Experimental report

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Title:	Magnetism and structure of e-Fe2O3films grown on gallium nitride buffer layer		
Research area:	Physics		

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Samples: Fe2O3 / GaN/ Al2O3

Instrument	Requested days	Allocated days	From	To
D17	8	6	06/06/2018	12/06/2018

Abstract:

The e-Fe2O3 phase is ferrimagnetic metastable iron oxide phase with huge magnetocrystalline anisotropy responsible for coercivity values exceeding 2 T in the nanocrystalline form and 1 T in epitaxial film. Moreover, the phase transition to the incommensurate spin structure has been observed in nanocrystalline e-Fe2O3 samples, but not in thin films. Besides this e-Fe2O3 exhibits ferroelectric properties and magnetoelectric coupling. It has been recently shown, that single crystal ferrimagnetic, antiferromagnetic and multiferroic layers of various iron oxides including the exotic e-Fe2O3 can be controllably grown by pulsed laser deposition beam epitaxy on GaN (0001) on sapphire substrates. Due to the specific shape of magnetization loops we suppose the existence of interfacial layer between e-Fe2O3 and GaN. The first polarized neutron reflectometry experiments at D17 setup (ILL) revealed the formation of 10 nm-thick interfacial layer with reduced magnetization as well as chemical composition. We propose to continue this research and to perform the layer-resolved magnetometry measurements for the samples grown at different conditions as a function of temperature.

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Magnetism and structure of ε-Fe_2O_3 films grown on gallium nitride buffer layer

Placing multiferroic layer with controllable magnetization/polarization into a contact with semiconductor is even more attractive due to additional functionality to control the optical, electronic and magnetic properties of heterostructure by applied voltage [1, 2]. One of the rare examples of material with spontaneous room-temperature magnetization and electric polarization is metastable ε-Fe_2O_3 [3]. Recently, crystalline layers of ε-Fe_2O_3 have been successfully synthesized on a number of oxide substrates and semiconducting GaN. Depending on growth parameters, the feasibility to synthesize the ε-Fe_2O_3, Fe_3O_4, α-Fe_2O_3 and γ-Fe_2O_3 phases on GaN(0001) has been demonstrated [4]. Moreover, stabilization of epsilon phase requires elevated growth temperature that leads to formation of a few nanometer-thick Ga-rich soft-magnetic transition layer at the interface between the iron oxide film and GaN substrate [5].

In present study, MgO buffer was used to eliminate Ga migration upon the film deposition. Stabilization of epsilon ferrite on MgO provides further opportunities to integrate this promising iron oxide phase with existing Fe/MgO-based devices. Polarized neutron reflectometry (PNR) technique was used to probe the structural and magnetic profiles of ε-Fe_2O_3/GaN and ε-Fe_2O_3/MgO/GaN heterostructures.

PNR experiment was performed at the D17 setup in the polarized time-of-flight mode. Sample temperature and magnetic field were controlled by 7 T vertical field cryomagnet with single-crystalline sapphire windows. Neutrons with wavelengths from 4 to 16 Å were integrated o maximize the incoming flux and keep the constant polarization 99%. Three different incident angles (0.8, 1.5 and 3.7 degrees) were chosen to access the Q_z range from 0.017 to 0.17 Å^{-1}. Intensity of the reflected beam was collected by two-dimensional ^{3}He position-sensitive detector, corrected for polarization efficiency, reflected beam divergence and wavelength resolution, and reduced by coherent summing method. Non-spin-flip reflectivity components R^+ and R^-, where +(-) denotes the incident neutron spins alignment parallel (antiparallel) to the direction of applied magnetic field, were acquired without polarization analysis. Supplemental x-ray reflectivity (XRR) measurements were performed using Panalytical X'Pert x-ray diffractometer at ILL.

