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In situ X-ray diffraction study of Co/Pd multilayers grown on Ta substrate during hydrogen loading

Senthilnathan Mohanan,[a,*] Maxim Smetanin,[b] Jörg Weissmüller[b] and Ulrich Herr[a]

[a]*Institute of Micro and Nanomaterials, Ulm University, 89081 Ulm, Germany*
[b]*Institute of Nanotechnology, Forschungszentrum Karlsruhe, 76021 Karlsruhe, Germany*

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For Co/Pd multilayers deposited on Ta the volumetric strain upon hydrogen loading is determined from the measured lattice parameter using Hooke's law. We found that the hydrogen completely diffuses out of the multilayer after exposing the sample to an argon atmosphere or to air, whereas it remains dissolved in the substrate. This causes a biaxial in-plane stress in the multilayer. We observed a change in the hydrogen solubility in the multilayer for varying in-plane stress.

Keywords: Biaxial stress; Co/Pd multilayer; Hydrogen solubility; X-ray diffraction

Co/Pd multilayers (MLs) exhibiting perpendicular magnetic anisotropy (PMA) are promising candidates for perpendicular recording media. In earlier work [1], we reported that hydrogen loading leads to an isotropic lattice expansion of Ta substrate, which results in a large in-plane biaxial tensile stress in the magnetic thin films. This stress significantly increases the PMA of the Co/Pd ML system due to the additional magnetoelastic energy contribution [2]. It is well known that Pd can be easily loaded with hydrogen. Moreover, Okamoto et al. showed that the structural changes upon hydrogenation and dehydrogenation of the Co/Pd/Co trilayer system would also contribute towards the PMA [3]. In their work the substrate did not absorb hydrogen. Here, we report experiments with Co/Pd MLs on a Ta substrate that absorbs hydrogen along with the multilayer. This leads to a different state of strain, which we investigate by in situ X-ray diffraction (XRD) during hydrogen loading and unloading.

The Co/Pd MLs were prepared on 125 μm thick Ta substrates (99.95% purity) at room temperature by DC magnetron sputtering. The base pressure of the system was 5×10^{-8} mbar and the Ar pressure during sputtering was 4.4×10^{-3} mbar. The polycrystalline Co/Pd MLs consisting of 30 bilayers of Pd (0.96 nm)/Co (t nm) were grown on one side of the substrate after deposition of a 9-nm thick polycrystalline Ta buffer layer and were protected by a 3-nm thick Ta cap layer. Two sets of samples with Co layer thickness, t, of 0.26 or 0.53 nm were prepared. The opposite side of the Ta substrate was coated with a 100-nm thick Pd layer to catalyze the dissociation of H_2 molecules into their atomic entities [4], which diffuse into the underlying Ta substrate. An environmental chamber for hydrogen loading and temperature control was mounted in a Philips X'pert X-ray diffractometer outfitted with a Mo X-ray tube and a solid-state detector. Before supplying hydrogen, the loading chamber was flushed with pure Ar and the temperature was raised to 70 °C. Then, the hydrogen loading was carried out in a continuous flow of H_2 at 1 bar. The loading was carried out at 70 °C in order to stay in the single-phase (αTa) region of the Ta–H phase diagram [5]. According to the equilibrium Ta–H phase diagram at 1 bar, one would expect an increase in the H concentration up to about 42 at.% under the conditions used here. However, in the current study it was found that the concentration of hydrogen in the Ta substrate increases continuously with the loading time, but stays below the equilibrium concentration during the loading times applied here.

Figure 1 shows the in situ X-ray scattering curves of the sample with $t_{Co} = 0.26$ nm under various environmental conditions. Several groups have reported that Co initially grows in the face-centered cubic (fcc) structure on (1 1 1) Pd surfaces which facilitates the coherent growth of the Co/Pd MLs [6,7]. This coherency can be observed from the occurrence of a single Co/Pd (1 1 1)

* Corresponding author. E-mail: senthilnathan.mohanan@uni-ulm.de

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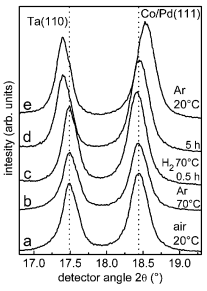

Figure 1. In situ XRD scans of the sample: (a) in air at 20 °C, (b) in Ar atmosphere at 70 °C, (c) 30min after introducing hydrogen into the loading chamber, (d) after 5h of hydrogen loading, and (e) after cooling down to room temperature in Ar atmosphere.

peak in the XRD patterns. When comparing the spectrum of the sample measured in air at room temperature (a) and in Ar atmosphere at 70 °C (b) there is no observable Co/Pd (1 1 1) peak shift upon increasing the temperature to 70 °C. Upon introducing hydrogen at 70 °C (c) the Co/Pd (1 1 1) peak shifts to lower 2θ angles (after 30 min). This confirms a noticeable expansion of the average Co/Pd lattice spacing due to hydrogenation. But there was no observable change in the Ta peak position at this time, which indicates that the hydrogen diffuses into the Co/Pd multilayer prior to the Ta substrate. After exposing the sample to hydrogen for 5 h (d) there was an observable shift in the Ta (1 1 0) peak to lower 2θ angles. This indicates lattice expansion of the Ta substrate due to the incorporation of hydrogen. After flushing out the hydrogen with Ar and cooling down to room temperature, there was a large shift of the Co/Pd (1 1 1) peak to higher 2θ values, indicating a lattice contraction of the Co/Pd MLs (e).

