Growth and morphology of thin Fe films on flat and vicinal Au(111): a comparative study

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Abstract. The epitaxial growth of Fe on flat and vicinal Au(111) was investigated with scanning tunneling microscopy (STM) and low-energy electron diffraction (LEED) in a comparative study. Below a critical film thickness we found a pseudomorphic growth behavior of Fe on flat and vicinal Au(111). Above the critical Fe overlayer thickness a phase transition from fcc(111) to bcc(110) occurs on both surfaces. As a result of this phase transition rectangular-shaped crystallites are formed on the surface. The predominant orientation of the crystallites is along all \(\langle 01\overline{1}\rangle_{\text{fcc}}\) directions for Fe on flat Au(111). For Fe on vicinal Au(111), we observed that no crystallites are orientated along the step edges. For Fe on flat Au(111), we found that the phase transition starts at a higher Fe overlayer thickness but a lower Fe coverage is required before the transition is completed in comparison with Fe on vicinal Au(111). Our results obtained with STM and LEED allow us to directly link the growth directions of the bcc(110) crystallites with twofold symmetry to the crystallographic directions of the substrate surface with hexagonal symmetry.

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1. Introduction

The growth of ultrathin films is governed by various parameters, e.g. substrate temperature, lattice mismatch between adlayer and substrate and step density. In the case of magnetic films, the growth behavior has a strong influence on the magnetic properties of these films. In this contribution we focus on the differences between the growth of Fe on flat and vicinal Au(111). Fe on flat Au(111) has already been studied by various groups [1]–[5]. A structural phase transition from fcc(111) to bcc(110) has been observed by scanning tunneling microscopy (STM) [3] and transmission electron microscopy (TEM) [1]. Recently, the growth of Fe on vicinal Au(111), namely Au(788), was investigated by Shiraki et al with STM [6, 7]. This study was restricted to submonolayer thicknesses [6]. No information about the topography of a wider range of Fe overlayer thicknesses on vicinal Au(111) can be found in the literature. Especially, the phase transition from fcc(111) to bcc(110), which has an influence on the magnetic properties of Fe on Au(111), has not been studied so far for Fe on vicinal Au(111).

Clean vicinal Au(111) surfaces with a variety of miscut angles were extensively studied by Rousset et al [8]. It was found that some vicinal Au(111) surfaces exhibit large and regular terraces. These surfaces are stable against facetting and show a surface reconstruction. The reconstruction of the vicinal Au(111) surfaces stems from the characteristic reconstruction of flat Au(111): the Au(111) surface exhibits a $22 \times \sqrt{3}$ reconstruction, which is due to a non-uniform compression of the surface along the [110]-direction [9, 10]. Domain boundaries separate the fcc regions from fault-stacked hcp regions. These domain walls are so-called discommensuration lines. Strain minimization leads to a secondary surface reconstruction, i.e. the so-called herringbone reconstruction. The discommensuration lines are aligned in a zigzag pattern across the surface. The lines cause a small corrugation of 0.2 Å with a periodicity of 30 nm [8].

When the surface is cut under a miscut angle larger than 1°, the herringbone reconstruction cannot exist in its original form, because its periodicity is larger than the terrace width. Depending on the direction in which the crystal is polished under the miscut angle, different surface reconstructions and microfacets can arise. In our work, the crystal was cut along the [211]-direction. This results in a formation of {111}-microfacets. On this type of vicinal surface the discommensuration lines run perpendicular to the step edges [8]. The terrace width distribution of vicinal Au(111) with surface reconstruction is about two to three times smaller
than for vicinal Cu(111) of comparable terrace width [11, 12]. This reflects a strong repulsive step–step interaction, probably resulting from the reconstruction.

As mentioned above, the system Fe on flat Au(111) has been studied to a great extent. The main interest was the exploration of the magnetic properties of fcc Fe which differ from the bcc bulk properties. Especially, the strong out-of-plane anisotropy of fcc Fe was considered interesting for magnetic data storage and retrieval. For the fabrication of one-dimensional (1D) magnetic nanostructures, the vicinal Au(111) surfaces are promising templates due to the step-flow like growth behavior reported for Fe on Au(788) [7].

In principle, any vicinal surface can serve as a template for growing nanostructures [13], but with vicinal Au(111) a surface with extraordinarily regular terraces is at hand. This is an important requirement for the creation of a regular array of nanostructures. Wide terraces are favorable in order to grow wide detached stripes to overcome the superparamagnetic limit at a given temperature. Due to the particular interest in wide terraces, we performed our growth study on a vicinal Au(111) surface with even wider terraces than on Au(788). We characterized both the early growth behavior and the growth behavior of thicker films on this surface.

This paper is organized as follows: experimental details are given in section 2. Section 3.1 presents our results concerning the early growth behavior of Fe on flat and vicinal Au(111). Section 3.2 deals with the phase transition from fcc(111) to bcc(110). The phase transition was monitored with STM. In analogy to studies of the phase transitions of Fe on flat Cu(111) [14], we explain the observed differences in the growth behavior for Fe on flat and vicinal Au(111) beyond the phase transition with the fitting of the rectangular bcc(110) unit cells onto the hexagonal substrate surface. Low-energy electron diffraction (LEED) measurements are employed to verify the structural model.

2. Experiment

The experiments were performed in an ultrahigh-vacuum (UHV) multifunction apparatus with a base pressure of less than $3 \times 10^{-11}$ mbar. Details of the experimental setup are given in [15].

