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Transport of 75–1000 eV electrons in metal–insulator–metal devices

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Metal–insulator–metal nanostructures with a 50 nm silver film and a 30 nm aluminum film separated by a few nanometer insulator layer (aluminum oxide) are irradiated with a focused e^--beam (diameter ≤500 μm) with kinetic energies in the range of 75–1000 eV. Impact angle and energy dependence of the e^--beam induced electron emission from oxide covered aluminum and from silver show a good coincidence with previous results. The e^--beam induced internal device current measured between the aluminum and the silver film, on the other hand, is found to be independent of the primary electron energy and impact angle. The results suggest that external electron emission may have to be included in the interpretation of the internal transport currents.

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1. Introduction

e^--beam induced scattering processes and electronic excitations have been intensively studied in the literature for more than 100 years [1–5]. One of the reasons for this research activity is that electron scattering and a possibly accompanying energy loss influence the image formation process in the transmission electron microscope (TEM) [6] as well as in the scanning electron microscope (SEM). In the SEM, bulk specimen can be studied using the beam reflected from the sample for the image formation [7]. In reflection direction, the beam induced electron emission from a specimen may contain back scattered primary electrons which are either elastically reflected or exhibit small characteristic energy losses (further called BSE), and secondary electrons (further called SE) with usually a broad energy distribution at energies below 50 eV excess energy (which is the energy above the work function of the material) [8–10]. Backscattered electrons and secondary electrons have different trajectories and therefore lead to different images in SEM microscopes and care has to be taken especially if the metrology of nanostructures is examined [11,12] since the spreading of BSE and SE at the edges of nanostructures differs due to the energy dependence of the mean free path [13].

The primary kinetic energy in the e^--beam of the experiments discussed here is identical with the lower limit of several 10 eV for the electrons in low voltage SEM experiments; therefore the external electron emission in the experiments discussed here can be discussed in the same manner as in LVSEM images. In this work, we present a study of electron reflection and transmission through a thin metal film, which is realized as the top electrode of a metal–insulator–metal (MIM) sandwich-like nanostructure. In such a device, two forms of electron emission phenomena can be observed, namely (i) backscattering or secondary electron emission at the vacuum–metal surface irradiated by the primary e^--beam (in the following be referred to as “external emission”) and (ii) transport of charge carriers through the internal barrier formed by the insulating layer separating the two metal electrodes (in the following referred to as “internal emission”). While the external emission signal can only comprise electrons possessing enough energy to overcome the surface barrier (i.e., the work function of the top metal), the internal current can contain contributions of electrons as well as defect electrons (“holes”), leading to a measurable current across the MIM device. It should be noted that charge carriers contributing to this current do not necessarily have to possess excitation energies above the internal energy barrier but may in principle tunnel through the barrier because the insulating film is

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thin (<5 nm). Transport of high energy electrons can also be studied in MIM devices with thicker oxide films ($d > 7$ nm) [14,15]. Independent on the oxide thickness, the shape of the internal barrier in MIM devices (its height is ≈2/3 of the silver films work function in our case) may be modified by applying a bias voltage between the two metal electrodes, thereby opening the possibility to control the spectral transmission characteristics for the internal emission process.