Figure 2 summarizes the change in out-of-plane interplanar spacing of the Ta substrate and the Co/Pd MLs as calculated from the Bragg reflection positions as a function of time. The plot can be divided into the following three regions:

(1) Hydrogen is introduced into the chamber, which is maintained at 70 °C. No significant change in the lattice spacing of the Ta substrate is observed but an increase in the out-of-plane lattice spacing of Co/Pd due to the diffusion of hydrogen in the ML is found. In our earlier work, we reported on the observation of a linear 1:1 transfer of in-plane strain from the Ta substrate to the Co/Pd MLs up to a strain of 0.6%, which showed that the Co/Pd ML is laterally clamped to the Ta substrate [2]. Therefore, although a volumetric expansion of the Co/Pd takes place, the Co/Pd ML can only expand in the direction perpendicular to the film plane.

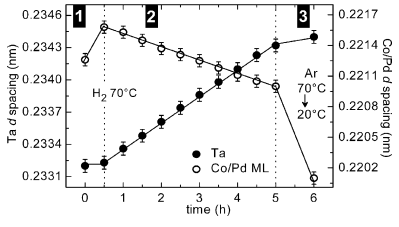

Figure 2. Lattice spacing of the Ta substrate and the Co/Pd MLs calculated from in situ XRD scans measured during hydrogen loading.

(2) A linear lattice expansion of the substrate with time due to the hydrogen loading is observed. Since the diffusion coefficient of hydrogen in Ta at 70 °C is about $4 \times 10^{-6}\,\mathrm{cm^2\,s^{-1}}$ [8], it would take only a fraction of a minute for the hydrogen to diffuse through the entire Ta substrate of 125 μm thickness. The slow kinetics therefore indicates that the absorption rate is limited by the dissociation of H_2 at the surface of the Pd layer and the incorporation into the Ta substrate. At the same time, a decrease in the out-of-plane lattice spacing of the Co/Pd ML is found. This is readily understood as follows: the isotropic lattice expansion of the Ta substrate leads to an in-plane biaxial tensile stress in the Co/Pd MLs. This causes the observed shift of the Co/Pd (1 1 1) peak to higher 2θ values due to the Poisson's contraction perpendicular to the sample plane and the accompanying reduction in the lattice spacing. The linear change in the lattice spacing of the Co/Pd MLs with the Ta lattice spacing shows that the deformation is still elastic. The decrease in the out-of-plane lattice spacing also shows that any possible volume expansion of the Co/Pd MLs is overcompensated by the lattice contraction due to the biaxial strain induced by the Ta substrate.

(3) The loading chamber is flushed with Ar and the temperature is reduced to room temperature. The concentration of hydrogen in the Ta substrate ($c_{H/Ta}$) can be estimated from the equation (in the case of isotropic lattice expansion), $c_{H/Ta} = 3\varepsilon/0.155$. The maximum value of $c_{H/Ta}$ during the entire loading was found to be 0.1, which is well below the value of 0.16 up to which the tantalum hydride exists in a single phase. This is consistent with the XRD observation of a single α-TaH phase upon cooling to room temperature. During flushing the environmental chamber with Ar, the partial pressure of hydrogen decreases continuously until the chamber is filled with pure Ar. A small increase in the Ta lattice spacing is observed during that procedure, indicating that the Ta was still absorbing a small amount of H. The latter occurs when the partial pressure of H_2 in the gas mixture stayed above the equilibrium pressure of H_2 at the corresponding concentration of hydrogen in the Ta substrate ($c_{H/Ta} = 0.1$) (which is below 10^{-4} mbar according to Ref. [9]). This observation is also consistent with earlier reports that hydrogen desorbs from Ta only at an elevated temperature of about 500 °C [10]. On the other hand, there is a significant reduction in the Co/Pd lattice spacing, indicating that the hydrogen desorbs from the Co/Pd MLs. As the corresponding volume reduction of the Co/Pd MLs is again hindered by the substrate, the lattice parameter can only contract in the direction perpendicular to the film plane.

The decrease in the lattice spacing of the Co/Pd MLs during unloading of hydrogen (region 3) was found to be much larger than the previous increase during loading (region 1). A similar behavior has been observed for the sample with a thicker ($t_{Co} = 0.53$ nm) Co layer. This indicates that some additional amount of hydrogen is dissolved in region 2. In order to estimate the change