The flat Au(111) crystal was cleaned by Ar$^+$-ion bombardment with an energy of 600 eV and subsequent annealing to 1000 K. The sputtering and annealing cycles were repeated until no contamination could be detected with Auger electron spectroscopy (AES) within the detection limit of 1–5% of a monolayer (ML) depending on the impurity. At this point the STM and LEED measurements revealed the typical herringbone surface reconstruction of an Au(111) surface [9, 10]. STM revealed that the width of the terraces is larger than 200 nm. An STM measurement of the clean Au(111) surface is shown in figure 1.

The vicinal Au(111) crystal was cleaned by Ar$^+$-ion bombardment with an energy of 600 eV and subsequent annealing to 815 K. We used a lower annealing temperature than in the case of the flat Au(111) surface to avoid the risk of destroying the regular step array. The sputtering and annealing cycles were repeated until no contamination was detectable with AES. The LEED pattern exhibits the characteristic splitting of the diffraction spots: the spot splitting perpendicular to the steps is caused by the miscut angle of the vicinal surface. The spot splitting along the steps is caused by the surface reconstruction [8]. The reconstructed vicinal Au(111) surface shows discommensuration lines running perpendicular to the step edges in a ‘V’ shape on the terraces [6]. After more than a hundred sputtering and annealing
cycles STM measurements showed straight and regular spaced terraces (see figure 2) with monatomic steps, which did not alter anymore by further cleaning procedures described above. The terrace width in the measurement of figure 2 is 6.3 nm on average. By analyzing many STM pictures at different positions on the crystal surface we found an average terrace width of $\langle W \rangle = 6.1 \pm 0.8$ nm, which corresponds to a vicinal surface with Miller indices (23 25 25) and a miscut angle of 2.3$^\circ$. The terraces of the Au(23 25 25) surface are reconstructed in the same way as on the Au(788) surface and the steps exhibit the same $\{111\}$-microfacets, but the terrace width is 6.1 nm (25 atomic rows) instead of 3.8 nm (16 atomic rows). Please note how regular the terraces are in width. This surface is therefore well suited for growing wide and well-ordered ferromagnetic 1D nanostructures.

To avoid possible contaminations from a crucible, Fe was evaporated from a high purity rod by a water-cooled electron beam evaporator. During evaporation the pressure was better than $5 \times 10^{-11}$ mbar. The sample was held at room temperature (RT) during Fe deposition. We used quantitative AES to calibrate the Fe evaporation rate by growing Fe on Cu(001). In the literature, it was observed that Fe grows not only layer-by-layer on Cu(001) but that it can also grow bilayer-by-bilayer [18, 19]. After the first one to two bilayers the growth proceeds layer-by-layer. In order to avoid a possible error in the calibration of the growth rate, we performed additional calibration measurements with the magneto-optical Kerr effect (MOKE) and STM. We used the well-known transition of the easy magnetization direction from out-of-plane to in-plane between 10 and 12 ML [17] in our MOKE study in order to clarify whether we evaporated layer-by-layer or bilayer-by-bilayer on Cu(001). Additionally, we determined the growth rate with STM by measuring the coverage of a submonolayer film at several positions on the sample. We found that a layer-by-layer growth is present at a deposition rate of less than 0.1 ML min$^{-1}$ in contrast to a bilayer-by-bilayer growth at rates of more than 1 ML min$^{-1}$ [18, 19]. The low deposition rate was chosen to favor a good growth of Fe nanostructures as will be discussed below.

Figure 1. STM measurement of flat Au(111). The image exhibits the characteristic herringbone reconstruction. The bright zigzag lines are the discommensuration lines that separate the fcc-stacked regions from the fault hcp-stacked regions (tunnelling parameters: tunnel voltage $U_T = -0.5$ V and tunnel current $I_T = 0.5$ nA). For STM data processing we used WSxM [16] throughout this contribution.
Figure 2. STM measurement of vicinal Au(111). The $\bar{[211]}$ direction is the step-downward direction. (a) Topography of the surface ($U_T = 1.4$ V, $I_T = 0.5$ nA). The regular steps of the substrate are clearly visible. The white line marks the position of the profile, which is shown in (b). From the profile the average terrace width can be deduced. In this measurement, it is 6.3 nm. (c) Same STM measurement as shown in (a), but with the global plane subtracted. (d) From various STM measurements taken at multiple positions on the crystal the mean terrace width $\langle W \rangle$ was determined to be $\langle W \rangle = 6.1 \pm 0.8$ nm, which translates to the Miller indices of (23 25 25) for the surface orientation and 25 atomic rows per terrace. The terrace width distribution is very narrow in comparison with other vicinal metal surfaces with similar terrace width.

As we will see, Fe grows on flat and vicinal Au(111) in a non-ideal layer-by-layer growth. Therefore, the thickness in ML is defined as the number of supplied Fe atoms with respect to the number of surface atoms of the substrate (ML equivalent). To transfer the calibration from Fe/Cu(001) to Fe/Au(111), the numbers of ML were multiplied by a factor of 1.11 for the coverage of Fe on Au(111) to take into account that the atomic density is 11% less ($N_{Cu} = 15.3$ atoms nm$^{-2}$ and $N_{Au} = 13.8$ atoms nm$^{-2}$).
Figure 3. STM measurements of Fe on flat Au(111) for different Fe overlayer thicknesses. (a) 0.2 ML Fe ($U_T = 0.5$ V, $I_T = 0.1$ nA). The triangular-shaped islands are characteristic for the pseudomorphic growth of Fe on an fcc(111) substrate. The edges of the islands are aligned along the closed packed (01̅1)$_{\text{fcc}}$ directions. (b) 1.3 ML Fe ($U_T = 2.5$ V, $I_T = 0.1$ nA). Since the spacing along the [211]$_{\text{fcc}}$ direction is smaller than along [01̅1]$_{\text{fcc}}$, the coalescence of the Fe islands takes place along [211]$_{\text{fcc}}$. (c) 1.7 ML Fe ($U_T = 2.0$ V, $I_T = 0.5$ nA). At this Fe coverage, the islands start to coalesce also along [01̅1]$_{\text{fcc}}$.

