Morphology and magnetism of ultrathin Fe films on Pd(100)

X. F. Jin*
Max-Planck Institut für Mikrostrukturphysik, 06120 Halle, Weinberg 2, Germany
and Surface Physics Laboratory, Fudan University, Shanghai 200433, China

J. Barthel, J. Shen,† S. S. Manoharan,‡ and J. Kirschner
Max-Planck Institut für Mikrostrukturphysik, 06120 Halle, Weinberg 2, Germany
(Received 30 April 1999)

Growth and magnetism of ultrathin Fe films on Pd(100) have been studied *in situ* by reflection high-energy electron diffraction, scanning tunneling microscope, and magneto-optical Kerr effect technique. It is found that the growth at the initial stage is in the layer-by-layer mode and the magnetic hysteresis loops are detectable only when the Fe films are thicker than 1.3 monolayer (at 300 K) and 0.5 monolayer (at 55 K). [S0163-1829(99)11139-1]

Epitaxial growth of ultrathin magnetic metals on nonmagnetic substrates and the evolution of the magnetic properties of these systems have attracted a lot of interest during the last two decades.[1,2] In these magnetic systems, the reduced dimensionality and the broken symmetry at the surface or interface give rise to striking and new magnetic properties, such as perpendicular anisotropy, enhanced magnetic moment, and low-dimensional critical behavior. Besides these the system Fe/Pd(100) shows additional attraction due to its exotic exchange coupling leading to the Fe-induced magnetization of Pd at the interface.[3–6]

The 4.2% lattice misfit between bcc Fe ($a_0 = 0.287$ nm) and fcc Pd ($a_0 = 0.389$ nm, $a = 0.275$ nm, where a is the side of the primitive surface mesh), and the -8.4% misfit between fcc Fe ($a_0 = 0.359$ nm) and Pd make the occurrence of epitaxy in this system both ambiguous and complex.[7] While there is a general agreement on the pseudomorphic growth at room temperature of thick Fe on Pd(100),[3,7,8] there are some unresolved questions about the morphology of the initial stage growth of Fe/Pd (100).[3,7,9–11] From their Auger electron spectroscopy (AES) measurement Bader and co-workers declared that the growth was in a layer-by-layer mode,[3] and further proposed from their photoemission measurement that the surface morphology in the submonolayer regime has a random distribution of Fe atoms (or small clusters of atoms) on the Pd(100) surface, as opposed to the usual picture of two-dimensional (2D) island formation in the initial stage of film growth.[9,10] However, it was argued by Jona and co-workers from their low-energy electron diffraction (LEED) study that no evidence for the layer-by-layer growth mode was found, but the growth mode rather appeared to involve pseudomorphic epitaxial flat-topped Fe islands of unequal heights in the initial stages of growth. In addition, it was reported more recently, by Carriere and co-workers from their AES study, that an island-growth mode starting from submonolayer was realized,[11] clearly in contradiction with that of Bader *et al.* Therefore, it is still very controversial whether the initial stage growth is in a layer-by-layer mode[3,9,10] or in an island mode.[7,11] We believe that this situation arises from the fact that there have been no direct experimental observations that are capable of pinning down the problem.

Another controversy that has not been resolved about the Fe/Pd(100) system is whether or not the submonolayer Fe films on Pd(100) grown at room temperature are ferromagnetic at room temperature.[3,6,11] It was found by Bader and coworkers from their magneto-optical Kerr effect (MOKE) measurement that ferromagnetic hysteresis loops were detected for Fe films with thickness from 0.6 monolayer.[3] It was argued by Gudat and co-workers from their spin-resolved photoemission measurement that the system showed nonvanishing spin polarization only after Fe films are thicker than 1.3 monolayer.[6] It was reported more recently by Carriere and co-workers from their magnetic circular x-ray dichroism measurement that the onset for the magnetic signal laid between 1 and 1.5 monolayer.[11] Obviously this controversy must arise from the deviation of thickness calibration in the different experiments. Such a deviation in film thickness might be only a minor problem in many other systems, but it is indeed very important in this Fe/Pd(100) system owing to the fact that a correct description of its critical behavior depends strongly on the film thickness. Therefore, we believe that it is necessary to have a more accurate thickness calibration for the Fe/Pd(100) system and to check again the magnetism in the submonolayer regime.

