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X-ray resonant magnetic scattering study of spontaneous ferrimagnetism

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Abstract
The authors report on an x-ray resonant magnetic scattering measurement of ferromagnetic order, in the absence of an external magnetic field, using resonant dipole scattering at the Gd L2 edge of GdMn2Ge2. Measurements of the ferromagnetic order of the Gd sublattice were accomplished using undulator radiation at the Advanced Photon Source and polarization analysis of the diffracted beam to significantly reduce the charge scattering background. The magnetic origin of the resonant intensity was confirmed by its temperature dependence and the angular dependence of the scattering cross section.

Keywords
Physics and Astronomy, X-ray scattering, Polarization scattering, Polarization, Magnetic resonance, Polarizability

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Comments
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X-ray resonant magnetic scattering study of spontaneous ferrimagnetism

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The authors report on an x-ray resonant magnetic scattering measurement of ferromagnetic order, in the absence of an external magnetic field, using resonant dipole scattering at the Gd L_2 edge of $GdMn_2Ge_2$. Measurements of the ferromagnetic order of the Gd sublattice were accomplished using undulator radiation at the Advanced Photon Source and polarization analysis of the diffracted beam to significantly reduce the charge scattering background. The magnetic origin of the resonant intensity was confirmed by its temperature dependence and the angular dependence of the scattering cross section. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2739403]

X-ray resonant magnetic scattering (XRMS) is rapidly becoming a standard probe of microscopic magnetic structure, particularly for compounds which are not amenable to neutron measurements. Due to the much weaker magnetic scattering strength compared to charge scattering, however, these investigations have generally been limited to antiferromagnetic compounds, where the magnetic reflections are not coincident with the positions of charge reflections. While there have been XRMS experiments carried out on ferromagnetic materials, in these studies the magnetic signal has been derived from the change in diffraction peak intensity realized upon switching the polarity of an externally applied magnetic field (e.g., flipping ratios) (Refs. 1 and 2) as, for example, experiments performed to investigate ferromagnetism in CoPt alloys[3] and EuS.[4] The interference between resonant magnetic and charge scattering contributions to reflectivity has also been used to investigate thin film in the soft x-ray regime, taking advantage of large resonant enhancements at the Fe L edges.[5]

All of these measurements were performed in the presence of an external magnetic field designed to maximize the magnetic/charge interference by aligning the magnetic moments along the field direction. In certain instances, including investigations of systems with weak magnetic anisotropy or weak ferromagnetism, the magnetic moment direction and/or magnetic structure may be perturbed by the external magnetic field itself and one is particularly interested in probing the spontaneous magnetic state of the compound without any applied magnetic field. For example, for both Gd^{3+} and Eu^{2+} ions the orbital angular momentum $L=0$, and the strong magnetic anisotropy generally associated with crystal electric field (CEF) effects is absent. Intermetallic compounds containing these species are ideal candidates for investigations of the weaker magnetic anisotropy associated with dipole interactions and the 5d band effects.[6,7] The naturally occurring isotopes of these ions strongly absorb neutrons so that x-ray scattering provides an important alternative for magnetic structural studies in these cases. We also point out that zero field scattering measurements may be of importance to studies of weak ferromagnetism arising from the lock-in of the magnetic modulation vector to the commensurate value in compounds such as RNi_2B_2C (R=Tb,Er).[8,9] The absence of an applied field can exclude any weak ferromagnetic response that results from the external magnetic field itself. Here, we demonstrate the zero-field measurements of ferromagnetism in a rare earth compound, $GdMn_2Ge_2$, by XRMS.

$GdMn_2Ge_2$ is a member of the RT_2X_2 (R=rare earth, T=transition metal, and X=Si,Ge) family of the ternary intermetallic compounds. It crystallizes in the body-centered tetragonal $ThCr_2Si_2$ structure with space group $I4/mmm$ (D_{4h}^{17}) and undergoes several magnetic transitions with temperature.[10,11] Below $T_N=365$ K, the Mn moments order antiferromagnetically along the c axis, while the Gd moments remain disordered. As temperature decreases to $T_t=97$ K, the magnetic structure transitions to a ferrimagnetic state. In this transition, the Mn sublattice changes from antiferromagnetic to ferromagnetic and the Gd sublattice from paramagnetic to ferromagnetic, with the Gd and Mn moments coupled antiparallel to each other. In both the ferri- and antiferromagnetic structures, the magnetic moments are aligned along the c axis.

