

Evidence of magnetic structure contribution to electron backscatter diffraction

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Diffraction experiments on crystalline solids provide information about the arrangement of regularly spaced planes within a material, thereby facilitating study of defects within those planes. The most commonly studied planes are those associated with the arrangement of atoms, but particles with non-zero magnetic moments (such as neutrons and electrons) can also diffract off of additional planes corresponding with a material's magnetic structure.

Shull and colleagues in the 1950s established neutron diffraction as the premier method for simultaneously studying the atomic and magnetic structures of materials, famously using this technique to observe MnO superlattice reflections in the antiferromagnetic (AFM) state that disappear above the ordering temperature [1].

Electron diffraction (ED) is a well-established technique in both scanning (SEM) and transmission (TEM) microscopy used for studying atomic structure, and there exist a variety of methods for imaging magnetic domains and textures with SEMs and TEMs [2], but to date little attention has been paid to the possibility of using ED to also probe magnetic structure. TEM experiments have demonstrated that ED can detect AFM superlattice peaks in NiO [3], though Coulomb interactions mean the observed ratio of atomic to magnetic scattering intensities can be as high as ~10,000-to-1, whereas ~1-to-1 is typical for neutrons. This ratio will vary from material to material depending on atomic and magnetic structure factors, so whether NiO is the exception or the rule remains to be seen. Regardless, given the relative accessibility of electron and neutron beams, there is significant value in exploring the general viability of ED to probe magnetic structures and their defects.

The standard modality for ED experiments in an SEM is electron backscatter diffraction (EBSD), which has rarely ever been used to examine magnetic materials near their ordering temperatures [4]. Here we present results of temperature dependent EBSD experiments on two isostructural single crystal samples: ferrimagnetic Y₃Fe₅O₁₂ (YIG) and paramagnetic Gd₃Ga₅O₁₂ (GGG).

Data were collected with an EDAX Velocity Super EBSD camera on a Thermo Fisher Quattro Field Emission SEM with the high vacuum heating stage and EBSD heat shield. Both samples were coated with a 4nm layer of carbon in a Leica ACE 600 coater to reduce charging. The microscope was run at 30kV with a current of 16nA, a stage tilt of 70 degrees, and a constant working distance of 16mm. The patterns were collected at a resolution of 640x480 pixels with a camera exposure time of 7 ms from an area of 78 microns by 68 microns with a step size of 0.7 microns in a square grid pattern. The 10,416 individual patterns collected at each temperature were averaged to produce a single representative pattern. The temperature was varied non-monotonically in order to mitigate any systematic errors arising from carbon coating degradation and/or general temperature-dependent impacts on Kikuchi pattern intensity. Data were collected at 297K, 800K, 750K, 400K, 350K, 700K, 675K, 375K, and 550K, in that order, with a minimum wait time of 30 minutes to allow the sample temperature to stabilize. These values were selected to provide several well-separated temperatures above and below 560K, which is the magnetic ordering temperature of YIG.

In the null case where magnetic structure has zero observable effect on Kikuchi pattern intensity, any intensity variations due to temperature should be similar for magnetic and non-magnetic materials alike. Under these conditions, subtracting averaged patterns for any two temperatures should result in similar differences in intensity with no consequence for being above or below the magnetic ordering temperature.

To test this hypothesis, we used ImageJ to compare every possible combination of Kikuchi patterns for each material by subtracting higher temperature patterns from lower temperature ones, then examined the results. A representative example is provided in Figure 1, which shows the averaged patterns for YIG at 800K (Fig 1a) and 297K (Fig 1b), along with the resulting difference in raw intensity (Fig 1c) and then with enhanced contrast/brightness (Fig 1d).

After assessing the subtraction results for all possible temperature points, a clear trend emerged: For GGG, very little difference in Kikuchi band intensity was observed when comparing any two temperatures, no matter how far apart. For YIG, little to no intensity variation was observed when comparing temperature points that were either both above or both below the ordering temperature (excluding 550K). These two groupings each comprise six possible combinations of "low-minus-low" or "high-minus-high" comparisons (e.g, 400K and 297K, or 800K and 700K). In contrast to this result, a very clear difference in

intensity was seen when comparing the 16 possible combinations of temperature points across the ordering temperature (“low-minus-high”, e.g., 800K and 297K). The averaged results of these comparison groupings are presented in Figure 2a-f.

In conclusion, we observe differences in temperature dependent Kikuchi pattern intensity between magnetic YIG and non-magnetic GGG. Specifically, the YIG Kikuchi patterns in the magnetically ordered state contain additional intensity not present in the disordered state, while no such difference is seen in GGG. These results support rejecting the null hypothesis in favor of the conclusion EBSD is sensitive to the magnetic structure of materials.

