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Monitoring growth of ultrathin films via ion-induced electron emission

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H^+, He^+, and N^+ ions with energy of 25 keV are scattered under a grazing angle of incidence from a clean and flat Cu(001) surface during deposition of ultrathin Co films. Making use of the ion-induced emission of electrons allows us to monitor growth of thin films via simple measurements of target current or from energy spectra of emitted electrons. The method provides excellent signals and is also applicable in the regime of poor layer growth.

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The physics of ultrathin films plays an important role in fundamental research as well as technological applications. As a prominent recent example, we mention the "giant magneto resistance" (GMR) effect[1,2] present in ultrathin films showing antiferromagnetic couplings. This effect is the basis for the function of reading heads used in modern magnetic hard disk drives or in sensor technology.[3] In particular for fundamental research, the preparation of defined ultrathin films in the monolayer (ML) regime is crucial. An established method is epitaxial growth of thin films via "molecular-beam epitaxy" (MBE) (Ref. 4), where atoms from an evaporator source are deposited on monocrystalline substrates.

In the characterization of films, monitoring of growth plays an essential role, since thickness and growth mode determine decisively the structure and the functioning of the system. A widely used technique to inspect growth of thin films in the monolayer regime is "reflection high-energy electron diffraction" (RHEED) (Ref. 5), where keV electrons are scattered from the surface of the film under a grazing angle of incidence. In a simple picture, the intensity of reflected electrons depends on the "smoothness" of the film surface. Then, e.g., for "layer-by-layer growth" the morphology changes periodically with coverage, and intensity oscillations for reflected electrons are observed. Aside from the powerful features of RHEED, diffraction phenomena make the general interpretation of data a nontrivial problem.

As alternative probes for monitoring film growth, scattering experiments with other microscopic particles are feasible which show a dependence of the backscattered yield on the morphology of the film surface. An interesting alternative to RHEED is scattering of fast atoms or ions instead of fast electrons.[6–10] This technique is similar to RHEED, however, it bears the advantage that the projectile trajectories can be described classically in terms of pure kinematical concepts.[11] For growth of thin semiconductor and metal films, grazing scattering of keV ions has been proven to be a powerful tool to study details on growth mode, island densities, critical island size, etc.[6–12] As an example, we mention studies on growth of ultrathin Co films on Cu(001), where a deviation from a monotonic Arrhenius type of dependence for the island density as a function of inverse growth temperature is observed.[13] This unusual behavior of film growth is attributed to intermixing of film and substrate atoms at elevated temperatures as predicted from density-functional theory (DFT) calculations and kinetic theory.[14]

In this Communication we present a simple method to monitor growth of ultrathin films using ion beams. Instead of recording intensities of projectiles for specular reflection, we propose to make use of electron emission phenomena induced during ion impact on the film surface. A similar procedure was proposed earlier for secondary electrons induced by impinging keV electrons,[15] where oscillations of the electron yield as function of coverage for layer-by-layer growth were observed. This yield shows only a weak variation of a few percent with coverage so that this technique did not find a wider application. Here we observe for the emission of electrons induced by light ions, pronounced variations of electron yields as function of film coverage. Based on our observation we propose a unique method to monitor growth of ultrathin films which has the potential of excellent signals and is simple in use.

In the experiments we have scattered light ions (H^+, He^+, N^+) with 25 keV energy from the target surface under a grazing angle of incidence Φ_{in} of typically 1°–2°. The target was a clean and flat Cu(001) substrate prepared by cycles of grazing sputtering with 25 keV Ar^+ ions and subsequent annealing. Ultrathin Co films were grown on the substrate using an electron-beam evaporator (Omicron, EFM3) with typical deposition rates of about 0.2 ML/min. For grazing collisions of keV ions with a surface, scattering proceeds in the regime of planar surface channeling,[16] where projectiles are specularly reflected from the topmost layer of the surface. At a distance of 85 cm behind the target, reflected projectiles were recorded by means of a channeltron detector with a 0.5-mm aperture. The current at the target during ion bombardment was measured with a nA meter. Energy spectra of emitted electrons were taken with a CLAM2 spectrometer (Fisons Instruments).

In Fig. 1 we present data for the initial growth of Co on a clean Cu(001) surface at room temperature (300 K) and at 243 K. The curves in the lower panel represent the intensity of specularly reflected 25 keV He^+ ions as function of Co coverage which reveals an oscillatory structure. This behavior results from a periodic change of the surface morphology during layer growth and can be analyzed using classical trajectory computer simulations and modeling of the surface structure.[11] In a simple interpretation, intensity oscillations—observed in a similar manner for keV electrons using RHEED—can be understood by the reduced reflectivity of the "rough" surface of an open layer. Whenever a layer is

