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The use of MIM tunnel junctions to investigate kinetic electron excitation in atomic collision cascades

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Abstract

A novel technique is introduced to investigate the kinetic excitation of electrons in a solid by bombardment with energetic ions. The sample is prepared as a metal-insulator-metal (MIM) tunnel junction which opens the possibility to detect hot electrons with excitation energies well below the vacuum barrier. The excitations produced by the projectile impact onto the top electrode are detected as a tunnel current into the underlying base electrode. By varying the top electrode thickness, the elastic transport of hot electrons towards the tunnel junction can be studied.

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1. Introduction

If an energetic particle impinges onto a solid surface, its kinetic energy dissipates within the solid by means of elastic collisions (“nuclear stopping”) and electronic excitation processes (“electronic stopping”). For impact energies in the keV range it is well known that nuclear stopping dominates the energy loss experienced by the projectile, thus generating an atomic collision cascade. Along the pathways of the cascade, however, a part of the kinetic energy of the primary ion as well as of the recoil atoms is converted into electronic excitation by means of inelastic processes. The excited electrons undergo cascades of collisions in the solid among each other until their energy degrades into heat. Some electrons receive enough energy to overcome the surface barrier and are emitted into the vacuum, giving rise to secondary electron emission. Kinetic ion induced electron emission (KEE) has been investigated in great detail [1–4]. In particular the yields, i.e. the average number of emitted

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electrons per impinging projectile, the emission statistics as well as energy distributions of the ejected electrons have been measured for many projectile-target combinations [5–7]. These experiments, however, are limited to electrons with excitation energies above the vacuum level. For metals, this restricts the range of observable excitation energies to relatively large values of several eV above the Fermi level. Electrons with excitation states located between the Fermi and the vacuum level remain within the solid and undergo further collisions until they finally thermalize. On the other hand, these weakly excited electrons are of particular interest because the prevailing mechanisms of kinetic electron excitation predict predominantly low energy excitation. Direct energy transfer by binary projectile- (or recoil atoms-) electron collisions [4], for instance, is expected to produce low excitation energies because of the huge mass difference of the collision partners. Electron promotion in close collisions between two atoms, as a second possible excitation mechanism [4], may in principle also populate higher lying states, which will, however, quickly relax due to extremely fast electron–electron interaction. Both mechanisms will therefore lead to occupation probability distributions peaking at energies close to the Fermi level, a fact which has recently been confirmed by energy loss experiments on fast neutral atoms grazingly scattered from a metal surface [8].

To gain more insight into the excitation and transport mechanisms of hot electrons produced by fast ion impact onto a solid surface, we use a novel detection technique, where the ion bombarded sample is prepared as a metal-insulator-metal (MIM) layer stack. The idea is that excited electrons, generated in a shallow sub-surface region of the top metal layer by ion impact, travel ballistically to the metal-oxide interface, tunnel through the thin oxide layer and are detected as an ion bombardment induced tunnel current in the underlying metal substrate. In order to allow elastic transport of the electrons to the tunnel junction, the top electrode thickness has to be of the order of the inelastic mean free path of the excited electrons, which is of the order of 10 nm for excitation energies around 1 eV [9] and increases with decreasing energy [10].

2. Experimental

The experiments are carried out in an ultrahigh vacuum chamber with a base pressure of about 10^{-9} mbar. The primary ions are generated by a commercial ion gun delivering a focused and pulsed inert gas ion beam with energies between 5 and 15 keV and a current of a few hundred nA impinging under 45° with respect to the surface normal. The sample is a MIM junction produced ex situ by evaporating a 20 nm thick aluminum electrode onto an insulating glass substrate. In an electrochemical treatment described in detail elsewhere [11,13], the aluminum is locally oxidized to form an Al_2O_3 overlayer of about 2.7 nm thickness. In a last step, a polycrystalline silver layer of 20 nm thickness is vapor deposited on top of the oxide layer. Note that this electrode constitutes the ion bombarded metallic surface under investigation here. From SRIM2003 [12] calculations for 10-keV Ar^+ ion impact onto silver, we calculate nuclear and electronic stopping powers of $S_n = 75.5$ eV/Å and $S_e = 17.2$ eV/Å, respectively. The corresponding mean range of 4.9 nm results in a negligible penetration probability ($<10^{-3}$) of projectile ions into the underlying oxide layer.

