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Preparation of smooth Fe (001) on MgO (001)

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Abstract. The high mobility of metal adsorbates on perfect oxide surfaces favours an island-growth mode, which results in a granular morphology for metallic thin films. At low temperature, a fast substrate coverage is achieved due to the reduced mobility which, on the other hand, inhibits the development of an atomically smooth surface. In this study we used a controlled procedure to grow smooth epitaxial films of a few nanometres of Fe on MgO (001). The development of the dynamic conductivity as a measure of morphological roughness was observed during metal deposition by means of in-situ infrared spectroscopy. At various steps of thin-film preparation we used low-energy electron diffraction (LEED) for a characterisation of the crystalline surface quality. With ex situ atomic force microscopy (AFM) we investigated the surface topology of the prepared films with respect to smoothness. For example, for a controlled-growth 2-nm film, we observed a sharp LEED pattern and a metallic dynamic conductivity, and we did not find the distinct grooves which are characteristic of a granular structure.

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Due to the small lattice mismatch (3.5%) between MgO (001) and Fe (001), this material combination offers the possibility to grow an epitaxial magnetic thin film on a non-magnetic insulator, which is a basic requirement for the study of electronic transport in a low-dimensional ferromagnet. At the present time, the interest in the investigation of planar tunnel junctions draws further attraction towards this system.

Almost ten years ago, Li and Freeman [1] calculated a strongly enhanced magnetisation for a monolayer of Fe on MgO (001). Much effort has been expended to verify this prediction. Actually, the strong tendency towards island growth [2–5], which is typical for most metal-on-insulator systems, has inhibited the verification. The deposition at high substrate temperature results in a coarse island structure, which leads to a pronounced granular morphology of continuous films [4]. Magnetic measurements on such continuous films find strong anisotropies [6] and a superparamagnetic behaviour [7]. At room temperature, a complete substrate coverage is achieved at a thickness of ~ 1 nm [2, 8]. This value is less than on other metal-on-insulator systems, but it is still too high to prove Li and Freeman's [1] prediction. In addition, the transport properties of these just-covering films are dominated by a strong (i.e. fast) electronic relaxation [8, 9] and the dc conductivity shows an abnormal temperature dependence [10]. At low temperature, the MgO surface is completely covered by deposition of ~ 0.3 nm iron. Nevertheless, it is probably the inclusion of defects due to the low adatom mobility that prevents a remanent homogeneous magnetisation [10] for these films.

We investigated the preparation of smooth Fe thin films by a controlled-growth procedure which is based on the knowledge of both the heteroepitaxial growth of Fe on MgO (001) for various substrate temperatures [2, 3] and the homoepitaxial growth on Fe (001) [11–13]. In addition, we took advantage of the observation that surface defects, due to crystal cleavage in air, e.g., help to completely cover the MgO substrate [8]. The controlled-growth procedure consists of a Fe pre-coverage of the MgO (001) at low adatom mobility followed by a smoothing second coverage at enhanced mobility. As we will show in this work, the choice of substrate temperatures and film thicknesses is crucial for the final smoothness of the film.

For a surveillance of thin-film growth during deposition in ultra-high vacuum (UHV) we used infrared (IR) spectroscopy, i.e. we measured the transmission at normal incidence. Due to the strong correlation between electronic transport and the morphology of a metallic thin film [3, 8, 9], these measurements allow us to identify the percolation threshold of the growing film or to detect an increase or decrease of metallicity, respectively. We examined surface crystalline order and crystalline orientation in reciprocal space by low-energy electron diffraction (LEED). The development of regular facets, which accompanies the growth of Fe islands on MgO at high adsorbate mobility or which occurs

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with Fe (001) homoepitaxy, should be revealed by a splitting of LEED spots [2, 12]. As a non-integrating method in real space, we used ex situ atomic force microscopy (AFM). The natural components of air prevent an atomic resolution but the long-range morphology can be examined [4, 5]. Our AFM studies of naked (i.e. without an additional protection layer) films of various thicknesses and grown at various substrate temperatures showed a clear variation in morphology.

