

Wednesday Afternoon, November 2, 2005

Magnetic Interfaces and Nanostructures

Room 204 - Session MI+EM-WeA

Magnetic Semiconductors

Moderator: A.T. Hanbicki, Naval Research Laboratory

2:00pm **MI+EM-WeA1 Effects Limiting the Formation of Ferromagnetic III@sub 1-x@Mn@sub x@V Alloys by Epitaxial Growth, J.K. Furdyna, INVITED**

III@sub 1-x@Mn@sub x@V alloys (e.g., Ga@sub 1-x@Mn@sub x@As), comprised of Mn@super +2@ incorporating substitutionally for the group-III element in the III-V lattice are captivating the attention of the scientific community worldwide because of the promise they hold for spin-electronic applications. Incorporation of Mn into the III-V lattice in sufficient concentrations to render the III@sub 1-x@Mn@sub x@V alloy ferromagnetic must be carried out by non-equilibrium low-temperature epitaxy, whereby Mn concentrations x approaching 0.10 can be attained. The ferromagnetism of these alloys occurs because, in addition to providing magnetic moments, the Mn ions also act as acceptors, thus supplying large concentrations of holes that mediate the ferromagnetic interaction between magnetic moments of the Mn ions. A mean field theory projects that the Curie temperature $T@sub C@$ should scale as the product of the Mn concentration x and of the hole density p . Thus, in principle, one should expect above-room-temperature ferromagnetism for large values of the $x \cdot p$ product. Our research on Ga@sub 1-x@Mn@sub x@As and In@sub 1-x@Mn@sub x@Sb has shown, however, that the Fermi energies achievable in these materials cannot exceed a certain maximum $E@sub Fmax@$, corresponding to a maximum hole concentrations $p@sub max@$. This occurs because the relationship between the creation energies for negatively-charged defects (such as the desired substitutional Mn acceptors Mn@sub III@, e.g. Mn@sub Ga@ or Mn@sub In@) and positively-charged defects (such as the unwanted interstitial Mn double donors, Mn@sub I@) is controlled by the Fermi energy. When $E@sub F@$ in the III@sub 1-x@Mn@sub x@V reaches $E@sub Fmax@$ due to the increasing free hole density, further formation of Mn@sub III@ becomes energetically unfavorable, and a high concentration of compensating Mn@sub I@ defects begins to form. The creation of Mn@sub I@ is deleterious to the ferromagnetism for multiple reasons: (1) compensation by the double Mn@sub I@ donors reduces the hole concentration, (2) interstitial Mn is RKKY-inactive (due to negligible p-d exchange), and (3) Mn@sub I@ forms antiferromagnetic pairs with Mn@sub III@, reducing further the density of Mn ions that contribute to the ferromagnetism of the III@sub 1-x@Mn@sub x@V alloy. Thus any increase of the Mn@sub I@ concentration automatically leads to lowering the value of $T@sub C@$. Ion-channeling experiments directly reveal this type of interstitial Mn creation whenever p approaches $p@sub max@$ due to a high Mn concentration. In this talk we concentrate on showing that substitutional vs. interstitial incorporation of Mn in III@sub 1-x@Mn@sub x@V alloys is determined by the Fermi level during the growth process itself, no matter what is the source of holes that establish the value of $E@sub F@$, and independent on the spatial location of the acceptors with respect to the magnetic Mn ions. To demonstrate this, we discuss two types of growth experiments that allowed us to vary the Fermi level independently of the Mn concentration, namely, experiments on Be co-doping of III@sub 1-x@Mn@sub x@V alloys; and on modulation doping of Al@sub 1-y@Ga@sub y@As/Ga@sub 1-x@Mn@sub x@As/Al@sub 1-y@Ga@sub y@As heterostructures by Be. Having established causes for the limit which nature imposes on incorporating substitutional Mn ions at the Group-III sites in III-Mn-V alloys, I will then discuss possible strategies for circumventing this obstacle, with an eye on increasing the Curie temperature of these novel ferromagnetic semiconductors.

