LETTER TO THE EDITOR

Hydrogen assisted growth of Fe/V superlattices

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Abstract

The functionality of magnetic heterostructures is strongly affected by the interface structure and morphology as well as by the crystalline quality. Using Fe/V superlattices, serving as model systems for exchange coupling and for superconducting spin valves, we show that the excellent interfacial properties can be further improved by hydrogen assisted growth. The role of the hydrogen thereby is twofold. It reduces thickness fluctuations and improves the crystalline quality. Sputter deposited Fe/V superlattices grown at a partial pressure of 2×10^{-6} mbar are atomically flat and possess a residual resistivity ratio which is significantly improved as compared to those of conventionally grown samples.

(Some figures in this article are in colour only in the electronic version)

Due to their unique magnetic and superconducting properties Fe/V multilayers are model systems for the investigation of interface magnetism [1], interlayer exchange coupling [2] and superconducting spin valves [3]. In particular, the realization of the latter requires smooth interfaces to guarantee high interface transparency for the superconducting pair density function, and a superconducting correlation length ξ_S of the order of the thickness d_S of the superconducting film. Fe/V turned out to be a promising system, as epitaxial Fe/V superlattices (SLs) can be grown with very high structural quality and smooth interfaces [1, 4]. Recently, a superconducting spin valve was realized using a thick V layer ($d_V = $ 16–30 monolayers (ML)), grown on a (Fe(2 ML)/V(13 ML)) $\times$ 20 SL [3]. Depending on the relative orientation of the magnetization vectors in adjacent Fe layers, the onset of superconductivity in the thick V film differs by as much as 120 mK. To increase this effect, the structural and chemical quality of the samples have to be further improved.

    We demonstrate that hydrogen assisted growth simultaneously leads to a reduced interface roughness and to a higher residual resistivity ratio (RRR), which is closely related to ξ_S. Two different kinds of samples were investigated. First, to study the influence of the hydrogen on the interface quality we have grown a series of [Fe(2 ML)/V(16 ML)] $\times$ 30 SLs by sputter
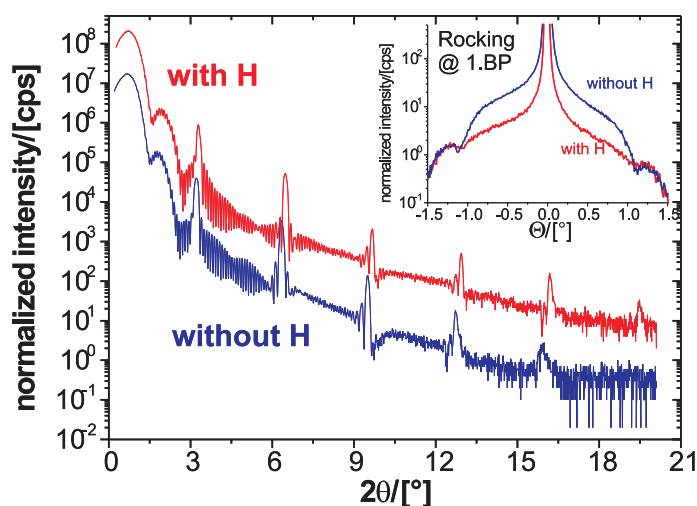
Figure 1. Small angle x-ray reflectivity scans recorded at $E = 8.048$ keV. The upper curve displays the reflectivity of the sample sputter deposited in a partial H pressure of 2×10^{-6} mbar at $T = 320$ °C. The lower curve shows the reflectivity of the reference sample grown without H assistance, also at $T = 320$ °C. The scans are shifted along the intensity axis for clarity. The inset shows the diffuse scattering recorded at the first SL peak on an enlarged scale.

deposition. Second, to further address the quality of the crystalline properties we prepared single, 27 nm thick V layers. All substrates (MgO(001)) originate from the same batch. A description of the sputter system used and of the hydrogen free growth process is given by Isberg *et al* [4]. Hydrogen partial pressures of up to 2×10^{-6} mbar were applied to the sputter gas (Ar 7 mbar). At each chosen hydrogen pressure we prepared a series of samples at substrate temperatures between of 270 and 320 °C. As the solubility of H in Fe and in V is negligible at the chosen pressures and temperatures [5], no H is expected to be incorporated during the growth process. The structural characterization was carried out using small angle x-ray reflectivity (XRR) and x-ray diffraction (XRD) at the wiggler beamline W1.1 at the Hamburg Synchrotron Laboratory in Hamburg, Germany.