In this report, we present our results on the Fe/Pd(100) system prepared at room temperature, studied by *in situ* reflection high-energy electron diffraction (RHEED), scanning tunneling microscope (STM), and magneto-optical Kerr effect technique (MOKE). The experiment was carried out in a multifunctional ultrahigh vacuum system (base pressure 3×10^{-11} mbar) equipped with RHEED, STM, cylindrical mirror analyzer (CMA)-based Auger electron spectroscopy (AES), low-energy electron diffraction (LEED), and magneto-optical Kerr effect (MOKE) measurement. Prior to Fe deposition, the clean Pd(100) surface was prepared by cycles of 1 keV argon-ion bombardment at 300 K until no contaminations are detectable by AES, followed by sputtering at 600 K for 15 min and annealing at 950 K for 30 min. This cleaning procedure is repeated until a sharp (1×1) LEED pattern is observed, meanwhile large atomically flat terraces are seen by STM. The Fe was evaporated from a

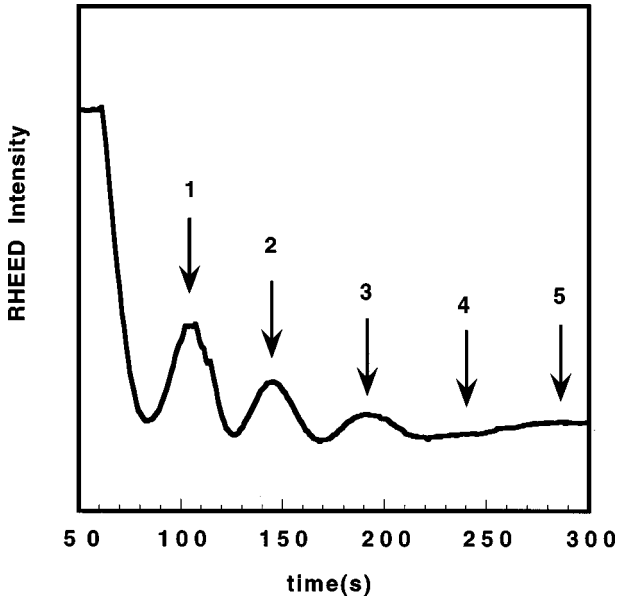

FIG. 1. RHEED intensity oscillations measured at the specular spot during the growth of Fe on Pd(100). The period of the oscillations corresponds to the time required to form one atomic layer.

BaO crucible heated by *e*-beam bombardment. The pressure was kept better than 2×10^{-10} mbar during the evaporation. The evaporation rate of Fe on Pd(100) is cross checked by quartz microbalance, STM, and RHEED oscillations. Because of the careful calibration the Fe film thickness provided in this work, the accuracy of the film thickness is determined to be within ±0.1 ML, which is considerably better than those provided by either AES kinks[3] or the quartz microbalance only.[6,11]

The growth of Fe on a Pd(100) substrate was checked first *in situ* by the development of RHEED patterns as a function of Fe coverage. The patterns show that the Fe growth follows the Pd(100) square mesh very closely, no reconstructions are visible, in agreement with the previous studies mentioned above. The growth mode of the Fe film was determined by following simultaneously the intensity changes of the (1,0), (0,0), and $(-1,0)$ diffraction spots during the growth. According to the interlayer spacing of the body-centered-cubic (bcc) Fe (0.1435 nm), the RHEED scattering angle was set to correspond to the second anti-Bragg reflection. As shown in Fig. 1 for the RHEED intensity measured at the specular spot as a function of Fe coverage, three well-defined RHEED oscillations are clearly seen at the initial stage of Fe deposition. The observation of RHEED oscillations here demonstrate unambiguously that the initial growth stage of Fe on Pd(100) is indeed in the layer-by-layer mode. However, the fast decay of the oscillation amplitudes also implies that the surface roughening happens at very early stage, which partly explains why the island-growth mode was proposed in other studies.[7,11]