2:40pm **MI+EM-WeA3 Structural Properties, Lattice Dynamics, and Optical Properties of GaMnN, W.E. Fenwick, M.H. Kane, M. Strassburg, A. Asghar, S. Gupta, H. Kang, Georgia Institute of Technology; Z. Hu, Georgia State University; S. Graham, Georgia Institute of Technology; U. Perera, N. Dietz, Georgia State University; I.T. Ferguson, Georgia Institute of Technology**

Dilute magnetic semiconductors (DMS) show promise as spintronic materials because of their electrical and magnetic properties. E.g., GaMnN exhibit ferromagnetism (FM) above room temperature (RT). Application of such materials for RT spintronic devices requires an understanding of the origin of this magnetism, which is still under debate in the literature. Knowledge of the structural properties is essential to determine the origin

of the RT FM in GaMnN. Therefore this work provides structural and optical studies of GaMnN to reveal the crystalline quality, lattice dynamics, and some fundamental properties such as the optical constant. Increasing Mn concentration significantly affects long-range lattice ordering. The observation of a local vibrational Raman mode at 669 cm^{-1} combined with the slight excess of metal components in the growth process and the incorporation of Mn acceptor states favors the formation of nitrogen vacancies. Such vacancies form shallow donor complexes and thus contribute to self-compensation, which may hamper carrier mediation. Raman spectroscopy also revealed a disorder-induced mode at 300 cm^{-1} . The intensity of both modes was found to be weaker by more than one order of magnitude compared to GaMnN grown by MBE or prepared by ion-implantation. This is a consequence of the improved MOCVD growth conditions. Crystalline integrity and the absence of major second phase contributions were confirmed by high resolution X-ray diffraction studies. Atomic force microscopy showed that optimized annealing conditions suppressed the formation of Mn-rich precipitates on the surface. Further investigations on the lattice dynamics and the determination of the optical constants were enabled by infrared reflectance spectroscopy. The GaN E1(TO) phonon frequency linearly increases with Mn composition, which is expressed by $(558 + 2.7x) \text{cm}^{-1}$. Meanwhile the peak values of the infrared dielectric functions of the GaMnN decrease with increasing Mn concentration.

3:00pm **MI+EM-WeA4 Epitaxial Growth of Ferromagnetic Mn@sub 3-delta@Ga Thin Films on Wurtzite GaN(0001) by Molecular Beam Epitaxy, E.D. Lu, M.B. Haider, R. Yang, C. Constantin, G. Pokharel, D.C. Ingram, A.R. Smith, Ohio University**

Magnetic metal and/or alloy films on III-V semiconductor substrates have attracted considerable interest due to the potential applications for magnetic/spintronic materials and devices, especially as spin electron injection sources for spin-sensitive heterostructures as well. The binary MnGa alloy is one of several promising metallic ferromagnetic candidates with CuAu-I type ordering. It is a face-centered tetragonal (fct) structure with lattice constants $a = 3.897 \text{ \AA}$, $c = 3.58\text{-}3.65 \text{ \AA}$ dependent on content of at.% Mn between 55-60%. The epitaxial ferromagnetic Mn@sub 3- $\tilde{\gamma}$ @Ga(111) thin films have been grown on wurtzite GaN(0001) substrates with Ga polar surface by molecular beam epitaxy through controlling the substrate temperature and flux ratio of manganese to gallium during the growth. Prior to growing Mn@sub 3- $\tilde{\gamma}$ @Ga thin films, the commercial MOCVD GaN substrates were directly heated for clean up and refreshed by growth of GaN layer by radio plasma MBE. The growth and structure of the Mn@sub 3- $\tilde{\gamma}$ @Ga thin films are monitored in situ by reflection high energy electron diffraction (RHEED). The RHEED pattern was spotty at the initial stage and gradually became streaky, indicating surface roughness at the beginning and finally a smoother surface at the end. Combined RHEED and ex situ XRD results have revealed the primarily structure of the CuAu $\tilde{\gamma}$ I type fct Mn@sub 3- $\tilde{\gamma}$ @Ga thin films grown with (111) plane lying on GaN(0001) plane; due to double lattice constant of the Mn@sub 3- $\tilde{\gamma}$ @Ga(111) plane along [11-2] direction is a good match with that distance along [11-20] direction of GaN(0001) (less than 4% mismatch), the epitaxial relationship of the fct Mn@sub 3- $\tilde{\gamma}$ @Ga is (111)[01-1] MnGa // (0001)[1-100]GaN and (111)[11-2] MnGa // (0001)[11-20]GaN. Rutherford Backscattering Spectroscopy (RBS) has also confirmed composition of the Mn@sub 3- $\tilde{\gamma}$ @Ga thin films with the ratio of Mn to Ga about 1.5 to 1.