1 Experimental

Our in situ experiments were performed with a combination of a vacuum Fourier-transform infrared (FTIR) spectrometer (Bruker IFS 66 v/S) and a UHV chamber ($< 2 \times 10^{-10}$ mbar) [14]. This set-up allows infrared-transmission spectroscopy during thin-film growth under UHV conditions and surface-structure analysis by LEED. The monitored metal-vapour flux from an electron-beam evaporator hit the MgO (001) surface at an angle of 37.5° with respect to the surface normal. The film thicknesses are calculated from the deposition rate (typically 0.15 nm/min assuming Fe bulk density), which was calibrated with a quartz microbalance. For this experiment, IR-transmission spectroscopy from 1500 cm^{-1} to 5000 cm^{-1} with a resolution of 32 cm^{-1} was performed at normal incidence during Fe deposition. While 100 scans were sampled to give one spectrum the film thickness increased by ~ 0.02 nm.

The MgO (001) substrates were prepared in air by cleavage of $7 \times 7 \times 15$ mm^3 sized single crystals. After cleavage the substrates were transferred to high vacuum within a few minutes and were baked for 20 h at ~ 473 K. After transfer to UHV they were degassed at 723 K for 2 h. Then, the substrates showed the typical LEED pattern (see Fig. 1a). The Fe films were prepared on these surfaces by two subsequent depositions at various temperatures. For example, for a 2 nm thick film, an initial layer of 1 nm thickness was grown while the substrate was held at room temperature. Then, the temperature was raised to 670 K and Fe was deposited up to the final thickness.

Ex situ AFM (Park Scientific Instruments) measurements of the prepared films were made in the contact mode with 5 nN repulsion using a commercial Si tip with a radius of ~ 10 nm and an internal angle of 10°.

2 Results

The results of our IR optical measurements during growth of the initial layer and the second layer are shown in Fig. 2. The selection of measured spectra in Fig. 2a demonstrates that IR transmission is reduced in the whole spectral range with increasing thickness of the initial layer. Nevertheless, the film remains transparent with more than 70% transmission up to thicknesses above 1.5 nm, which allows its characterisation by means of IR transmission. From all the subsequently measured spectra, which are normalised to the transmission measured prior to deposition, we use the values at 2000 cm^{-1}

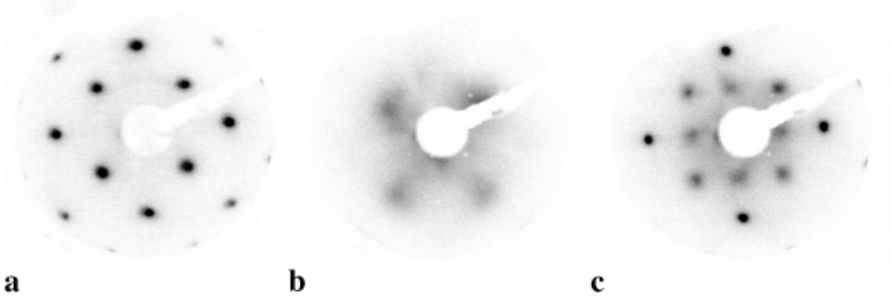

Fig. 1a–c. LEED patterns of **a** MgO (001), **b** 1 nm Fe/MgO (001) and **c** 2 nm Fe/MgO (001). Details of preparation are described in the text. The kinetic energy is 137 eV

