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# Fe/Cu(100)—a test case for the understanding of epitaxially grown magnetic thin films

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Properties of ultra-thin films deviate from those of the corresponding bulk material already because of the mere reduction of dimensionality and, related to that, the reduction of symmetry. This applies to all types of collective phenomena as, e.g. geometrical structure, electronic properties and magnetism [1]. Also, the alternating combination of different film material stacked perpendicular to the surface can lead to completely new physical phenomena as, e.g., the giant magnetoresistance (GMR) effect, i.e. the enhanced sensitivity of the electrical resistivity to external magnetic fields. Within only about one decade after its discovery, the first GMR-based read-head devices appeared in commercial hard disks, and the near future might bring even non-volatile magnetic random access memory (MRAM) devices based on this technology [2,3]. For these reasons, thin films of magnetic materials have attracted a great deal of research attention.

Yet in addition to the reduced dimensionality, there is another factor with tremendous impact on the film properties: When the film is grown on a crystalline substrate it tends to assume the latter's lateral periodicity. Such *pseudomorphic growth* is realized when the energy costs for the distortion of the film's native lattice is overbalanced by the energy gained by the formation of the film–substrate interface. This is frequently the case when the substrate's binding-potential surface is strongly corrugated. In these cases, the thin film is an artificial material whose surface parallel lattice parameter deviates from that of the bulk material (usually accompanied by a tetragonal distortion). A completely different crystal structure may even result. An example for the first case is Ni on Cu(100), which simply continues the substrate lattice parameters up to a thickness of about 20 monolayers (ML) before it gradually converts towards Ni's bulk structure. In the whole pseudomorphic range, the film exhibits a constant, laterally expanded lattice parameter (≈2.5%) and likewise contracted layer distances (≈3%) [4]. In contrast, cobalt—whose native structure at room temperature

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is hexagonally close-packed (hcp)—exhibits pseudomorphic growth in face-centred cubic (fcc) structure on Cu(1 0 0) up to several tens of monolayers [5].

Compared to these structurally very simple systems, the epitaxial growth of iron on Cu(1 0 0) turned out to be a rather exceptional case. At room temperature, bulk iron crystallizes in a body-centred cubic (bcc) structure (α-Fe) with a lattice parameter of $a_{\mathrm{Fe}}^{\mathrm{bcc}} = 2.87$ Å appearing hardly compatible with the dimensions of the fcc Cu substrate ($a_{\mathrm{Cu}} = 3.61$ Å). On the other hand, the high temperature (1185–1667 K) modification of iron with fcc structure (γ-Fe) would fit to the Cu(1 0 0) substrate with nearly no mismatch at all. From that, the growth of practically undistorted fcc-Fe films was anticipated, offering the chance to experimentally study its magnetic properties, which were postulated by ab initio calculations to depend crucially on the fcc lattice parameter [6]. So it came as a surprise that a variety of different geometrical structures and also different magnetic states were found dependent on film thickness. (Also there is some influence of the preparation procedure.) Due to the richness of phases observed, Fe/Cu(1 0 0) has become an important prototype system for studying the intimate coupling between structure and magnetism. As such, it has attracted a great deal of attention, being the subject of hundreds of publications within the last 20 years. It certainly represents the most intensely investigated epitaxial system in this field.

A first classification for room temperature film growth (to which we restrict this discussion) was given by Thomassen et al. [7]. There are three regimes: In regime 1 (up to 4 ML Fe coverage), the whole film is ferromagnetic with the magnetization directed perpendicular to the surface. Rather complex superstructures ($(4 \times 1)$ and $(5 \times 1)$) are involved, which are due to the lattice distortion throughout the whole film and which feature expanded layer spacings, as revealed through quantitative low-energy electron diffraction (LEED) analyses by Müller et al. [8]. In regime 2 (up to about 10 ML coverage), the body of the film assumes a practically undistorted fcc-structure and is covered by a reconstructed $(2 \times 1)$-periodic overlayer whose spacing to the next layer is expanded [9]. It is only the top (bi-)layer which is found to be ferromagnetic within this regime. Finally, exceeding 10 ML coverage (regime 3), the whole film converts into iron's native structure with the film's bcc-[1 1 0] direction parallel to the surface (''Pitsch orientational relationship'') and with the magnetization direction in-plane.