3:20pm **MI+EM-WeA5 Nanostructure of Ferromagnetic Mn-implanted Si, C. Awo-Affouda¹, M. Bolduc, K.A. Dunn, M.B. Huang, F. Ramos, G. Agnello, B.L. Thiel, V.P. LaBella, University at Albany-SUNY**

Semiconductor devices which exploit the spin of the electron hold great potential to produce devices with increased functionalities. Making conventional semiconductors ferromagnetic via ion implantation of Mn will aid in fabricating future spintronic devices. We recently demonstrated that ferromagnetism can be achieved via Mn-ion implantation of n-type and p-type Si wafers. A Curie temperature greater than 400K was observed for the p-type samples. The structure of the implanted material was investigated in order to identify the source of the ferromagnetism. SIMS depth profiling of the as-implanted samples showed a typical Gaussian shape profile of Mn atoms in the silicon lattice, which peaks at 250 nm. Post-implant annealing was performed to heal the damage from the implantation process and resulted in a strong redistribution of the Mn atoms. Furthermore diffraction contrast TEM of the annealed samples revealed nanometer size precipitates distributed throughout the implanted region, along with a large band of dislocation and stacking faults. Selected

¹ Falicov Student Award Finalist

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area diffraction patterns gave strong evidence that these phases are $\text{MnSi}_{1.7}$ crystallites. We will discuss the role of these precipitates on the observed ferromagnetism.

3:40pm **MI+EM-WeA6 Ferromagnetic Properties of Mn-implanted Si**, *M. Bolduc, C. Awo-Affouda, M.B. Huang, F. Ramos, V.P. LaBella*, University at Albany - SUNY

Integrating spintronic device concepts with silicon may enable new possibilities for fabrication. In addition, theoretical calculations have predicted ferromagnetic ordering in Mn-doped group-IV semiconductors. This potential has motivated the search for a Si-based ferromagnetic semiconductor. We demonstrate that p-doped and n-doped Si crystals can be made ferromagnetic above room temperature through Mn-ion implantation. 300-keV Mn^{+} ions were implanted at doses of $(1-10) \times 10^{15} \text{ cm}^{-2}$ reaching peak concentrations of (0.1-0.8) at.% as measured through SIMS profiling. Ferromagnetic hysteresis loops were obtained using a SQUID magnetometer at temperature of (10-300) K, yielding a saturation magnetization of 0.1-0.7 emu/g-sample. The Curie temperature is found >400 K with carrier concentration dependence. We will report on the dependence of the magnetic properties on the post-implant annealing temperature and Mn concentration. These results will be discussed in comparison with other ion implanted or MBE grown group-IV ferromagnetic semiconductors.

4:00pm **MI+EM-WeA7 Growth and Magnetic Properties of Doped ZnO Epitaxial Films and Nanocrystal**, *S.A. Chambers*, Pacific Northwest National Laboratory and Univ. of Washington; *A.C. Tuan*, Pacific Northwest National Laboratory; *K.R. Kittilstved, D.R. Gamelin*, University of Washington **INVITED**