The two metal electrodes are both 2 mm wide and orientated at 90° with respect to each other. Both electrodes therefore overlap across an area of 2×2 mm^2 where the tunnel junction is formed. The electrical characteristics of the MIM junction are shown in Fig. 1. A bias voltage was applied between the Al and the Ag electrode by means of a programmable potentiostat and the resulting current through the oxide barrier was measured with a three step current-to-voltage converter connected to the Al electrode. For the characterization of the system, a voltage ramp was applied from −0.5 V to 0.5 V with a scan rate of 200 mV/s. The resulting current shows a hysteresis which is due to the charging current of the system capacitance. With an oxide film thickness of 2.7 nm, a dielectric constant of $\varepsilon = 8$ and a total area of 4×10^{-2} cm^2, the specimen should have a theoretical capacity of 104 nF which is verified by impedance spectroscopy [13]. With a scan rate $dV/dt = 200$ mV/s, one expects a charging current of 20 nA which is in good agreement with the data

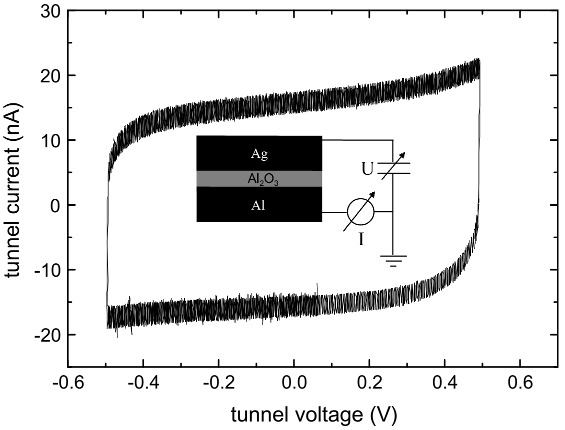

Fig. 1. Dynamic *I–V* curve characterizing the electrical properties of the MIM-junction measured at a scan rate of 200 mV/s.

in Fig. 1. At voltages $|V_t| > 300$ mV the current increases due to the superposition of charging and actual tunneling currents. Curves like the one displayed in Fig. 1 are recorded frequently during data acquisition in order to ensure that the MIM junction is not modified or destroyed by the ion bombardment.

3. Results and discussion

In a first set of experiments to characterize its electrical response to ion bombardment, the MIM-junction was exposed to a focused and pulsed 10 keV Ar^+ ion beam with a spot size of about 200 μm diameter and a current of 190 nA. The pulse length was 10 ms at a repetition rate of 5 Hz. No bias voltage was applied across the junction to eliminate any dc currents. The ion beam was aimed at different lateral positions of the sample to separate trivial charging and discharging effects of the MIM's capacitance from actual tunnel currents. If the impact point of the ion beam is located on the silver electrode above the center of the tunnel junction, current pulses of about 55 nA amplitude are observed in phase with the primary ion pulses, the polarity of which shows that electrons are flowing from the top silver electrode through the oxide into the aluminum electrode. When the ion beam is moved along the silver electrode to the edge of the junction area, the tunneling current decreases since only a part of the ion beam still illuminates the junction, the remaining part hitting outside the junction area and therefore not inducing any tunneling current. If the beam is located on a spot of the silver electrode which is completely outside the tunnel junction area, no tunneling current can be observed at all. These observations provide clear evidence that the measured current cannot be induced by simple charging effects of the MIM capacitance. When the ion beam hits the bare aluminum electrode far away from the junction, both the sign and the magnitude of the current pulses change, since now the measured signal simply constitutes the usual neutralization current of the Ar^+ ion beam.

A second experiment was performed to investigate the influence of the inevitable damage and removal of material from the top metal layer induced by sputtering. In principle, a significant reduction of the silver film thickness during data acquisition is expected to modify the measurement because the path length of the excited electrons between the collision cascade volume and the oxide interface is reduced. With decreasing film thickness, one would therefore intuitively expect an increasing tunneling current due to the decreasing influence of relaxation processes occurring during transport of the originally produced excitation distribution to the junction interface.

In order to avoid such effects, it is necessary to use a pulsed primary ion beam with a temporal pulse length as short as possible compatible with the response time of the employed current to voltage converter. Experiments with varying primary ion pulse lengths have shown that the shortest possible pulse duration for our actual setup is approximately 500 μs. To estimate the stability of the measured signal under these conditions, the tunnel current is shown as a function of the accumulated primary ion fluence in Fig. 2. Every displayed data point has been acquired by averaging the tunnel current over two primary ion pulses, resulting in 1 ms bombardment of the top MIM electrode per point. The total bombarding time accumulated during the entire measurement amounts to 1 s. The primary ion fluence is calculated from the known primary ion current and the spot size of the beam on the sample.

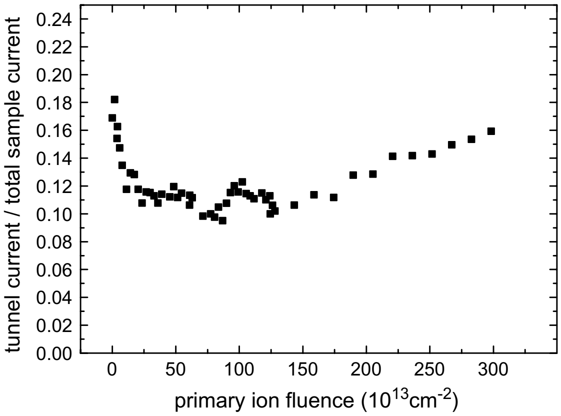

Fig. 2. Measured tunneling current as a function of accumulated total primary ion fluence. The decrease at the beginning is due to removal of surface contamination, after a constant region the signal increases because of degrading silver thickness.

