Electronic Materials and Processing
Room 2003 - Session EM+MI-WeA

Magnetic Semiconductors
Moderator: S.A. Chambers, Pacific Northwest National Laboratory

2:00pm **EM+MI-WeA1 Spectroscopy and Magnetism of Oxide Diluted Magnetic Semiconductors*, D.R. Gamelin,*** *K.R. Kittilstved, W.K. Liu,* University of Washington **INVITED**

Diluted magnetic semiconductor (DMS) nanostructures are pivotal architectural elements in many proposed spintronics devices. DMSs of ZnO are being intensely investigated for their potential use in spintronics technologies, but such applications have been hindered by their inconsistent magnetic properties. This talk will present our group's recent advances in the use of targeted chemical perturbations to manipulate high-Tc spin ordering in ZnO DMSs predictably and controllably.@footnote 1,2,3,4@ Apart from the technological advantages of reliable, controllable, and even switchable high-Tc ferromagnetic semiconductors, these experiments are motivated by the new fundamental insights they provide into the microscopic mechanisms behind magnetic ordering in this class of materials. The results from concomitant optical, magneto-optical, structural, X-ray absorption, and magnetic data collected on the same materials will be used to evaluate dopant electronic structure contributions to magnetic ordering, and the findings will be discussed in the context of current theoretical models of high-Tc ferromagnetism in oxide DMSs. @FootnoteText@ @footnote 1@Kittilstved, K. R.; Liu, W. K.; Gamelin, D. R. "Electronic Structure Origins of Polarity Dependent High-Tc Ferromagnetism in Oxide Diluted Magnetic Semiconductors." Nature Materials, 2006, 5, 291-297.@footnote 2@Kittilstved, K. R.; Norberg, N. S.; Gamelin, D. R. "Chemical Manipulation of 300K Ferromagnetism in ZnO Diluted Magnetic Semiconductors." Phys. Rev. Lett., 2005, 149049.@footnote 3@Kittilstved, K. R.; Gamelin, D. R. "Activation of High-Tc Ferromagnetism in Mn2+:ZnO using Amines." J. Am. Chem. Soc., 2005, 127, 5292-5293.@footnote 4@Schwartz, D. A.; Gamelin, D. R. "Reversible 300K Ferromagnetic Ordering in a Diluted Magnetic Semiconductor." Advanced Materials, 2004, 16, 2115-2119.

2:40pm **EM+MI-WeA3 Deposition of Doped ZnO by Pulsed Laser Deposition Utilizing Novel Ablation Targets*, T.C. Kaspar,*** *T. Droubay, S.M. Heald, V. Shutthanandan, P. Nachimuthu, C.M. Wang, S.A. Chambers,* Pacific Northwest National Laboratory*; K.R. Kittilstved, C.A. Johnson, K.M. Whitaker, D.R. Gamelin,* University of Washington

Zinc oxide (ZnO) is a promising material for optical, electro-optical, magneto-optical, and spintronic applications. The desired properties of ZnO are obtained by doping the material, in which case distribution and substitution of the dopants is of prime importance. Doped ZnO nanoparticles can achieve the necessary dopant dispersion and speciation to create high-quality material exhibiting, for example, the room temperature ferromagnetism necessary for spintronic applications. However, these nanoparticles may not be as useful in practical devices as high-quality epitaxial films. To this end, we have explored the deposition of epitaxial Co- and Mn-doped ZnO thin films by pulsed laser deposition (PLD), utilizing well-characterized doped ZnO nanoparticles as the basis for the PLD ablation target material. Ablating a target which already contains the dopant in the desired substitutional environment in ZnO should allow full substitution of the dopant into the epitaxial film and reduce or eliminate the formation of secondary phases. Initial results on Co-doped ZnO films deposited on Al@sub 2@O@sub 3@(0001) from a Co:ZnO nanoparticle target indicate that smooth, epitaxial ZnO films can be obtained. Co K-edge x-ray absorption near edge spectroscopy and extended x-ray absorption fine structure show full oxidation of Co to Co(II) for all oxygen pressures explored (5x10@super -5@ - 5x10@super -2@ Torr), with substitution of Co for Zn in the ZnO lattice. Issues associated with the nanoparticle ablation targets, such as target densification using low-temperature processing, particle and droplet ejection during ablation, and localized dopant diffusion in the ablated region, will be discussed.