Single crystals of $GdMn_2Ge_2$ were grown at the Ames Laboratory using a high temperature flux growth technique.[12] The sample was mounted on a copper holder in a closed-cycle He displex refrigerator for XRMS measurements and enclosed in a Be cup with He gas in an attempt to optimize sample cooling in the presence of the intense undulator radiation available at the Advanced Photon Source. The surface of the sample was perpendicular to the c axis and the mosaic of the sample was approximately 0.01° full width at half maximum.

The XRMS experiments were performed on the 6ID-B undulator line in the MUCAT Sector at the Advanced Photon Source using a double-crystal Si(1 1 1) monochromator to select the incident photon energy. The incident beam energy was tuned to the maximum in the scattered intensity at the Gd L_2 resonant dipole ($E1$) transition, corresponding to $E=7.933$ keV. A bent mirror was used to focus the beam vertically and reduce the higher harmonic content of the beam, and pyrolytic graphite (PG) was used as an analyzer for the scattered photons. The incident beam was confined to a cross section of 0.2×0.2 mm^2 using slits.

Since, for ferro- or ferrimagnetic ordering, the magnetic scattering is collocated with the normal charge scattering, and is five to six orders of magnitude weaker, the key to observing the magnetic signal is to significantly reduce the

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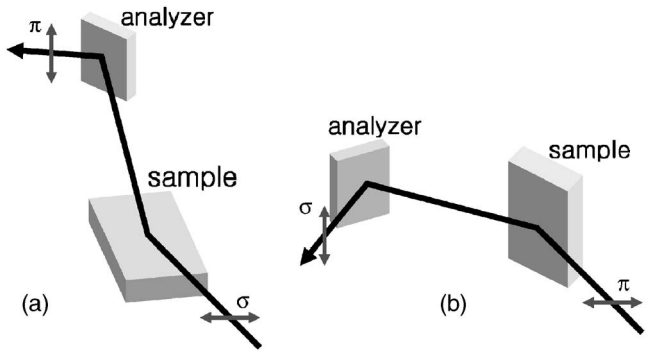

FIG. 1. Schematic views of geometries used in the experiment. (a) σ-to-π geometry and (b) π-to-σ geometry.

charge contribution by taking advantage of the difference in polarization between the magnetic and charge scatterings. This is possible since charge scattering does not rotate the polarization of the diffracted beam with respect to the incident beam, while resonant dipole scattering rotates the polarization of the diffracted beam by 90°.[13]

A six-circle diffractometer was used to perform measurements in both the vertical and horizontal scattering geometry, as shown in Fig. 1. The incident x-ray beam from the storage ring is linearly polarized in the horizontal plane. In the vertical scattering geometry of Fig. 1(a), the polarization of the incident beam is perpendicular to the scattering plane (σ polarization). The polarization of the charge scattering contribution to the diffraction peak intensity is unchanged from the incident beam polarization and, in principle, is reduced relative to the π-polarized magnetic scattering contribution due to the polarization factor of the analyzer by a factor of $\cos^2 2\theta_{\text{analyzer}}=0.000\,56$. This arrangement defines the so-called σ-to-π geometry. In the horizontal scattering geometry of Fig. 1(b), the polarization of incident beam is parallel to the scattering plane (π polarization) and the analyzer selects the scattered photon polarization perpendicular to the scattering plane (σ polarization). For this π-to-σ geometry, in addition to the polarization selection of the analyzer, the charge scattering is further reduced by the polarization factor ($\cos^2 2\theta_{\text{sample}}$) of the scattering from the sample. Therefore, the charge scattering for reflections at sample scattering angles close to 90° can, in principle, be further reduced to a level approaching the resonant magnetic scattering contribution itself.