Figure 1a shows XRR measured at room temperature and neutron reflectivity curves R^+ and R^- measured at T = 5 K and applied magnetic fields B = 0.025 T (remanent state) and B = 2 T (saturated state) as a function of the momentum transfer vector. Due to significant electron density contrast between GaN, MgO and Fe_2O_3, a set of well-pronounced Kiessig fringes appears in the XRR curve, which is not the case for the direct contact of iron oxide layer with GaN substrate. Measured XRR and PNR curves were fitted using GenX software [6] simultaneously assuming the model consisted of GaN substrate, MgO, intermediate iron oxide layer with unknown density and ε-Fe_2O_3 layers. This stack corresponds to the minimal model for convergence of the fitting routine. The depth-profiles of the x-ray ρ_e and nuclear neutron ρ_n scattering length densities (SLDs) extracted from the refined model are shown in Fig. 1b. The profiles reflect the chemical composition and density of the layers in heterostructure and structural roughness of the interfaces. Notably, we observe the transition layer at the iron oxide/MgO interface with thickness of 105±10 Å and reduced x-ray and neutron nuclear SLDs compared to the main ε-Fe_2O_3 volume of the film. The root mean square (RMS) roughness of all interfaces of the heterostructure is below 15 Å.

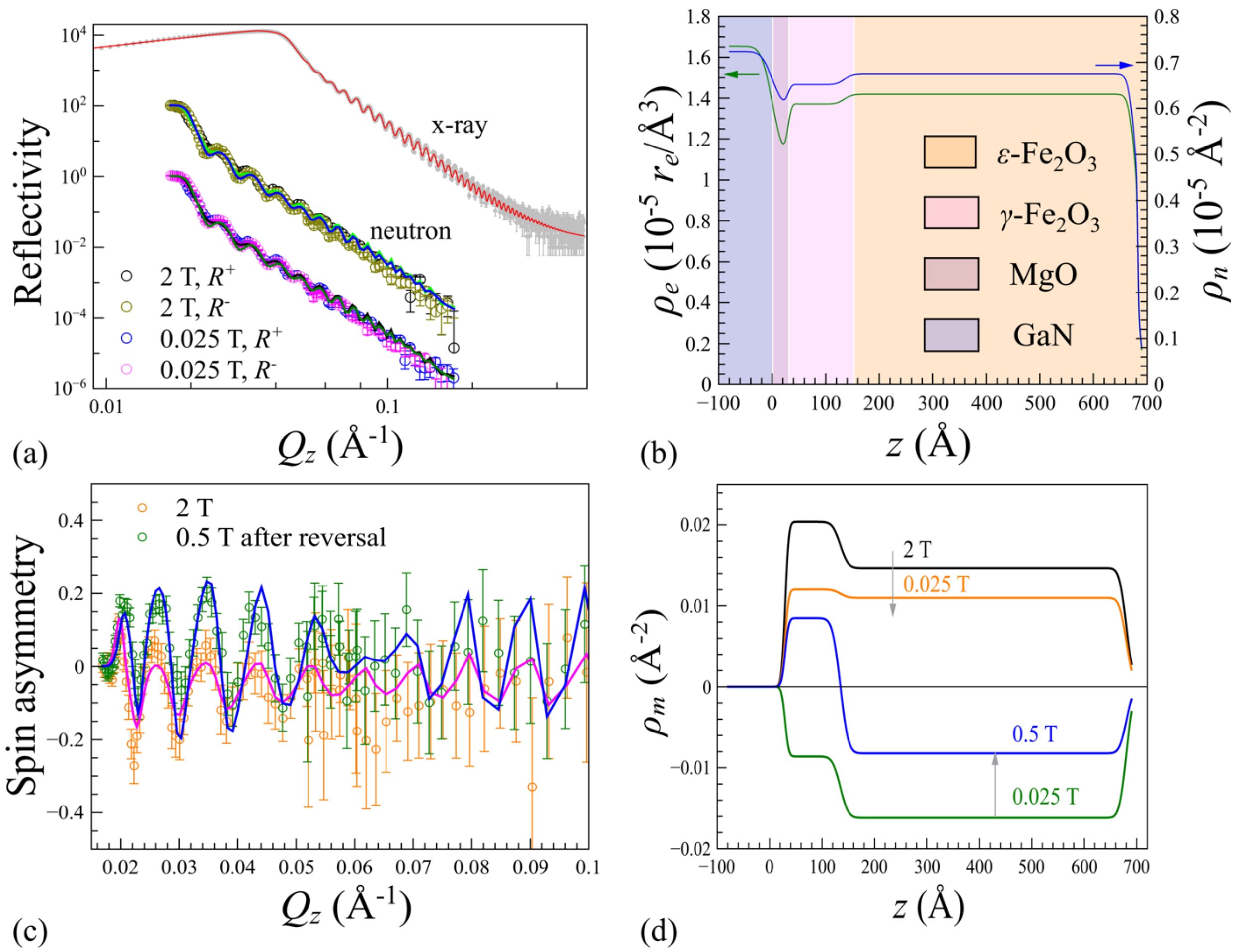

Figure 1 Measured (symbols) and fitted (solid lines) x-ray and neutron reflectivity curves as a function of momentum transfer (Q_z) on a logarithmic scale. X-ray scattering length density (SLD) ρ_e (black line), and neutron nuclear SLD ρ_n (red line) of ε-Fe_2O_3/MgO/GaN film as a function of the distance from the GaN layer surface (z) obtained from the fitting routine. The curves are shifted along vertical axis for clarity. (b). X-ray SLD ρ_e is given in the units of the classical electron radius r_e=2.8·10^{-15} m. PNR spin-asymmetry at applied magnetic fields B = 2 T and B = 0.5 T after magnetization reversal obtained from experimental data (symbols) and fitted models (solid curves). Neutron magnetic SLD ρ_m profile at B = 2 T, B = 0.025 T before and at B = 0.025 T, B = 0.5 T after magnetization reversal (d).