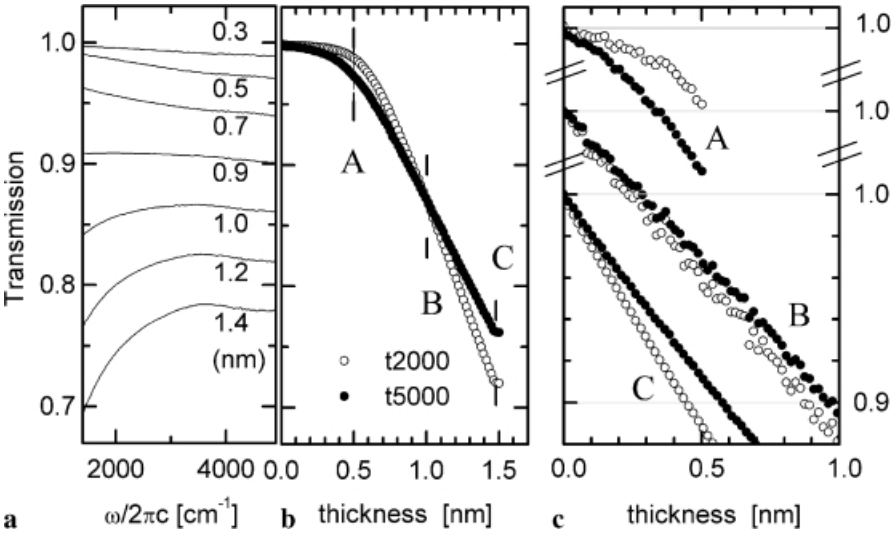

Fig. 2a–c. IR transmittance (at normal incidence) during growth of Fe on MgO (001) (see text). The labels A, B and C indicate variable thicknesses where the growth of the initial layer was stopped and the second deposition was started. **a** Selection of spectra (for film thicknesses as indicated) and **b** transmittance at 2000 cm^{-1} (*open circle*) and at 5000 cm^{-1} (*solid circle*) for growth of the first layer at 300 K. Unity means the same transmittance as the substrate. **c** IR transmittance during growth of the second layer at 670 K. Unity means the same transmittance as the underlying initial layer of 0.5 nm (A), 1 nm (B) and 1.5 nm (C) thickness

and 5000 cm^{-1} to characterise the development of absolute and spectral IR transmission with thickness (Fig. 2b and c). The transmission-versus-thickness curves for the initial layer indicate the negative dispersion (i.e. negative slope of transmission spectra in Fig. 2a) for the thinnest films and the IR optical crossover, i.e. the change to positive dispersion (positive slope) at ~ 1.0 nm. Around this thickness, the spectra show almost no frequency dependence (see Fig. 2a). For the second deposition the curves indicate an increase in dispersion, if starting from an at least 1 nm thick initial layer, but a decrease for an initial thickness of 0.5 nm (see Fig. 2c).

The LEED patterns in Fig. 1b and c were observed for the two steps of our controlled growth of a 2-nm Fe film. For the initial layer (1 nm deposited at ~ 300 K) we find an increased diffuse background and a few rather broad spots roughly at positions of the LEED pattern from the substrate surface. After deposition of the second layer (1 nm at 670 K) the (1×1) pattern re-appeared. Compared to the initial layer the pattern is sharper and the background is weaker.

Representative results from our ex situ AFM investigations of controlled-growth Fe/MgO (001) are shown in Fig. 3a for a smooth film of 2.0 nm thickness (1.0 nm at 300 K and 1.0 nm at 670 K) and, for comparison, in Fig. 3b for a rough film of 1.0 nm thickness (0.5 nm at 300 K and 0.5 nm at 670 K). The height range from black to white is 1.0 nm for both these scans over 160 nm $\times$ 160 nm. In the contour plot (Fig. 3c) of the surface of the 2-nm film, each line represents a height increase of iron monolayer thickness (0.144 nm). For ease of comparison of the different topologies, height profiles for the two films are given in Fig. 3d. They show that a change in height of about 0.5 nm is found on the 1-nm film within distances of ~ 10 nm while on the 2-nm film the same change extends over more than 40 nm.