3. Results and discussion

3.1. Growth of fcc Fe(111) on flat and vicinal Au(111)

For the growth of Fe on flat Au(111) it was observed that Fe nucleates at the corners of the herringbone reconstruction [2]–[4]. During subsequent dosing of Fe onto the surface, monatomic high islands are formed. This can also be observed in our STM measurements shown in figure 3(a). The shape of the islands is polygonal and nearly triangular. The edges are aligned along the closed packed (01̅1)$_{\text{fcc}}$ directions. The islands are atomically flat and the triangular shape reflects the fcc(111) symmetry of the substrate surface. Therefore, we observed that Fe grows pseudomorphically on the Au(111) substrate, which is confirmed by the LEED study presented in section 3.2.3 (see figures 13(a) and (b)) and was observed with STM [2, 3] and TEM [1] by other authors.

Due to the surface reconstruction of flat Au(111), the islands form a regular pattern (see figure 3(a)). For subsequent Fe dosing, the islands continue to grow laterally in all directions. Since the spacing between the islands along [211]$_{\text{fcc}}$ is smaller than along [01̅1]$_{\text{fcc}}$, the islands start to coalesce along the [211]$_{\text{fcc}}$ direction. This can be seen in figure 3(b) for a coverage of 1.3 ML Fe/Au(111). The islands are connected along the [211]$_{\text{fcc}}$ direction but not perpendicular to this direction (i.e. along [01̅1]$_{\text{fcc}}$). With increasing coverage the islands start to coalesce along [01̅1]$_{\text{fcc}}$ as well (see figure 3(c)).

For Fe on vicinal Au(111), the growth proceeds differently from the growth on flat Au(111). When adatoms are evaporated onto the vicinal surface, they start to move on the terrace until they are trapped at a step edge or meet with other diffusing adatoms. At a low evaporation rate or a high mobility, the probability for the adatoms to reach the step edge is higher than to form clusters or islands. In our case the evaporation rate of less than 0.1 ML min$^{-1}$ is sufficiently low to observe a good growth of Fe nanostructures at the step edges with a
Figure 4. The sketch illustrates how Fe grows on reconstructed vicinal Au(111) with \{111\}-microfacets. Note that the ratio between step height and terrace width is not drawn to scale. (a) Fragments of Fe chains resulting from the preferential growth of Fe on the fcc sites of the reconstructed terraces are present at the step edges. (b) Then the Fe grows over the hcp sites, the first row of Fe wets the step edge (1D coalescence). The undulated structure due to the different growth speeds on fcc and hcp is already visible. (c) The faster growing Fe bumps on the fcc sites almost reach the adjacent step edges. (d) The coalescence of Fe at the step edges has taken place (2D coalescence). The local thickness at the sites, where the coalescence has taken place, is higher than the average (nominal) thickness.

minimum of unwanted clusters formed by immobilized adatoms. The Fe atoms located at the step edges act as nucleation centers for the subsequently evaporated Fe adatoms and monatomic high Fe fragments are formed. The spacing between the fragments corresponds to the periodicity of the surface reconstruction, which is 7.2 nm [8]. The Fe adatoms grow preferentially on the fcc sites. Thus, the fragments are formed on these sites and during subsequent dosing of Fe onto the surface the fragments connect finally with each other occupying also hcp sites [7]. This will be called the 1D coalescence in the following sections. The growth behavior is illustrated schematically in figures 4(a) and (b). After the first row is completed, the growth of Fe still continues preferentially on the fcc-stacked regions [7] (see figure 4(c)), which produces undulated Fe stripes. A closer look reveals that triangular-shaped islands are formed (see figure 5). The island edges are aligned along the closed packed \{011\}_fcc directions. Between the islands the step edges are decorated with Fe stripes, which connect the islands along \{01\}_fcc. The growth mode can be best described as a 1D Stranski–Krastanov growth and proceeds similar as on Au(788) [7]. Just as in the case of Fe on flat Au(111) proceeds the growth
Figure 5. STM measurements of different Fe overlayer thicknesses deposited on Au(23.25.25) at RT. The $[\bar{2}11]$ fcc direction is the step-downward direction. (a) 0.2 ML of Fe ($U_T = 0.3$ V, $I_T = 0.5$ nA). The characteristic buckling of the Fe stripes is due to the surface reconstruction. The spacing between the centers of the structures is given by the surface reconstruction and amounts to 7.2 nm [8]. The growth of Fe on Au(23.25.25) proceeds in the same way as on Au(788) [7]. (b) 0.5 ML of Fe ($U_T = 0.3$ V, $I_T = 0.5$ nA). The average terrace width in this measurement is 6.0 nm. For these wide terraces the percolation limit is almost reached. The enhanced contrast between Fe and Au is due to chemical contrast.

We can conclude that the growth of Fe for submonolayer coverage proceeds pseudomorphically on flat and vicinal Au(111). In both cases, triangular-shaped islands are formed. The observed Fe overlayer thicknesses required for the coalescence of the triangular-shaped islands are different on flat and vicinal Au(111) due to the different island spacings. While the spacing of the nucleation centers on flat Au(111) is 7.5 nm along $[\bar{2}11]$ fcc and is given by the surface reconstruction, the spacing on vicinal Au(111) is given by the terrace width (here: 6.1 nm). Furthermore, the growth proceeds preferentially on the fcc sites of the reconstructed terraces. Thus, the Fe islands reach the step edges faster than for an equal growth on fcc and hcp sites. Along the [01\bar{1}] fcc direction the spacing is given by the surface reconstruction on both surfaces ($\approx 11.5$ nm for flat Au(111) and 7.2 nm for all vicinal Au(111) surfaces exhibiting discommensuration lines perpendicular to the step edges).