3:00pm **EM+MI-WeA4 Room Temperature Ferromagneticism in Fe Implanted ZnO Nanotips*, R.A. Bartynski,*** *D. Hill,* Rutgers University*; D.A. Arena,* National Synchrotron Light Source*; P. Wu, Y. Lu, J.F. Al-Sharab, F. Cosandey,* Rutgers University

Transition metal- (TM-) doped ZnO is a promising candidate dilute magnetic semiconductor for room-temperature spintronics applications. Controlled synthesis of nanoscale structures of these materials offers the possibility to develop low-dimensional spin-dependent electronic devices. We have grown well-aligned ZnO nanotips on SiO@sub 2@/quartz substrates using MOCVD. The tips were subsequently Fe-doped to a dose 5 x 10@super 16@ cm@super -2@ using ion implantation at 200 keV. The magnetic, structural, and chemical properties of both as-implanted and post-implantation annealed nanotips were studied using SQUID magnetometry, electron energy loss (EELS) and X-ray Energy Dispersive (EDS) spectroscopy in high resolution transmission electron microscopy (HRTEM), X-ray diffraction (XRD) and soft X-ray absorption spectroscopy (SXAS). The as-implanted tips were ferro-magnetic at room temperature with a saturation moment of ~ 0.2 μ@sub B@/Fe-ion, a remnant magnetization of ~0.03 μ@sub B@/ion, and a coercive field of ~150 Oe. The tips exhibit a core-shell structure with a high concentration (~ 8%) of Fe in the first ~ 10 nm, and about 3% in the interior, with roughly equal concentrations of Fe@super +2@ and Fe@super +3@ oxidation states. Post implantation annealing to 700C for 10 minutes improves crystallinity, produces a more uniform ~ 5% concentration of Fe, and increases the Fe@super +3@:Fe@super +2@ ratio, but significantly reduces the saturation magnetization. However, the tips remain ferromagnetic up to at least room temperature. The reduction in magnetic response, despite the increased magnetic moment/ion expected from the increased Fe@super +3@ concentration, suggests that the redistribution of Fe ions dominates the ferromagnetic coupling in the system. @FootnoteText@ * Supported by NSF Grant ECS-0224166.

3:20pm **EM+MI-WeA5 X-ray Characterization of Oxide-based Magnetic Semiconductors*, Y.U. Idzerda,*** *A. Lussier, J. Dvorak, A. McClure, M. Liberati, J. Holroyd,* Montana State University*; E. Arenholz,* ALS/LBNL*; S.R. Shinde, S.B. Ogale, T. Venkatesan,* University of Maryland, College Park **INVITED**

Although the evidence for magnetic semiconductors (not simply semiconductors which are ferromagnetic) is compelling, there is much uncertainty in the mechanism for the polarization of the carriers, suggesting that it must be quite novel. Recent experimental evidence suggests that this mechanism is similar to the polaron percolation theory proposed by Kaminski and Das Sarma,@footnote 1@ which was recently applied specifically to doped oxides by Coey et al.@footnote 2@ where the ferromagnetism is driven by the percolation of polarons generated by defects or dopants. We have used X-ray absorption spectroscopy at the L-edges and K-edges for low concentrations transition metal (TM) doped magnetic oxides (including TiO@sub 2@, La@sub 1-x@Sr@sub x@O@sub 3@, HfO@sub 2@, and In@sub 2@O@sub 3@). We have found that in most cases, the transition metal assumes a valence consistent with being at a substitutional, and not interstitial site. We have also measured the X-ray Magnetic Circular Dichroism spectra (including TM doped GaN and GaAs systems). Although these materials show strong bulk magnetization, we are unable to detect a robust dichroism feature associated with magnetic elements in the host semiconductor. In the cases where a dichroism signal was observed, it was very weak and could be ascribed to a distinct ferromagnetic phase (TM metal cluster, TM oxide particulate, etc.) separate from the host material. This fascinating absence of a dichroic signal and its significant substantiation of important features of the polaron percolation model may help to finally resolve the issue of ferromagnetism in magnetically doped oxides. @FootnoteText@ @footnote 1@ Kaminski and S. Das Sarma, Physical Review Letters 88, 247202 (2002).@footnote 2@ J. M. D. Coey, M. Venkatesan, and C. B. Fitzgerald, Nature Materials 4, 173 (2005).

4:00pm **EM+MI-WeA7 Characteristics of Ti@sub 1-x@Co@sub x@O@sub 2@ Thin Films Deposited by MOCVD*, A. McClure,*** *A. Kayani, M. Liberati, J. Dvorak, R.J. Smith, Y.U. Idzerda,* Montana State University*; E. Arenholz,* Lawrence Berkeley National Laboratory

