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Step decoration during deposition of Co on Ag(001) by ultralow energy ion beams

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A possibility for decorating atomic steps on single-crystal surfaces by using ultralow energy ion beams is reported. Isotopically pure ion beams are produced by a mass separator and subsequently decelerated by an electrostatic lens. The lens was designed to allow sweeping of the ion beam in order to obtain a uniform deposition over a large area. The preferred sites of single Co atoms on Ag are investigated with *in situ* scanning tunneling microscopy measurements. A clear indication is found that by increasing the energy of the deposited Co to several electron volts, an enhanced Co decoration of the Ag steps is induced. This technology opens perspectives for an increasing number of elements which can form self-organized nanostructures such as atomic wires on vicinal crystal surfaces. © *1999 American Institute of Physics.* [S0003-6951(99)03533-0]

In thin film growth a variety of deposition parameters such as the growth temperature, the deposition rate, and the energy of deposited species have a crucial influence on the resulting physical properties of the thin film. Nowadays, it is straightforward to vary the temperature of the substrate over a wide range with commercially available systems. Also the deposition rate can easily be varied over several orders of magnitude, e.g., by modifying the dissipated power of an electron beam gun and control of the evaporation rate with a quadrupole mass spectrometer setup. One of the growth parameters which is less well understood is the energy of the deposited particles. Varying the energy of the deposited species in a controlled manner is a very tedious job but can be done by using ion beams for which the accelerating voltage in principle can be varied.[1,2]

Here we describe a low energy ion beam deposition system and report its first results. Using *in situ* scanning tunneling microscopy we evidence the possibility to deposit ions at a sufficiently low energy so that the surface of the single-crystal substrate does not experience noticeable damage during deposition. Moreover we observed step decoration by Co on Ag(001) using ultralow energy ion beams (several electron volts). Using thermal deposition of Co on Ag with molecular beam epitaxy (MBE) (hence with typical deposition energies on the order of a few tens of milli-electron-volts) in similar growth conditions no such decoration effects can be observed, indicating the importance of the energy of the deposited species for the decoration process. The problem of a small sample size when using a focused ion beam can be overcome by sweeping the ion beam over larger areas. We indicate that the method has potential applications for the discovery of new substrate/film combinations which exhibit the phenomenon of step decoration and which can thus lead to fabrication of quasi-one-dimensional wires. To our knowledge step decoration has only been obtained previously for Co on Cu(111), Fe on Cu(111), and Fe on W(110).[3–5]

When an ion hits a surface it is usually not a gentle encounter because of the relatively high energies which are used in surface science [several kilo-electron-volts to a few mega-electron-volts as compared to the surface energy (a few electron volts)]. Depending on the energy of the ions, processes such as channeling along an array of atoms of the crystal, scattering from the surface, implantation, or sputtering events can be observed.[6] In ion beam deposition techniques such as ion beam sputtering the energy of the deposited elements exhibits a distribution. This energy distribution shows a pronounced maximum at an energy which is equal to one-half that of the surface binding energy. The average energy of the sputtered particles can be up to an order of magnitude larger since the distribution is asymmetric with higher population on the high-energy side of the maximum.[7] In other words, the atoms are deposited with an energy on the order of a few up to a few tens of electron volts. To investigate what is at the origin of the difference between films produced by, e.g., MBE and ion beam sputtering, a deposition method which allows a controlled energy variation is needed. The deposition method that we have used is capable of accurately controlling the energy in the region which is of interest for ion beam sputtering and thus will allow for a detailed investigation of the energy dependence of the sputter deposition process.