3.2. Phase transition from fcc(111) to bcc(110) Fe

As described in the previous section, Fe starts to grow pseudomorphically on Au(111). The fcc structure is a metastable phase for Fe at RT. The native RT structure of Fe is bcc.
The stress building up during the pseudomorphic growth results in a phase transition from fcc(111) to bcc(110) for larger overlayer thicknesses [1]–[3]. This structural phase transition and the corresponding disruption of the pseudomorphic growth mode was observed with STM [2, 3]. The following sections deal with this phase transition. In section 3.2.1, we present our results obtained with STM, where we observed the phase transition in a change of the topography by the appearance of rectangular-shaped structures, so-called crystallites. From our STM measurements we deduced, how the rectangular bcc(110) unit cells fit onto the flat and vicinal Au(111) substrate (see section 3.2.2). The obtained model is then verified with LEED results in section 3.2.3.

3.2.1. Phase transition observed with STM. For Fe films on flat Au(111) the film structure changes above 1.7 ML (see figure 6). At 2.5 ML, the triangular-shaped islands are almost no longer visible in the STM measurements due to the coalescence along [211]_{fcc} and [011]_{fcc}. Crystallites, which are already present to a small extent at 1.7 ML, grow in number and size with increasing Fe overlayer thickness. For an Fe coverage of 3.0 ML, the crystallites cover the major part of the surface. For higher Fe overlayer thicknesses, the crystallites start to grow together. This results in longer crystallites, e.g. for 10 ML as shown in figure 6(e). The crystallites are atomically flat and predominantly orientated along the (011)_{fcc} directions. The disappearance of the triangular-shaped islands and the appearance of the Fe crystallites marks the end of the pseudomorphic growth, since the symmetry of the substrate is no longer reflected by the shape of the islands. The rectangular-shaped crystallites resemble the twofold symmetry of bcc(110) and reflect the phase transition from fcc(111) to bcc(110).

The early fcc growth behavior of Fe on Au(23 25 25) at RT was discussed in section 3.1. The development of the topography with increasing Fe overlayer thickness is shown in figure 7: with increasing film thickness the Fe starts to grow over the step edges, i.e. along [211]_{fcc}. Due to the preferential growth on the fcc sites, the 2D coalescence takes place at these distinguished sites as illustrated in figures 4(c) and (d). Crystallites start to form at a coverage of 1.0 ML and grow in number and size with increasing coverage. In comparison with the crystallites formed on flat Au(111), the crystallites are smaller for the same coverage. This can be seen by comparing figures 6 and 7. For 3.7 ML Fe on Au(23 25 25) (see figure 7(b)) is the size of the crystallites similar to those found on flat Au(111) for 3.0 ML of Fe (see figure 6(c)) and the size of the crystallites found on Au(111) for 4.2 ML (see figure 6(d)) compares well to the size of the crystallites on Au(23 25 25) for coverages between 5.0 and 5.8 ML of Fe (see figures 7(c) and (d)). For higher Fe overlayer thicknesses, the size difference vanishes. Although the Fe crystallites appear for a smaller overlayer thickness on the vicinal surface, the development of the crystallites regarding their length and width is impeded in comparison with Fe on flat Au(111) and, therefore, a higher Fe overlayer thickness is required until the crystallites become dominant on the Au(23 25 25) surface.

At first glance, the topography of thick Fe films on Au(23 25 25) is quite similar to the topography of thick Fe films on Au(111) but significant differences do exist: on flat Au(111), all three (011)_{fcc} directions are preferential growth directions of the crystallites in equal parts, while on the vicinal surface, the growth along the (011)_{fcc} direction, i.e. along the step edges, is not present. The crystallites are elongated at angles of ±30° with respect to step edge normal, i.e. along the [110]_{fcc} and [101]_{fcc} directions. This results in a more ripple-like topography.

The observed differences can be explained as follows: the growth path of Fe on flat and vicinal Au(111) is governed by the surface reconstruction. For flat Au(111), the Fe nucleates at
Figure 6. STM measurements of Fe on flat Au(111) for different Fe overlayer thicknesses ($U_T = 0.3$ V, $I_T = 0.5$ nA). (a) For an overlayer thickness of 1.7 ML and higher, small crystallites begin to form as an indication of the phase transition from fcc(111) to bcc(110). (b) For 2.5 ML, the Fe islands are connected with each other along $[\overline{2}11]_{fcc}$ and $[01\overline{1}]_{fcc}$. For this reason, the triangular shape of the islands is no longer visible in the STM measurements. The Fe crystallites cover only a minor portion of the surface at this Fe coverage. The pseudomorphic growth is disrupted for this and higher overlayer thicknesses. (c) At 3.0 ML, the crystallites are clearly visible but the surface is not covered with crystallites completely. With increasing coverage, the surface is more and more covered with crystallites. (d) and (e) At and above a coverage of 4.2 ML, only crystallites are visible at the surface. The crystallites are predominantly orientated along the closed-packed $(01\overline{1})_{fcc}$ directions.