Polycrystalline anatase thin films of Ti@sub 1-x@Co@sub x@O@sub 2@ were prepared on TiO@sub 2@ (50nm)/Si (111) substrates using liquid delivery metal organic chemical vapor deposition (MOCVD). This growth technique allows for the arbitrary variation of the Co concentration. The precursors for these growths were titanium isopropoxide and Co(TMHD)@sub 3@ dissolved in tetrahydrofuran. These films were characterized by X-ray absorption spectroscopy (XAS) and X-ray magnetic circular dichroism (XMCD) to determine the phase of the TiO@sub 2@ host, the Co valency, and the presence of Co clusters. Film thicknesses and Co dopant concentrations were determined from Rutherford backscattering (RBS). A vibrating sample magnetometer (VSM) revealed room temperature ferromagnetism, allowing for a determination of the moment per Co atom vs. Co concentration. I will discuss these results in the context of the bound magnetic polaron model. Recent work@footnote 1@ suggests that this model may only need oxygen vacancies and

ferromagnetism may occur with nonmagnetic doping. Our experimental results on Pt:TiO@sub 2@ and its lack of ferromagnetism will also be presented. @FootnoteText@ @footnote 1@Osorio-Guillén, J., Lany, S., Barabash, S. V. and Zunger, A., Phys. Rev. Lett., 96, 107203 (2006).

4:20pm **EM+MI-WeA8 Detection of Nanometer-Sized Inclusions in Annealed Ga@sub 1-x@Mn@sub x@As from Atypical Scaling Behavior of the Anomalous Hall Coefficient**, ***H.K. Choi***, *S.S.A. Seo, W.O. Lee, Y.S. Oh, K.H. Kim, T.W. Noh, Y.D. Park,* Seoul National University, Korea

We report on the anomalous Hall coefficient (R@sub s@) and longitudinal resistivity (@rho@@sub xx@) scaling relationship (R@sub s@=c@rho@@sub xx@@super n@) on three series of annealed LT-MBE Ga@sub 1-x@Mn@sub x@As epilayers (x ~ 0.55). After growth, Mn@sub Ga@ were varied by low temperature annealing from 200°-350°C. Our report of the scaling relationship can provide new methods to detect metallic secondary phases could not be observed in HRXRD and SQUID magnetometer. As-grown samples exhibit scaling parameter n of 1, which can be attributed to extrinsic skew scattering origins of the anomalous Hall Effect@footnote 1@ or to AHE attributed to phonon-assisted hopping between localized states in the impurity band.@footnote 2@ For annealing temperatures near the optimal (~ 250°C), we find n ~ 2 to be consistent with recent theories on the intrinsic origins of AHE in Ga@sub 1-x@Mn@sub x@As.@footnote 3@ For annealing temperatures above the optimum, we note n > 3, which atypical behavior cannot be explained in terms of AHE from a DMS system. This atypical behavior is similar to nanometer-sized super-paramagnetic particles in a paramagnetic matrix such as CoAg granular systems.@footnote 4@ This observation from AHE measurements agree well with optical spectroscopy measurements with observed characteristic features attributable to spherical resonance from metallic inclusions.@footnote 5@ @FootnoteText@ @footnote 1@ J. Smit, Physica (Ultrecht) 21, 877 (1955).@footnote 2@ A. A. Burkov and L. Balents, PRL 91, 057202 (2003).@footnote 3@ T. Jungwirth, Q. Niu, and A. H. MacDonald, PRL 88, 207208 (2002).@footnote 4@ Peng Xiong et al., PRL 69 (22), 3220 (1992).@footnote 5@ S.S.A. Seo et al., APL 82, 4749 (2003); S.S.A. Seo et al., JAP 95, 8172 (2004).

4:40pm **EM+MI-WeA9 Effect of Growth Conditions on the Magnetic Properties of GaGdN**, ***J.K. Hite***, *R.M. Frazier, R.P. Davies, G.T. Thaler, C.R. Abernathy, S.J. Pearton,* University of Florida*; J.M. Zavada,* Army Research Office

Due to the increasing interest in spintronics, many attempts have been made at incorporating spin-based technology into the existing semiconductor technology, with a recent focus on rare earth doped GaN. GaGdN layers were grown by gas source MBE under a broad range of thicknesses and Gd cell temperatures. Magnetic measurements obtained using a SQUID magnetometer showed ferromagnetic behavior at room temperature. Magnetization of the material was dependent both on dopant cell temperature and crystalline quality. The Gd concentration was under the detection limit of secondary ion mass spectroscopy, and from the highly insulating nature of the films is estimated to be on the order of 10@super 16@ atom/cm@super 3@. In addition, the GaGdN films were also co-doped with Si at varying Si cell temperatures. In contrast to GaGdN, the co-doped material was conductive, with resistivities reaching 0.04 @ohm@-cm. Room temperature ferromagnetism was also retained, some of which exceeded that of the singly doped films. No evidence of second phases was seen in x-ray diffraction. These materials may be useful in the development of devices such as magnetic tunnel junctions and spin valves. This work is supported by the Army Research Office under W911-NF-04-10296.

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