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2005 J. Phys.: Condens. Matter 17 L493

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LETTER TO THE EDITOR

Magnetic powder diffraction from GdNi_2Ge_2 using x-ray resonant magnetic scattering

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Received 19 September 2005, in final form 6 October 2005

Published 1 November 2005

Online at stacks.iop.org/JPhysCM/17/L493

Abstract

We describe the first successful magnetic x-ray powder diffraction measurement using resonant dipole scattering at the L edge of a rare earth compound. Measurements of the antiferromagnetic ordering in GdNi_2Ge_2 were accomplished using undulator radiation at the Advanced Photon Source and polarization analysis of the diffracted beam to significantly reduce the charge background. Our results agree well with recent resonant magnetic scattering measurements on single crystals of this compound.

X-ray resonant magnetic scattering (XRMS) has become a useful complement to neutron diffraction determinations of magnetic structures. This is especially true for investigations of magnetic order in rare earth intermetallic compounds that contain neutron opaque elements such as Gd, Eu, and Sm [1–3], and for detailed investigations that require the high intrinsic collimation of synchrotron x-ray sources for increased angular resolution, without significant degradation in photon flux [4]. A notable weakness of the XRMS technique, however, is that even with the relatively strong resonant enhancement of magnetic peaks obtained at the L edges of rare earth elements, the magnetic scattering is still four to five orders of magnitude weaker than the charge scattering. This has, until now, required the use of single crystals in x-ray investigations, and precluded investigations of the magnetic structure of rare earth compounds by x-ray powder diffraction methods. Neutron powder diffraction measurements of magnetic structure are an invaluable tool for the initial determination of antiferromagnetic ordering especially for complex structures where more than one magnetic wavevector, or magnetic structure, may be identified.

A significant first step towards the realization of magnetic x-ray powder diffraction (MXP) was taken a decade ago by Collins *et al.*, using the strong uranium M_4 dipole resonance in antiferromagnetic UO_2 [3]. The integrated intensity for the (102) magnetic reflection was approximately 0.2% of the (220) charge reflection. This resonant enhancement produced a magnetic diffraction peak intensity of about 20 counts s^{-1} over a fluorescent

and charge scattering background of approximately $400 \text{ counts s}^{-1}$. Unfortunately, the huge M edge resonances for the actinide elements occur at rather low energies (the U M_4 edge is at 3.727 keV), severely restricting the accessible range of momentum transfer for scattering measurements. Whereas the L edges of rare earth ions are well suited to diffraction studies, ranging in energy from 6 to 10 keV, the L edge resonant enhancement is typically three to four orders of magnitude weaker than that of the actinide M edges, requiring a different approach to the measurement of magnetic powder diffraction.

To realize a significant gain in the signal-to-noise ratio for magnetic peaks, the measurement described here takes advantage of the change in polarization of the diffracted beam arising from the resonant dipole scattering cross-section [5]. As described below, by using the appropriate analysing crystal the charge scattering background can be effectively suppressed by the polarization selection of the analyser, and the fluorescent background is reduced through the energy discrimination of the analyser. Using this method on a powder sample of GdNi_2Ge_2 with undulator radiation at the Advanced Photon Source, we obtain a signal-to-background ratio of 2:1, sufficient for measuring several of the strong magnetic powder diffraction lines and investigating the temperature dependence of the sublattice magnetization in this incommensurate antiferromagnetic compound.

The RNi_2Ge_2 (R = rare earth element) compounds crystallize in the ThCr_2Si_2 body centred tetragonal structure (space group $I4/mmm$). Nickel is non-magnetic and the magnetic ordering of the R ion is determined by the indirect Ruderman–Kittel–Kasuya–Yosida (RKKY) exchange interaction, mediated by the conduction electrons [6]. Recent XRMS measurements at the Gd L_2 absorption edge of a single crystal of GdNi_2Ge_2 demonstrated that this compound transforms from a collinear AFM structure for $16 \text{ K} < T < T_N = 27.5 \text{ K}$ to a canted spiral structure for $T < 16 \text{ K}$. The magnetic moment for both structures lies in a plane tilted at approximately 10° from the basal plane of the tetragonal structure [7]. Over the entire temperature range, the magnetic structure remains incommensurate with the chemical lattice, so the magnetic ordering along the c -axis is described by the wavevector $(0 \ 0 \ \tau)$, where τ ranges from 0.790 at T_N to 0.808 at $T = 10 \text{ K}$.

