However, the magnitude of the effect is not the same. While the tunneling yield decreases by about a factor of 10 between normal and grazing incidence, the decrease in total excitation energy is only by a factor 2, regardless of whether the contribution of projectiles, recoils or both is considered.

As a consequence, one has to conclude that neither the total deposited excitation energy nor its depth distribution as calculated by SRIM are sufficient to explain the impact angle dependence of the internal emission yield data measured here. We attribute this finding to the fact that SRIM does not include transport of the generated excitation and therefore cannot account for the time and space dependence of the excitation energy density created within the solid. In fact, a simple model interpreting the measured tunneling yields in terms of a locally and temporarily treated electron gas in the top metal electrode has revealed that the kinetic internal emission detected here must be generated by a local hot spot of only a few femtoseconds duration featuring electron temperatures of the order of $10^4$ K [8]. Moreover, model calculations based on molecular dynamics simulations of the cascade dynamics have shown such a hot spot to be formed by the projectile traversing the surface layer immediately upon impact [11–14]. Interestingly, the energy density which is generated at the surface that way should be independent of the projectile impact angle, since the larger energy loss per unit depth experienced by an obliquely incident projectile is deposited into a correspondingly larger surface area and, hence, into a larger volume. As a consequence, it is tempting to assume that each projectile that is actually penetrating the surface produces roughly the same average internal emission yield. In this picture, the observed decrease of the measured tunneling yield would simply be attributed to the fact that more projectiles are backscattered from the surface under oblique incidence. In order to examine that notion, Fig. 4 shows the fraction of projectiles calculated by SRIM to actually penetrate the surface as a function of the impact angle. It is evident that the angle dependence of this quantity is surprisingly similar to that of the data depicted in Fig. 1. Moreover, the magnitude of the reduction between normal and oblique incidence as well as the observed independence of the impact energy are consistent with the experimental observation. Although this picture clearly cannot explain all details of the measured curves, it still provides a reasonable qualitative explanation of the observed impact angle dependence of the kinetic tunneling yield. A more sophisticated interpretation of the measured data requires more detailed model calculations of the time and space dependent excitation energy density profile within the solid along with a proper account for the transport of hot electrons and holes to the junction. Calculations of this kind are currently under way in our lab.

4. Conclusions

The kinetic internal electron emission yields observed in a MIM tunnel junction under bombardment with energetic particles exhibit an interesting dependence on the projectile impact angle. Being largely independent of impact angle between normal and about 40° incidence, the yield falls strongly with increasing impact angle under more oblique incidence conditions. Attempts to understand this behavior in terms of the total excitation energy as calculated by Monte Carlo codes such as SRIM 2006 are not successful, since this approach does not describe time and space dependence of the
excitation energy density generated by the projectile impact. However, it is demonstrated that a simple estimate of the fraction of backscattered projectiles – which can easily be calculated by SRIM – is capable to qualitatively account for the measured impact angle dependence. This finding indicates that each projectile impact generates roughly the same average internal emission yield regardless of impact angle. This notion is consistent with the interpretation of previously measured internal electron emission yields as well as with model calculations of the kinetic excitation process. However, more sophisticated theoretical work is clearly needed to quantitatively explain the remaining differences between experimental data and predicted impact angle dependence.

Acknowledgement

The authors are greatly indebted to the Deutsche Forschungsgemeinschaft for financial support in the frame of the Sonderforschungsbereich 616 “Energy Dissipation at Surfaces”.

References