Since 2001, researchers around the world have been involved in a vigorous search for new ferromagnetic oxide semiconductors with Curie temperatures above ambient. Such materials are vitally important for the practical realization of spintronics. Two wide bandgap oxide semiconductors have been of particular interest - TiO_2 anatase and ZnO. A number of claims of room temperature ferromagnetism (RTFM) in these host oxides with various dopants have been made. However, some of the results were based on poorly characterized material, often containing magnetic secondary phases, leading to illegitimate claims. Even for well characterized materials which are phase-pure magnetically doped oxides, the mechanism(s) of magnetism remain largely undetermined. We have used oxygen plasma assisted metal organic chemical vapor deposition along with direct wet chemical synthesis and spin coating to prepare $\text{Co}_{1-x}\text{Zn}_x\text{O}$ and $\text{Mn}_{1-x}\text{Zn}_x\text{O}$ epitaxial and nanoparticle films. Co(II) and Mn(II) substitute for Zn(II) in the wurtzite lattice in materials synthesized by both methods. Room temperature ferromagnetism in epitaxial Co:ZnO films can be reversibly activated by diffusing in Zn, which occupies interstitial sites and makes the material n-type. O-capped Co:ZnO nanoparticles, which are paramagnetic as grown, become ferromagnetic upon being spin coated in air at elevated temperature. Likewise, spin-coated N-capped Mn:ZnO nanoparticle films also exhibit room temperature ferromagnetism. However, the inverse systems, N-capped Co:ZnO and O-capped Mn:ZnO , are entirely paramagnetic when spin coated into films in the same way. Unfortunately, the nanoparticle films are not sufficiently conductive to perform magneto-transport measurements. Instead, we have carried out a detailed analysis of optical absorption, photovoltage, and magnetic circular dichroism spectra. This analysis reveals that the resonances $\text{Co(I)} \leftrightarrow \text{Co(II)} + e^-$ and $\text{Mn(III)} \leftrightarrow \text{Mn(II)} + h^+$ are energetically favorable, consistent with strong hybridization of Co (Mn) with the conduction (valence) band of ZnO. In contrast, the resonances $\text{Mn(I)} \leftrightarrow \text{Mn(II)} + e^-$ and $\text{Co(III)} \leftrightarrow \text{Co(II)} + h^+$ are not energetically favorable. These results indicate that Co(II) -derived states will strongly interact only with the conduction band, whereas Mn(II) -derived states interact strongly only with the valence band. These spectral results are consistent with the observed ferromagnetism in Co:ZnO (Mn:ZnO) being mediated by electrons (holes).

This work was performed in part in the Environmental Molecular Sciences Laboratory, a national scientific user facility sponsored by the Department of Energy's Office of Biological and Environmental Research and located at Pacific Northwest National Laboratory. This work was supported by the US Department of Energy, Office of Science, Office of Basic Energy Sciences, Division of Materials Science and Engineering Physics. Work at UW was funded by the NSF (DMR-0239325 and ECS-0224138) and the Research Corporation.

4:40pm **MI+EM-WeA9 Effects of Differing Mn@Ga/Mn@I@ on the Anomalous Hall Effect in (Ga,Mn)As**, *Y.S. Kim, H.K. Choi, Z.G. Khim*, Seoul National University, Korea; *S.H. Chun*, Sejong University, Korea; *Y.D. Park*, Seoul National University, Korea

We report on the effect of differing ratios of substitutional and interstitial Manganese ($\text{Mn}_{\text{Ga}}/\text{Mn}_{\text{I}}$) on the Anomalous Hall Effect (AHE) in low temperature molecular beam epitaxy prepared (Ga,Mn)As diluted magnetic semiconductors. As-grown (Ga,Mn)As epilayers with Mn content from 2.4% ~ 6.1% exhibit ferromagnetic ordering below temperatures ranging from 60 to 110 K. Relatively differing $\text{Mn}_{\text{Ga}}/\text{Mn}_{\text{I}}$ ratios were achieved by careful annealing at moderate temperatures as evident in the differing resistivities (as low as 2.5 m $\Omega\text{-cm}$) / Hall carrier concentrations (as high as $\sim 8 \times 10^{20} \text{ cm}^{-3}$) as well as T_{C} (as high as 150 K) with optimal annealing temperature found to be 250°C. AC-magnetotransport measurements conducted from 5 K to 300 K and with applied magnetic fields ranging to ± 7 Tesla indicate similar field dependence of resistivity, AHE, and metal-insulator-like transition near T_{C} as reported by others. T_{C} 's as found from Arrott plots from the resulting AHE measurements agree well with direct SQUID magnetometry measurements. Log-log plots of $\rho_{\text{xy}}/d\rho_{\text{xy}}/dH$, and $\rho_{\text{xy}}/M_{\text{S}}$ vs. ρ_{xx} , cumulated from AHE measurements at temperatures below T_{C} of differing as-grown Mn content and annealing conditions, indicate skew scattering to be the dominant mechanisms for AHE in (Ga,Mn)As regardless of as-grown Mn content or $\text{Mn}_{\text{Ga}}/\text{Mn}_{\text{I}}$ ratios and possibly regardless of presence of non-magnetic or magnetic Mn-rich nano-clusters. We will also discuss the results in light of theory of AHE in clean (Ga,Mn)As and experimental reports of AHE in DMS systems with ferromagnetic nano-clusters.

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