We confirm earlier reports [4, 6] that best results for the hydrogen free growth are obtained at a substrate temperature of 320 °C. At this temperature XRR shows well defined Kiessig fringes [7] and SL reflections, which gradually disappear at higher growth temperatures, indicating increasing interface roughness and/or intermixing. Figure 1 compares XRR scans obtained from samples grown at 320 °C with (sample 1) and without H assistance (sample 2), recorded at a fixed photon energy of 8.048 keV. The effect of the H assisted growth can be seen qualitatively in the following three features of the reflectivity curves: first, sharp SL peaks up to the sixth order are clearly visible for sample 1, while for sample 2 only five broader SL peaks are observed. Second, the total thickness oscillations for sample 1 persist up to the third SL reflection, while for sample 2 they are damped out after the second SL reflection. Third, the overall intensity and the intensity of the SL reflections drops off faster for sample 2 than for sample 1. Obviously, the sample grown under hydrogen atmosphere possesses a sharper optical contrast between the metal layers, indicative of well defined interfaces. As discussed by Isberg *et al* for the H free growth [4] the samples possess an average interface roughness of 1–2 Å, corresponding to one-atom steps. An improvement by H assisted growth would lead to more extended atomically flat terraces between two steps, resulting in larger in-plane coherence length. This is confirmed by scans of the diffuse background recorded at the first SL reflection.
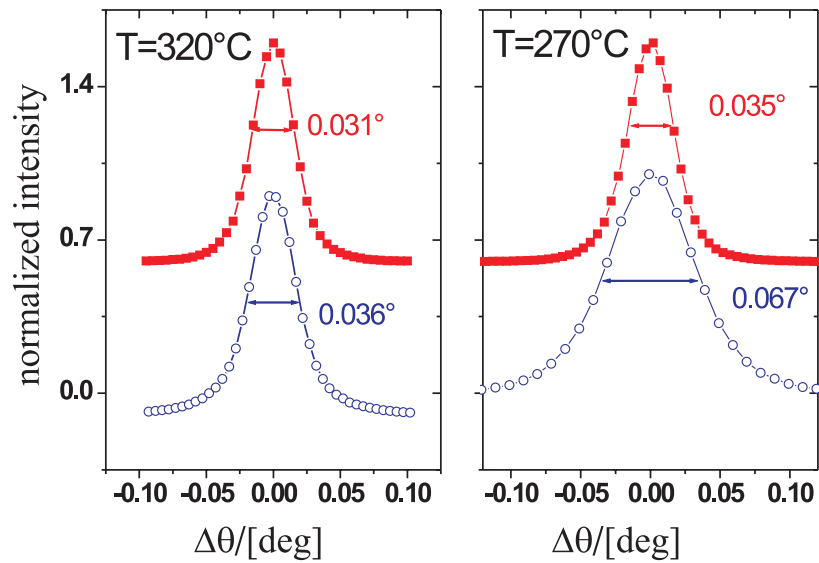
Figure 2. Transverse scans through the (002) superlattice Bragg reflection, recorded at $E = 8.048$ keV for samples grown at $320\,°C$ (left) and $270\,°C$ (right). The upper curves correspond to samples grown with a partial H pressure of 2×10^{-6} mbar. The lower curves show the corresponding scans of the reference samples grown without H assistance.

The transverse scans depicted in the inset to figure 1 clearly show a smaller angular width and a strongly reduced diffuse contribution to the scattering signal for sample 1 as compared to the reference sample. The scan from the reference sample is taken from a reciprocal space map recorded around the first SL reflection, showing a strong resonant diffuse scattering sheet, indicative of conformal roughness [8]. Its width corresponds to a correlation length of 150 nm, while the average correlation length from the H assisted growth sample equals 300 nm.

High angle x-ray diffraction indicates that also the crystallinity improves upon H assisted growth. Longitudinal scans along the growth direction show the structural (002) SL Bragg reflection accompanied by satellite peaks up to the sixth order and numerous Laue oscillations which are again more pronounced for sample 1 than for sample 2. Also the mosaicity of the SL decreases. Especially at the lower growth temperature of $270\,°C$, the benefit of H assisted growth becomes evident. The measured full width at half-maximum (FWHM) of the transverse scans through the (002) Bragg peak markedly decreases from $0.067°$ to $0.035°$, respectively. Taking the instrumental broadening of about $0.01°$ into account, this results in a reduction of more than 50%. At higher growth temperature the effect is less pronounced. Figure 2 displays the respective scans. The value $0.031°$ achieved with H assisted growth at $320\,°C$ is the best result obtained in this series and compares very well with those for the best metallic superlattices reported in the literature [9, 10].