The foregoing conclusions about the surface roughening as well as the layer-by-layer growth mode in the Fe/Pd(100) system are based on the RHEED measurement—a technique providing the information in terms of the reciprocal space. However, if the arguments are correct, they should also be confirmed by the STM measurement—a technique enabling direct observation in real space. Figure 2 contains a set of

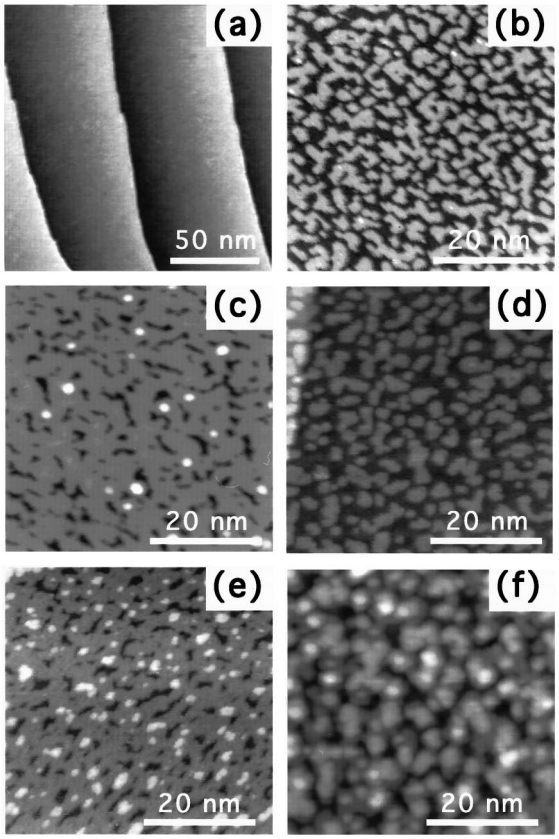

FIG. 2. A set of STM pictures as a function of Fe coverage on Pd(100). (a) Clean Pd(100) surface, (b)–(f) corresponds to the Fe film thickness at 0.5, 0.9, 1.4, 1.9, 6.5 ML.

STM pictures that clearly show the development of the surface morphology of the Fe/Pd(100) system. In order to have an internal calibration of the vertical distance, some well-defined monatomic steps of the clean Pd(100) surface are shown in Fig. 2(a). The rest of the figures show the changes of surface morphology caused by Fe deposition. The first point to be noticed is that, as shown in Fig. 2(b), only monatomic high-Fe films are found on the Pd(100) substrate when the Fe film is 0.5 ML thick, which confirms our RHEED result that the growth of Fe on a Pd(100) substrate is in 2D-layer mode in the submonolayer regime. In addition, it is obvious to see in this figure that such a 2D film consists of a large amount of randomly distributed 2D clusters. This result together with more STM pictures taken at lower Fe coverage (not shown here) do confirm the earlier conclusion obtained by Bader *et al.*, i.e., the surface morphology in the submonolayer regime is characterized by a random distribution of Fe atoms (or small clusters of atoms) on the Pd(100) surface. When the Fe coverage reaches 0.9 ML, an overall flat monolayer Fe film is seen, as shown in Fig. 2(c). Meanwhile, it is also noticed that there are little uncovered areas (black dots) of the clean Pd surface as well as a few second layer of Fe clusters (bright dots). The overall feature of the second layer growth of Fe is similar to that of the first layer, i.e., it can still be called a 2D-layer growth mode, as shown in Fig. 2(d) for 1.4-ML Fe coverage. However, the surface morphology apparently becomes rougher than that of the first layer of the growing Fe film, which can be clearly recognized in Fig. 2(e). Compared to Fig. 2(c), Fig. 2(e) shows