The understanding that the basic film structure in regimes 1 and 2 is fcc (though heavily distorted) was challenged by recent scanning tunnelling microscopy (STM) investigations. For iron films in regime 1, Biedermann et al. [10] revealed zig-zag-like stripe arrangements with much larger lateral atomic displacements than ever expected. The local structure appears as a result of shearing the fcc-lattice by about 14° and so strongly resembles a bcc(1 1 0) surface which, however, is substantially strained in order to maintain an epitaxial relationship to the fcc(1 0 0) substrate. This is why the authors denote this phase as ''nano-martensitic'' [11]. Their assumption that the whole film undergoes such a shear deformation (which cannot be seen directly in STM) is corroborated by recent ab initio calculations by Spišák and Hafner [12] who revealed a shear instability for fcc-Fe films leading the atomic arrangements found by STM. Also, a re-analysis of the LEED data for the $(4 \times 1)$ structure used in Ref. [8] and applying a model based on the published STM data (which was not considered before) led to an at least equally satisfactory fit of model intensities to the experimental spectra [13]. With today's knowledge, this latent ambiguity can be traced back to both the reduced sensitivity of conventional LEED analyses with respect to lateral parameters as well as to the strong parameter couplings frequently occurring in multi-parameter data fitting. [1]

Biedermann et al. shed new light on this important Fe/Cu(1 0 0) system in this issue [14], which, as Biedermann et al. expressed elsewhere [11] may necessitate a new paradigm for understanding this prototypical magnetic thin film system. They extend their structural studies to an experimentally broad characterization

[1] For the LEED-analyses of the $(4 \times 1)$ and $(5 \times 1)$ structures, as many as 16 and 40, respectively, parameters had to be varied independently. With the computer power available at that time, this was at the limits of feasibility. Therefore, only ''reasonable'' models were tested. A detailed investigation of parameter interdependencies for these systems would be very demanding even nowadays.

of the Fe film in regime 2, with surprising results. They combined very detailed STM investigations with atomic resolution performed at various temperatures in the range 5–300 K with high-standard quantitative LEED analyses. It was necessary for the authors to use cryogenic STM imaging in order for them to detect the true ground-state structure of the fcc-Fe film's surface: ($(2 \times 2)$p4g rather than $(2 \times 1)$p2mg). At higher temperatures this structure is hidden to STM by rapid fluctuations of domain boundaries. Furthermore, its symmetry leads to an extreme weakening of intensities (to below the typical detection limit) just for its characteristic spots. From LEED intensity analyses based on the new model, its detailed geometrical parameters were determined as well. The result agrees with a former analysis for this phase [9] in that the subsurface part of the film is of fcc structure. This implies that the bcc-like layers of regime 1 have transformed to fcc. Disagreement occurs with respect to the arrangement of the top layer atoms, which Biedermann et al. find are laterally shifted in a direction rotated by 45° with respect to the former model. In their new model, lateral bond angles result which are very similar to those observed in regime 1. Also, due to the rotation of the shift direction directly towards the second-layer atoms, the origin of the expanded top-layer spacing has to be re-interpreted. While it was formerly attributed to the repulsion between ferromagnetically coupled fcc-Fe surface layers, it may now be traced back to steric reasons as frequently observed for surface reconstructions. They find the bond lengths between the first and second layer atoms are reduced, instead of being overall expanded as in the earlier model.

These results of the present article by Biedermann et al. [14] seem to modify only some atomic coordinates within an already existing structural model. However, they add the last parts into a long-standing structural puzzle. In doing so, they open up a completely different view of the structural diagram of Fe films on Cu(1 0 0) and provide a new understanding of the relationship between thin-film structure and magnetism of this important model system. Obviously, the community has to discard the idea that there is ferromagnetic fcc-Fe in the phase regimes of Fe/Cu(1 0 0). In contrast, it seems to be proven that whenever ferromagnetism appears in the whole of the films (regimes 1 and 3) or in part of it (regime 2), the local atomic arrangement is simply related to iron's native bcc-structure with similar bond angles, bond lengths and the reduced coordination involved. On the other hand, by no means can the films be regarded as regular bcc-Fe (with the exception of regime 3, of course). They are heavily strained and folded in regime 1, and the "bcc-like" surface in regime 2 is—strictly speaking—nothing else than a reconstructed overlayer. It is therefore not surprising that physical properties like the electronic structure or the size and direction of the magnetization lack correspondence to native bcc-Fe. Nevertheless, all future interpretation of those effects has to be based on these new structural insights. The crystallographers have done their part and the ball is now in the field of electronic structure and magnetism.

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