Ion-beam-induced nanosmoothening and conductivity enhancement in ultrathin metal films

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We present a systematic in situ study of the effect of postdeposition low-energy (200 eV) ion bombardment on resistance and surface topography of ultrathin iron (<50 Å) and copper (<130 Å) films. The ion-beam-induced nanosmoothening occurs while material is being removed and gives rise to an initial decrease in resistance followed by a steady increase as the film is subsequently uniformly eroded. The shunt resistance associated with the resistance decrease is found to be independent of the thickness of the underlying film, thus indicating that the conductivity enhancement is due primarily to surface modification. © 2005 American Institute of Physics. [DOI: 10.1063/1.1861953]

Ion-assisted deposition refers to the technique in which a beam of noble gas ions, usually argon, with energies less than 1 keV, is simultaneously incident on a thin film during growth. The most prominent consequences of ion-assisted film growth include densification, modification of nucleation and growth, interface mixing, defect generation, and changes in topography and surface roughness. In contrast to this general technique of ion-assisted deposition, some investigators have reported on the use of a sequential technique in which thin films are first deposited and then, after deposition, exposed to an ion beam. Results of this approach include ion-bombardment-induced nanostructuring of Cu(001) surfaces, the formation of reproducible ripple structures on Si(001) and Ag(110) and (001) surfaces, and the roughening or smoothing of Si(001) and SiO2 surfaces where the result depends on ion type, energy, and angle of incidence. At the moderately higher energies of a few keV, interlayer exchange coupling in Fe/Cr/Fe trilayers can be controlled with He ion bombardment, and microscopic holes can be filled using pulsed Ar ion beams in a process called “ion-beam sculpting.” Many of these experiments thus provide strong evidence of ion-beam-induced nanoscale matter transport on solid-state surfaces, a process that promises to be useful in applications requiring nanotextured surfaces and interfaces.

We report on an additional and unexpected effect of ion milling on the resistance of ultrathin films. By monitoring in situ the resistance of ultrathin films of Fe and Cu that have been prepared in high vacuum and then exposed to an argon ion flux, we find, as shown in Fig. 1, that the resistance initially decreases to a substantially lower value and then increases as more material is removed from the film. The initial resistance decreases have been measured to be as large as a factor of ~25 for Cu and ~2 for Fe. The experiments are performed in UHV conditions, and the sheet resistance of the films is monitored in situ without breaking vacuum between the film growth and subsequent ion-beam exposure steps. The incident noble gas ions are chemically unreactive and simply transfer a fraction of their kinetic energy to the atoms in the film, a process which, as discussed earlier, can result in a significant modification of the surface morphology of the film and hence its physical properties. Using atomic force microscopy (AFM), we confirm an ion-beam-induced smoothening of our films and find that there is a reproducible correlation of the reduction in sheet resistance with ion-beam parameters (beam voltage and current density) and initial starting sheet resistance.

Thin films of Fe and Cu were, respectively, grown using rf magnetron sputtering and thermal sublimation through square-shaped shadow masks onto clean glass substrates at room temperature. Contact leads comprising Au(400 Å)/Cr(100 Å) bilayers were predeposited onto the glass substrate before loading into the chamber. Since ultrathin films are extremely air sensitive, the experiments were carried out with base pressures typically less than 5 × 10-9 Torr. The Ar ions had energies of 200 eV and a flux density of ~1014 cm-2 s-1. The sheet resistance of the films

![Graph](https://via.placeholder.com/150)

FIG. 1. Plot of sheet resistance (left-hand axis) and thickness (right-hand axis) as a function of time for an ultrathin Cu (upper panel) and Fe (lower panel) film. At time = 0 the ion beam is turned on. The resistance and thickness, measured simultaneously, show that as material is being steadily removed the resistance initially decreases to a minimum and then increases.

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was measured using a nonswitching van der Pauw technique \(^8\) with the four leads arranged to make symmetric contact at the corners of a square. This arrangement allows us to simultaneously measure both components \((R_1\) and \(R_2)\) of the resistance and to assess film homogeneity \((|R_1-R_2|)\) during growth. Film thickness was measured by a calibrated quartz crystal thickness monitor placed in close proximity to the sample and varies from 15 to 45 Å for the Fe films and 75 to 130 Å for the Cu films.

As shown in Fig. 1, the resistance of a Cu (top panel) and Fe (bottom panel) film undergoes a pronounced decrease, which is initiated at time \(t=0\) when the beam is first directed onto the sample. The resistance reaches a broad minimum and then begins to rise after about 50 s. This is an unusual result because the ion beam is expected to erode the film at a constant rate, as indicated by the linear decrease of the accumulated thickness of the material deposited onto the thickness monitor (right-hand axes). Apparently, during initial stages of milling, the resistance decreases as material is being removed. As discussed subsequently, we attribute this decrease to an ion-beam-induced smoothening of the film. If the ion beam is turned off, the resistance remains stable near that point. Auger electron spectroscopy of an ion-milled film compared with that of a pure film does not show the presence of impurity contaminants that might be inadvertently sputtered on the film by the ion beam and thus lower its resistance.

