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Preparation and analysis of epitaxial Fe monolayers buried in Pd

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Abstract. Pd almost fulfils the Stoner criterion to be ferromagnetic. In order to understand effects of ferromagnetic inclusions in Pd it is of great interest to study in detail the influence of an ultrathin Fe layer in Pd. Here we report on the preparation and characterization of epitaxial Pd containing Fe layers of a few monolayer thickness. The growth procedure on Ag buffered GaAs(001) substrates was optimized by in-situ Low Energy Electron Diffraction (LEED) and Auger Electron Spectroscopy (AES) to obtain chemical purity and monocrystallinity of the layers with smooth interfaces. By using a variable deposition temperature during the growth of the first Pd layer on the Ag buffer we were able to avoid interdiffusion between Ag and Pd. X-ray- and polarized neutron reflectometry revealed a very well defined interface between the Pd and Fe on top, but an intermixing zone between Fe and Pd on top. With decreasing Fe layer thickness we have observed an increasing magnetization compared to bulk Fe moments.

1. Introduction

Proximity effects in magnetic multilayers are expected to be particularly pronounced for ultrathin ferromagnetic layers in direct contact to magnetically polarizable metals, namely Pd. The basic underlying principles are far from being understood, while these proximity effects might have important consequences for the design of certain spintronic devices. Therefore, we want to analyse both the influence of an ultrathin ferromagnetic layer on a polarizable environment and the transition of 3D to 2D behaviour of the ferromagnetic layer by reducing its thickness down to a few monolayers. The situation is different from ultrathin ferromagnetic layers deposited on isolating substrates, because here the Fe atoms are embedded in a metallic matrix, where the band structure extends over a 3-dimensional system, even if the chemical structure is 2-dimensional. Also the influence of the Pd environment back on the ferromagnetic Fe is subject of the investigation. The results of this experiment might ultimately be relevant for an improved design of spintronic devices, where stabilization of ferromagnetism in ultrathin ferromagnetic films is required.

It has been shown that concentrations of less than $2\,\%$ Fe in bulk Pd lead to an up to three times bigger macroscopic magnetic moment than one would expect resulting from the Fe alone [1]. In addition, the radius of influence of each single Fe atom in the Pd bulk can be as large as $50\,\text{Å}$. It was also investigated how the magnetization of a ferromagnetic substrate is changed, if Pd is deposited monolayer by monolayer [2][3][4]. These experiments revealed that Pd is polarized when deposited on Fe, Co or Ni. Even though these effects seem to be much smaller

than reported on Fe diluted in bulk Pd, there are some hints that the effect depends a lot on the quality of the layer structure. The range of influence of the ferromagnetic substrate on Pd was biggest in the samples where layer by layer growth was observed. Beyond that, it has been shown that in better structured Pd/Fe multilayers up to 4 monolayers of Pd at the interface are magnetically influenced by Fe with a maximum moment of $0.4\,\mu_B$ per Pd atom in the monolayer directly in contact with the ferromagnet [5]. These experiments still do not reproduce effects of the order of the "Giant moments" in bulk Pd. It thus can be expected that even better structured multilayers lead to even bigger effects. In addition, by reducing the Fe layer thicknesses down to the monolayer regime one can investigate the coupling of the Fe to the Pd environment on both sides and - in case of roughness - 3-dimensionally, which should give a similar situation to the bulk Pd containing Fe impurities.

Thus we now want to explore the spatial and magnetic structure of an epitaxially grown well structured Pd/Fe multilayer system with best possible interfaces. In this paper we present an optimized growth procedure of epitaxial Pd containing Fe monolayers together with the structural analysis by LEED, AES and x-ray-reflectometry and the magnetic characterization by SQUID magnetometry and polarized neutron reflectometry.