A 50 keV ^{59}Co ion beam is produced by the Leuven Ion Separator and is decelerated by an electrostatic lens which is mounted in a differentially pumped ultrahigh vacuum (UHV) section of the beamline. A Faraday cup and beam profile monitor are installed to monitor the current and shape of the beam entering the lens. Deflection plates can be used to steer and/or sweep the beam. In Fig. 1, a schematic view of the deceleration stage is shown. It consists of seven ring-shaped electrodes with increasing diameter toward the last electrode. The sample, which is incorporated in the latter one, can be inserted by a sample transfer stick which is also used as a Faraday cup behind the lens. The high decelerating potential of +50 kV is derived from the high voltage terminal and supplied to the last electrode (and the sample). The different electrodes are connected over 500 MΩ resistors to create a potential which decreases stepwise toward the ground. The

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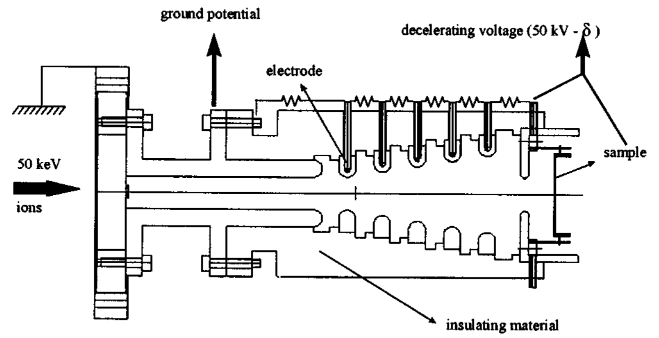

FIG. 1. Cross-sectional view of the ion beam deceleration optics. The voltage δ can be varied between a few volts and 1 kV and allows one to modify the incident particle energy.

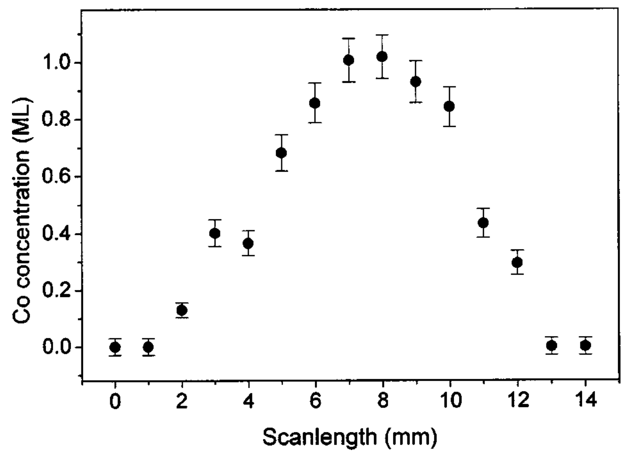

FIG. 2. Lateral Co thickness profile for a layer deposited with 10 eV on a Ag surface as determined with Rutherford backscattering spectroscopy.

applied high voltage is directly derived from the ion source anode potential reduced by an additional voltage which allows the ions to reach the sample with an energy variable from 1 keV to 1 eV. Hence, the ions are accelerated toward the entrance of the lens and subsequently decelerated in the lens to reach the sample with the desired energy. The lens itself was designed using the SIMION program, which calculates both the electrostatic field and the ion trajectories.[8] The simulations were performed for a 50 keV incoming ^{59}Co beam which was decelerated to energies ranging from 50 down to 1 eV. Two parameters are found to be crucial for optimal beam transport. First, a nearly perfect axial passage of the ion beam through the lens is important to avoid a strong increase of the beam spot size at the target. We used a beam with diameter of about 4 mm in a lens with electrodes varying in diameter from 20 up to 50 mm. Second, we observed that the behavior of the ion beam inside the lens is extremely sensitive to the position of its focal point. An optimal focus position is situated inside the lens a few centimeters in front of the sample position. Experimentally optimum beam settings are obtained by using a Faraday cup behind the lens while no high voltage was applied. This results in an axial beam with a focus at the second Faraday cup. Second, high voltage is applied to the lens and the focus of the beam is shifted toward the lens while optimizing the beam in the second Faraday cup. The ion current and thus the deposition rate is determined by integration of the signal at the beam profile monitor. We typically used a current of 150 nA, which gives a deposition rate of about 0.002 Å/s.