The corners of the herringbone reconstruction and the triangular-shaped Fe islands grow laterally in all directions. For Au(23 25 25), the Fe nucleates on the fcc sites, which are separated from the hcp-stacked regions by the discommensuration lines. Fe grows preferentially on the fcc sites and, therefore, the Fe starts to grow over the step edges at these sites with increasing coverage. The Fe connects with the Fe on the adjacent terrace, forming a 2D coalescence. There the local thickness is increased as illustrated in figure 4(d). As a result, the crystallites appear at a lower average (nominal) thickness than on flat Au(111) and grow along the $[\overline{1}10]_{fcc}$ and $[101]_{fcc}$ directions. When the surface is covered with Fe, the surface reconstruction of Au(23 25 25) is not transferred to the Fe surface but the growth direction of the crystallites
Figure 7. STM measurements of various Fe overlayer thicknesses deposited on Au(23 25 25) at RT ($U_T = 0.3$ V, $I_T = 0.5$ nA). The $[\bar{2}11]_{fcc}$ direction is the step-downward direction. For 1.0 ML, crystallites begin to form on the surface. With increasing coverage, the dominance of the crystallites increases until, at a coverage of 5.0 ML, the surface is completely covered with Fe crystallites. The crystallites are significantly smaller in comparison with the crystallites formed on flat Au(111) at the same nominal coverage.

is maintained even for overlayer thicknesses as high as 20 ML (highest overlayer thickness studied). The crystallites maintain their shape and suppress the formation of crystallites along the step edges of the vicinal Au(111) surface (i.e. along $[01\bar{1}]_{fcc}$). It is surprising that the increase in length and width of the crystallites with increasing Fe overlayer thickness on Au(23 25 25) is impeded in comparison with the crystallites on flat Au(111), although they appear at a lower (nominal) thickness. This unexpected behavior can be described within the same picture as the orientation of the crystallites. The slower growing Fe on the hcp sites wets the step edges but does not contribute to the formation of the crystallites until the 2D coalescence occurs. After the coalescence, the Fe adatoms contribute to the development of the crystallites aligned $\pm 30^\circ$ to the normal of the step edges. This explains the impeded formation of Fe crystallites on the vicinal Au(111) surface in comparison with the Fe crystallites on the flat Au(111). The impeded formation of the crystallites is the equivalent of an impeded phase transition from fcc(111) to bcc(110).

In order to learn more about the growth behavior of Fe on flat Au(111) and Au(23 25 25), we analyzed the portion of each layer present at the surface for different Fe overlayer
Figure 8. Fe on flat Au(111): the bar plot shows to which percentage each layer is closed. This STM analysis allows the characterization of the growth front. The area used was $100 \times 100 \text{nm}^2$. The nominal thickness given by the number below the STM image was calibrated by AES and MOKE, which average over an area of about $1 \text{mm}^2$.

thicknesses. The results for Fe on flat Au(111) are shown in figure 8. The early growth of Fe on flat Au(111) proceeds pseudomorphically in a quasi layer-by-layer mode. For higher Fe overlayer thicknesses the growth front consists of three (1.7 ML), four (5 ML) and up to five layers (see figures 8(c) and (d)). The numbers of layers contributing to the film surface is similar to other systems, which grow quasi-layer-by-layer, e.g. Co/Cu(001) for Co films grown at RT [20]. Furthermore, the crystallites are atomically flat. This shows that the Fe still grows in layers, with atomically flat terraces with the size of the crystallites. This was also observed in an STM study of up to 3 ML [3]. Ideal layer-by-layer growth reproduces the topographic properties of the substrate. This is not the case here, since the pseudomorphic growth is disrupted and the crystallites begin to form, but the rectangular shape of the crystallites is reproduced layer-after-layer. The growth of Fe on flat Au(111) can be best described as a non-ideal layer-by-layer growth because a new layer starts before the previous layer is completed. We observed this growth mode for the entire Fe film thickness region studied (up to 10 ML).

For Fe on vicinal Au(111), the growth front was also analyzed by STM and the same growth behavior as for Fe on flat Au(111) was found (see figure 9). For 3.7 ML, the growth front exposes three layers. Similar to Fe on flat Au(111), the number of exposed layers increases with increasing coverage. For 20 ML Fe on vicinal Au(111), five layers are exposed (see figure 9(c)). As observed for Fe on flat Au(111) the crystallites are atomically flat. From this observation and the analysis of the exposed layers, it can be concluded that the growth also proceeds in a non-ideal layer-by-layer growth after the 2D coalescence (up to 20 ML, which is the highest Fe overlayer thickness studied).
Figure 9. Fe on vicinal Au(111): the bar plot shows to which percentage each layer is closed. This STM analysis allows the characterization of the growth front. The area used was 100 × 100 nm². The (slightly deviating) nominal thickness given by the number below the STM image was calibrated by AES and MOKE, which average over an area of about 1 mm².

3.2.2. Structural model of the phase transition from fcc(111) to bcc(110). The stress, which results from the mismatch between the pseudomorphic fcc Fe on Au(111) and the stable bcc(110) phase, can only be accumulated for a few atomic layers. Consequently, the pseudomorphic fcc(111) Fe film can only be maintained for few layers. This is the driving force for the phase transition from fcc(111) to bcc(110). As depicted in section 3.2.1, the growth is not pseudomorphic any more and the described rectangular-shaped crystallites appear, which represent relaxed bcc Fe with (110) surface orientation.

