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Anomalous Magnetic Ground State in NiMn$_2$O$_4$ Thin Films

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Abstract Body: Bulk NiMn$_2$O$_4$ (NMO) is an intriguing magnetic material which exhibits two different ferrimagnetic phases as a function of temperature (a collinear moment configuration below 110K and a canted moment configuration below 70K). Surprisingly, as-grown epitaxial NMO films exhibit only one magnetic transition at 60K, while annealed films recover the two magnetic transitions seen in bulk. To study the magnetic ground state of the as-grown systems, NMO films between 10-850 nm were grown by pulsed laser deposition on (110)-oriented MgAl$_2$O$_4$ and SrTiO$_3$ substrates, and twin samples were annealed in air. X-ray diffraction and Rutherford backscattering confirm that all films grow epitaxially with the spinel crystal structure and 1:2 Ni to Mn stoichiometry. Since the magnetic properties of spinels are determined by the interaction between the transition metal cations, we investigate the cause for the differences in magnetic transition temperatures by studying the properties of the Ni and Mn cations. Cation site occupancies, valences, and magnetic properties were studied by resonant X-ray scattering (RXS), X-ray absorption spectroscopy (XAS) and X-ray magnetic circular dichroism (XMCD) techniques. RXS indicates that the cation site occupancies of the as-grown films do not differ from bulk. However, RXS and XAS show that a large increase in the octahedral Mn$^{4+}$ concentration coincides with the change from one to two magnetic transitions. Additionally, while the Ni XMCD lineshapes are similar between the as-grown and annealed films, the Mn XMCD lineshapes show distinct differences. Thus the decrease in the Mn$^{4+}$ cations disrupts the double exchange that dictates the high temperature collinear moment phase. Consequently, the as-grown NMO films exhibit a new ground state where the high temperature magnetic phase is quenched and the low temperature phase is preserved.


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