At first sight, it is apparent that the tunneling current decreases at the beginning of the measurement. Because the MIM junction is prepared ex situ, the silver surface is initially contaminated. It is well known from kinetic electron emission experiments [2] that the observed electron currents are very sensitive to surface contaminations like, for instance, oxide layers. The presence of such layers generally tends to increase the observed electron emission yields. As seen in Fig. 2, a similar trend is observed for the internal tunneling current as well. The transient measured in the ion fluence interval between zero and $2 \times 10^{14}\,\mathrm{cm}^{-2}$ is therefore presumably induced by the removal of a surface contamination layer. After this initial cleaning, the surface conditions appear to be stable for a fluence interval extending to about $2 \times 10^{15}\,\mathrm{cm}^{-2}$. During this interval, the ion bombardment induced damage is obviously still negligible, leading to the observation of a stable tunnel current. Towards larger fluences, the erosion of the silver film starts to increase the measured tunnel current.

For a more quantitative discussion, the primary ion fluence f_p depicted in Fig. 2 can be converted into eroded depth

$$d = \frac{f_p \cdot Y}{n}, \tag{1}$$

where n denotes the atom density of the silver target ($5.85 \times 10^{22}\,\mathrm{cm}^{-3}$). The quantity Y denotes the sputter yield, i.e. the average number of surface atoms removed per projectile impact, which for the prevailing conditions of 10-keV Ar^+ ion impact onto silver can be estimated as $Y \sim 10$ atoms/ion [14]. As a result, we estimate that a total of about 5 nm has been removed from the top silver layer during the entire experiment displayed in Fig. 2. The stable fluence range below $2 \times 10^{15}\,\mathrm{cm}^{-2}$ therefore corresponds to the removal of about 3 nm or 12 atomic layers of silver, whereas the fast initial transient of the measured signal clearly corresponds to the removal of a surface contamination layer of one or two monolayers thickness.

In order to further study the influence of the top layer thickness on the tunnel current, the result of a similar experiment performed with longer primary ion pulse duration (5 ms) is shown in Fig. 3. Technically, the tunneling current is recorded as a function of total ion bombardment time, and the abscissa is rescaled into layer thickness by means of $d(t) = d_0 - \dot{d} \cdot t$, where d_0 is the original layer thickness of 20 nm. During the entire experiment, the top film thickness is decreased to approximately 9.5 nm. The solid dots in Fig. 3 represent the measured tunneling current normalized to that of the impinging ion beam. The solid line represents a least square fit of the attenuation function

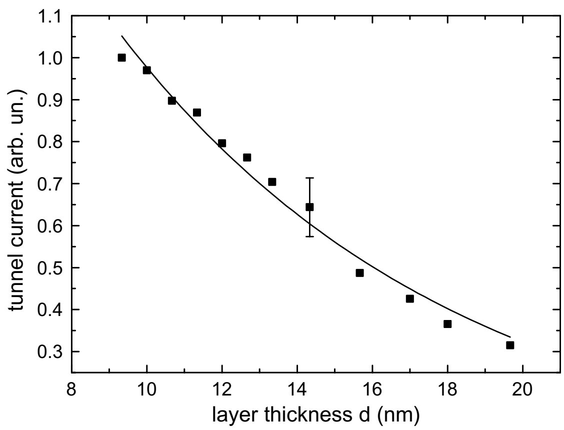

Fig. 3. Measured tunneling current as a function of the silver top electrode thickness as calculated from the primary ion fluence and literature data of the sputter yield.

$$I_t(d) = I_t^0 \cdot \exp\left(-\frac{d}{\lambda}\right) \quad (2)$$

which describes the probability for an electron to reach the silver-oxide interface without being inelastically scattered. From this fit, a mean free path λ of the hot electrons – averaged over all relevant excitation energies – can be determined as $\lambda \sim 9$ nm in agreement with the expectation described in Section 1.

4. Conclusion

The measurement of tunneling currents across a buried junction provides a unique and exceedingly simple method to obtain information about low energy electronic excitations produced under bombardment of a solid surface with energetic ions. It is demonstrated that the a priori unavoidable damage induced by the projectile impact does not prevent useful measurements and, most importantly, does not destroy the electric properties of the tunnel junction. Our preliminary results indicate that a total ion fluence of more than 10^{15} cm^{-2} can be tolerated before the measured signal is significantly influenced by the sputter induced surface erosion. The interpretation of the data presented in Figs. 2 and 3 described above must of course be cross-examined by a systematic variation of the initial top layer thickness of the MIM structure. In the same fashion as done in ion induced electron emission experiments, it is furthermore necessary to distinguish between kinetic and potential excitation processes by varying the kinetic impact energy and/or charge state of the projectiles. Corresponding experiments are currently under way in our laboratory.

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