Single crystals of GdNi_2Ge_2 were grown at the Ames Laboratory using a high temperature flux growth technique [6]. These crystals were finely powdered, pressed into a pellet, and mounted on a copper holder in a closed-cycle He dilplex refrigerator for the powder diffraction measurements. The sample was enclosed in a Be cup with He gas in an attempt to optimize sample cooling in the presence of the intense incident beam from the undulator. Nevertheless, there remained significant local heating of the sample within the illuminated volume of the sample, resulting in a shift of the sample temperature ($\approx 2.5 \text{ K}$) with respect to the values measured by the sensors. This apparent shift of approximately 2.5 K is consistent with our previous experience with samples exposed to the full monochromatic undulator beam in this temperature range. In previous single-crystal studies of GdNi_2Ge_2 , for example, the magnetic peak intensity was strong enough to allow attenuation of the incident beam in order to avoid local sample heating. With attenuation, a T_N of 27.5 K was measured. With exposure to the full beam, the measured T_N dropped to approximately 25 K.

The XRMS powder measurements were performed on the 6ID-B undulator beamline at the Advanced Photon Source (APS) at Argonne National Laboratory. The incident photon energy was selected by double-crystal Si(1 1 1) monochromator. A bent mirror was used to suppress the higher order harmonics and focus the beam, producing a beam size at the sample position of approximately 0.2 mm vertical by 1.0 mm horizontal. The resonant scattering measurement was carried out with the incident radiation linearly polarized parallel to the horizontal scattering plane (incident π -polarization). The energy was tuned to the maximum scattered intensity at the dipolar $E1$ resonance, which occurs just above the Gd L_2 edge, at $E = 7.934 \text{ keV}$.

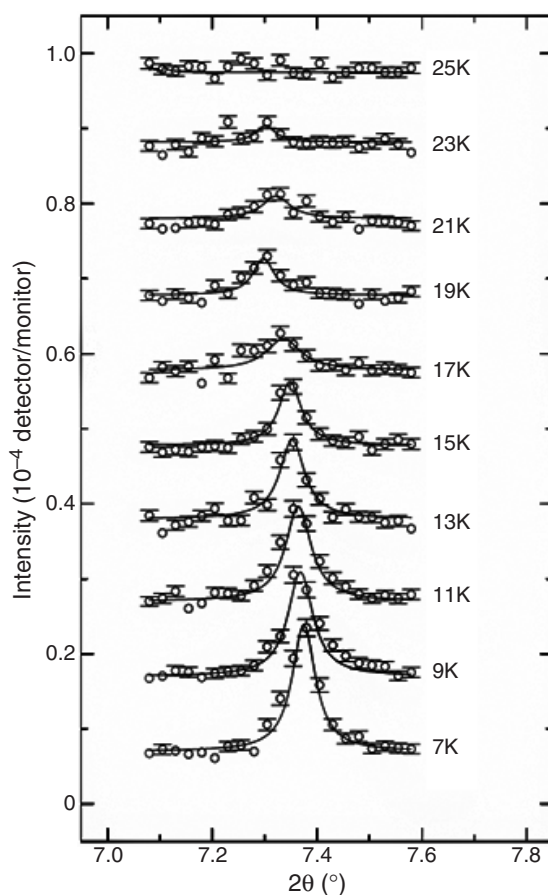


Figure 1. Evolution of the $(0\ 0\ \tau)$ magnetic satellite as a function of temperature.

For charge scattering, the polarization of the diffracted beam remains unchanged (π -polarization), whereas for resonant dipole scattering the diffracted beam polarization vector is perpendicular to the scattering plane (σ -polarization) [5]. Therefore, by making an appropriate choice of analyser crystal after the sample, oriented to diffract the beam perpendicular to the scattering plane, the charge scattering background can be effectively suppressed by roughly a factor of $\cos^2(2\Theta_{\text{analyser}})$ relative to the dipole resonant magnetic scattering. A $\text{Ge}(3\ 3\ 3)$ analyser was chosen for this purpose since $2\Theta_{(3\ 3\ 3)} = 91.73^\circ$ for the $\text{Gd}\ L_2$ edge. In addition, the relatively tight energy resolution of the $\text{Ge}(3\ 3\ 3)$ analyser, tuned for elastic scattering at $E = 7.934\ \text{keV}$, does not pass the fluorescence radiation at lower energy, thereby reducing this contribution to the background.

In figure 1 we show the evolution of the $(0\ 0\ \tau)$ magnetic satellite as a function of temperature. With the APS storage ring operating at 7 GeV and 100 mA, the maximum count rate at our base temperature was approximately $2\ \text{counts}\ \text{s}^{-1}$ on a background of approximately $1\ \text{count}\ \text{s}^{-1}$ using the configuration described above. Energy scans through the $\text{Gd}\ L_2$ edge display a distinct peak at the expected position for dipole resonant scattering. We note that the apparent Néel temperature of 25 K is approximately 2.5 K lower than that found for single-crystal samples, but this discrepancy arises from the local heating effect described above.

