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Room Temperature Photo-induced Magnetization of Spinel $(Mn,Zn,Fe)_3O_4$ Thin Films.

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A photomagnetic material changes its magnetization upon illumination with light. Spinel ferrites with multiple Fe valences have been identified as a promising class of photomagnetic materials, though the photo-induced magnetization is mostly observed at low temperatures making it less attractive for applications. We report on the observation of photo-induced magnetism (PIM) in textured thin films of the spinel $Mn_xZn_yFe_{3-x-y}O_4$ ($0.225 \leq x \leq 0.525$, $0.5 \leq y \leq 0.6$) at and above room temperature. Samples with grain sizes of approximately 15-50 nm were grown by pulsed laser deposition at room temperature on Si substrates. The samples show a room temperature photomagnetic effect that appears to be strongly correlated to the zero crossing of the magnetocrystalline anisotropy (MCA) at approximately room temperature.[1] An inverse correlation exists between crystallite size and the extent of the photo-induced magnetization. At a surface or interface, the symmetry is reduced which increases the effect of the MCA on the system. Smaller nanocrystallites, which have larger surface to volume ratio, thus have an increased dependence on MCA and exhibit a larger photomagnetic effect.

Epitaxial films of the same stoichiometry deposited on $MgAl_2O_4$ substrates do not exhibit this photomagnetic effect since their magnetic anisotropy is dominated by magnetostrictive effects.

We have examined the PIM at a variety of wavelengths in the near UV and visible spectrum. We observe a constant PIM between approximately 2-2.5 eV, and an increased PIM at approximately 2.75-3.5 eV. We attribute the photomagnetic effect near 2 eV to an optically induced inter-valent charge transfer (IVCT) between a magnetically hard Fe^{2+} and a magnetically easy Fe^{3+} on octahedral sites, which serves to increase the magnetic moment at low fields. This effect is enhanced when the magnetic anisotropy is easily perturbed, such as a zero crossing of the magnetic anisotropy. The increased PIM

above 2.75 eV is most likely due to additional IVCT mechanisms between the Mn cations or the Fe and Mn cations. We have also measured soft X-ray magnetic circular dichroism (XMCD) spectra with and without incident HeNe laser light (1.96 eV) to determine the relative distribution of Fe^{2+} and Fe^{3+} on the octahedral and tetrahedral sites. We observe an enhanced dichroism at the Fe^{2+} and Fe^{3+} in octahedral sites indicating the importance of the IVCT mechanism to the photomagnetism of these samples. Comparison of our XMCD data to multiplet simulations indicates that our samples are first known instance of Mn^{2+}_{Oh} cations in a ferrimagnetic material.

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[1] K. Ohta, *J. Phys. Soc. Japn.* **18** 685 (1963)