To illustrate the discussion above, Fig. 2 shows energy scans taken through the dipole resonance at the Gd L_2 absorption edge in both the σ-to-π [Fig. 2(a)] and the π-to-σ [Fig. 2(b)] geometries above and below the ferrimagnetic ordering temperature of T_t=94 K. The subtraction (bottom panels) of the higher temperature data from the lower temperature data, in both cases, yields a well defined diffraction peak corresponding to the ferromagnetic contribution. How-

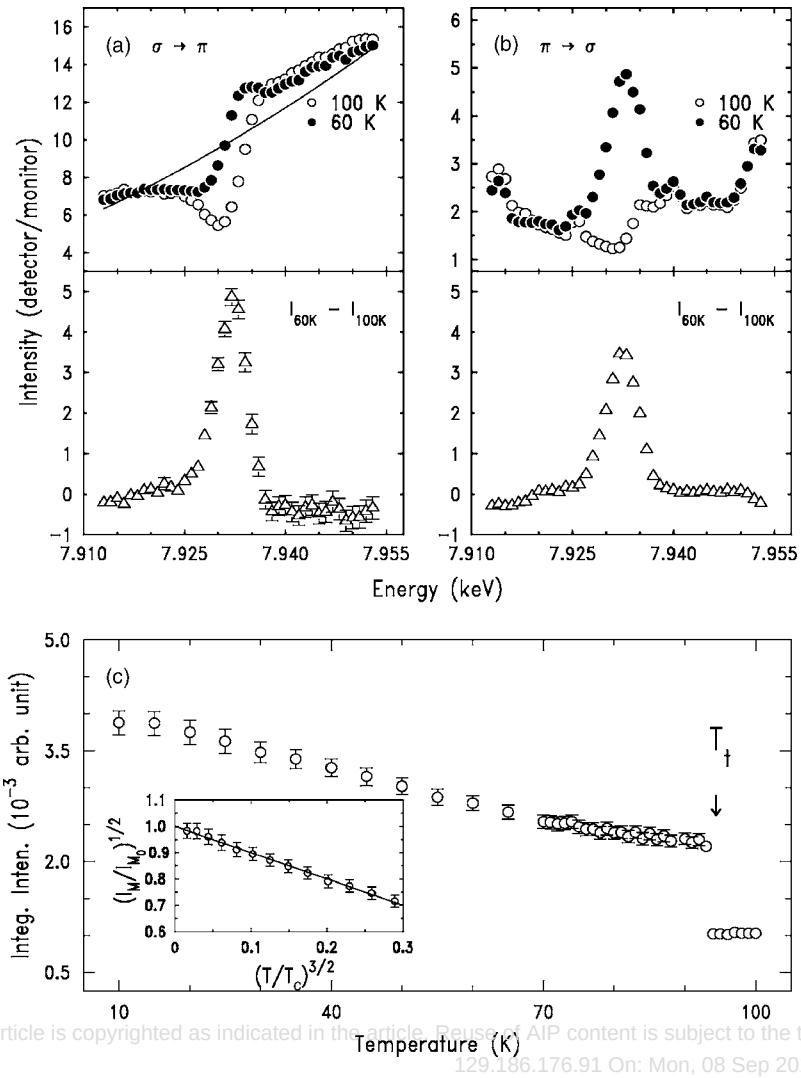

FIG. 2. (a) Energy scans for the (0 0 10) reflection at 60 and 100 K, and the differences of the intensities between two temperatures with σ-to-π geometry and (b) π-to-σ geometry. The solid line represents the polarization factor of the analyzer, $\cos^2 2\theta_{\text{analyzer}}$. (c) Temperature dependence of the integrated intensity of the (0 0 10) reflection with the π-to-σ geometry. The inset plots $(I_M/I_{M_0})^{1/2}$ where I_M is the integrated intensity of the magnetic reflection obtained by subtracting the measured intensity above the transition temperature from the intensity at each temperature and I_{M_0} is the estimated magnetic intensity at T=0 K. This is proportional to the local magnetization of Gd. The line represents Bloch's $T^{3/2}$ law $[M/M_0=1-(T/T_c)^{3/2}]$.

ever, there are clear differences in the raw data taken in these two geometries that bear further discussion.

In the σ-to-π geometry, the charge scattering is mainly σ polarized and the "charge background" is quite sensitive to the polarization factor of the analyzer. Indeed, the polarization factor changes quite rapidly for small changes in the incident photon energy for analyzer scattering angles close to 90°, giving rise to the observed slope in the background in the top panel of Fig. 2(a).