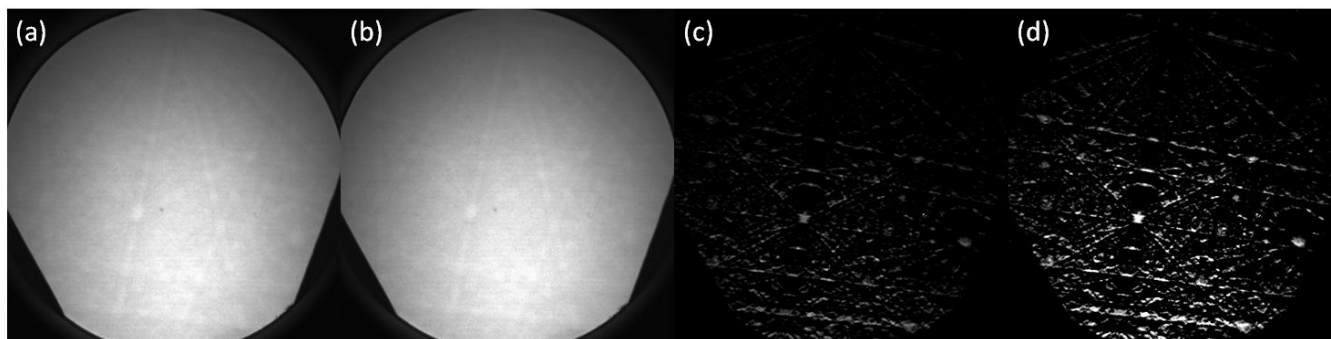


Figure 1. Figure 1: Averaged image of 10,416 raw YIG Kikuchi patterns collected at (a) 800K and (b) 297K. Subtraction of (a) from (b) is presented in (c), with enhanced contrast and brightness in (d) for clarity.

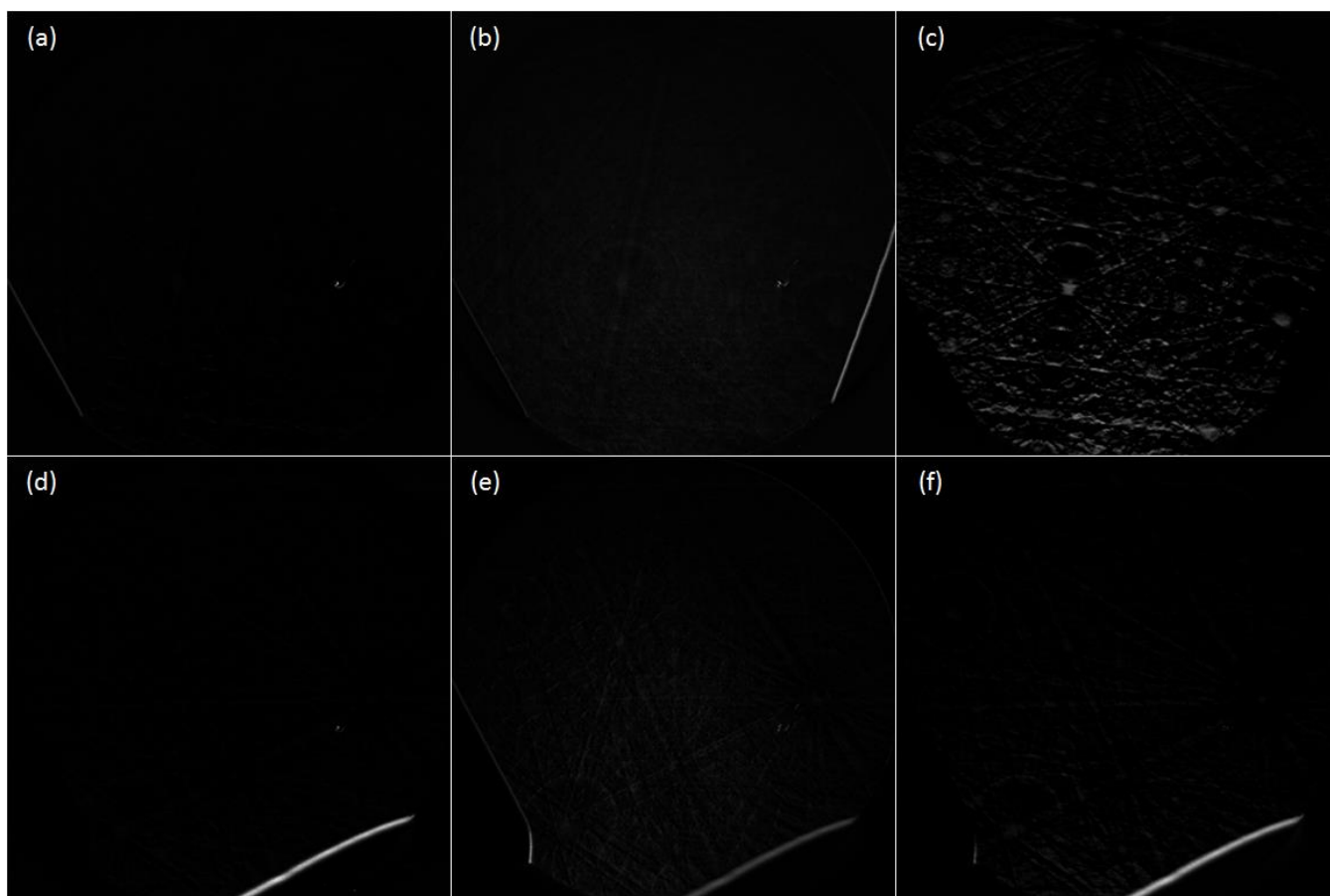


Figure 2. Figure 2. Averaged differences between Kikuchi patterns at different temperatures for YIG (a-c) and GGG (d-f), with six each “low minus low” comparisons in (a) and (d), six each “high minus high” in (b) and (e), and 16 each “low minus high” in (c) and (f).

References

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