The in-plane magnetization profile of the heterostructure is encoded in the spin-asymmetry function $(R^+-R^-)/(R^++R^-)$. Measured and fitted spin-asymmetry ratios as a function of Q_z are shown in Fig. 1c for the following conditions: 1) in the saturation field B = +2 T and 2) in B = +0.5 T after the sample training in B = -2 T (corresponding to the lower branch of hysteresis loop). In the latter case the spin asymmetry ratio does not show complete reversal compared to the saturation state, indicating partial switching of in-plane magnetization. Magnetic contribution to the neutron SLD ρ_m can be easily converted to the magnetization units: $\rho_m = 2.853\cdot10^{-9}M$ Å^{-2}, where M is given in emu/cm^3 units. Fitted model reflects the magnetic separation of the iron oxide film into two sub-systems: main ε-Fe_2O_3 layer with saturation magnetization of $M_s \approx 50$ emu/cm^3 and interfacial layer with $M_s \approx 70$ emu/cm^3 (Fig. 1d). This situation is drastically different to the ε-Fe_2O_3/GaN heterostructure where magnetically degraded $GaFeO_3$ layer was observed at the interface due to the interfacial Ga diffusion [5]. Interestingly, at 5 K the magnetization switching of the interface layer takes place at B = +0.5 T, while magnetically hard ε-Fe_2O_3 layer is in the remnant state.

In conclusion, in the present work we have shown first to our knowledge epitaxial stabilization of the ε-Fe_2O_3 thin film on MgO(111) surface by pulsed laser deposition. In contrast to previously investigated ε-Fe_2O_3/GaN(0001) system the thermal migration of Ga atoms from the substrate is successfully blocked by the MgO barrier. Interestingly, an intermediate phase is formed at the initial stage of the iron oxide growth. A complimentary combination of structural characterization and polarized neutron reflectometry techniques unambiguously identified this phase as cubic γ-Fe_2O_3.

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[4] S. Suturin, et al., Physical Review Materials 2, 073403 (2018).

[5] V. Ukleev, et al., Scientific Reports 8, 8741 (2018).