The superior structural quality of samples with H assisted growth also results in a larger residual resistivity ratio (RRR). Figure 3 compares the temperature dependence of the specific electrical resistance for two Pd capped single V films prepared with and without H assistance at a constant substrate temperature of $320\,°C$. Both samples display a decreasing resistance with decreasing temperature until they become superconducting at 4.38 K (w/o H) and 4.42 K (with H), respectively. For thin V films the transition temperature scales strongly with the thickness, explaining the reduced T_c as compared to bulk V (5.32 K) [11]. The difference of 40 mK is
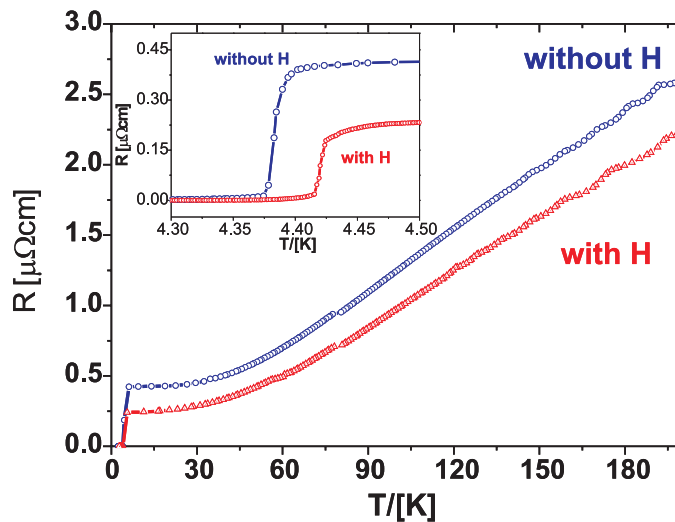
Figure 3. Temperature dependence of the specific electrical resistivity for two Pd capped thick V films. The upper curve shows the conventionally grown sample, the lower one the H assisted growth sample. The inset shows in detail the temperature region around the critical temperature.

a minor effect and may originate from the slightly different V thicknesses of the two samples. The striking difference is the strong increase by 50% of the RRR from 9 to 14 and consequently an increased superconducting correlation length ξ_S. From the increased RRR we infer that H assisted growth reduces the defect density in the film and that no H impurities remain in the sample after growth.

The observed improvement of the structural and transport properties is beyond the natural run-to-run deviations in this series and markedly better than for other samples reported so far [4, 6]. We attribute the improvement properties of the samples through H assisted growth to two effects. First, H may act as a surfactant, causing an increased surface self-diffusion during growth. Second a cleaning of the surface at the growth front occurs due to the reduction of impurities, especially oxygen. Self-diffusion is one of the governing parameters of thin film or SL deposition, as a high surface mobility generally promotes smooth layer by layer growth [12]. Deposition at elevated temperatures to increase the surface mobility is often limited by the increased probability of interface alloying. At higher temperatures the H sticking coefficient decreases and the desorption rate increases. Thus for a fixed H partial pressure, H assisted growth is more effective at lower temperatures, compensating for the reduced surface mobility.

Hydrogen has been extensively used as a surfactant for the growth of semiconductors such as GaAs and Si [13–15]. Thereby the hydrogen atoms passivate the semiconductor surfaces and reduce the density of the interface states by saturating dangling bonds. Asahi *et al* concluded that H_2 promotes step-flow growth for Ga on a GaAs via an increased diffusion length [16].

Also for metals, adsorbed hydrogen is known to influence the kinetics of self-diffusion by modifying the surface energy barriers and should thereby also affect the growth of metallic films. However, no general trend has been observed so far, whether hydrogen promotes or inhibits the self-diffusion. While it has been reported to enhance the self-diffusion on Pt(110) [17], Be(0001) [18], Rh(100) and Rh(311) [19], it suppresses the self-diffusion on Pt(100) [20]. The effect appears to be dependent on the local geometry of the diffusion path. Obviously, for the system Fe/V, the presence of hydrogen supports smooth interfaces as can be seen from the XRR data. Additionally, we have evidence for chemically cleaner growth

conditions, resulting in a reduced number of defects in the bulk of a thick, H assisted deposition V layer.

In summary, the effect of H as a surfactant during sputter growth of epitaxial Fe/V SL on MgO(001) substrates was examined. Structural and transport measurements clearly show the superior quality of the samples in terms of thickness fluctuations, crystallinity and residual resistivity ratio. These results can be explained by an increased surface mobility of the metal atoms during growth and a chemical reduction of impurities in the presence of H at the growth front.

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