Epitaxial growth of metastable Pd(001) on bcc-Fe(001)

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Abstract: Epitaxial growth of metastable Pd(001) at high deposition temperatures up to a critical thickness of 6 monolayers on bcc-Fe(001) is reported, the critical thickness being depending dramatically on the deposition temperature. For larger thicknesses the Pd film undergoes a roughening transition with strain relaxation by forming a top polycrystalline layer. These results allow to make a correlation between previously reported unusual magnetic properties of Fe/Pd double layers and the crystallographic structure of the Pd overlayer.

The epitaxial growth and the structure of layered systems containing 3d magnetic metals covered by nonmagnetic films have been intensively studied during the past years [1-4]. Fe/Pd multilayers are of particular interest because of an abnormally large Pauli susceptibility of the Pd-atoms. Recently magnetic properties of Fe films with Pd overlayers were investigated [5]. The measured anisotropy constants showed a linear change with increasing Pd-thickness up to 5.5 monolayers (ML) with suddenly following saturation.
However, the epitaxial growth of Pd on Fe(001), in particular in case of low (few ML) Pd coverage, needs further investigation. Celinski et al. grew monocrystalline Pd/Fe(001) double layers on bulk Ag(001) at room temperature (RT) and they speculate about a fcc structure of the Pd layer [6]. In subsequent work Fullerton at al. found a face-centered structure of 6.9 ML thick Pd with a tetragonal distortion of c/a=0.89 [7]. Recent results [8] indicate that in two-atomic Pd layers adjacent to Fe surface layer, the Pd atoms have a local structure which differs from face centered cubic. We report on high temperature epitaxial growth of a metastable Pd-phase on bcc-Fe(001) for low Pd coverage. A direct connection between structure of Pd-films and magnetic properties of Fe/Pd double layers is demonstrated.
The sample preparation was performed in a UHV system by means of molecular beam epitaxy. The *in situ* analysis of the surfaces has been performed by Auger electron spectroscopy (AES) and low energy electron diffraction (LEED). The films were deposited onto GaAs(001) substrates, covered by a fcc Ag(001)-buffer layer. The preparation procedure is reported elsewhere [5, 9]. Pd wedges were grown on bcc Fe(001) at a growth temperature of 540-590 K with a growth rate of 0.02 ML per second. After *in situ* characterization the samples were capped with a Cr or Ag overlayer. The high growth temperature plays a decisive role for forming an epitaxial Pd layer on Fe. At lower temperatures the roughness of the Fe surface impedes epitaxy, because of the difference in the vertical lattice spacing of bcc-Fe (0.14 nm) and fcc-Pd (0.2 nm). In fact, every monolayer step on the Fe surface disturbs the growth dramatically. We therefore believe, that at high enough growth temperatures the first Pd layers adjacent to the Fe surface form a metastable phase with a smaller vertical lattice spacing to accommodate the vertical misfit. The magnetic properties of the Fe/Pd double layers were investigated at RT by Brillouin light scattering from thermally excited spin waves.
Successive LEED patterns using an e-beam energy of 98 eV observed during the growth of the Ag(001)/Fe(001)/Pd(001)-system are shown in Fig. 1. The LEED patterns of the Ag(001) buffer (Fig. 1a) are almost ideal with very sharp spots. However, the LEED patterns of the as-deposited Fe film (Fig. 1b) are not perfect. The (11)-spot, *e.g.*, is sharper than the (10)-spot. With increasing beam energy the widths of the spots oscillate, the phase shift between the (10)- and (11)-spot being about 180°. It indicates a rough Fe-film surface. Fig. 1c shows the LEED patterns of the Fe film annealed at 570 K for 300 s. After annealing, the LEED patterns indicate a c(2×2)-reconstruction of the Fe(001) surface, which also becomes much smoother. A thorough AES examination does not reveal any contamination of the Fe surface, except tiny traces (≤0.03 ML) of Ag. Fig. 1d shows the LEED patterns for a 3ML-thick Pd film, grown on Fe at 540 K, which attests the high-quality growth of Pd(001) on Fe(001). For thicker Pd films the LEED patterns deteriorate, indicating polycrystalline growth. Good coverage of the Fe layers by Pd and lack of interdiffusion of Fe into Pd, grown on the Fe film at T≤590 K is attested by the exponential decay of the Fe Auger peak (47 eV) intensity with increasing Pd thickness t_{Pd}.

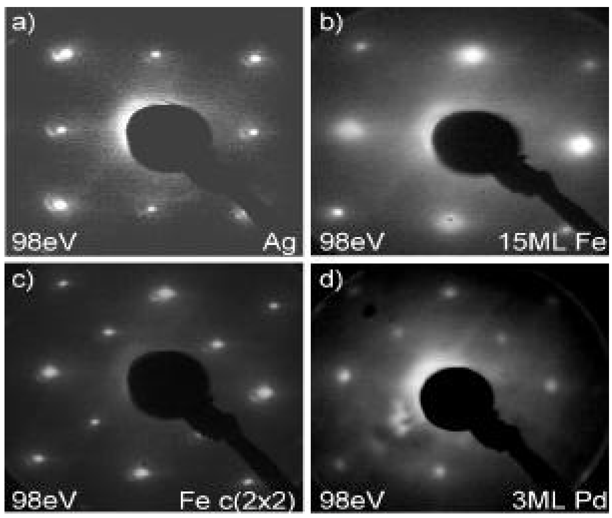

Fig. 1. LEED patterns during the successive growth of the Fe/Pd structure on a Ag buffer measured at an e-beam energy of 98 eV. *a)* Ag buffer, *b)*15ML Fe films as deposited, *c)* the same film after annealing, showing a c(2×2) reconstruction, *d)* 3ML Pd on Fe.

