Iron Nanoparticles in Fe/GaN

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In this work we present the formation of Fe-alloy nanoparticles upon the deposition of Fe onto GaN(0001) surfaces *via* metalorganic chemical vapor deposition. The growth has been monitored *in-situ* by means of spectroscopic ellipsometry and the samples have been characterized *ex-situ* by means of high-resolution x-ray diffraction, high resolution transmission electron miscroscopy, atomic force microscopy and SQUID.

Introduction

In the last years physical processes governed by spin-dependent phenomena have been intensively studied. Ferromagnetic metals such as iron are natural sources of spin-polarized electrons, and semiconductors have shown to be an ideal host for the transport and manipulation of spins. Therefore, structures like Fe/GaAs, Fe/AlGaAs and Fe/GaN are of high interest. Recently spin injection efficiencies up to 50% in heterostructures like Fe/GaAs [1] have been demonstrated. Theoretical calculations have predicted the spin relaxation lifetime in GaN as being three orders of magnitude longer than in GaAs, arising great interest on the hybrid Fe/GaN system for future quantum information processing.

So far the Fe/GaN heterostructure has been achieved by depositing Fe and GaN separately in two different steps. In this work we report the first attempts of fabrication of Fe films on (0001)GaN fully accomplished via metalorganic chemical vapor deposition (MOCVD). Special attention has been devoted to the Fe nucleation on the nitride surface and the growth has been monitored in-situ via spectroscopic ellipsometry (SE) and ex-situ via high-resolution x-ray diffraction (HRXRD), atomic force microscopy (AFM), high resolution transmission electron microscopy (HRTEM) and SQUID magnetrometry for the structural and magnetic characterization of the structures respectively.

A systematic study of the effect of Fe deposition onto the (0001)GaN templates has been performed, with particular attention to the early stages of Fe nucleation. Several series of samples were fabricated at different Fe source flux (50 – 400 sccm) and different substrate temperature (150 – 1020 °C). Insights into the surface roughness and its evolution with Fe deposition could be gained by routinely performing in-situ SE upon growth. SIMS studies gave information on the diffusion of Fe into the GaN buffer and a considerable effort has been devoted to the identification of a suitable window of growth parameters allowing a compromise between the substrate temperature required to ensure an efficient performance of the metallorganic ferrocene source and the unwanted diffusion of Fe into the buffer.

Experimental Procedure

All samples have been grown in an AIXTRON 200RF horizontal reactor MOCVD system using Trimethylgallium (TMGa), ammonia (NH_3) and Ferrocene (Cp_2Fe) as precur-

sors. The GaN(0001) samples were fabricated using a standard procedure of substrate nitridation, GaN nucleation layer deposition at 540 °C and 1 μ m GaN Buffer deposition at a constant pressure of 200 mbar and 1020 °C in N₂ atmosphere. Afterwards, Fe was directly deposited on top at 200 °C under N₂ atmosphere.

Results

Surface roughening became evident after studying the samples with atomic force microscopy. These measurements clearly show the changes in the GaN surface morphology with Fe deposition. As can bee seen in the images reported in Fig. 1, there is a significant increment in surface roughness with increasing deposition time, and after 60 minutes of Fe growth we can observe the presence on the sample surface of hexagonal structures, which disappear for longer deposition time.



Fig. 1: AFM images of GaN, and Fe/GaN samples with Fe deposition times of 30 min (top right), 60 min (bottom left), and 120 min (bottom right).

TEM studies carried out on the samples presenting the hexagonal reconstructions on the surface revealed the presence of nanoparticles distributed few nanometers below the surface as reported in Fig. 2. The nanoparticles size is of several nanometers and energy dispersive spectra (EDS) confirmed the presence of Fe inside the particles.

Previous HRTEM studies on (Ga,Fe)N samples uncovered the presence of Fe-rich nanocrystals embedded into the GaN matrix for samples with Fe concentrations above the solubility limit at the given growth conditions [2]. The comparison of these Fe-rich nanocrystals with the nanoparticles found in the Fe/GaN samples will contribute in shading new light on the origin of the magnetic response of the Fe/GaN samples.

Figure 3 summarizes the SQUID magnetization curves, measured at 200 K for a Fe/GaN structure (where Fe has been deposited for 60 minutes onto the GaN surface) compared with a (Ga,Fe)N sample. The magnetic response of (Ga,Fe)N shows a pro-

nounced hysteresis loop – generally assigned to a ferromagnetic behavior –, while the Fe/GaN structure only shows two paramagnetic contributions: a temperature-dependent Curie component at low temperatures and a temperature-independent one above 30 K.



Fig. 2: HRTEM image of sample with 60 minutes Fe deposition, showing the presence of Fe-rich nanoparticles close to the surface.

The paramagnetic signal coming from the Fe/GaN sample suggests the formation of antiferromagnetic clusters (non metallic nanoparticles) close to the surface, tentatively identified as FeN clusters. This result can be supported by the HRTEM images, from where we can state that the nanoparticles are located *below* the surface rather than on the surface.



Fig. 3. Comparison of magnetic response vs. magnetic field at 200 K for (Ga,Fe)N and Fe/GaN.

References

- [1] C.Adelmann et. al, Phys. Rev. B 71 (2005) 121301.
- [2] A. Bonanni et al., Phys. Rev. B (to be published).