Ag substrates were prepared in a Riber MD*S*32 molecular beam epitaxy facility. A 1000-Å-thick Ag single crystal was deposited on polished MgO(001) held at ambient temperature. The MgO substrate was chemically cleaned in isopropanol and annealed to 600 °C for 30 min. To enable smooth and epitaxial growth of Ag on MgO(001) a 50 Å Cr buffer layer was first grown on MgO at 175 °C. The epitaxial relations are Ag[100](001)//Cr[110](001)//MgO[100](001). The reflection high-energy electron diffraction (RHEED) pattern after growth reveals the presence of some remaining roughness. Annealing to 200 °C in the ultrahigh vacuum (UHV$<5\times10^{-11}$ mbar) and subsequent gently cooling down allows the quality of the Ag substrate to further improve until a sharper RHEED pattern was observed.

The Ag substrate was then transferred *in vacuo* from the growth chamber to adjacent surface analysis chambers with facilities for Auger spectroscopy and scanning tunneling microscopy (UHV$<2\times10^{-10}$ mbar) to further characterize the surface quality. The Ag surfaces are concluded to be extremely clean since no traces of any impurities could be observed using Auger electron spectroscopy. The clean surface shows monoatomic steps (2 Å high) between terraces of several hundreds of angstroms average width. The atomic steps of the MBE grown Ag film have a varying distance and orientation. This can be due to the presence of small angle grain boundaries. The mosaic spread of such films as measured by x-ray diffraction rocking curves is typically 0.5°. All images are taken with the sample at ambient temperature. Ion beam depositions were performed in an UHV chamber (UHV$<1\times10^{-10}$ mbar). A layer of about 1 monolayer (ML) Co was deposited with a 10 eV ion beam. To protect the sample from oxidation a 20 Å Ag epilayer was MBE deposited on top of this Co layer immediately after the ion beam deposition.

The Ag capping layer also avoids sputtering of the Co layer during subsequent Rutherford backscattering spectrometry (RBS) measurements, which were used to determine the lateral homogeneity of the deposited layer. The measurements were performed with a 2 MeV ^{4}He$^+$ ion beam, and a scattering angle of 172°. Figure 2 shows a lateral thickness profile of the deposit. Within the experimental error, the profile is nearly uniform over a region of about 4 mm, which is

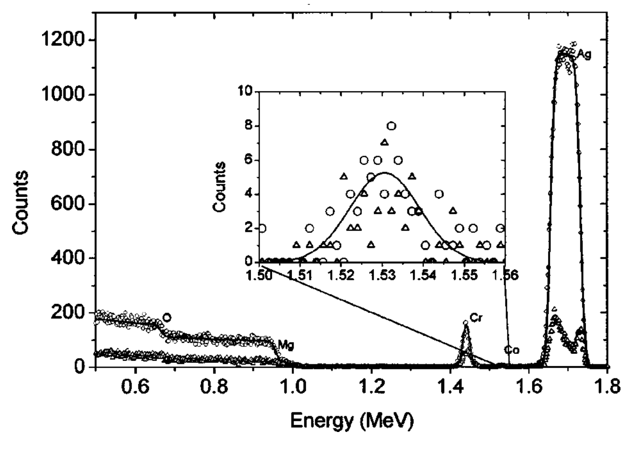

FIG. 3. 2 MeV ^{4}He$^+$ random and aligned Rutherford backscattering spectrum of 1 ML Co deposited with 10 eV on Ag(001)/Cr(001)/MgO(001).