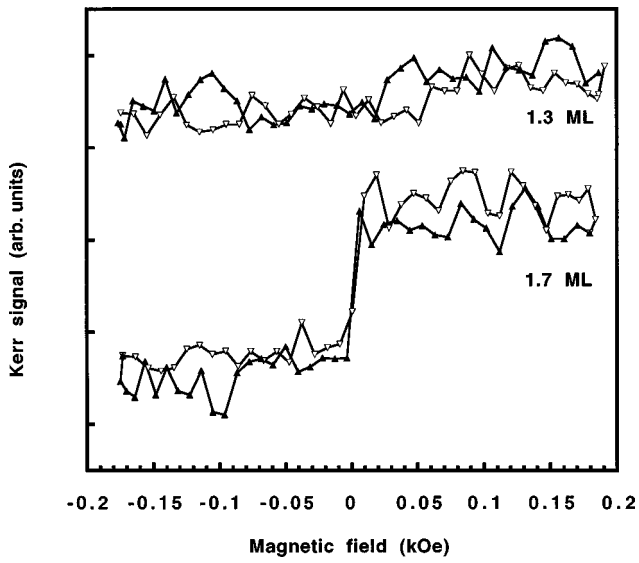

FIG. 3. The onset of ferromagnetism measured at room temperature by MOKE technique.

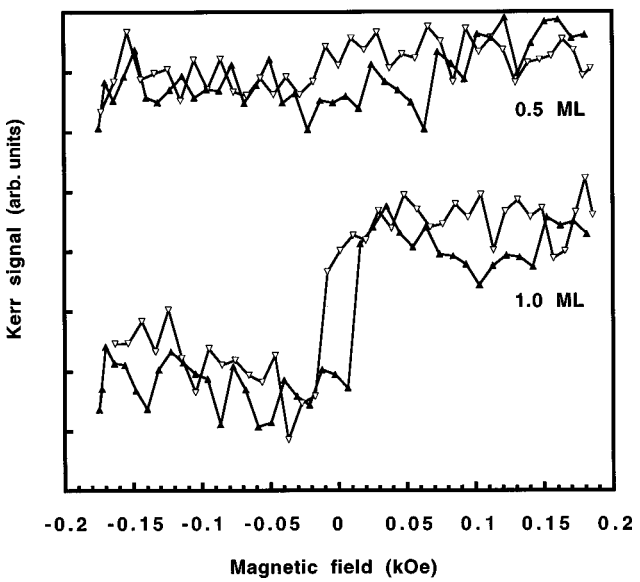

FIG. 4. The onset of ferromagnetism measured at 55 K by MOKE technique.

that more Fe atoms floating on the topmost surface rather than to fill in the available spaces on the inner surface. Starting from the third layer, the surface morphology becomes worse and worse as the Fe growth continues. Figure 2(e) shows a picture for 6.5-ML Fe coverage, which would rather be called the flat-topped Fe islands of unequal heights, as declared by Jona and co-workers, instead of the layer-by-layer grown film. In fact it is this effect of surface roughening that causes the above described decay of the RHEED oscillation amplitudes. Based on our RHEED and STM results, we conclude that the submonolayer growth of Fe on Pd(100) proceeds in the 2D-island mode, and the overall growth feature of the next two monolayers of Fe is still in the layer-by-layer growth mode. However, the surface morphology of the Fe/Pd(100) system becomes worse and worse as the Fe coverage is increased, and the layer-by-layer growth mode seems not a good description for the Fe film growth after 3 ML. This result has clarified the previous controversy in the literature. Furthermore, the random-site-occupancy model proposed by Bader and co-workers for the submonolayer morphology of Fe on Pd (100) has been verified more profoundly by our STM observations.

After the clear picture of the film growth has been established, it is also interesting to investigate the development of magnetism in the system as a function of film thickness. For the Fe film prepared at room temperature, we are going to provide the onsets of the magnetic hysteresis loops detected by MOKE technique at room temperature and 55 K, respectively. It should be mentioned that the sensitivity of our MOKE has been cross checked with those used by Qiu and Bader[12] and it turns out to be the same as theirs. In a first step, we have carried out the MOKE measurement on the Fe/Pd(100) system at room temperature. As shown in Fig. 3, no ferromagnetic hysteresis loops can be observed at and below 1.3 ML of Fe, but a loop is indeed seen at 1.7 ML. Therefore, the onset of ferromagnetism of the Fe/Pd(100) system at room temperature, as a function of Fe film thickness, happens at a Fe coverage between 1.3–1.7 ML. This finding supports those results by Gudat *et al.*[6] and Carriere *et al.*[11] but contradicts that of Bader *et al.*[3] An interesting question to be asked now is whether the lack of ferromagnetism at room temperature is caused by the fact that the measuring temperature is higher than the film's Curie point T_c. Hence, in a second step the onset of ferromagnetism of the Fe/Pd(100) system is checked again at 55 K, as shown in Fig. 4. It is seen that the onset of ferromagnetism at this temperature is between 0.5–1.0 ML. Interestingly, from our foregoing STM results it is about the coverage range that the percolation of Fe monolayer clusters happens. Presumably, it is this percolation of the first Fe monolayer clusters that leads to the onset of ferromagnetism at the system of Fe/Pd(100), although no direct experimental evidence exist so far. This will be an interesting problem awaiting for further study. Based on our more accurate calibration of the Fe film thickness, we conclude that the onset of ferromagnetism of the Fe/Pd(100) system is between 1.3–1.7 ML at room temperature and is between 0.5–1.0 ML at 55 K, which has clarified the previous controversy about this problem.

