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Published in:
MC 2011 Kiel

Publication date:
2011

Citation (APA):
Kovács, A., Schaffer, B., Moreno, M. S., Kasama, T., Craven, A. J., & Dunin-Borkowski, R. E. (2011). Evidence for molecular N2 bubble formation in a (Ga,Fe)N magnetic semiconductor. In MC 2011 Kiel: Microscopy Conference 2011 (pp. M2-P523). DGE – German Society for Electron Microscopy.

Evidence for molecular N_2 bubble formation in a (Ga,Fe)N magnetic semiconductor

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Keywords: EELS, STEM, (Ga,Fe)N

Fe-doped GaN semiconductors are of interest for combining the properties of semiconductors and magnetic materials [1]. Depending on the growth temperature used, Fe can either be distributed homogenously in the GaN host lattice or it can accumulate in the form of Fe-N nanocrystals. As a result of the small size of the nanocrystals and the sensitivity of Fe-doped GaN to specimen preparation for electron microscopy, the formation and physical properties of Fe-N nanocrystals in GaN are not yet fully understood.

The (Ga,Fe)N samples examined below were grown by metalorganic chemical vapour deposition [1] and studied in cross-sectional geometry using several transmission electron microscopy (TEM) techniques. Great care was taken during TEM specimen preparation to minimize Ar ion-beam induced artefacts. Fe-N nanocrystal formation was observed in samples that had been deposited at temperatures higher than 850°C. Interestingly, most of the Fe-N nanocrystals were found to be associated with closely adjacent void-like features in both TEM and scanning TEM (STEM) images, as shown in Figure 1(a). Here, we use aberration-corrected STEM and electron energy-loss spectroscopy (EELS) to show that these features are bubbles filled with molecular N_2. In order to interpret our experimental results, we calculate the N K spectrum for GaN using self-consistent real-space multiple-scattering calculations using FEFF 9.05, which allows to include the experimental conditions.

A dedicated STEM EELS measurement was performed across a single nanocrystal (shown in Fig. 1 (b)) embedded in the GaN host. A 100 kV acceleration voltage and a distributed-dose acquisition routine [2] was used to either minimize or control electron beam induced damage during the experiment. Figure 2 (a) shows selected N K edge spectra recorded from the nanocrystal, the adjacent N_2-containing region and the GaN host. The N K edge shows a typical three-peaked structure between 400 and 407 eV. Figure 2 (b) shows the experimental spectrum acquired from the bubble alongside with the experimental spectrum of N_2 [3] and simulation of a GaN spectrum.

Figure 3 shows the result of an experiment that provides direct evidence for the presence of N_2 gas by using the electron beam to puncture the bubble adjacent to the Fe-N nanocrystal and to release the gas into the microscope. We used a static sub-Angstrom beam with a current of about 350 pA to make a hole in the specimen exactly at the position of the bubble, while recording EELS spectra every 40 seconds. The intensity of the characteristic first peak of the N K edge at 401 eV was observed to decrease suddenly when the gas was released. The quantitative determination of the pressure of the N_2 gas in the bubble from the recorded EELS spectra is in progress.

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4. This work was supported by the “FunDMS” Advanced Grant of the European Research Council within the “Ideas” 7th Framework Programme of the European Comission. EPSRC is acknowledged for financial support under grant reference EP/D040205/1.

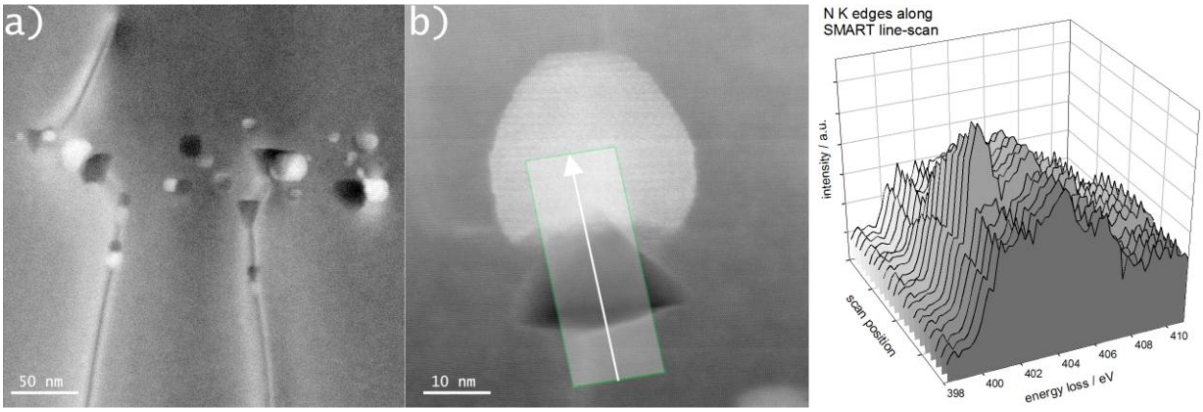

Figure 1. (a) HAADF STEM image of Fe-N nanocrystals and N_2 bubble in GaN. The nanocrystals appear with bright contrast. (b) Higher magnification image and averaged line-scan EELS measurement acquired from a single nanocrystal-bubble pair from the area indicated by the box. The EELS spectra show systematic changes in the N K edge fine structure.