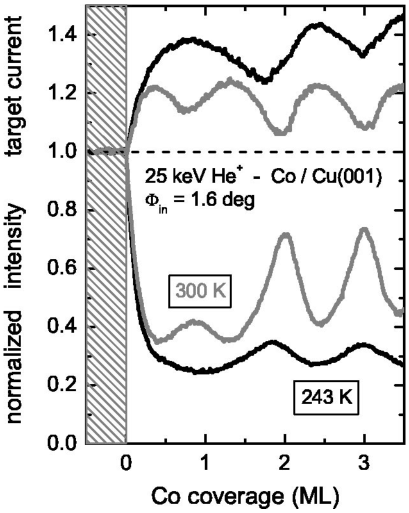

FIG. 1. Normalized intensity (lower panel) and target current (upper panel) as function of Co coverage on Cu(001) for scattering of 25 keV He$^+$ ions under $\Phi_{in}=1.6°$. Gray curve: growth temperature 300 K, black curve: 243 K.

completed, the intensity for reflected particles reaches a maximum and an oscillatory variation with coverage is observed. Important details on growth mode, island density, critical cluster size, etc., can be derived from those data recorded *in situ* and in real time.[7–10] For the present conditions, we conclude a type of initial bilayer growth at 243 K and for consecutive layers, as well as for the complete 300 K data, a layer-by-layer growth.

The curves in the upper panel in Fig. 1 represent measurements of the uncompensated target current (normalization: 30 nA=1) which consists of the ion current of the incident beam and of the additional current owing to emission of electrons during ion impact on the surface (total electron yield is about 5 here). These measurements are simpler to realize, because—instead of a scattering experiment—a current from the target to ground has to be measured only. The target current for grazing scattering of 25 keV He$^+$ ions as a function of Co coverage reveals also an oscillatory structure. However, these oscillations show a reversed behavior compared to the intensity of scattered projectiles. For the "smooth" surface of a completed layer electron emission and target current are reduced, whereas for the rougher surface of an open film layer these quantities are enhanced.

For the mechanisms of this experimental method, we give some qualitative arguments. We interpret the increase of the target current (i.e., enhanced ion-induced emission of electrons) for the "rough" surface of uncompleted layers by enhanced contributions of binary collision of incident ions with individual atoms at the surface. Contributions from this type of collision are suppressed for a smooth surface, where under channeling conditions the projectiles are steered in a sequence of small-angle scattering events with large impact parameters relative to individual target atoms.[16,17] These conditions are substantially changed for a surface with defects, since defect structures affect trajectories during channeling and give rise to binary encounters under small impact parameters. In such "violent" collisions, projectiles can undergo large angular deflections which will also enhance the probability of projectiles to cross the vacuum-solid interface and to penetrate into the subsurface region. Here projectiles probe higher electron densities than in the selvedge of the surface, and might have longer trajectories than for specular reflection in front of the surface plane.

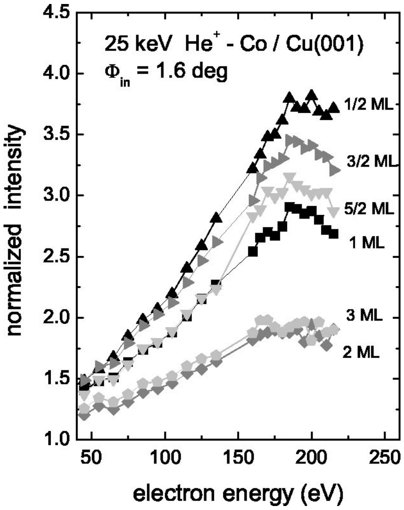

FIG. 2. Normalized intensity of emitted electrons as a function of electron energy for impact of 25 keV He$^+$ ions on Co/Cu(001) under $\Phi_{in}=1.6°$ for different Co coverages.

Support for this interpretation of data comes from recent studies on the number distributions for emitted electrons observed with a surface barrier detector biased to a high voltage (some 10 kV) (Ref. 18) for grazing impact of light atoms and ions on a flat and clean surface. In these experiments, we could demonstrate that the enhanced emission of electrons for an azimuthal orientation of the beam along low index crystallographic directions in the surface plane stems from fractions of projectiles which have penetrated into the subsurface region of the target.[19] These projectiles give rise to emission of a higher number of electrons per ion and dominate the overall increase of the total electron yield. Our measurements with this technique reveal similar effects for changes of surface morphology and support the interpretation.

For the experiments outlined above, main contributions to the target current result from electrons emitted to vacuum with a finite kinetic energy. We have investigated via electron energy spectroscopy the effect of film growth on the energy distribution of emitted electrons. We recorded electron spectra for impact of 25 keV He$^+$ ions on the Cu(001) substrate and on films with a Co coverage of up to 3 ML grown at room temperature. In Fig. 2 we have plotted the electron intensity normalized to the data for the clean substrate as a function of electron energy for intervals of 1/2 ML. A striking feature is a pronounced increase of the intensity for larger electron energy. The variation of intensity for half filled and completed ML increases also with electron energy.

