Building Nanospintronic and Nanomagnetic Structures: Growth, Manipulation, and Characterization at the Atomic Scale

NSF NIRT Grant 0304314

PIs: **Arthur R. Smith, Saw-Wai Hla, Nancy Sandler, and Sergio Ulloa**
Ohio University, Department of Physics and Astronomy, and
Nanoscale and Quantum Phenomena Institute, Athens, OH 45701

The field of *spintronics* is centered around taking advantage of the spin degree of freedom of electrons to create multi-functional devices, e.g. devices which integrate electronic and magnetic components into a single chip. Such possibilities make this an exciting field, and one with significant challenges. Under intense investigation for many years is the possible integration of magnetic metals with semiconductors (i.e. Fe/GaAs). Alternatively, the class of materials known as dilute magnetic semiconductors (DMSs) has the potential for homostructural electronic and magnetic layers in which spin-polarized carriers mediate ferromagnetism. For practicality, such a material should have a Curie temperature $T_C > 300K$. Magnetic-doped GaN has been predicted to be such a material.[1]

Initial studies,[2] including some work of our own, [3] concentrated on Mn as the magnetic impurity in GaN. More recently, however, attention has shifted toward Cr as a magnetic dopant of choice in GaN, based on both theoretical and experimental studies.[4] To date, most of the work done in these areas has focused attention on bulk magnetic properties. Yet for future spintronic devices, the nanometer scale will become increasingly important.

The objectives of this NIRT project are centered around the experimental and theoretical exploration of magnetic dopants in GaN films and particularly in the surface layers of GaN.[5] Ultimate goals include not only growing and characterizing *at the atomic scale* magnetic and spintronic nanostructures, but also using atom manipulation to create artificial magnetic and spintronic nanostructures. The surfaces are prepared by thermal evaporation (molecular beam epitaxy) using Cr and Ga effusion cells and a plasma source for N. Without removal from ultra-high vacuum (UHV), we then use scanning tunneling microscopy (STM) to study the properties of these magnetic-doped GaN surfaces. Multiple theoretical tools are also available in this project. For example, we have been using the technique known as SIESTA to model both wurtzite and cubic GaN surfaces. In the case of *c*-GaN (001), we recently reported the first STM images of a Ga tetramer reconstruction, and SIESTA was successfully used to model the experimental data.[6]

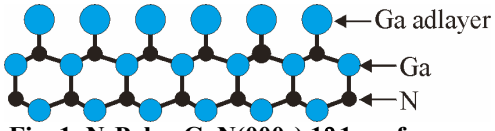

Fig. 1 N-Polar GaN(000) 1?1 surface

A number of results have been reported for the magnetism of Cr-doped GaN.[4] As part of our study of ternary CrGaN, we have also grown and studied binary CrN(001); this material has a rock-salt type structure, characteristic of many of the transition metal nitrides.[7] In our work on ternary CrGaN, the structural and magnetic properties have been studied vs. the growth conditions. We have grown N-polar CrGaN on sapphire(0001) with a 2-5 % Cr/Ga under 3 growth conditions: N-rich, metal-rich, and Ga-rich. This N-polar face of *w*-GaN is known to have an intrinsic 3_3 reconstruction which forms on top of the 1_1 Ga adlayer. A model of the 1_1 Ga adlayer of clean N-polar *w*-GaN is shown in Fig. 1. Normally, in GaN growth in Ga-rich conditions, Ga adatoms will be found atop the Ga adlayer.

Our results so far indicate that Cr incorporation occurs under all 3 growth conditions. Moreover, in all 3 conditions, we observed a 3_3 reconstruction. To investigate further the effect of Cr on the surface, a series of experiments were performed. In one, sub-monolayer amounts of Cr were deposited on top of the clean GaN 1_1 surface, and a 3_3 reconstruction formed. Thus, Cr atoms appear to behave similar to Ga adatoms under Ga-rich conditions.