For the π-to-σ geometry, there should be an additional suppression of the charge background (by roughly a factor of 10^3) at the position of the (0 0 10) reflection because the incident beam polarization is in the scattering plane and the sample scattering angle is close to 90°. However, the incident beam is not perfectly polarized in the horizontal plane. The incident σ polarized component (approximately 0.1%) gives rise to charge scattering from the sample which is also σ polarized and passes through the analyzer unattenuated by the sample or the analyzer. This represents the major contribution to the charge background in this scattering geometry. Note that since the polarization analyzer is set to diffract σ-polarized light in this geometry, the sloped background evident in Fig. 2(a) is absent in Fig. 2(b). In the σ-to-π geometry, the charge scattering is mostly σ polarized because the incoming π polarization from the imperfect polarization is suppressed by the polarization factor of the sample. For both scattering geometries, the suppression of the charge scattering relative to the resonant magnetic contribution is approximately a factor of ~1000.

In order to confirm the magnetic origin of the peaks in Fig. 2, several tests were applied to the data. First, we note that there are additional peaks shown nearby the resonant peak in energy scans of Fig. 2(b). We have determined that these arise from multiple charge scattering contributions since (i) they do not exhibit any temperature dependence and (ii) the distribution and intensity of these peaks change dramatically as the sample is rotated about the normal to the crystal surface (in an azimuth scan) as expected for multiple scattering. In contrast, the diffraction peaks observed in the bottom panels of Figs. 2(a) and 2(b) exhibit the temperature dependence shown in Fig. 2(c), for the π-to-σ scattering geometry, with a first order ferrimagnetic to paramagnetic transition at T_t=93 K. For $T \ll T_t$, the inset of Fig. 2(c) demonstrates that the temperature dependence of the magnetization (proportional to the square root of the magnetic peak intensity) is well described by Bloch's $T^{3/2}$ law[14] up to 70 K and is consistent with the behavior of an isotropic Heisenberg spin system. The deviation from Bloch's $T^{3/2}$ law near T_t might be the influence of Mn moments.

Unlike previous observations of ferromagnetic ordering by resonant magnetic scattering in an applied field, the present experiment is not based upon the interference between charge and magnetic scatterings but rather measures the resonant magnetic scattering contribution itself. The magnetic scattering cross sections described by Hill and McMorrow,[13] may be used to determine the magnetic moment direction for Gd in this compound.[15] The resonant scattering intensities were obtained for (0 0 4), (0 0 6), (0 0 10), and (0 0 12) reflections in the σ-to-π geometry by subtracting the measured intensity above the transition temperature from that obtained from the sample at 10 K, and are plotted in Fig. 3. The general trend of the measured angular dependence agrees with a magnetic moment direction along the c axis. The magnetic diffraction does not distinguish the magnetic moment along [0 0 1] and [0 0 $\bar{1}$] so that the magnetic domain structure does not affect the magnetic intensity.

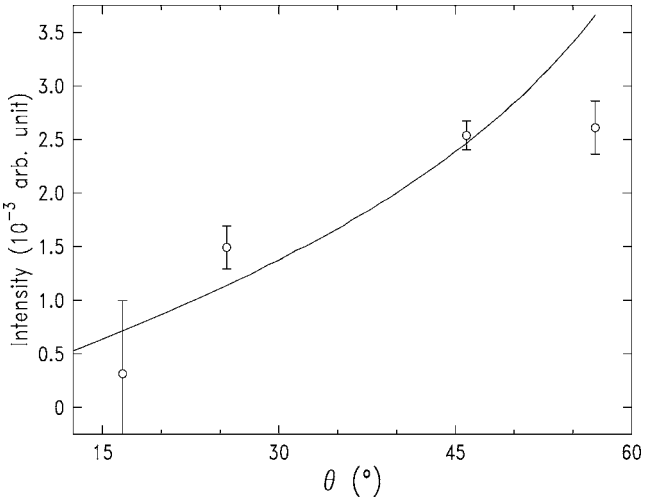

FIG. 3. Scattering angle dependence of magnetic intensity. The solid line represents theoretical prediction for a moment direction aligned along the c axis ($I \propto \sin^2\theta/\sin 2\theta$).

In summary, our result shows that it is possible to measure the ferromagnetic response at the rare earth L edges in the absence of an external magnetic field by using linearly polarized x rays and the XRMS technique. This offers the potential for measurements of spontaneous magnetism with element specificity.

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