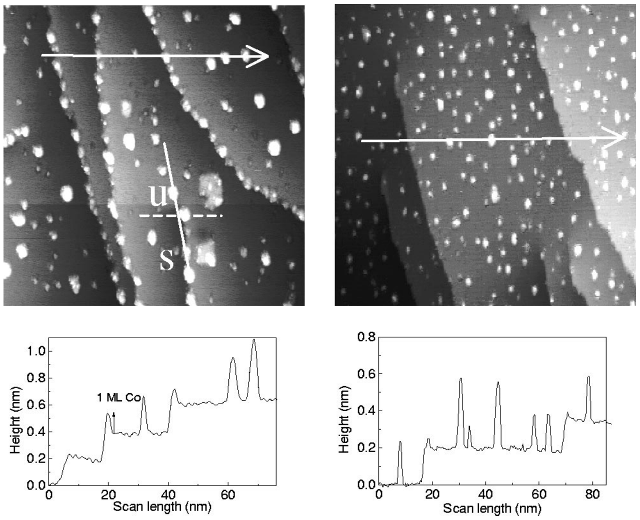

FIG. 4. STM image and height profile along the white arrow of 0.1 ML Co deposited on Ag(001) with energy of 15 eV (left panel) and deposited with thermal energy (right panel). In the ion beam deposited layer the upper Ag steps are decorated with Co islands of various heights. Co islands nucleate also near a screw dislocation along the step direction both in the unslipped (*u*) and the slipped (*s*) region.

exactly the length over which the ion beam was swept during deposition. Due to the finite width of the ion beam the Co coverage does not abruptly drop to zero but exhibits a gradual decrease on the sides.

The crystalline quality of the layers is determined prior to and after the Co deposition from channeling measurements. We find no evidence of any damage to the Ag epilayer (see Fig. 3). The minimum yield in the Ag layer is equal to 12%. There is nearly no He channeling observed in the Co signal (see inset Fig. 3). Due to the large lattice mismatch between Ag and either fcc or hcp Co, and due to their difference in the surface free energy, the Co layer does not grow pseudomorphically in a layer by layer mode. This probably results in highly strained, distorted islands which show poor registry with the Ag substrate and thus do not show a clear channeling effect.

In the left-hand panel of Fig. 4 a scanning tunneling microscopy (STM) topography image for a nominal coverage of 0.1 ML Co deposited at ambient temperature using a ^{59}Co ion beam with an energy equal to 15 eV is shown. One clearly observes two types of nucleation sites: decoration of steps on the upper side of the terraces and islands in the terraces. For this nominal Co coverage of 0.1 ML, the islands do not have a uniform height distribution—confirming that the Co islands do not grow layer by layer. At the upper side of the steps, the islands are mostly 1 ML high while those formed on the terraces are higher (see Fig. 4, left-hand panel). Co islands also nucleate along the step both in the slipped and the unslipped part of the crystal near a screw dislocation. Seemingly the stress field of the dislocation is sufficient to act as a nucleation center for Co island growth. At a deposition energy of 15 eV, the Co islands preferably nucleate in substrate regions where the strain caused by the island can be easily accommodated by a relaxation of the underlying substrate. This is, e.g., the case for Co on the upper side of a step where the underlying Ag step atom has a larger lateral freedom.

The importance of the energy of the incident Co particles for the step decoration process is determined by comparing the ion beam deposited Co layers with thermally deposited Co. The right-hand panel of Fig. 4 shows a STM topograph of 0.1 ML Co deposited at a rate of 0.02 Å/s on Ag(001) by MBE. The complete absence of step decoration is striking. Due to the low surface mobility of thermally deposited Co atoms on Ag, the Co islands are on average larger in number and smaller in size than for an ion beam deposited layer.

Finally, we indicate that this deposition method allows us to investigate the magnetic properties of Co nanostructures formed at steps on vicinal crystal surfaces by emission Mössbauer spectroscopy measurements. Using this ultralow energy ion beam deposition, ^{57}Co can be incorporated in, e.g., atomic wires. The sensitivity of emission Mössbauer spectroscopy is several orders of magnitude better than absorber Mössbauer spectroscopy and will allow the investigation of magnetic ordering and magnetic anisotropy of atomic wires.

In conclusion, we have shown with STM that Co atoms deposited on Ag by ultralow energy ion beam deposition with a deposition energy of 15 eV do not damage the Ag surface. Due to their excess kinetic energy compared to thermally deposited atoms, the Co ions have sufficient surface mobility to reach substrate regions with a low nucleation barrier, resulting in a strong preference for decorating Ag steps on the upper side. We have shown that the growth morphology of submonolayer films of Co on Ag can be changed drastically by increasing the Co energy by several orders of magnitude with respect to thermal deposition. This clearly shows the importance of the energy of the deposited species. Using this method one can accurately control the energy of the deposited material and this will allow a detailed analysis of the energy dependence of the deposition process.

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