Magnetic properties and structure of sputter-deposited UN layers

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Although magnetism of amorphous and nanocrystalline systems is a fast developing field, there is only very limited information on magnetic properties of U compounds as a function of their microstructure. Few studies indicate that for ferromagnets like UGa_2 [1] or UFe_2 [2] the amorphous state leads to a strong suppression of T_C and μ_S. Our research concentrates on variations of magnetic properties of a band 5*f* antiferromagnet UN as a function of microstructure, affected by varying conditions of sputter deposition.

UN layers were prepared by a reactive sputtering in Ar atmosphere containing N_2. Choosing a proper partial pressure of N_2, one can achieve either the formation of UN or U_2N_3 [3]. The final stoichiometry is checked by XPS. For the magnetic study, UN was deposited on quartz glass substrates, having susceptibility weak and *T*-independent in the whole range below 300 K. This allowed the magnetic signal of the layer (≈ 1 mg UN) to be subtracted from that of the substrate when using a squid magnetometer.

The deposited UN layers turned out to be stable on air. Although XPS showed oxidation of few uppermost atomic layers, the lack of cracks prevented the degradation of the layers, which keep a mirror surface even after many months, and no UO_2 could be detected by X-ray diffraction experiments done at glancing angle of incidence.

By XRD, we studied so far layers deposited at substrate temperatures T_s between -200°C and 300°C. At all temperatures, a large lattice deformation was observed caused by a compressive residual stress within the layer. The largest residual stress of -4.3 GPa was found at T_s = -200 °C and it decreases with increasing T_s, as seen in Fig.1

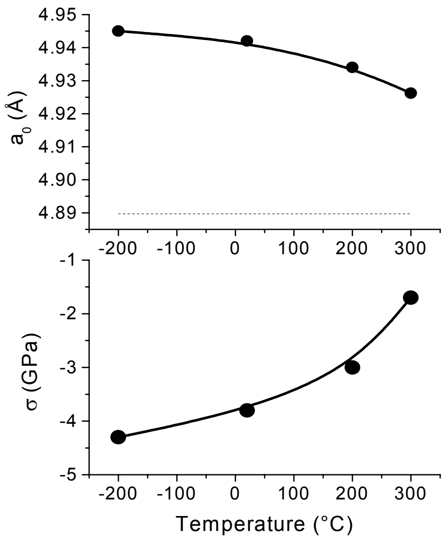

Fig. 1. Stress-free lattice parameter (top) and residual stress (bottom) as a function of the synthesis temperature. Dashed line in the upper figure shows the intrinsic value of the lattice parameter in UN (4.8897 Å).

At low temperatures, the UN layers are far from equilibrium. This is clear from the difference of the stress-free lattice parameter from its intrinsic value (Fig. 1), which decreases with increasing T_s. The layers have the texture {111} (perpendicular to the substrate). The degree of the preferred orientation decreases with increasing T_s. Noteworthy is the observed strong anisotropy of the lattice deformation, which is typical for thin films of some *fcc* nitrides, if they form columnar grains perpendicular to the surface [4]. The mean crystallite size for T_s = -200°C is about 230 Å, but exceeds 500 Å for the high-*T* synthesis. In view of the columnar shape of the crystallites, we may assume that their lateral size is much smaller than the mean size, whereas their length can reach the layer thickness (> 1000 Å). There is a high density of structure faults in the films, as it follows from a high microstrain (inhomogeneous variations in the interplanar spacing) at all temperatures, which decreased from $12*10^{-3}$ for T_s = -200°C to $10*10^{-3}$ for 300°C. The increase of the crystallite size and the decrease of the microstrain with increasing T_s were expected because the elevated temperature stimulates both the growth of crystallites and the diffusion of structure faults towards the grain boundaries.

Magnetic susceptibility of UN is normally characterized by a pronounced maximum at T_N = 53 K (see Fig. 2). In the paramagnetic range, it is described by a large negative Θ_p = -247 K and a reduced effective moment 2.66 μ_B [5]. Sputter deposited UN layers do no exhibit the anomaly around 50 K. Instead, $\chi(T)$ increases monotonously with decreasing T. A comparison of $\chi(T)$ in various fields indicates a small ferromagnetic component developing gradually below $T \approx 100$ K (Fig. 3). Generally, it can be attributed to not fully compensated AF coupling at grain boundaries (the grain boundaries represent a non-negligible volume fraction) and/or at numerous defects. This situation affects the susceptibility even in the paramagnetic state by contributing by an extra term with a small μ_{eff} but $\Theta_p \geq 0$ K. Magnetic history phenomena (difference of FC and ZFC regimes) show that the blocking of the weakly ferromagnetic clusters in random positions is appreciable for the high-T deposition, while low-T-deposited layers are magnetically "soft". Finally, for the highest T_s (400°C), an anomaly around 50 K emerges, meaning probably a partial recovery of the bulk UN magnetism.

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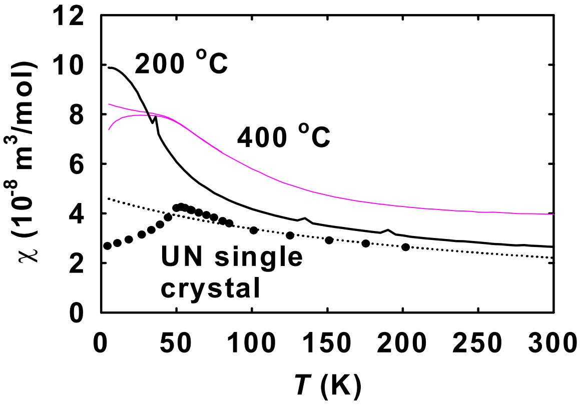

Fig. 2. Temperature dependencies of magnetic susceptibility of the UN layers synthesized at T_s = 200°C and 400°C compared with the bulk data, taken from Ref. 5.

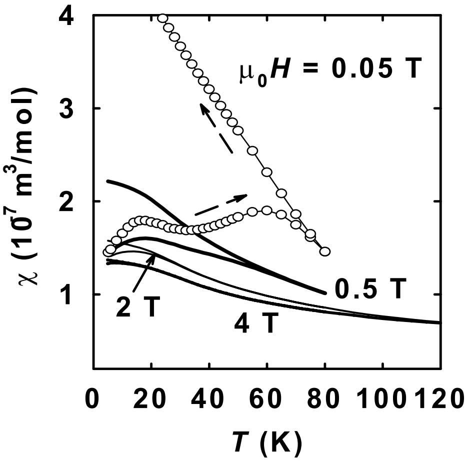

Fig. 3: Temperature dependence of magnetic susceptibility of UN synthesized at T = 300°C. The arrows indicate ZFC and FC regimes for the measurement in $\mu_o H$ = 0.05 T. Increasing magnetic field reduces the FC-ZFC difference, which becomes barely noticeable for $\mu_o H$ = 4 T. The ferromagnetic component developing gradually below 100 K corresponds to ≈ 0.01 μ_B/f.u. at T = 5 K.

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