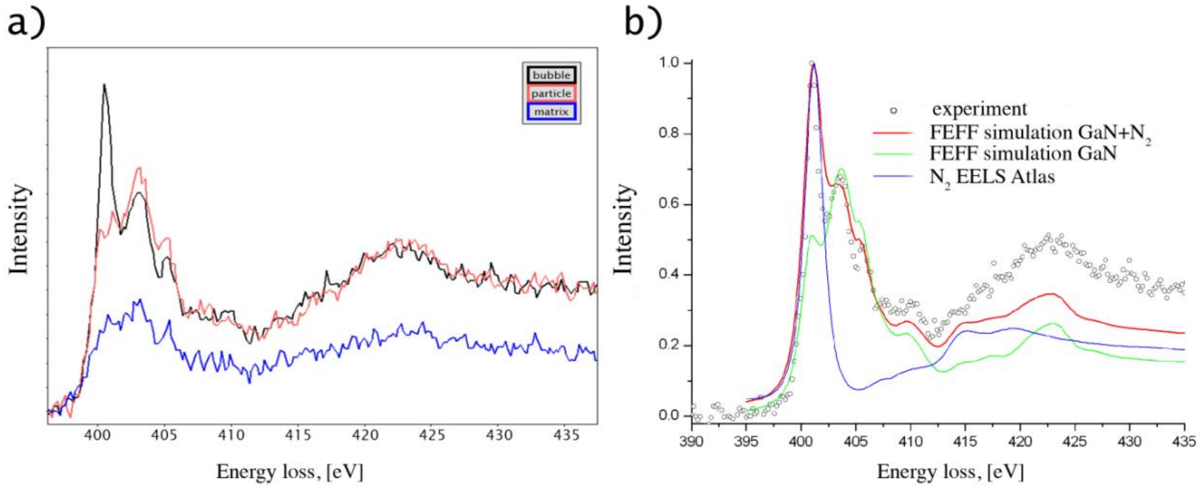

Figure 2. (a) Background-subtracted N K edge spectra measured from the Fe-N nanocrystal, N_2 bubble and GaN host lattice shown in Fig. 1. (b) Experimental spectra acquired from the bubble alongside an experimental measurement from N2 from the EELS Atlas, and simulated spectra calculated for GaN.

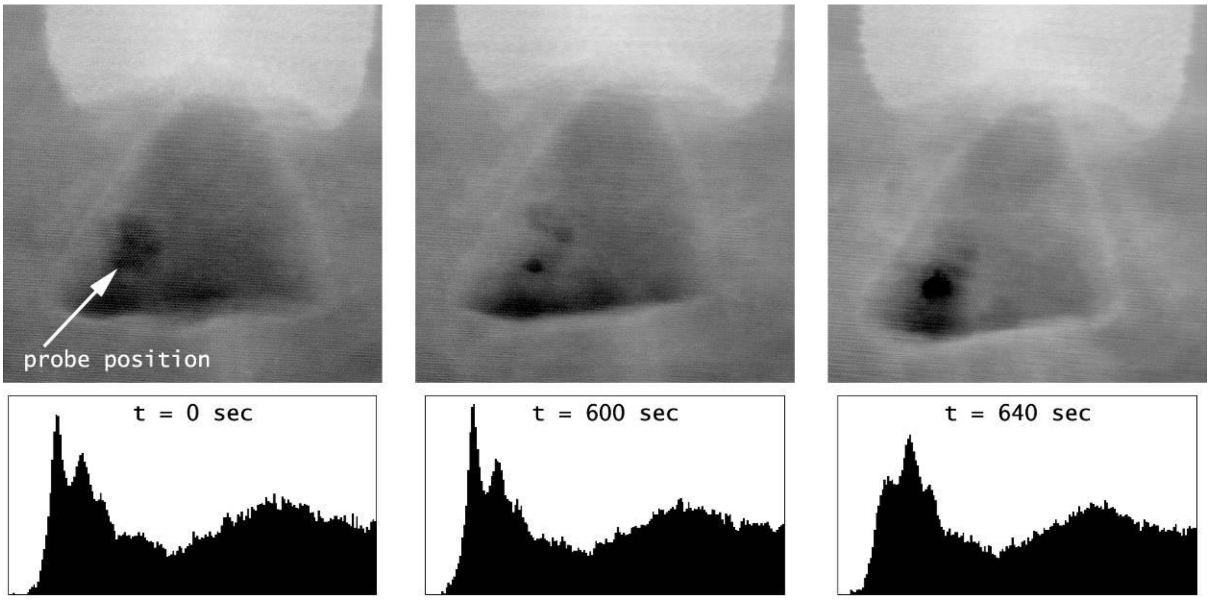

Figure 3. Time series of HAADF STEM images and N K edge measurements recorded while drilling a hole through the GaN using a stationary, focused electron probe. After 10 minutes a hole connects the bubble to the microscope vacuum and the N_2 gas is released. The intensity of the first peak in the spectrum is then reduced significantly.