3 Discussion

The results of our IR optical measurements during initial Fe deposition reveal the island-growth mode of Fe on MgO (001). As shown in an earlier work [8, 9], in the case of iron the optical crossover indicates a film thickness just beyond the percolation threshold, where the film already completely covers the substrate. As mentioned above, this is achieved at a smaller thickness on MgO cleaved in air compared to MgO cleaved in UHV [8]. From our experiments we also know that the time of exposure of MgO to air, the actual humidity and other contaminants sensitively influence the initial iron growth. As the films presented in this work were grown on substrates which, after cleavage in air, were quickly transferred to high vacuum (within 30 min), they should have a lower contamination than, e.g., thin films on chemically polished substrates.

From our calculations [8, 9] we know that the decrease of IR transmission with increasing thickness is dominated by the

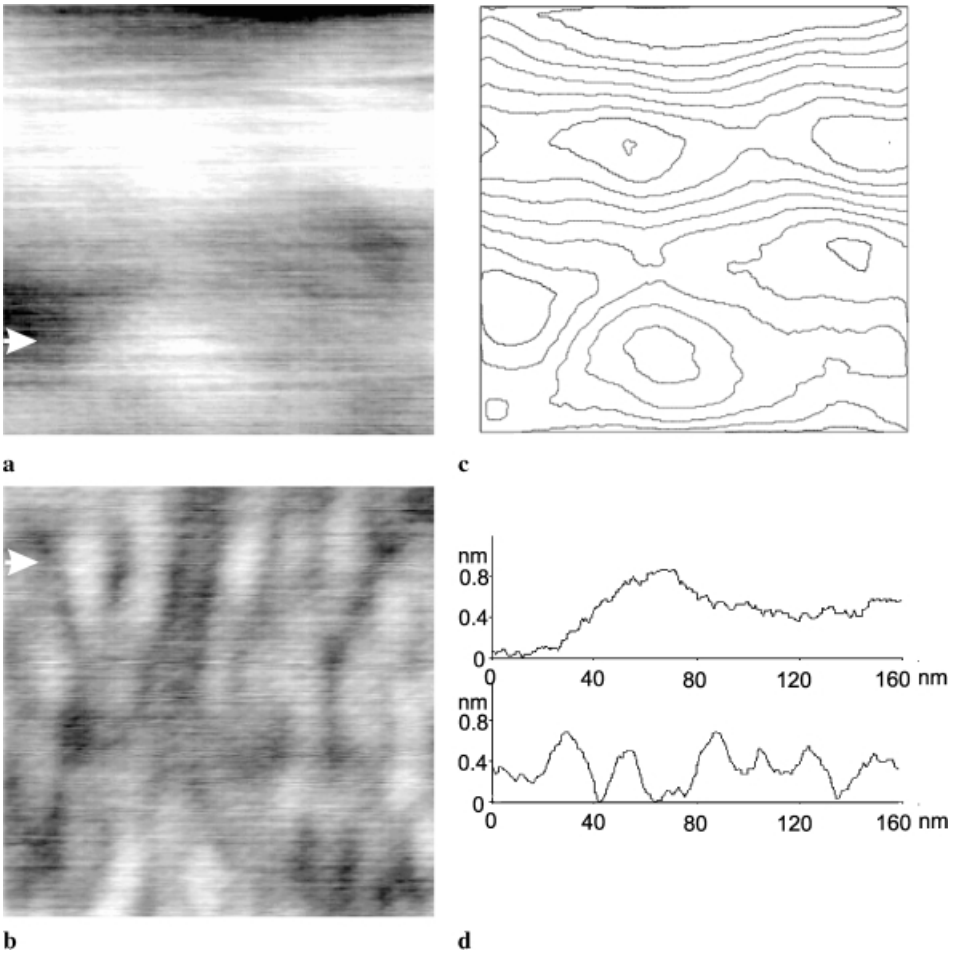

Fig. 3a–d. AFM image of **a** 2.0 nm Fe/MgO (001) and **b** 1.0 nm Fe/MgO (001) grown up to half the thickness at 300 K and then at 670 K. The scanned area is 160 nm $\times$ 160 nm and the height range from black to white is 1 nm. A linear background has been subtracted from the data. **c** Contour plot of the shown 2.0 nm Fe/MgO (001) image. The height difference between two neighbouring lines is 0.144 nm, which corresponds to the atomic step height. **d** Height profiles from the images in **a** and **b** along lines as indicated by the *arrows*