2. Sample preparation and in-situ characterization

Samples were grown by MBE on GaAs(001) substrates covered by a $5\,\text{Å}$ thick Fe seed layer (to determine the growth direction of Ag to be (001)) and a $1500\,\text{Å}$ thick Ag buffer layer. In-situ characterization was done by LEED and AES. The substrate preparation procedure is well known and optimized [6][7]. LEED analysis confirmed epitaxial growth of Pd on Ag at all temperatures between room temperature and $200\,°\text{C}$. In addition, at higher temperatures sharper LEED spots indicate higher crystalline quality. In contrast, AES revealed interdiffusion of Ag into the deposited Pd layer at temperatures higher than $100\,°\text{C}$. To obtain both well structured and pure Pd layers, a variable deposition temperature was applied. It was shown that starting the deposition at room temperature, then after the first monolayers (about $10\,\text{Å}$) increasing the temperature to $120\,°\text{C}$ for the rest of the layer leads to high quality monocrystalline and pure Pd thin films (see fig. 1). It was also shown that postannealing above $120\,°\text{C}$ is not possible without interdiffusion of Ag and Pd. Therefore, all further layers must not be grown at a temperature above $120\,°\text{C}$, which nevertheless seems to be sufficient for growing monocrystalline multilayer stacks: Even very thin Fe layers (down to single monolayers) deposited at $120\,°\text{C}$ on Pd showed monocrystallinity (LEED) and stability over long time scales (AES). We did not observe any hint for interdiffusion between Fe and Pd even after postannealing at $120\,°\text{C}$ for 20 hours. We have grown $[\text{Pd}_{150\,\text{Å}}/\text{Fe}_{x\,\text{Å}}]_{10}/\text{Pd}_{150\,\text{Å}}$ multilayers with x=150,10,4,2,1 at a substrate temperature of $120\,°\text{C}$ for all layers except the first one, where the variable temperature scheme described above was applied.

3. Ex-situ characterization

We have taken polarized neutron, x-ray-reflectometry and SQUID magnetometry data. The polarized neutron reflectometry (PNR) was performed at the TREFF reflectometer of the JCNS at the FRM II research reactor. The neutron wavelength was fixed at $4.73\,\text{Å}$. The efficiencies of the polarizer, analyzer and both spin flippers are above $98\,\%$. Figure 2 shows as an example x-ray-reflectometry and PNR data for the multilayer and the simulations fitted to these data. Especially the analysis of the $[\text{Pd}_{150\,\text{Å}}/\text{Fe}_{150\,\text{Å}}]_{10}/\text{Pd}_{150\,\text{Å}}$ multilayer has shown that it is necessary to introduce a mixed Pd/Fe layer on top of every Fe layer to obtain a good agreement between measurement and simulation. On the other hand, between a Pd layer and a Fe layer on top a sharp interface is present as was also indicated by the AES in-situ characterization. The thickness of the mixed layer for the sample is about $10\,\text{Å}$. Therefore, for all the Fe layers of
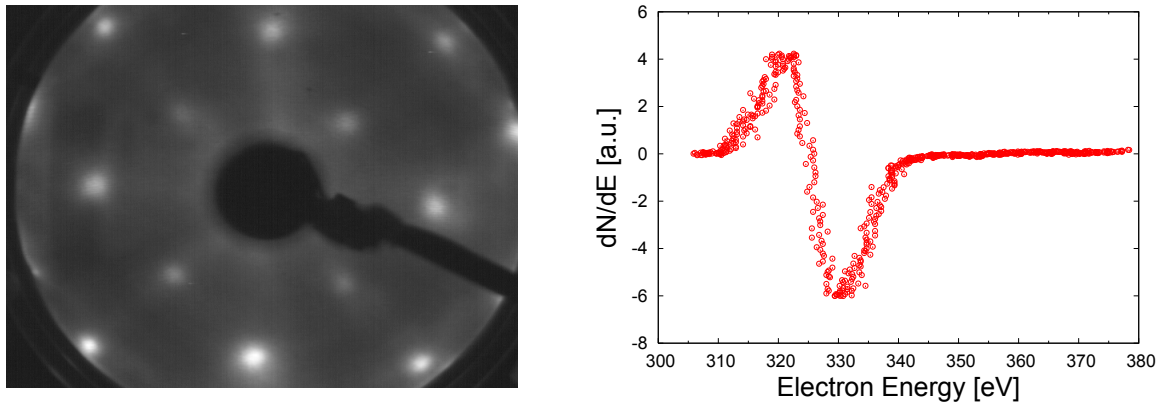
Figure 1. Left: LEED pattern of a 150 Å thick Pd layer deposited on a Ag buffered GaAs substrate with a variable deposition temperature; right: Auger electron spectrum of the same layer. The main peak of Pd at 331 eV is clearly visible, no peak is seen at 350 eV which is the energy of the main peak of Ag.

