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Surface order dependent magnetic thin film growth: Fe on GaN(0001)

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Abstract

X-ray photoemission spectroscopy and X-ray magnetic circular dichroism have been used to study the growth process, chemical composition and magnetic character of iron deposited on ordered and disordered GaN(0001) surfaces. On the (1×1) ordered surface the Fe grows uniformly but with disruption to the substrate surface, subsequently nitrogen desorbs from the surface, some of which diffuses into the Fe overlayer. The film is magnetically fractured, with high magnetic coercivity and broad switching fields. Conversely, the gallium rich disordered surface protects the underlying substrate from any disruption and initially induces non-uniform growth, the Fe clusters coalesce at ~12 Å, to produce a uniform film with desirable magnetic characteristics. Films beyond this point (>12 Å) indicate sharp hysteresis loops with low coercivities. For the resultant film (36 Å) we measure a magnetic moment of 2.02 μ_B, in agreement with bulk bcc iron (2.068 μ_B).

Keywords: Metal–semiconductor interfaces; Magnetic thin films; Wide band gap semiconductors

Magnetic transition metals with a T_c above room temperature grown on semiconductor substrates offer a viable platform to create semiconductor based spin current devices [1]. Investigating the characteristics of such interfaces will help to understand and ultimately avoid interfacial spin scattering.

Gallium nitride and its related ternary compounds typically have large defect densities (to the order of 10^{10} cm^{-2} [2]). Despite this, recent GaN based spin coherence measurements indicate spin current lifetimes of ~20 ns at 5 K [3]. Calculations have also predicted that because of the large conduction band density of states and the small spin–orbit interaction, spin lifetimes in GaN maybe up to three orders of magnitude longer than in GaAs [4]. The ability to sustain long spin lifetimes is necessary for device fabrication where relatively long length scales are required, thus GaN presents itself as a strong potential candidate for such devices.

Studies of thin metal-film growth on GaN have typically focused on ohmic contact formation. Nickel deposition on GaN(0001) was investigated in detail by Bermudez et al. [5]. Nickel was found to grow uniformly whereupon the surface structure degraded rapidly with increasing coverage. Bermudez also investigated the initial deposition of Mg on GaN surfaces, introducing the concept of 'free' Ga produced by Mg–Ga exchange that resulted in isolated surface gallium [6]. This gallium displacement is a prominent feature of metal deposition on GaN surfaces [6,7].

In this experiment, iron was deposited on both reconstructed and disordered GaN surfaces to investigate the comparative effects of substrate surface order on the growth, chemical and magnetic characteristics of the over-layer. The experiment was performed at beamline 4IDC at the Advanced Photon Source, which produces intense circularly polarized photons between 400 eV and 3 KeV [8]. Iron was deposited in situ on MOCVD grown *n*-GaN by e-beam evaporation. The deposition rate was calibrated using a quartz crystal balance. The substrates were kept at 150 K during the Fe deposition. Low energy electron diffraction (LEED) indicated the presence of

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surface order or lack thereof. Core-level (CL) photoemission spectroscopy investigated the surface chemistry during Fe deposition. The magnetic character of the Fe film was studied by X-ray magnetic circular dichroism (XMCD). Hysteresis loops using XMCD were obtained using total electron yield (TEY) at the Fe L_3 edge and applying a sweeping magnetic field within the film plane. The total magnetic moment, m_T, $(m_L + m_S)$ of the final films was measured employing sum-rule analysis of the XMCD across the Fe $L_{3,2}$ edges [9].

X-ray magnetic circular dichroism is the difference between spectra taken with both left and right incident photon helicity on a magnetically aligned sample. The same effect may be obtained by holding the incident helicity and switching the sample magnetization direction as illustrated in the inset of Fig. 1 [10]. The total electron yield measured XMCD was taken with the sample in magnetic remanence. Both TEY spectra across the Fe $L_{3,2}$ edges and their difference i.e. the XMCD, are plotted in Fig. 1. The XMCD signal has been treated to account for a 35° angle between the sample magnetization and the incident polarized light, as well as the degree of polarization of the incident photons (96%). In order to measure the magnetic moment m_T, the electron occupancy of the 3d states is required. A bulk Fe electron occupancy of 6.61 was used from previously published work [11].