In conclusion, the clear RHEED oscillations together with the STM images demonstrate unambiguously that the initial stage growth of Fe on Pd(100) is in the layer-by-layer mode. Under a careful and accurate thickness calibration, it is realized that the magnetic hysteresis loops are detectable only after the Fe films are thicker than 1.3 monolayer at room temperature but thicker than 0.5 monolayer at 55 K.

The authors gratefully acknowledge numerous invaluable discussions with Professor U. Gradmann. One of the authors, X. F. Jin, acknowledges the MPI Halle for financial support during his stay. He also acknowledges the partial support from the National Natural Science Foundation (Grant Nos. 19625410 and 19734002), the Education Ministry Foundation for the Yangtse Project, Hong Kong Qiushi Science Foundation, Huo Ying-dong Education Foundation, Shanghai Applied Physics Center, and the QMX Project.

*Author to whom correspondence should be addressed. Present address: Department of Physics, Fudan University, Shanghai 200433, China. FAX: 86-21-65104949. Electronic address: xfjin@fudan.ac.cn

†Present address: Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, TN 37831-6057.

‡Present address: Department of Chemistry, India Institute of Technology, Kanpur 208016, India.

[1] L. M. Falicov, D. T. Pierce, S. D. Bader, R. Gronsky, K. B. Hathaway, H. J. Hopster, D. N. Lambeth, S. S. P. Parkin, G. Prinz, M. Salomon, I. K. Schuller, and R. H. Victora, J. Mater. Res. **5**, 1299 (1990).

[2] F. J. Himpsel, J. E. Ortega, G. J. Mankey, and R. F. Willis, Adv. Phys. **47**, 511 (1998).

[3] C. Liu and S. D. Bader, J. Appl. Phys. **67**, 5758 (1990).

[4] C. Li, A. J. Freeman, H. J. Jansen, and C. L. Fu, Phys. Rev. B **42**, 5433 (1990).

[5] Z. Celinski, B. Heinrich, J. F. Cochran, W. B. Muir, A. S. Arrott, and J. Kirschner, Phys. Rev. Lett. **65**, 1156 (1990).

[6] O. Rader, E. Vescovo, J. Redinger, S. Blügel, C. Carbone, W. Eberhardt, and W. Gudat, Phys. Rev. Lett. **72**, 2247 (1994).

[7] J. Qiunn, Y. S. Li, H. Li, D. Tian, F. Jona, and P. M. Marcus, Phys. Rev. B **43**, 3959 (1991).

[8] E. Vescovo, O. Rader, and C. Carbone, Phys. Rev. B **47**, 13 051 (1993).

[9] C. Liu and S. D. Bader, J. Magn. Magn. Mater. **99**, L25 (1991); Phys. Rev. B **44**, 2205 (1991); J. Vac. Sci. Technol. A **9**, 1924 (1991).

[10] K. J. Strandburg, D. W. Hall, C. Liu, and S. Bader, Phys. Rev. B **46**, 10 818 (1992).

[11] X. Le Cann, C. Boeglin, K. Hricovini, and B. Carriere, Thin Solid Films **275**, 95 (1996); X. Le Cann, C. Boeglin, B. Carriere, and K. Hricovini, Phys. Rev. B **54**, 373 (1996).

[12] Z. Q. Qiu (private communication).