The smoothest CrGaN surfaces are obtained under Ga-rich conditions. Interestingly, excellent magnetic properties were also found for the Ga-rich CrGaN growth. Shown in Fig. 2(a) is a magnetization vs. temperature measurement for a CrGaN sample grown with Ga:N flux ratio = 110% and Cr/Ga flux ratio = 2.1%. The substrate temperature was 700 ºC, and final sample thickness was ~ 5800 Å. Both zero-field cooled (ZFC) and field cooled (FC) (H=100 Oe) magnetization curves show sizeable magnetization, and the distinct separation of the curves below 300K clearly indicates ferromagnetism. While the ZFC and FC curves nearly join at 300 K, the Curie temperature T_C >> 300K. For example, a ferromagnetic magnetization curve typically should drop off steeply near T_C, and that point is not yet reached for this sample at 300K. A magnetization vs. applied field loop acquired at 300K for this same sample is shown in Fig. 2(b). Having also a very small coercivity and a small remnant field, this very promising magnetic result suggests that CrGaN as a spintronic material worthy of further investigation, in particular at the nanoscale.

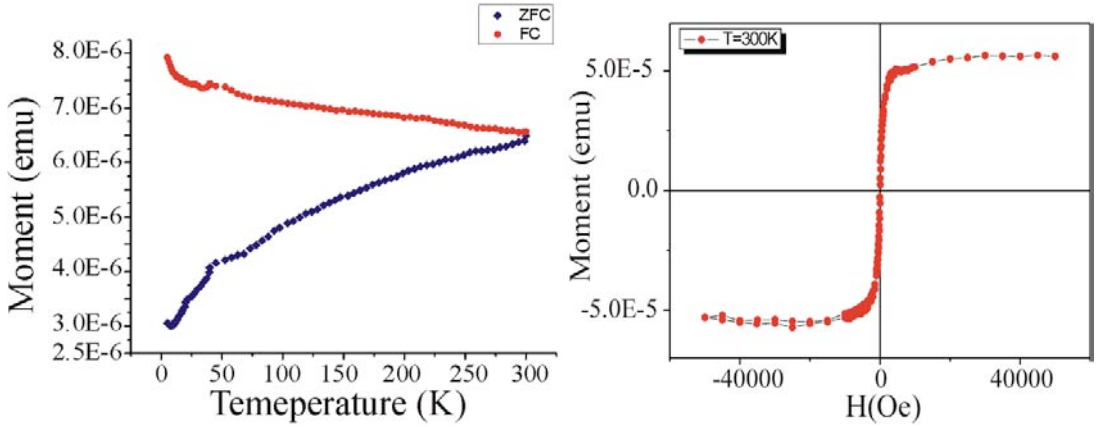

Fig. 2. (a) Zero-field cooled and field-cooled magnetization of CrGaN; (b) Magnetization vs. applied field.

To study the growth of Cr-doped GaN at the atomic scale, thin (5 ML thick) layers of CrGaN were grown on a GaN layer with Cr/Ga flux ratios of 5% and 20%. The surfaces were then examined with UHV STM. As shown in Fig. 3(a), the image of 5% CrGaN shows a surface having the known 3_3 reconstruction, corresponding to cations in every third adatom site. In this image, the brighter features suggest Cr atoms, and the Cr atoms appear to be distributed throughout the surface. Also, it appears that Cr adatoms are located at Ga adatom sites.

The STM image of 20%CrGaN tells a different story. As seen in Fig. 3(b), the STM image shows the well-ordered 3_3 structure separated by linear and dot-like nanostructures. These nanowires and nanodots are attributed to Cr precipitation at the surface. The nanowires clearly form along the 3-fold surface symmetry directions. We expect these spontaneously formed Cr nanostructures to have interesting magnetic properties of their own.

As the next step, we plan to study the magnetic properties of these surfaces and nanostructures using spin-polarized STM (SP-STM), which is highly sensitive to the surface magnetic structure. Also, we plan to apply atom manipulation techniques to create artificial nanostructures on these surfaces.

In parallel with our work on CrGaN, we have been developing our SP-STM capability using a model antiferromagnetic nitride test surface – Mn_3N_2 (010). This surface is ideal as a testing ground for SP-STM due to its row-wise anti-ferromagnetic surface structure. In our earlier paper, we reported the atomic-scale magnetic contrast, along with a method to separate the magnetic and non-magnetic components of the STM image.[8] Along with our experimental SP-STM work, we have collaborations with both U.S. and German theorists using first-principles calculations (i.e. density functional theory) to simulate the SP-STM images. We have recently reported the importance of taking into account the strong directionality of the hybrid *p-d* orbital lobes in the atomic-scale SP-STM images, as opposed to averaging the spin-polarization over the atomic sphere.[9] We have also studied the tip-sample voltage-dependence of the SP-STM images. We found experimentally that a reversal of the magnetic contrast between _ and _ atomic rows occurred at sample voltage of ~ +0.4 V [10] (see also images in our associated *NanoHighlight*). Interestingly, the first-principles SP-STM simulations find that the contrast reversal is only explained by a reversal in the spin-polarization of the STM tip. We have also reported during the last year that the effective spin polarization of the STM tip is affected by just the few atoms or cluster of atoms at the tip apex.[9]