The relative decrease in the resistance of our ultrathin films due to ion milling is strongly correlated with their initial surface roughness. Figure 1 shows that for Cu and Fe films with similar initial sheet resistances near 5 kΩ, the minimum resistance reached during the ion-milling process is more than a factor of 10 lower for Cu (~200 Ω) than it is for Fe (~2.5 kΩ). An important insight into the cause of this difference is provided by our ex situ AFM studies, which show that Cu films grown by thermal sublimation have an rms roughness larger by a factor of ~3 than Fe films grown by sputtering. We have also compared the roughness of films not exposed to an ion beam with films ion milled close to the resistance minimum. As shown in Fig. 2, this comparison for a typical Cu film reveals that the ion-milled film (lower panel) has an rms roughness of 10 Å compared to 30 Å for an unmilled film (upper panel). The smoothening effect is also confirmed in AFM images of Fe films, which, with their initially smoother topography, exhibit a smaller reduction (factor of 1.2) in rms roughness. For even smoother films such as Gd and Pd, which become conducting almost immediately after deposition begins, we do not observe an initial resistance decrease.

There are also a pronounced increase in homogeneity associated with ion milling. If the films are homogeneous, then \(R_1\) and \(R_2\) should both show the same time dependence and have similar magnitudes. However, a thin film grows through various stages, starting with nucleation of isolated grains, then coalescence of the grains, and finally formation of a homogeneous system of well-connected grains. At the early stages of growth, the local resistance is extremely sensitive to local variations in temperature, incident flux, and pressure. Any gradients in these quantities can give rise to long-length-scale anisotropies in the electrical resistance and hence significant differences in \(R_1\) and \(R_2\). In our experiments, \(R_1\) and \(R_2\) can differ by factors as large as 3 for Fe and 20 for Cu films. However, when these “inhomogeneous” films are exposed to the ion beam, both \(R_1\) and \(R_2\) individually decrease to approximately the same minimum. Thus, the anisotropy measured by \(|R_1-R_2|\) and the total sheet resistance as measured by the van der Pauw combination of \(R_1\) and \(R_2\) simultaneously decrease.

We surmise that two competing processes are occurring during the ion irradiation process: (1) sputter erosion by the impinging ions preferentially removes atoms from the film at high points where they have less coordination with neighbors and hence less binding energy and (2) the nanoscale transport of material from high points (peaks) near grain centers to low points (valleys) between adjacent grains. The resulting nanosmoothening process dominates in the initial stages of the ion exposure, resulting in a decrease in resistance and an increase in homogeneity. From a microscopic point of view, the decrease in resistance can be attributed to a variety of mechanisms including: a transition from the diffuse surface scattering of rough films to specular surface scattering of smooth surfaces, \(^9\) the removal of foreign surface absorbates, \(^1\) and the filling in of high-resistivity weak spots at grain boundaries. Equivalently, one can interpret the results in terms of percolation, where the ion milling gives rise to a restructuring of the grains and a concomitantly lower
critical thickness for the onset of conductivity. Ion-beam-induced grain growth\textsuperscript{12} is probably not relevant because our incident ion energies are too low. At all stages of milling, the erosion of the film at a constant rate is occurring and, as shown in Fig. 1, this process eventually dominates over the smoothening process when the resistance starts to rise.

Using the parallel resistance formula, $R_{\text{p}}^{-1} = R_{1}^{-1} + R_{b}^{-1}$, we model the ion-beam-induced decrease in sheet resistance from an initial value $R=R_{0}$ to a minimum value $R=R_{m}$ as equivalent to connecting a shunt resistance $R_{s}$ in parallel with $R_{i}$. In Fig. 3(a) we plot the calculated values of $R_{s}$ versus $R_{i}$ for 22 different Fe films. We note the interesting result that $R_{s}$ (indicated by the horizontal dashed lines) is constant and on the order of 4500 $\Omega$ for ultrathin films with $R_{i}>1500$ $\Omega$ and constant, and on the order of 1400 $\Omega$ for thicker films with $R_{i}<1500$ $\Omega$. Figure 3(b) shows the dependence of $R_{s}$ on the ion exposure time $\Delta t$ needed to reach the minimum resistance $R_{m}$. Since the beam flux is constant for all the experiments, $\Delta t$ is proportional to the total number of ions incident on the films or, equivalently, the ion dose. We find that $\Delta t$ behaves similarly to $R_{i}$. The crossover in both plots near $R_{i}=1500$ $\Omega$, corresponding to a thickness of around 25 Å, most likely reflects a change in film morphology in which a smaller ion dose ($\propto \Delta t$) gives rise to a larger shunt resistance.

The observation of a constant shunt resistance that is independent of the underlying film implies that the initial resistance decrease is due primarily to surface modification. In the initial stages of ion bombardment, in which pronounced resistance decreases are observed, the lateral transport of material and the associated nanosmoothening dominates over the removal of material. With continued milling, the film is uniformly etched away and the resistance increases. While these techniques are clearly applicable to fundamental studies of thin films where the resistance can advantageously be externally tuned with an ion beam, they may also have applicability to the preparation of polycrystalline surfaces prior to the formation of tunnel barriers or to the improvement of interfaces in metallic bilayers or superlattices.

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