But how does the rectangular bcc(110) unit cell fit onto the hexagonal substrate? The bcc(110) Fe unit cells can fit in the so-called Nishiyama–Wassermann (NW) orientation onto the fcc(111) substrate [21, 22] as illustrated in figure 10. The planes of substrate and film are parallel to each other, thus (110)bcc || (111)fcc. Furthermore [001]bcc is parallel to ⟨01¯1⟩fcc and [110]bcc is parallel to ⟨211⟩fcc (2D matching). The smallest lattice mismatch between bcc(110) and fcc(111) Fe on Au(111) is realized in this orientation. The lattice mismatch is only 0.5% along [001]bcc corresponding to ⟨01¯1⟩fcc (in-plane nearest-neighbor distance 0.288 nm for bcc Fe.
Figure 10. Illustration of the 2D matching between the rectangular bcc(110) and fcc(111) Fe unit cells. Since the Fe(111) grows pseudomorphically on the Au(111) substrate, the sketch also represents the Au(111) unit cell. The mismatch between the unit cells is 0.5% for [001]_{bcc} ∥ ⟨01̅1⟩_{fcc} and 18.7% for [110]_{bcc} ∥ ⟨211⟩_{fcc}. The planes of the fcc substrate and the bcc film are parallel, i.e. (110)_{bcc} ∥ (111)_{fcc}. The sketch shows one of the three possible equivalent matchings in the Nishiyama–Wassermann orientation [21, 22].

and 0.287 nm for pseudomorphic fcc Fe), but 18.7% along the [̅110]_{bcc} corresponding to 〈̅211〉_{fcc} (0.406 nm for Fe and 0.499 nm for the second nearest-neighbor spacing of pseudomorphic fcc Fe) [3].

Another orientation of bcc(110) onto fcc(111), called Kurdjimov–Sachs [22, 23], is illustrated in figure 11. For this orientation, the planes of substrate and film are also parallel and the [̅110]_{bcc} direction is parallel to the ⟨01̅1⟩_{fcc} directions (1D matching). The angle between the [110]_{bcc} and the ⟨211⟩_{fcc} directions for the KS matching is 5.25°.

In the NW orientation, the Fe bcc(110) unit cell fits onto the flat Au(111) substrate in three equivalent orientations (see figure 12(a), white atoms) and in the KS orientation in six orientations (see figure 12(a), black atoms)². This picture illustrates the origin of the three dominant orientations of the Fe crystallites on flat Au(111). The orientations given by the KS orientation deviate only by ±5.25°. Deviations of the crystallites from the preferential growth direction of the order of a few degrees can be found in the STM measurements, but it cannot be decided from these measurements, to which extent this is the result of the KS orientation or a result of the statistic variations of the growth process. TEM measurements [1] and combined reflection high-energy electron diffraction and grazing-incidence surface x-ray diffraction [5] confirm the coexistence of NW- and KS-orientated bcc(110) unit cells on Au(111). The amount of NW- and KS-orientated domains is not accessible from our STM measurements, but the

² This illustration was done in analogy to Fe/Cu(111) by Shen et al [14].
Figure 11. Illustration of the 1D matching between the rectangular bcc(110) and fcc(111) Fe unit cells (see also figure 10). The $\langle \bar{1}1\bar{1} \rangle_{\text{bcc}}$ direction is parallel to the $\langle 01\bar{1} \rangle_{\text{fcc}}$ directions. The angle between the $\langle 110 \rangle_{\text{bcc}}$ and $\langle \bar{2}11 \rangle_{\text{fcc}}$ directions is 5.25°. The planes of the fcc substrate and the bcc film are parallel to each other, i.e. $\langle 110 \rangle_{\text{bcc}} \parallel \langle 111 \rangle_{\text{fcc}}$. The sketch shows one of the six possible matchings in the Kurdjimov–Sachs orientation [23].

KS-orientated unit cells (1D matching) builds up more stress in the film than the NW-oriented unit cells (2D matching). Therefore, the KS domains should be less favorable than the NW domains as suggested for Fe/Cu(111) [14].

On the vicinal Au(111) surface, crystallites are present for 1.0 ML of Fe and above (see section 3.2.1). The orientation of the crystallites is the same as for Fe on flat Au(111) except for the growth direction $\langle 01\bar{1} \rangle_{\text{fcc}}$, i.e. along the step edges, which is not present for Fe on Au(23 25 25). Nevertheless, the appearance of the crystallites on the vicinal Au(111) surface is an indication of the phase transition because the rectangular shape and the growth along the $\langle 01\bar{1} \rangle_{\text{fcc}}$ and the $\langle 110 \rangle_{\text{fcc}}$ directions can be explained by the NW and/or KS fittings of the bcc(110) to fcc(111) (see figures 10 and 11). This means that, for the vicinal Au(111) surface, one NW orientation of the bcc(110) unit cells and two corresponding orientations in the KS mode are not present as illustrated in figure 12(b).

This circumstance allows us to gain insight into the crystallographic properties of the crystallites: from the STM measurements we know that the crystallites are orientated under an angle of ±30° toward the step edge normal. The long side of the crystallites could be either parallel to the $\langle 001 \rangle_{\text{bcc}}$ or to the $\langle 110 \rangle_{\text{bcc}}$ direction, but only the growth along the $\langle 001 \rangle_{\text{bcc}}$ direction allows the formation of the crystallites, which exhibit an elongation of ±30° toward the step edges. This seems reasonable since the mismatch for $\langle 001 \rangle_{\text{bcc}}$ parallel to $\langle 01\bar{1} \rangle_{\text{fcc}}$ is only 0.5% instead of 18.7% for $\langle 110 \rangle_{\text{bcc}}$ parallel to $\langle \bar{2}11 \rangle_{\text{fcc}}$ (see figures 10 and 11). The growth proceeds therefore along the direction in which the minimum of stress is built up. For the system Fe on flat Au(111) this was also suspected by Voigtländer et al [2] but with the use of a vicinal Au(111) substrate we were able to directly identify the crystallographic growth direction of the crystallites.
Figure 12. Illustration of the matching between the bcc(110) Fe unit cells on (a) flat and (b) vicinal Au(111). For simplicity, the lattice mismatch was neglected. Note for the vicinal surface: the terrace width in this sketch was chosen to illustrate the orientations of the bcc(110) unit cells and does not correspond to the actual terrace width of Au(23 25 25), which is 25 atomic rows. (a) For bcc(110) Fe, six orientations of the unit cells are present in the KS orientation (black atoms) and three orientations for the NW growth mode (white atoms). (b) For bcc(110) on Au(23 25 25) we found from our STM measurements that only four of the six KS and two of the three NW-orientated unit cells are realized for the crystallites (see section 3.2.1).