We make use of the intensity at specific electron energies to study growth at room temperature. In Fig. 3 we show normalized intensities [integrated over electron energies (145±10 eV)] for impact of 25 keV He$^+$ ions (solid curve)

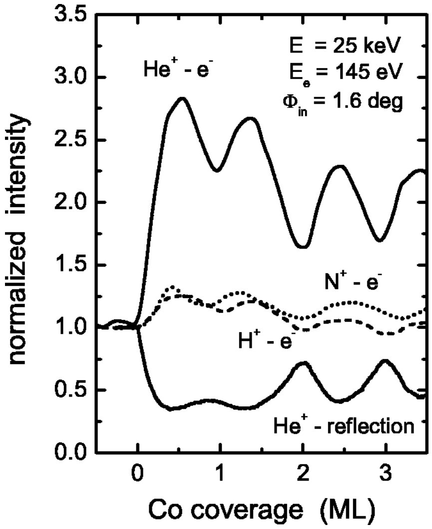

FIG. 3. Normalized intensity of electrons (energy 145 eV) as a function of Co coverage for impact of 25 keV H^+ (dashed curve), He^+ (solid curve), and N^+ ions (dotted curve) from Co/Cu(001) film under $\Phi_{in}=1.6°$. Lower solid curve: intensity of specularly reflected He^+ ions.

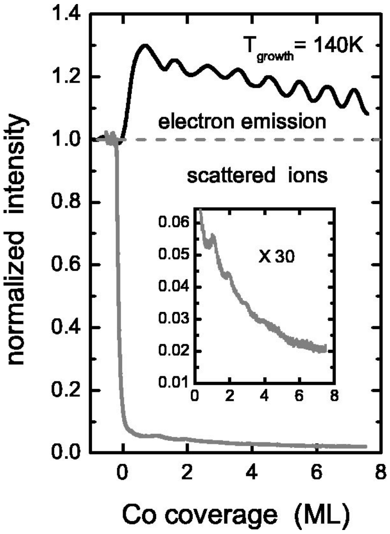

FIG. 4. Upper panel: Normalized intensity of emitted electrons (energy 100 eV) as a function of Co coverage for impact of 25 keV H^+ ions under $\Phi_{in}=1.6°$ from Co/Cu(001) film grown at 140 K. Lower panel: Same, but for specularly reflected ions. Inset: enlarged scale.

as a function of Co coverage. We also plot data for incident protons (dashed curve) and N^+ ions (dotted curve) and from scattering experiments with He^+ ions (solid curve, cf. Fig. 1). We observe pronounced oscillations with He^+ projectiles which clearly exceed the variation of the signal for the two other ions and also for the intensity of reflected projectiles. For smaller angles of incidence this effect is even stronger.

The origin for this observation is not clear at present. We state that electrons emitted with kinetic energies around 145 eV in collisions of 25 keV He^+ ions with a metal surface can only be understood in terms of binary collisions under small impact parameters. The energy transfer in collisions of He^+ with conduction electrons (maximum velocity, Fermi velocity) is about 60 eV here.[20] So higher electron energies can only stem from collision with electrons bound in atoms (Compton momentum profile).[21] For the smooth surface of a completed layer, collisions with small impact parameters are suppressed owing to channeling; however, for uncompleted layers or deposition different from layer growth the probability for binary collisions with individual atoms at the film surface is substantially enhanced. From the data we conclude that this process depends particularly on surface morphology for He^+ ions.

The enhanced signal observed for emission of electrons with relatively high energies bears the potential to apply this method in cases where ion reflection has shown its limits to monitor growth at low temperatures. As an example we present in Fig. 4 data for deposition of Co atoms on Cu(001) at a temperature of 140 K. In the lower panel of Fig. 4 we display the intensity of specularly reflected 25 keV H^+ ions from the film surface which shows a pronounced decay with Co coverage.[22] For initial growth, weak indications for growth oscillations can be identified, however, with further coverage the oscillations vanish (see enlarged scale in inset), and the number of ML deposited on the substrate can be estimated only via the flux from the evaporator source. This is different for the intensity of 100 eV electrons induced by 25 keV H^+ ions (upper panel of Fig. 4). The oscillation amplitudes are reduced compared to pure layer growth, but we resolve here oscillations up to 8 ML and possibly beyond this regime.

At low temperatures, RHEED also shows oscillations for a number of metal systems [Cu/Ag(100), etc.],[23] partly ascribed to a variation of the step density of the film. At present, we would tentatively interpret our finding in a similar manner, where inspection of data reveals almost constant oscillation periods (cf. Fig. 4) and would allow one to follow quasilayer coverage of the film.

Finally, we mention that the modification of growth and film surface by incident ions was checked to be on a negligible level by a controlled variation of the ion-beam current. Use of light ions with low currents (1 nA up to some 10 nA) result in negligible sputtering effects here. On the other hand, enhancement of current densities and use of heavier projectiles bear the potential to apply concepts of *ion-beam-assisted deposition* (IBAD) (Ref. 24) during film growth.

In summary, we propose electron emission to study real-time growth of ultrathin metal films via grazing scattering of keV ions from the film surface. In its simplest variant, the method can be applied by recording the target current with a current meter. In general, one might apply this method using a low intensity beam from an ion gun available in most UHV setups for sputter cleaning. Detection of energy-resolved electrons provides interesting features. For electrons induced by fast ions with energies of about 100 eV we observe pronounced oscillation amplitudes. At low temperatures in the regime of poor layer growth, we could still monitor specific numbers of ML deposits.

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