thickness smaller than 10 Å, we assume that the complete Fe layers are mixed with Pd with a gradually increase of the Pd concentration from the bottom to the top of the Fe layers.
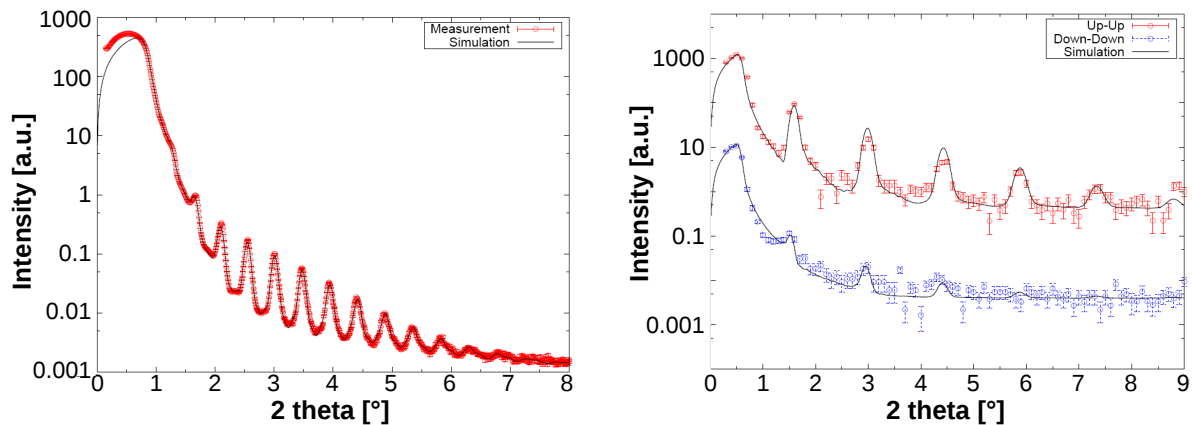

Figure 2. Left: X-ray reflectometry measurement and simulation of the $[Pd_{150\,Å}/Fe_{10\,Å}]_{10}/Pd_{150\,Å}$ multilayer; right: PNR measurement and simulation of the $[Pd_{150\,Å}/Fe_{10\,Å}]_{10}/Pd_{150\,Å}$ multilayer (Up/up channel is multiplied by 100).

By introducing a mixed layer very good agreement between experimental data and the simulated model can be obtained for all the samples. On the other hand this hampers the accurate determination of the amount of Fe in the samples. The superlattice period for all samples is higher than expected from the sample preparation which is just a constant calibration offset factor of the quartz crystal oscillator used in the MBE to control growth rate and layer thicknesses. As Pd and Fe are deposited from the same position in the MBE chamber this calibration factor is the same for both materials. Therefore, the assumption was made that

the ratio of Pd to Fe amount in the samples are given by the nominal value, e.g. 15 to 1 for the $[\mathrm{Pd}_{150\,\text{Å}}/\mathrm{Fe}_{10\,\text{Å}}]_{10}/\mathrm{Pd}_{150\,\text{Å}}$ multilayer. To estimate the Fe content in the multilayers the superlattice period that could be determined very accurately by reflectometry should also contain Fe and Pd of the same ratio. For the analysis of the SQUID data we used the resulting values for the Fe content to calculate a reference ferromagnetic moment (RFM) that these amounts of bulk Fe (magnetic moment per atom: $2.2\,\mu_B = 2.0 \cdot 10^{-20}$ emu) would cause (see table 1).

The magnetic scattering length density $Nb_{m,Fe}$ for the Fe layers in the $[\mathrm{Pd}_{150\,\text{Å}}/\mathrm{Fe}_{10\,\text{Å}}]_{10}/\mathrm{Pd}_{150\,\text{Å}}$ sample determined by PNR is very close to the bulk value of Fe $Nb_{m,Fe,bulk}$ ($Nb_{m,Fe}/Nb_{m,Fe,bulk} = 0.92 \pm 0.06$) and in very good agreement with the SQUID results. In addition, the mixed layers in this multilayer have scattering length densities below the Fe bulk value. However, this value does crucially depend on the choice of the position of the interface: Due to the gradual transition from Fe to Pd at the interface, the thickness of the mixed layer can be chosen in a range of several Å and similar good fits can be obtained by changing the other parameters accordingly. Therefore, it is very difficult to extract detailed quantitative information about the magnetic structure of the intermixing layer and compare them with the SQUID results. The same holds for the other samples with thinner Fe layers, where it must be assumed that the complete Fe layers are mixed with Pd. For these samples we also obtained magnetic scattering length densities below the Fe bulk values, but the big uncertainties caused by the intermixing hinder a sensible comparison with the SQUID data.