3.2.3. Phase transition observed with LEED. We performed LEED measurements to gain direct information about the crystallographic properties of Fe on flat and vicinal Au(111). The differently orientated domains of bcc Fe(110) should give rise to various satellite spots in the LEED pattern. The results of the LEED study for Fe on flat Au(111) and Fe on Au(23 25 25) can be seen in figures 13 and 14, respectively. The LEED measurements were performed at an electron energy of 110.8 eV. The different intensities of the LEED spots on the left- and right-hand side are due to a slight tilt of the sample in this direction. We made the following observations:

1. For up to 2.0 ML of Fe on flat Au(111) the LEED pattern is identical with the pattern of the clean surface. This is an additional indication of the pseudomorphic growth of Fe on flat Au(111) for up to 2.0 ML.
2. With increasing Fe overlayer thickness the spots broaden. The shape of the spots changes from round spots to crescent-shaped stripes with a curvature toward the (0,0)-spot for an Fe coverage larger than 2.0 ML (see figure 13(c)). This is interpreted as an appearance of (unresolved) satellite spots.
3. The transformation of the LEED spots into stripes is completed at a coverage of 4.5 ML (see figure 13(f)). Above this coverage only the intensity of the LEED streaks increases.
Figure 13. LEED images of Fe on flat Au(111) for various Fe overlayer thicknesses ranging from 0 to 8.0 ML. The energy of the primary electrons was $E_0 = 110.8$ eV.

The LEED patterns obtained from Fe on Au(23 25 25) are very similar (see figure 14):

1. Up to a coverage of 2.0 ML the LEED pattern resembles the pattern of the substrate (see figures 14(a) and (b)) as a result of the pseudomorphic growth. For higher Fe overlayer thicknesses the LEED spots transform into similar structures as observed on flat Au(111).
2. The transformation also starts at an overlayer thickness $>2.0$ ML (see figure 14(c)).
3. The transformation is completed at a coverage of 6.0 ML (see figure 14(f)). For higher overlayer thicknesses only small intensity changes are observed. The length of the LEED streaks no longer changes.
4. While the LEED pattern for Fe on flat Au(111) shows a sixfold symmetry, the LEED pattern for Fe on Au(23 25 25) exhibits only a twofold symmetry. The left and right streaks...
Figure 14. LEED images of Fe on vicinal Au(111) for various Fe overlayer thicknesses ranging from 0 to 8.0 ML. The energy of the primary electrons was $E_0 = 110.8$ eV.

are as long as in the case of Fe on flat Au(111) but the other four LEED streaks are shorter. The origin of this difference is the missing orientation of the crystallites along [011]_{fcc} as will be demonstrated in the following.

The LEED patterns were simulated using the lattice parameters for Au(111) and bcc Fe(110). Figure 15 shows the result of the simulations. The simulations were performed with the assumption of NW, KS and NW + KS orientations for Au(111) and Au(23 25 25). In the latter case, it was taken into account that not all possible orientations of the bcc(110) unit cells are realized on the vicinal surface. Therefore, fewer satellite spots are expected in the LEED pattern for Fe on vicinal Au(111). The simulations fit well to the measured LEED patterns. Thus, the presence of shorter streaks for the system Fe on vicinal Au(111) in comparison with Fe on flat Au(111) can be explained by the missing bcc(110) domains. The sixfold symmetry of Fe on

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Figure 15. Simulation of the LEED patterns for Fe on flat Au(111) and Fe on Au(23 25 25) using the lattice constants of Au(111) and bcc Fe(110). The simulation was done for Fe on flat Au(111) and Fe on Au(23 25 25) separately in order to take into account the missing growth direction of the crystallites on the vicinal Au(111) surface. We used either domains orientated in the NW, KS- or NW + KS orientation in equal parts. The satellite spots are due to the differently orientated bcc(110) unit cells. The orientation of the unit cells and the spots can be directly related to each other by the color code used in the legend. The simulation of the LEED pattern resembles our LEED results. For the system Fe/Au(23 25 25) several satellite spots are missing in comparison with Fe on flat Au(111) in line with our LEED measurements, where four of the streaks are less pronounced and shorter.
flat Au(111) surface and the twofold symmetry of Fe on vicinal Au(111) surface observed with STM was retrieved in the LEED measurements.

From the comparison between the LEED measurements and simulations, we conclude that the measured streaks are too wide to be explained only by the NW-orientated growth of the bcc(110) unit cells. Therefore, we assume that the bcc(110) unit cells are present either in the KS- or in the NW+KS-orientated growth mode. Our growth study demonstrates the surplus of combined LEED and STM investigations.

4. Summary and conclusion

In this contribution, we characterized the development of the topography of submonolayer and ML Fe films on Au(111) and Au(23 25 25). We observed a pseudomorphic growth behavior of Fe on the flat and vicinal Au(111) up to a critical Fe overlayer thickness. For Fe on flat Au(111) the critical thickness is ≈1.7 ML and for Fe on vicinal Au(111) it is ≈1.0 ML. Since Fe grows in the same way on Au(23 25 25) as on Au(788) below the critical thickness [7], we assume that the growth behavior is the same on all vicinal Au(111) substrates with reconstructed terraces and \{111\}-microfacets steps.

