Electronic Supplementary Information (ESI)

Atomic force microscopy analysis

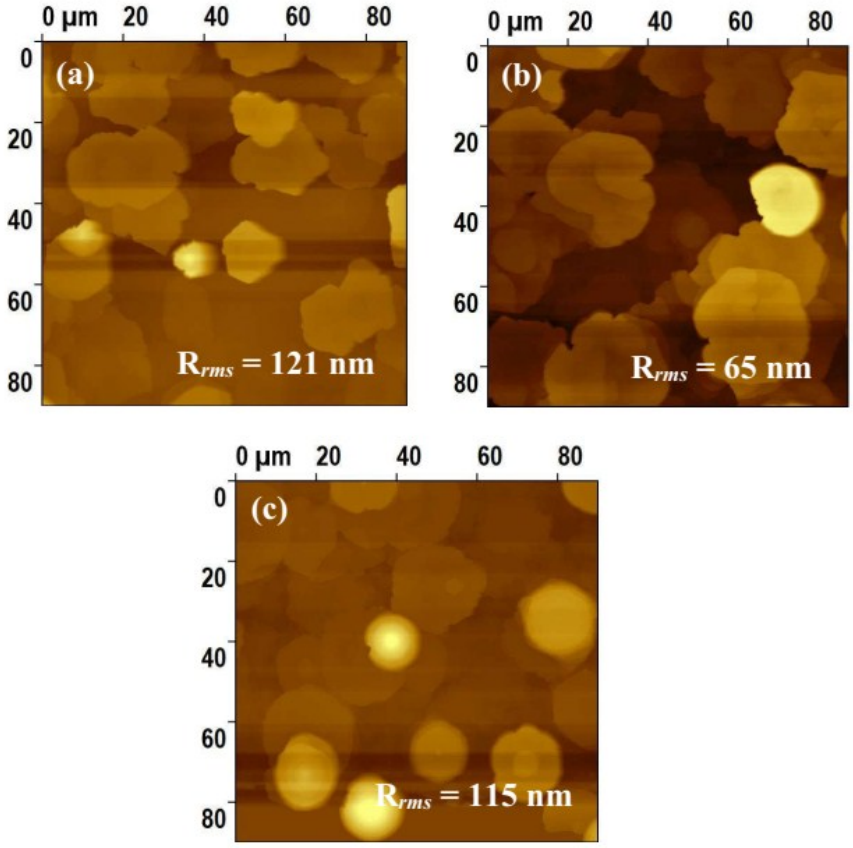

Figure S1. Atomic force microscopy (AFM) images of (100 x 100) µm^2 scan areas for sample (a) SMn0, (b) SMn225, and (c) SMn450.

We present some results on the effect of Mn co-doping on the surface morphology of the studied samples. Usually, the self-assembly of Fe_xN NCs results in a rough surface morphology. However, the efficiency of spin transfer in ferromagnet/non-magnet hybrid structures is enhanced by atomically flat surfaces and the use of Mn as a surfactant during the MOVPE process was proven to promote the layer-by-layer growth and to hinder aggregation.[1]

Information on the morphology on the sample surface is obtained from tapping mode atomic force microscopy (AFM) performed with a VEECO Dimension 3100 AFM set-up. Scans of (100 x 100) µm^2 measured on the samples are reported in Fig. S1(a) – S1(c) for samples SMn0, SMn225 and SMn450 respectively. A rough surface morphology, characteristic of Fe_xN NCs

embedded in a GaN matrix, is observed for all three samples under investigation. The root mean square roughness, R_{rms}, for samples SMn0, SMn225 and SMn450 are estimated to be ~121 nm, 65 nm and 115 nm, respectively. The reduction of R_{rms} for SMn225 from that of SMn0 indicates that introduction of Mn in the lattice assist in smoothening of the surface due to its role as a surfactant. However, for sample SMn450 with the highest Mn co-doping, the value of R_{rms} is estimated to be identical to that of the undoped sample SMn0. This behavior is attributed to the evolution of the secondary ε-Fe_3N phase alongside the incorporation of Mn in the γ'-$Ga_xFe_{4-x}N$ phase in this sample which leads to new defect centers that are the main reason for this mesa like overgrowth in δ-doped layers in MOVPE grown GaN.

[1] T. Devillers, T. Li, R. Adhikari, G. Capuzzo, and A. Bonanni, *Cryst. Growth Des*. 2015, **15**, 587 DOI: 10.1021/cg501144w

Additional TEM images

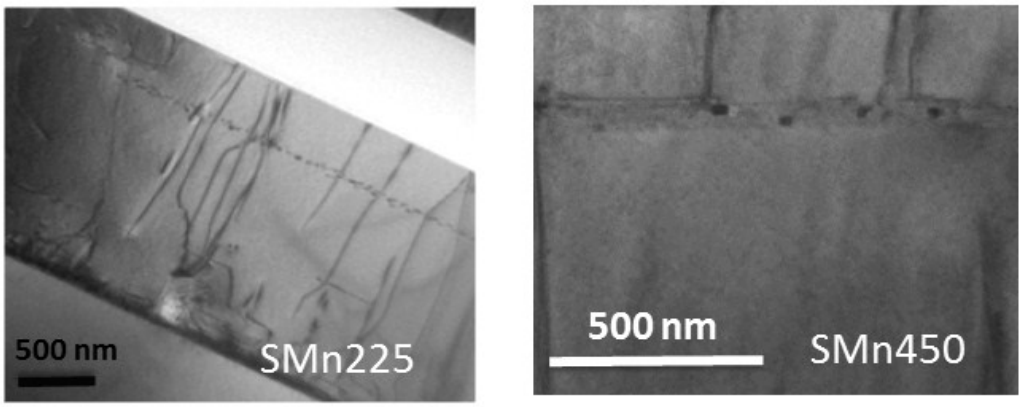

Figure S2. Cross-sectional TEM images of sample SMn225 (left) and SMn450 (right)