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PUBLICATION
B.J. Kirby, J.A. Borchers, J.J. Rhyne, K.V. O'Donovan, S.G.E. te Velthuis, S. Roy, C. Sanchez-Hanke, T. Wojtowicz, X. Liu, W.L. Lim, M. Dobrowolska, and J.K. Furdyna, "Magnetic and Chemical Nonuniformity in $Ga_{1-x}Mn_xAs$ Films as Probed by Polarized Neutron and X-Ray Reflectometry," *Phys. Rev. B,* **74**, 245304 (2006).

FUNDING
National Science Foundation; Missouri University Research Reactor Graduate Fellowship; U.S. Department of Energy

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Magnetic and Chemical Non-Uniformity in $Ga_{1-x}Mn_xAs$ as Probed with Neutron and X-Ray Reflectivity

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Artificial magnetic semiconductor materials could play a key role in future spin-electronics, or "spintronic" devices. We have used x-ray and polarized neutron reflectometry to study the mechanisms through which post-growth annealing increases the ferromagnetic transition temperature (T_C) of manganese-doped gallium arsenide. Our combined studies suggest that annealing liberates Mn from interstitial sites throughout the $Ga_{1-x}Mn_xAs$ film, and allows them to migrate to the film surface and oxidize – a process that drastically increases T_C, and alters the distribution of the magnetic moment.

Magnetic semiconductors are of great importance to the development of spin-electronics (spintronics) technology, as they can be used to exert magnetic control of electrical current in devices. While suitable ferromagnetic semiconductors cannot be found in nature, non-magnetic semiconductor materials can be made ferromagnetic by replacing a fraction of the atoms in the crystal lattice with magnetic atoms. For example, low-temperature molecular beam epitaxy is used to produce manganese-doped gallium arsenide ($Ga_{1-x}Mn_xAs$, x>10%), in which long-range order among Mn at Ga sites give rise to ferromagnetism. This material is attractive because GaAs is commonly found in modern electronics. However, room temperature ferromagnetism is necessary for practical device applications, but the ferro transition temperature (T_C) of $Ga_{1-x}Mn_xAs$ is only around 70 K. The unstable growth conditions that allow for Mn to enter Ga sites also lead to small concentrations of Mn at interstitial sites in the lattice. These unwanted interstitial Mn impurities are known to oppose ferromagnetic ordering, and are partially responsible for the low T_C of as-grown $Ga_{1-x}Mn_xAs$. However, T_C can be more than doubled by a careful post-growth annealing.

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Fully understanding how this annealing process works could lead to room temperature ferromagnetism in $Ga_{1-x}Mn_xAs$. To that aim, we used x-ray and polarized neutron reflectometry (PNR) to study how annealing altered a series of $Ga_{1-x}Mn_xAs$ thin films. Following growth, pieces were cleaved off and annealed – resulting in sets of as-grown/annealed pairs. **Figure 1** shows magnetic and chemical depth profiles for one such set, deduced from PNR measurements taken on the NG-1 Polarized Beam Reflectometer at the NIST Center for Neutron Research. For this set, annealing was found to increase T_C from 60-120 K. The as-grown models are in blue, the annealed are in red. The chemical profiles are shown above the break in the vertical axis, and the magnetic profiles are shown below. Annealing is observed to increase the net magnetization, change the depth distribution of magnetic moment, and add a layer of foreign material to the film surface. Since the chemical profile is flat, the as-grown magnetization gradient cannot be explained by changes in the concentration of Mn at Ga sites, but can be explained by small changes in the depth-dependent concentration of interstitial Mn impurities.

Energy-dependent x-ray reflectometry measurements of these samples were taken on beamline X13A of the National Synchrotron Light Source at Brookhaven National Laboratory, and are shown in **Figure 2**. For the annealed piece, there are pronounced peaks at the oxygen and manganese edges that are absent for the as-grown piece. Since this technique is most sensitive to material near the film surface, these results indicate that the layer of foreign material added by annealing is rich in Mn and O. Non-scattering work by other researchers has shown evidence that the positive effects of annealing are due to removal of interstitial Mn. Combined, our neutron and x-ray results corroborate that idea, suggesting that annealing liberates Mn from interstitial sites throughout the $Ga_{1-x}Mn_xAs$ film, and allows them to migrate to the film surface and oxidize – a process that drastically increases T_C, and alters the distribution of magnetic moment.

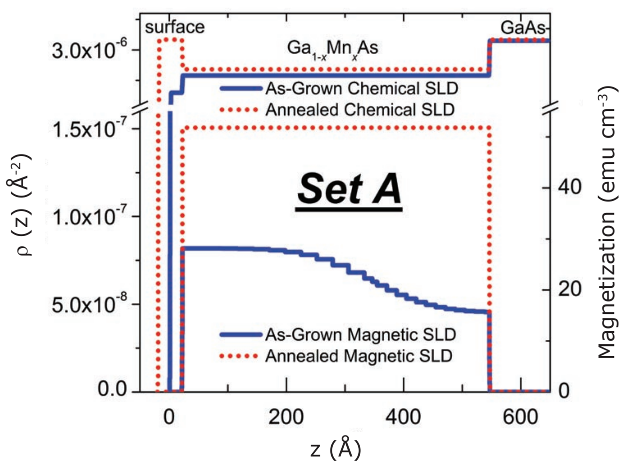

__Figure 1__. Scattering length density models of the as-grown (blue) and annealed (red) films, as deduced from polarized neutron reflectometry. The chemical depth profiles are shown above the break in the vertical axis, the magnetic depth profiles are shown below.

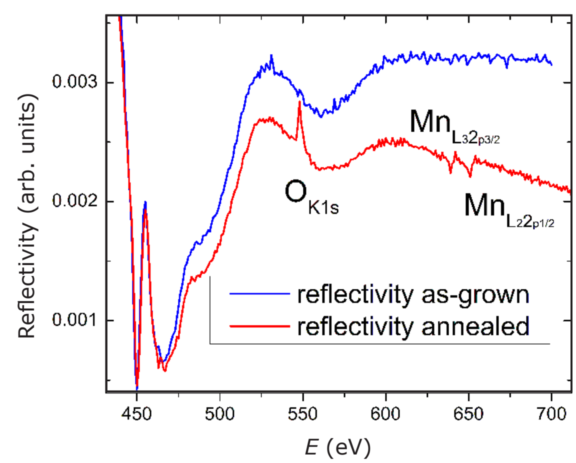

__Figure 2__. X-ray reflectivity for the as-grown (blue) and annealed (red) films taken at NSLS beamline X13A. The annealed film features pronounced oxygen and manganese peaks, while the as-grown film does not.