Advanced electron microscopy of Fe-N precipitates in epitaxial GaN layers

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Introduction

The origin of ferromagnetism in transition metal (TM) doped semiconductor arsenides, oxides and nitrides is highly controversial. Recent studies have underlined the need to understand the incorporation of magnetic ions into the host lattice [1], i.e., to study at the nanoscale whether the material is a dilute magnetic alloy, whether magnetic dopants have aggregated to form coherent TM-rich regions, or there is precipitation of secondary magnetic phases.

In this work, we employ advanced transmission electron microscopy (TEM) techniques to study the local structural, chemical and magnetic properties of Fe-doped GaN epitaxial layers.

Fe-doped GaN

Fe-doped GaN layers were grown by metalorganic chemical vapour deposition on (001) plane sapphire at a substrate temperature of 950°C. The growth conditions, the effect of growth temperature and the results of magnetic measurements are described elsewhere [3].

Structure characterisation

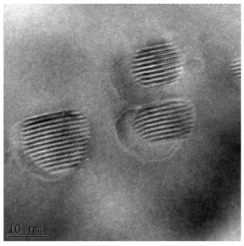
Cross-sectional (Ga,Fe)N samples were prepared using mechanical polishing and Ar ion milling with a Fischione 1010 system. Each cross-sectional specimen was finished by low-energy Ar ion milling at 500 eV in order to minimize sample damage.

TEM studies were carried out using FEI Tecnai G2 and FEI Titan microscopes, with the latter equipped with a monochromator, energy dispersive x-ray spectroscopy, electron energy-loss spectroscopy, probe aberration corrector and a biprism for electron holography.

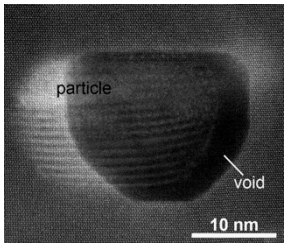
FEI Titan microscope at DTU Cen.

For low-temperature studies, a LN_2 cooling TEM specimenholder was used.

Fe-N precipitates in the GaN matrix were observed using several TEM techniques. Precipitates larger than 10 nm were associated with voids, while particles without voids typically had sizes of ≈5 nm.

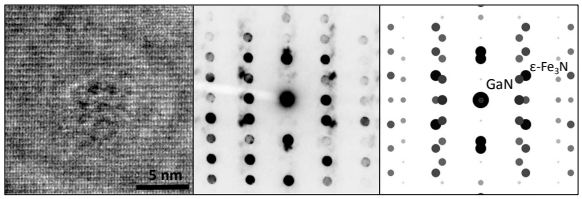


Bright-field TEM image of Fe-N precipitates and voids.



Scanning TEM annular dark-field image of a precipitate and an associated void.

Nano-beam diffraction was used to determine the structures of the precipitates



The precipitates were identified as hexagonal ε-Fe_3N (space group 182, a=0.474 nm c=0.44 nm). They were in epitaxial with the GaN with GaN[100](001)//ε-Fe_3N[210](001). The ε-Fe_3N [210] viewing direction was used to determine the lattice parameters of the particles along the *b*- and *c*- directions as 0.458 and 0.430 ± 0.002 nm, respectively. The lattice spacings are smaller than those reported in the literature. However, ε-Fe_3N is known to show a strong variation in lattice parameter with nitrogen content [4].

References

[1] A. Bonanni and T. Dietl, *Chem. Soc. Rev.* **39**, 528 (2010)
[2] P.A. Midgley and R.E. Dunin-Borkowski, *Nature Materials* **8**, 275 (2009)
[3] A. Navarro-Quezada et al., Phys. Rev. B **81**, 205026 (2010)
[4] T. Liapina et al. *Acta Materialia* **52,** 173-180 (2004)

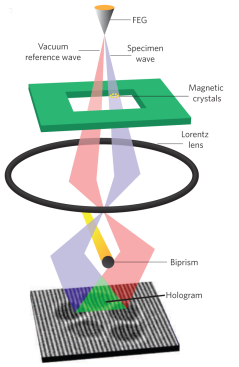
Off-axis electron holography

Off-axis electron holography involves the use of an electron biprism to overlap a high-energy electron wave that has passed through a TEM specimen with another part of the same electron wave that has passed only through vacuum [2].

The resulting interference fringe pattern can be interpreted to retrieve the phase modulation of the electron wave, which is described by the "Aharonov-Bohm" expression

$$\varphi(x,y) = C_E \int V(x,y,z)dz - \frac{\pi}{\phi_0}\int A_z(x,y,z)dz$$

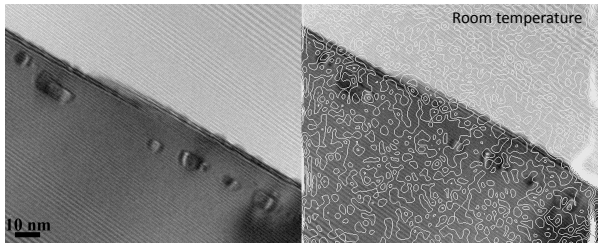
where V is the electrostatic potential, and A_z is the component of the magnetic vector potential in the electron beam direction z. Quantitative information about the magnetic properties of the sample is obtained from the phase image reconstructed from the hologram.



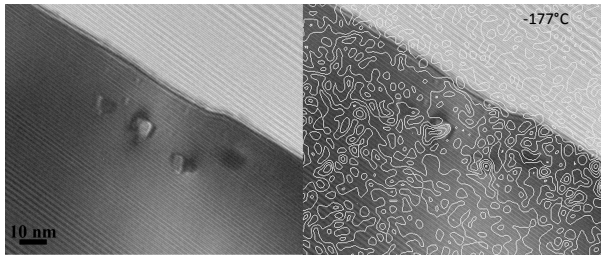
Schematic diagram of the experimental setup used for recording off-axis electron holograms [2].

Electron holography of Fe-N precipitates

Electron holograms were acquired in magnetic-field-free conditions after subjecting the specimen to the magnetic field of the microscope objective lens (≅2T) in opposite directions.



Electron hologram and corresponding magnetic induction map recorded at room temperature. The contours were generated from the magnetic contribution to the measured electron holographic phase shift, after subtracting the mean inner potential contribution to the phase.



Electron hologram and corresponding magnetic induction map recorded at -177°C.

Diffraction contrast and charging of the sapphire substrate made the electron holography study very difficult. A thin carbon layer was deposited on the sample surface to partially overcome the charging problem.

The present study revealed no detectable magnetic signal from the Fe-N particles. This conclusion is not yet definitive, and further electron holography studies are planned.

Summary

- Advanced electron microscopy has been used to identify and characterise ε-Fe_3N particles associated with voids in epitaxy with GaN.
- Electron holography was performed at room temperature and at -177°C. No magnetic signal was identified.
- The origin of the enigmatic magnetic properties of magnetic semiconductors will be studied in depth in more comprehensive electron holography study.