To prepare the substrate surfaces both were sputter cleaned in a nitrogen atmosphere at 5×10^{-5} Torr and annealed at ~850 °C to clean and reorder the surface. The procedure resulted in a clear (1×1)-(0001) ordered surface. The samples polarity (0001) indicates a Ga face [12]. One sample was subsequently sputtered to produce a disordered surface with no LEED pattern. Photoemission indicated that both surfaces were clean with only a trace of oxygen and no carbon.

Fig. 2 presents the relative integrated intensities of the Ga 3d and N 1s CL spectra with increasing Fe coverage for both samples. The data is plotted alongside the predicted decay for an ideal interface. For the ordered surface grown film, the Ga 3d CL decay closely matches the expected fall off, indicating uniform growth with little intermixing similar to what has been reported for the growth of Ni on GaN(0001) [5]. The hexagonal LEED pattern becomes diffuse and disappears after a few layers of Fe growth. The N 1s CL intensity shows a significantly sharper decay path than predicted, indicating that nitrogen desorbs from the surface upon Fe deposition. The N 1s signal plateaus at ~15 Å coverage with a small but finite intensity suggesting that a small quantity of nitrogen diffuses into the film during deposition.

For the disordered surface grown film, Fig. 2c shows the Ga 3d CL intensity larger than calculated, with further deposition the two converge at ~18 Å coverage. The data suggests three dimensional growth up to ~12 Å, and between ~12 Å and ~18 Å the iron clusters coalesce, forming a more uniform film. A similar growth sequence has been identified for Fe on GaAs(100) [13]. Fig. 2d plots the N 1s CL intensity, it shows a similar attenuation to that of the Ga 3d intensity. The surface disordering process leaves a gallium rich surface barrier protecting the underlying substrate nitrogen. Although the ordered surface has a Ga face, this is insufficient to protect the substrate surface. The Ga 3d–N 1s CL intensity ratio for the disordered surface is 22% larger than the ordered surface indicating the increase in Ga present on the disordered surface.

Fig. 3 shows Ga 3d CL spectra for both films. The spectra have been normalized to equal peak height. Comparison of the Ga 3d spectra for both clean surfaces in Fig. 3a indicates a lower binding energy feature for the disordered surface, indicative of the excess surface Ga in a different chemical environment from that in the substrate. In Fig. 3b, iron deposition on the ordered surface introduces a similar lower binding energy feature. With further deposition the line shape remains relatively constant, as 'free' Ga [6] becomes localized at the interface and protects the substrate from further exposure. For the disordered surface in Fig. 3c, there is no change in the line shape with Fe deposition as the Ga rich surface protects the substrate.

The normalized N 1s spectra for both samples are shown in Fig. 4. Spectra from the ordered surface show a broad lower binding energy feature with increasing Fe coverage, this indicates nitrogen from the substrate diffusing into the Fe overlayer. On the disordered surface, the FWHM remains constant throughout the deposition as the initial Ga rich surface provides a protective barrier and the substrate nitrogen remains unaffected.

The magnetic character of both films is considerably different as shown in Fig. 5a by the Fe L_3 XMCD hysteresis loops of both samples taken at the magnetic onset thickness. The iron film grown on the disordered surface demonstrates sharp stepped square loops from 12 Å and higher. While the difference in coercivity between the two loops shown may be partially due to different film thickness, an increase in the density of magnetic domains will increase the coercivity, as seen in the ordered surface grown film.

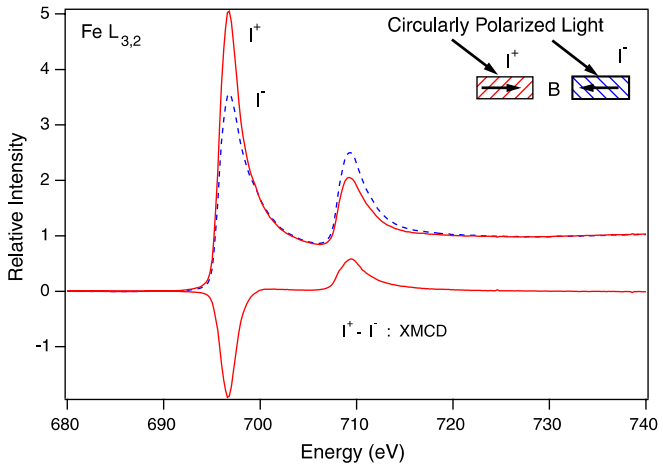

Fig. 1. Illustration of the Fe $L_{3,2}$ TEY spectra taken with circularly polarized X-rays, with anti-parallel applied magnetic fields.