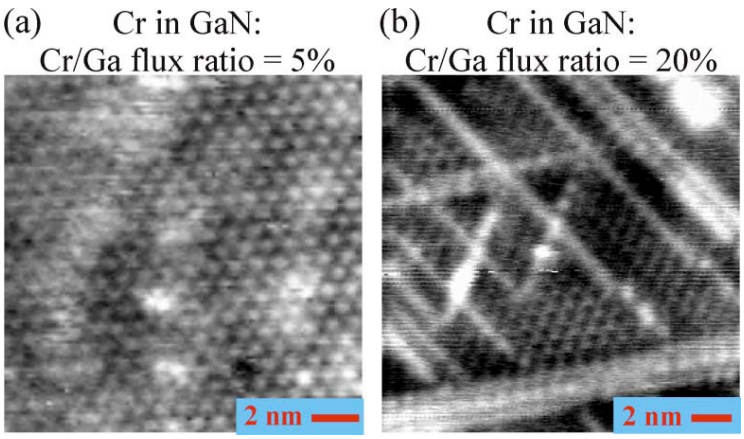

Fig. 3. CrGaN(000-1) Surfaces with different intended concentrations of Cr: (a) 5% intended Cr/Ga; (b) 20% intended Cr/Ga.

References

[1] T. Dietl, H. Ohno, F. Matsukura, J. Cibert, and D. Ferrand, Science 287, 1019 (2000).

[2] M. E. Overberg, C. R. Abernathy, S. J. Pearton, N. A. Theodoropoulou, K. T. McCarthy, and A. F. Hebard, Appl. Phy. Lett. 79, 1312 (2001).

[3] M. B. Haider, C. Constantin, H. Al-Brithen, H. Yang, E. Trifan, D. Ingram, A. R. Smith, C. V. Kelly and Y. Ijiri, Journal of Applied Physics 93(9), 5274 (2003).

[4] Hashimoto et al., Journal of Crystal Growth **251**, 327 (2003).

[5] For further information about this project link to http://nsnm.phy.ohiou.edu or email smitha2@ohio.edu or hla@helios.phy.ohiou.edu or ulloa@helios.phy.ohiou.edu or sandler@helios.phy.ohiou.edu.

[6] H. A. AL-Brithen, M. B. Haider, N. Sandler, A. R. Smith, and P. Ordejon, Scanning Tunneling Microscopy and Surface Simulation of c-GaN(001)-4x1 Ga Tetramer Reconstruction, submitted to *Physical Review Letters*.

[7] C. Constantin, M. B. Haider, D. Ingram, and A. R. Smith, Metal/Semiconductor Phase Transition in Chromium Nitride(001) Grown by rf-Plasma Assisted Molecular Beam Epitaxy, in press in *Applied Physics Letters* (2004).

[8] Haiqiang Yang, Arthur R. Smith, Margarita Prikhodko, and Walter R. L. Lambrecht, Atomic-Scale Spin-Polarized Scanning Tunneling Microscopy Applied To Mn_3N_2 (010), Phys. Rev. Lett. **89**, 226101 (2002).

[9] A.R. Smith, R. Yang, H. Yang, W.R.L. Lambrecht, A. Dick, and J. Neugebauer, Aspects of Spin-Polarized Scanning Tunneling Microscopy at the Atomic Scale: Experiment, Theory, and Simulation, Surf. Sci. **561**, 154 (2004).

[10] Arthur R. Smith, Rong Yang, and Haiqiang Yang, Bias-Voltage Dependence in Atomic-Scale Spin Polarized Scanning Tunneling Microscopy of Mn_3N_2 (010), Mat. Res. Soc. Symp. Proc. **803**, GG1.2 (2004).