increasing absorption of the growing film. For a metal and frequencies in the middle infrared this decrease is expected to be the faster the lower the frequency [15]. In metallic thin films, where metallic dynamic conductivity develops while passing through the percolation threshold [8, 9], this spectral behaviour may be used as an indicator for metallicity. We do so for the in situ characterisation of the growing films during the second deposition period. The main question for this second step of preparation is: which initial layer thickness is sufficient to grow a homogeneous metallic film?

Our IR data from the second deposition (at 670 K) show an increase of metal-like behaviour for an initial layer of at least 1.0 nm thickness. In contrast to this, a decrease is found for 0.5 nm initial thickness and a large amount of metal (> 3 nm) is necessary to achieve strong metallic IR absorption. Obviously, for growing a smooth thin film there is a critical thickness for the change from low-temperature deposition to high-temperature deposition. Unless the surface is completely covered by a metallic film, heating the sample to 670 K and continuing the metal deposition leads to strongly inhomogeneous films. The growth mode of Fe on MgO (001) [2, 3] corroborates this observation. Above room temperature, there is a high probability for an adatom on MgO (001) to cross the Fe island border line and get on top of an Fe island. This leads to a weak lateral growth of Fe islands (see Fig.6 in [2]), hampers the coalescence of the island morphology and delays the percolation transition [2, 3].

The films prepared from a metallic initial layer (≥ 1 nm thick and completely covering the substrate) show an IR spectral behaviour which points at an increase of metallic character. Obviously, in these cases the adatom mobility causes a smoothing of the granular initial layer. This is consistent with two facts known from Fe (001) homoepitaxy. Firstly, above 500 K adatoms overcome the Schwoebel barrier at terrace edges on Fe (001) [12] and allow a mass transport from higher terraces down to lower terraces. Secondly, at 670 K islands nucleate on Fe (001) with a density of $\sim 2.5 \times 10^{-11}\,\mathrm{cm}^{-2}$ [11], which corresponds to island distances of roughly 20 nm. A step-flow growth mode is expected for a surface where the distances between terrace steps are below this value. For the initial film with grains of about 4 nm diameter, both the mass transport and the step-flow growth at the higher temperature lead to a smoothing of the surface and to a more homogeneous film thickness. As indicated by the IR results from films with various initial layer thickness (see Fig. 2c), this is only true if the initial layer completely covers the substrate, which is not fulfilled for a too thin pre-coverage (< 1 nm in this work) or for a too high temperature for the second deposition. From other experiments, we know that Fe complete coverages (< 10 nm) of MgO (001) are unstable above 740 K [16], i.e. the complete coverage lacerates as indicated by IR spectroscopy and by AFM.

We have further IR optical information on the surface quality of our controlled-growth films (≥ 2 nm) from CO-adsorption experiments [17]. The measured IR absorption shows only one strong vibrational resonance for the CO stretch mode. This points to a high homogeneity of the Fe surface. For comparison, on the surfaces of as-grown Fe films on MgO (001) at room temperature a variety of resonance structures is found [14, 18].