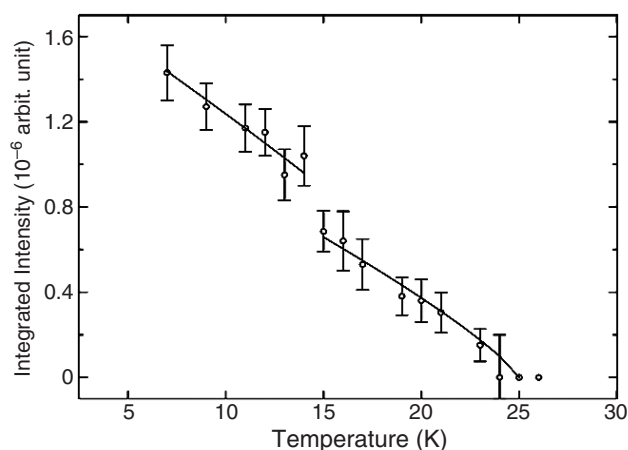


Figure 2. The temperature dependence of the integrated intensity of the $(0\ 0\ \tau)$ magnetic satellite. The lines describe power law fits to the data as described in the text.

In figure 2 we plot the temperature dependence of the integrated intensity of the $(0\ 0\ \tau)$ magnetic satellite. The solid line represents a power law fit to the data given by $I = A(1 - T/T_N)^{2\beta}$, where $\beta = 0.41 \pm 0.01$ with T_N fixed at 25 K. The break in the order parameter at approximately 14 K arises from the transition from the collinear structure at higher temperatures to the equal moment spiral structure at lower temperatures [7]. The solid line through the lower temperature data represents the same power law fit rescaled by a factor of 1.3. This change in the first-harmonic satellite intensity is consistent with the powder-averaged difference between a collinear structure that is ‘squaring up’ and an equal moment spiral magnetic structure.

In addition to the $(0\ 0\ \tau)$ peak, other magnetic satellites were measured at the base temperature of 7 K as well. Using the magnetic structure for the low temperature spiral phase of GdNi_2Ge_2 determined by the previous single-crystal measurements, the powder-averaged diffraction intensity for the $(0\ 0\ L\pm\tau)$ magnetic satellites may be written as

$$I \sim \mathbf{M} \frac{\frac{1}{2} \cos^2 \theta (1 + \cos^2 \alpha) + \sin^2 \theta \sin^2 \alpha}{\sin 2\theta} \quad (1)$$

where \mathbf{M} is the multiplicity of the reflection, 2θ is the scattering angle of the peak, and α ($=10^\circ$) is the angle between the tetragonal basal plane and the plane in which the magnetic moments lie. For the π to σ scattering geometry employed in this measurement, the polarization factor is unity, and the Lorentz factor, $\sin 2\theta$, is included in equation (1).

Using equation (1), table 1 compares the calculated and measured integrated intensities for the magnetic powder peaks measured in this experiment normalized to the $(0\ 0\ \tau)$ magnetic peak. The agreement between these values is reasonably good, showing consistency between the model proposed from the single-crystal measurements and the limited powder data presented here.

A key ingredient to the success of this experiment was the reduction of both charge scattering and the fluorescence background through the use of a polarization selection analyser with relatively tight energy resolution. Fortunately, a wide range of analyser crystals with scattering angles close to 90° are available for the energy range between 6 and 10 keV. For example, the scattering angle for the $\text{Ge}(3\ 3\ 3)$ analyser is well suited for measurements at the $\text{Gd}\ L_2$ and $\text{Ho}\ L_3$ resonances, while $\text{Al}(2\ 2\ 2)$ would be better suited for measurements at the $\text{Tb}\ L_3$ resonance [8].

Table 1. The calculated and measured integrated intensity of magnetic diffraction peaks in GdNi_2Ge_2 .

Magnetic peak	Calculated intensity	Measured intensity
$(0\ 0\ \tau)$	1	1
$(0\ 0\ 2 - \tau)$	0.66	0.8 ± 0.1
$(0\ 0\ 2 + \tau)$	0.28	0.25 ± 0.04

Although these measurements clearly demonstrate the potential for magnetic powder diffraction measurements at the L edges of rare earth elements using XRMS, there is clearly further work necessary with respect to the reduction of background and local sample heating by the incident beam. Nevertheless, the development of magnetic x-ray powder diffraction for investigations of the magnetic structures of rare earth compounds adds an important new tool, complementary to neutron diffraction techniques, for magnetic structure determinations. This is particularly true for those compounds that contain neutron opaque rare earth elements, such as gadolinium, or ligands, such as boron. Furthermore, the elemental selectivity of resonant magnetic scattering can be used to some advantage in measurements of the magnetic structure of multi-component, multi-magnetic-site compounds in the same manner as anomalous scattering measurements of charge structure. Finally, the high wavevector resolution available at third-generation synchrotron sources can help to reveal fine details of magnetic structures in complex materials.

Ames Laboratory is supported by the Department of Energy, Office of Science, under Contract No W-7405-Eng-82. The MUCAT sector at the APS is supported by the Department of Energy, Office of Science, through the Ames Laboratory Contract No W-7405-Eng-82. Use of the Advanced Photon Source is supported by the US Department of Energy, Basic Energy Sciences, Office of Science, under Contract No W-31-109-Eng-38.

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