The study performed here was motivated by a series of experiments, where MIM devices were used to detect excited charge carriers generated in the top metal film either by chemical surface reactions [16] or by irradiation with photons [17], electrons [18] or heavy particles [19–22]. With respect to the latter, the internal emission process was employed to investigate kinetic excitation, i.e., the transient production of hot excited charge carriers following the impact of a fast projectile particle (typically rare gas ions with some keV kinetic energy) onto a solid surface. In these experiments, the internal emission current was used to monitor the presence of hot charge carriers generated in an ion bombarded top metal electrode of typically 20–40 nm thickness, which was separated from the bottom metal electrode of similar thickness by a thin intermediate oxide layer (see e.g. Refs. [21–24]). The interpretation of the resulting data left an open question, namely as to which extent the detected ion induced internal emission current is caused by ballistic transport of high energy excitations produced from direct projectile-electron scattering events close to the bombarded surface or by excited charge carriers generated by a diffusion-like collective transport of low energy excitations to the metal–oxide interface. In order to address that question and shed more light on the transport mechanism of excited charge carriers in such a system, MIM devices like the ones used in the experiments mentioned above are now irradiated with electrons as primary particles. The kinetic energy of the e^--beam is varied in the range 75–1000 eV, and both the internal and external emission behavior following the electron impact are studied systematically. The MIM sandwich structure used here consists of a silver top electrode (thickness ≈50 nm) and an aluminum bottom electrode (thickness ≈30 nm) deposited on a glass substrate. The two metal films are separated by a thin aluminum oxide layer of ≈3 nm thickness. In order to ensure that the primary excitation is confined to the top silver layer, the kinetic energy of the irradiating e^--beam was kept well below 2 keV, since simulations using the Casino Monte Carlo package (see [25]) show that the transmission through a 40 nm silver film is negligible for energies below ≈1.5 keV. In the energy range studied here, secondary electron emission as well as backscattering of primary electrons into the vacuum are supposed to play a dominant role. Internal emission, on the other hand, requires the transport of excitation from the region close to the silver surface to the silver film/oxide interface and would therefore be small if the barrier height for internal and external emission were comparable. In the Ag|AlO_x|Al structures, however, the height of the internal barrier is 2.1–2.4 eV at the aluminum–aluminum oxide interface and 3.4–3.9 eV at the aluminum oxide–silver interface [26]. This corresponds to approximately 2/3 of the Ag work function [27]. Moreover, the barrier is thin enough to permit tunneling contributions of both electrons and holes at excitation energies below the respective barrier height, so that the measurement of internal emission currents represents a promising tool to study low energy transmission processes. The setup presented here allows for the measurement of internal and external electron emission processes at the same sample. The quantity measured in these experiments is the (external or internal) emission yield, which for the external emission case is easily defined as the average number of emitted electrons per impinging primary electron. For internal emission, the definition is more complicated, since electrons as well as holes can contribute to the measured internal current. The process can be described in terms of a two band tunneling process, where the electron and hole transport is mediated via the conduction and valence bands of the oxide, respectively [28,29]. We define the yield in this case as the average number of (negative) elementary charge equivalents transported from the irradiated to the non-irradiated metal electrode per impinging primary electron. In other words, excited electrons and holes flowing from the irradiated top metal into the underlying bottom metal film contribute to the measured internal current with opposite sign, thereby allowing the possibility of a measured internal emission current of zero when electron and hole transport annihilate.

2. Experimental setup

2.1. Sample preparation and electrical wiring

The ≈30 nm thick bottom aluminum electrode (lateral dimensions 18 mm·4 mm) of the metal–insulator–metal thin-film devices is thermally evaporated under ultra-high vacuum conditions on top of a glass substrate (size 18 mm·9 mm). The aluminum oxide film is formed by consumptive oxidation in an electrochemical droplet cell. The process is described in detail elsewhere [30], and the oxidation potential was adjusted to form a 3–4 nm oxide film. The oxide film is on the one hand thick enough to warrant a stable interface wall and on the other hand thin enough to allow tunneling of excited charge carriers. The electrical characteristics of the resulting device are such that for bias voltages of ±1 V between the two electrodes the DC tunnel current density across the barrier remains below 1 nA cm^{-2}.

Across the oxidized aluminum film, the top metal film (≈50 nm thick silver) is again thermally evaporated under ultra-high vacuum conditions with a lateral dimension of 8 mm·4 mm. This way, the active area of the resulting MIM device, i.e. the region where all three layers overlap and form a sandwich like structure, has a lateral dimension of 4 mm·4 mm and a total thickness of ≈80 nm. Depending on the desired experiment, either the current flowing into the top or bottom electrode was measured with this "probe" electrode kept at ground potential, while the other electrode was set to a desired potential in order to establish a certain bias voltage. To do this, the other electrode was connected to the voltage output of a potentiostat (Heka PG 510 with counter and reference connectors bridged), while the probe electrode was connected to the current meter function ("working" connector) of the potentiostat and kept at virtual ground potential. If the probe electrode was the top silver electrode, the measurement will be referred to as ("probe top"), experiments where the current into the bottom aluminum electrode were measured will be referred to as ("probe bottom"). The potentiostat was used since it is equipped with an internal voltage ramp generator which permits a fast characterization of the MIM device as well as monitoring the bias voltage dependence of the measured signals as described below.

The sample was irradiated with a DC e^--beam generated by a Kimball Physics ELG-2 electron gun. The electron kinetic energy was adjusted from 75 to 1000 eV, where the lower limit of 75 eV was chosen because the beam was found to be significantly less stable at lower energies. In all cases, the beam diameter at the sample surface was 500 μm or below and the primary electron current was kept in the range of 10–40 nA. The energy dependent beam characteristics (width and current) were characterized by means of a Faraday cup with an entrance aperture of 0.6 mm. Fig. 1(a) shows a schematic cross-sectional view of the sample along the x-direction across the top silver film through the center of the active area (not drawn to scale). The aluminum and silver films are not perfectly cuboid but trapezoidal at their edges due to the shadows of the evaporation masks, which were approximately 3 mm away from the substrates.

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