Apart from proving the epitaxial orientation of bcc Fe (001) films on fcc MgO (001), our LEED observations also corroborate the IR optical results. For 1-nm Fe/MgO (001) (grown at room temperature) the small number of spots, their broadness and the diffuse background point to scattering from different atomic terraces and from step edges. This finding corresponds to morphological roughness, which is expected for films close to the percolation transition [2, 3, 8, 9], and to atomic roughness, which is expected for growth at room temperature [11]. The situation is reversed on the surface of the 2-nm film. Sharp spots point to scattering from extended terraces. On variation of electron kinetic energy no splitting of these spots could be observed. This excludes the development of extended regular facets as found for thicker films of Fe on MgO (001) [12]. From our LEED experiment we also exclude the presence of large pinholes in the 2-nm film. We do not observe the distortions of the scattered intensity, which we know from incomplete coverages due to partial charging of the inhomogeneous substrate surface.

The final characterisation of the controlled-growth 2-nm films by ex situ AFM demonstrates their homogeneity by viewing their surface topology (Fig. 3a). Compared with the result of the 1-nm film (Fig. 3b), it appears smooth, i.e. it shows less variation in height on a larger length scale. The comparison of these two AFM images does also corroborate the interpretation of the IR-spectroscopy data of Fig. 2, i.e. for subsequent growth at room temperature and at 670 K, a total thickness of 1 nm (i.e. 0.5 nm at 300 K and 0.5 nm at 670 K) is insufficient for preparing a homogeneous film with a smooth topology. Furthermore, the height profile of the 1-nm film is in accordance with our IR measurements (Fig. 2c), which point at an incomplete coverage of the substrate with metal islands. There is no clear indication that the second deposition at the higher temperature (670 K) has smoothed the valleys between neighbouring grains.

Besides, in contrast to as-grown films (for substrate temperatures at room temperature [4] and above [5]), our controlled-growth 2-nm films do not show the pronounced granular structure, which is the result of island growth and hampered coalescence. For example, in AFM pictures of as-grown films [4] (at 300 K and 2-nm thick), grains of ~ 5 nm diameter are to be seen. These grains are separated by areas with a variation in height of > 1 nm on a lateral distance of < 1 nm. This is far above the roughness of our smooth films. A high roughness is found even for 10-nm films grown above 693 K [5]. The 10-nm Fe films show grains with < 44 nm diameter and, compared to our 2-nm film, a larger height variation (> 4 nm) on a much smaller lateral distance (< 10 nm).

From our experiments we deduce for the same initial layer thicknesses (1 nm), the smaller the final thickness (compared with 2 nm) the stronger the roughness. This should be due to topological reasons linked to the fact that the initial layer thickness of 0.5 nm is little beyond the critical thickness for a complete coverage of the substrate at 300 K [8]. Further deposition of Fe should only slightly increase the smoothness, which is due to the limitation of the step-flow growth mode. At 670 K islands nucleate on Fe (001) at distances of ~ 20 nm [11] which is a measure of the diffusion length. Therefore, a transition to a layer-by-layer growth mode (which maintains the actual topological roughness) starts when the terraces are much wider than this distance. Regarding the contour plot of Fig. 3c, we conclude that our 2-nm film has reached this limit on a large fraction of surface area.

The remaining long-range roughness with up to ~ 0.5 nm change in height on a distance of ~ 40 nm (this corresponds to 11 nm minimum width for atomic terraces, see Fig. 3d) is certainly not a fingerprint of heteroepitaxial growth kinetics. It is rather due to inhomogeneities of the substrate surface. Multilayer steps at distances down to 40 nm are known for cleaved MgO (001) [19].

4 Conclusion and summary

We have shown that the earlier proposed [2] preparation of thin Fe films leads to highly homogeneous films with atomically flat terraces. The thickness for changing from low-temperature (i.e. low mobility) deposition to high-temperature (i.e. high mobility) deposition is important to gain this smoothness. The remaining long-range roughness of the films should be due to inhomogeneities of the substrate surface. For the preparation of smooth films thinner than 2 nm, the mobility during the first deposition should be further reduced. In a very recent experiment we already took advantage of our smooth films for an investigation of the interaction of adsorbate vibrations with a metallic thin film [17].

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