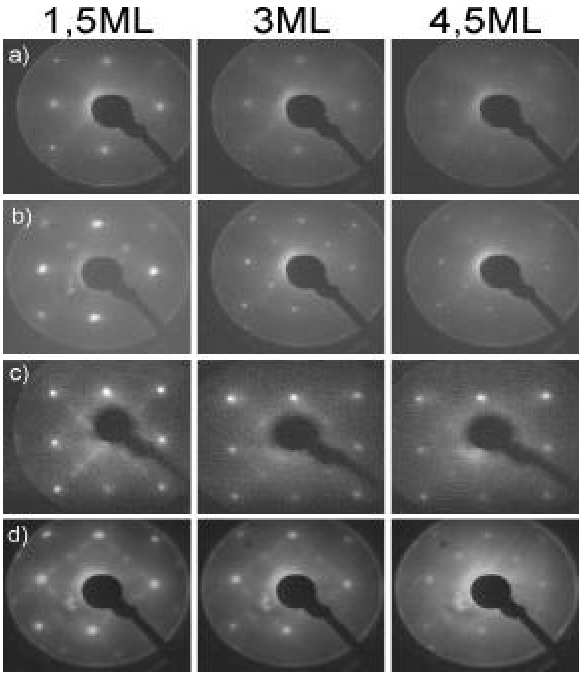

Fig. 2. LEED-patterns of the Fe/Pd structure at different Pd-thicknesses and various deposition temperatures. ***a)*** 300 K, ***b)*** 540 K, ***c)*** 570 K, ***d)*** 590K. Note: 4.5 ML are for ***a)***, ***b)*** and ***d)*** above the critical thickness.

From this fact one can conclude that the Fe film is nearly completely covered by Pd at $t_{Pd} \approx 2$ ML. There is no visible interdiffusion of Fe into the Pd layer as well. At higher growth temperatures an essential interdiffusion of Fe into Pd is seen.

Fig. 2 shows LEED patterns of four different Fe/Pd samples grown at various temperatures. A dramatic dependence of the critical thickness, which determine the transition from monocrystalline to polycrystalline growth, on the deposition temperature is visible. The critical thickness varies between 3ML and 6ML. The best deposition temperature is between 560 K and 570K.

The correlation between the crystallographic structure of the Pd overlayer and the magnetic anisotropy constants of Fe in the Fe/Pd(001) double layer system is clearly demonstrated by the spinwave frequency measurements. The spinwave frequencies, directly connected with the anisotropy constants [5], are shown in Fig. 3 versus t_{Pd}. All samples show a linear dependence of the frequencies versus the Pd-thickness for small t_{Pd} with approximately the same slope. However, the respective t_{Pd} corresponding to saturation are different for all samples. It is highest for the Pd-layer prepared at 570 K and lowest at 540 K.

The critical thickness at which the polycrystalline growth mode starts, dramatically depends on the deposition temperature as well. To demonstrate the correlation between crystallographic and magnetic properties the LEED patterns

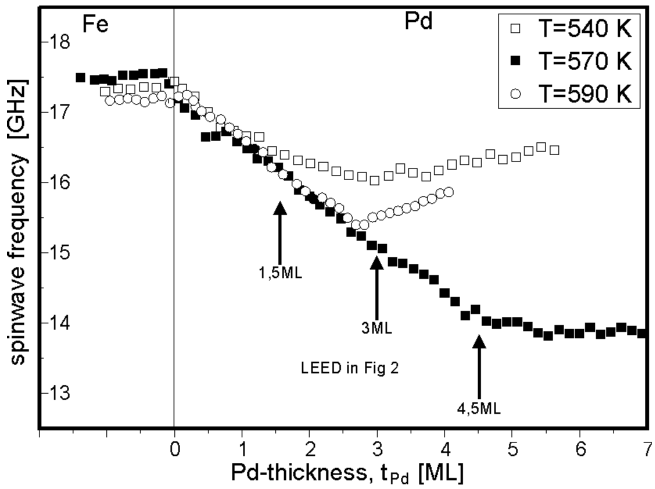

Fig. 3. Spin wave frequency for Pd wedges deposited on a 15 ML thick Fe film at different temperatures T_d (: T_d =540 K, : T_d = 570 K, O : T_d =590 K) as a function of the Pd thickness, t_{Pd}. t_{Pd}<0 stands for the uncovered Fe film and is used as a reference. Wedges were capped with a Cr layer for the ex-situ BLS measurements.

of Fig. 2 for t_{Pd}=4.5 ML are compared with the BLS measurements in Fig. 3. The LEED patterns of the sample grown at 570 K show still sharp spots. On the other hand, the saturation thicknesses of the two other samples are lower than 4,5 ML and the corresponding LEED patterns are very weak. On the basis of these findings we conclude that the origin of the observed linear change of the anisotropy constants is a strain in the Fe film, caused by an substantially stressed Pd layer. Under such conditions the stress is proportional to the Pd-thickness. At some critical thickness the growth of epitaxial metastable Pd-phase is destroyed and a polycrystalline Pd film which does not contribute to the strain is formed by further growth.

In conclusion, recently observed changes of the magnetic anisotropy constants of Fe(001) films covered by Pd are directly correlated with the growth modes of Pd.

Support by the Deutsche Forschungsgemeinschaft is gratefully acknowledged

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