Above the critical thicknesses we observed a phase transition from fcc(111) to bcc(110) with STM and LEED. In STM, the appearance of Fe crystallites marks the end of the pseudomorphic growth and, in LEED, satellite spots arise from these crystallites. The critical thicknesses for the phase transition, which we deduced from STM and LEED for the onset and end of the phase transition, are summarized in table 1. We found that the onset of the phase transition from fcc(111) to bcc(110) of Fe on Au(23 25 25) starts at a lower Fe coverage than for Fe on flat Au(111), but the development of the phase transition is impeded and therefore is completed at a higher Fe overlayer thickness on the vicinal surface.

From the STM and LEED measurements, we were able to deduce the orientation of the crystallites on the flat and vicinal Au(111) surface: while for Fe on flat Au(111) all \langle 01\bar{1}\rangle_{\text{fcc}} directions are preferential growth directions of the Fe crystallites, no crystallites were found along [01\bar{1}]_{\text{fcc}} for the system Fe on Au(23 25 25). That means that not all orientations for the bcc Fe(110) unit cells are present on the vicinal surface. This circumstance allows us to directly link the elongation direction of the crystallites to the Fe bcc(110) unit cell. The crystallites grow along the [001]_{\text{bcc}} direction. The unit cells are either orientated in a KS orientation or in a combination of NW and KS orientations.

It will be interesting to link the results obtained for the growth and morphology of Fe on flat and vicinal Au(111) to the magnetic properties of the studied systems for two reasons: firstly, the well-known spin reorientation transition for Fe on flat Au(111) is accompanied by the phase transition from fcc(111) to bcc(110). If the phase transition is impeded, the spin

| System                  | Fe/Au(111) | Fe/Au(23 25 25) |
|-------------------------|------------|-----------------|
| Onset of crystallites in STM | 1.7 ML     | 1.0 ML          |
| Dominance of crystallites in STM | 3.0–3.3 ML | 3.7–5.0 ML     |
| Onset of satellite spot formation in LEED | >2.0 ML   | >2.0 ML         |
| Ending of satellite spot formation in LEED | 4.5 ML     | 6.0 ML          |
reorientation transition for Fe on Au(23 25 25) may be impeded as well. Secondly, the easy in-plane magnetization direction should be different for both systems. Fe films on flat Au(111) exhibit a sixfold symmetry due to the alignment of the crystallites along all \(\langle 01\bar{1}\rangle_{\text{fcc}}\) directions, while the surface of Fe on Au(23 25 25) is twofold due to the missing growth along the \(\langle 01\bar{1}\rangle_{\text{fcc}}\) direction. These questions will be addressed in a future work.

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References

[1] Honjo G, Takayanagi K, Kobayashi K and Yagi K 1977 J. Cryst. Growth 42 98
[2] Voigtländer B, Meyer G and Amer N 1991 Surf. Sci. Lett. 255 L529
[3] Stroscio J, Pierce D, Dragoset R and First P 1992 J. Vac. Sci. Technol. A 10 1981
[4] Chambliss D D, Johnson K E, Wilson R J and Chiang S 1993 J. Magn. Magn. Mater. 121 1
[5] Dekadjevi D T, Hickey B J, Brown S, Hase T P A, Fulthorpe B D and Tanner B K 2005 Phys. Rev. B 71 054108
[6] Shiraki S, Fujisawa H, Nantog M and Kawai M 2004 Appl. Surf. Sci. 237 284
[7] Shiraki S, Fujisawa H, Nantoh M and Kawai M 2004 J. Electron Spectrosc. Relat. Phenom. 137–140 177
[8] Roussset S, Repain V, Baudot G, Garreau Y and Lecoeur J 2003 J. Phys.: Condens. Matter 15 S3363
[9] Wöll C, Chiang S, Wilson R J and Lippel P H 1989 Phys. Rev. B 39 7988
[10] Barth J V, Brune H, Ertl G and Behm R J 1990 Phys. Rev. B 42 9307
[11] Giesen M and Einstein T L 2000 Surf. Sci. 449 191
[12] Mugarza A, Mascaraque A, Repain V, Roussset S, Allmann K N, Himpsel F J, Koroteev Yu M and Chulkov E V 2002 Phys. Rev. B 66 245419
[13] Himpsel F J, Ortega J E, Mankey G J and Willis R F 1998 Adv. Phys. 47 511
[14] Shen J, Klaua M, Ohresser P, Jennichs H, Barthel J, Mohan C and Kirschner J 1997 Phys. Rev. B 56 11134
[15] Budke M, Allmers T, Donath M and Rangelov G 2007 Rev. Sci. Instrum. 78 113909
[16] Horcas I, Fernández R, Gómez-Rodríguez J M, Colchero J, Gómez-Herrero J and Baro A M 2007 Rev. Sci. Instrum. 78 013705
[17] Giergiel J, Shen J, Woltersdorf J, Kirilyuk A and Kirschner J 1995 Phys. Rev. B 52 8528
[18] Kief M T and Egelhoff W F 1993 Phys. Rev. B 47 10785
[19] Glatzel H, Fauster T, Scherzer B M U and Dose V 1991 Surf. Sci. 254 58
[20] Schneider C M, Schmid A K, Schuster P, Oepen H P and Kirschner J 1993 Magnetism and Structure in Systems of Reduced Dimension ed R F C Farrow, B Dieny, M Donath, A Fert and B D Hermann (New York: Plenum) p 453
[21] Nishiyama Z, Shimizu K and Sugino K 1961 Acta Met. 9 620
[22] Wassermann E F and Jablonski H P 1970 Surf. Sci. 22 69
[23] Kurdjumov G and Sachs G 1963 Z Phys. 64 325