With the SQUID magnetometer we have measured hysteresis curves at different temperatures between 2 and 350 K. Saturated ferromagnetic moments and coercive fields are decreasing with increasing temperature as expected for all samples (see fig. 3). In addition, the RFM is in very good agreement with these data lying between the saturated moments measured at 2 K and 350 K (see table 1). The low temperature data taken at 2 K reveal an increasing ratio of measured moment to RFM from a value of approximately 1 to 2.6 with decreasing Fe thickness. In contrast, these data suggest that in the samples with relatively thick Fe layers the effect on Pd is much smaller compared to that reported for the bulk Pd with Fe impurities.
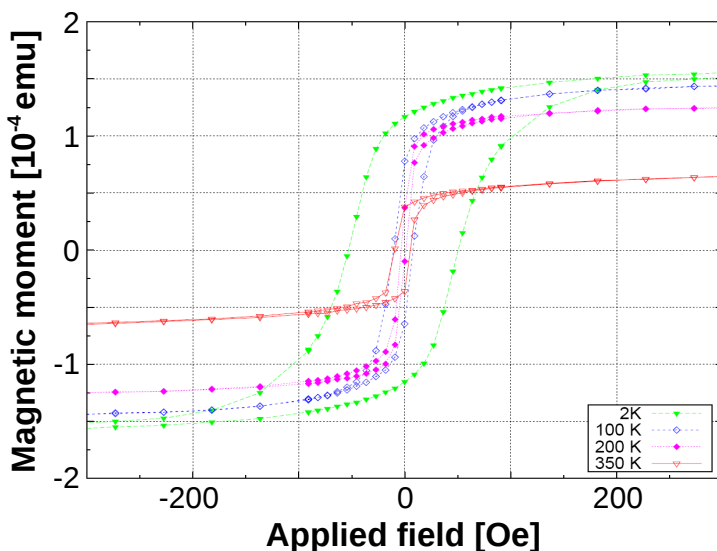

Figure 3. Temperature dependence of the hysteresis curves of the $[\mathrm{Pd}_{150\,\text{Å}}/\mathrm{Fe}_{2\,\text{Å}}]_{10}/\mathrm{Pd}_{150\,\text{Å}}$ multilayer (substrat signal is substracted).

4. Conclusions and Outlook

We have developed a procedure to grow pure, monocrystalline Pd layers on Ag buffered GaAs substrates. A variable temperature during growth of the first Pd layer was applied to avoid interdiffusion with Ag. We were able to deposit very thin, monocrystalline and stable Fe films

Table 1. Results of the macroscopic SQUID measurements (FM = ferromagnetic). The reference ferromagnetic moments (RFM) were calculated by the atomic moment per Fe atom in bulk Fe multiplied by the number of Fe atoms.

	Pd_{150}/Fe_{10}	Pd_{150}/Fe_4	Pd_{150}/Fe_2	Pd_{150}/Fe_1
superlattice period [Å] (reflectometry)	183.4 (2)	179.8 (2)	164 (1)	155 (1)
expected Fe layer thickness [Å]	11.4 (3)	4.7 (3)	2 (1)	1 (1)
RFM μ_{Fe} [10^{-4} emu]	4.9 (1)	2.0 (1)	1.0 (4)	0.4 (4)
FM moment at 2 K μ_{2K} [10^{-4} emu]	5.4063 (4)	2.2407 (3)	1.6054 (4)	1.0458 (6)
μ_{2K}/μ_{Fe} [10^{-4} emu]	1.10	1.12	1.61	2.61
FM moment at 350 K μ_{350K} [10^{-4} emu]	4.5871 (2)	1.9624 (1)	0.7504 (1)	0
μ_{350K}/μ_{Fe} [10^{-4} emu]	0.94	0.98	0.75	0

on Pd. The preparation of Pd/Fe multilayers was also possible even though x-ray- and polarized neutron reflectometry revealed an intermixing at the interfaces of Fe and Pd on top. Surprisingly, in the samples with Fe layers thicker than 2 Å the effect on Pd seems to be very small even though due to the intermixing the situation should be more similar to the situation in dilute bulk Fe/Pd alloys, where the "Giant Moments" were found, than in a perfect multilayer system, where both materials are spatially separated from each other. Nevertheless, the SQUID data yield some indication for an increasing magnetization with decreasing Fe layer thickness compared to the bulk values of Fe. Whether this increase is due to an increased Fe magnetic moment per atom or to an induced magnetization in Pd, could not be clarified due to the intermixing. To determine the exact magnetic structure of the sample and the amount of polarized Pd, X-ray Magnetic Circular Dichroism (XMCD) measurements are planned. In addition, the growth procedure of the multilayers will be optimized. A variable deposition temperature for all Pd layers might also avoid intermixing of Pd and Fe. Furthermore, polarized neutron measurements will also be done at the new reflectometer MARIA at FRM II, which will yield much more detailed information about the multilayer systems due to the higher signal/noise ratio available.

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