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Temperature dependence of the magnetic excitations in Gd*

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ABSTRACT

We have made inelastic neutron scattering studies of the magnetic excitations in gadolinium above and below the Curie temperature ($T_C = 293$ K). The constant Q measurements were made out to the Brillouin zone boundaries in the $[001]$ and $[100]$ directions in the temperature range from 9 to 593 K. The small q spin-wave energies re-normalize in approximate proportionality to the net magnetization and approach zero at T_C . There is very little broadening of these peaks except at temperatures very near T_C . At large q , however, there is appreciable broadening but a much slower decrease of spin-wave energies with increasing temperature. The large q peaks do not vanish at T_C ; instead, they persist as broad shoulders in the magnetic excitation spectra up to about $1.2 T_C$.

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INTRODUCTION

Metallic gadolinium is an isotropic Heisenberg ferromagnet with a Curie temperature of 293 K. The Gd atoms behave electronically as tripositive ions with half-filled f shells so there is a spin-only moment and very little anisotropy. This makes Gd an ideal candidate for neutron inelastic scattering studies of spin dynamics since the spin coupling is dominated by the exchange interactions. In earlier papers (1,2), we reported the spin-wave dispersion for Gd in the temperature range from 78 to 232 K. We have now extended these measurements to temperatures above T_C to determine if spin waves persist in the paramagnetic region for this Heisenberg system similar to those observed (3,4) in the itinerant systems Fe and Ni. Preliminary results are presented in this paper.

CROSS SECTION

The neutron cross section for inelastic scattering from a magnetic material is proportional to a scattering function $S(q, \omega)$ which is the Fourier transform of a spin-pair correlation function. The form that $S(q, \omega)$ assumes is fairly well understood for a Heisenberg ferromagnet only in the limits of $T \ll T_C$ and $T \gg T_C$. In the low temperature limit, $S(q, \omega)$ describes propagating spin waves and consists of a pair of Lorentzian shaped peaks at $\pm \omega(q)$ where $\omega(q)$ is proportional to

q^2 at small q . At high temperatures, $S(q, \omega)$ describes the paramagnetic spin correlations and consists of a single peak centered at zero energy with a q dependent shape and width. In the intermediate region, especially near T_c , the theoretical form of $S(q, \omega)$ is model dependent and, although several analytical expressions have been suggested (5-8), none of these are firmly established. For this preliminary analysis we have taken a modified version of an expression used in recent analyses of neutron data for EuO (6) and Pd_3Fe (7), which reproduces the observed thermal evolution of the magnetic excitation spectra. We take

$$S(q, \omega) \propto (1 - e^{-\hbar\omega\beta})^{-1} \left[\frac{\omega\Gamma}{(\omega - \omega_0)^2 + \omega^2\Gamma^2} + \frac{\omega\Gamma}{(\omega + \omega_0)^2 + \omega^2\Gamma^2} \right] \quad (1)$$

where the first term is the thermal population factor with $\beta = (kT)^{-1}$. The important parameters are the characteristic frequency, ω_0 , and a dimensionless damping parameter, Γ , which arises from spin wave-spin wave interactions. In the weak interaction limit where $\Gamma \ll 1$, this gives a pair of Lorentzian peaks at $\pm \omega_0$ with half widths of $\omega_0\Gamma$. With strong interactions so that $\Gamma \gg 1$, this goes over into a single Lorentzian centered at $\omega = 0$ with a half width of ω_0/Γ . A variety of spectral shapes is obtained for intermediate interaction cases.

EXPERIMENTAL

The sample was the same single crystal of ^{160}Gd that was used in the previous studies (1,2). Neutron measurements were made with a triple-axis spectrometer in the constant- Q mode of operation. Energy scans were made by varying the incident energy with fixed final energies of either 1.0 or 3.6 THz depending on the resolution requirements. Higher order wavelength contaminants were essentially removed from the scattered beam by the use of either Be or pyrolytic graphite filters. Measurements were made out to the Brillouin zone boundaries in the $[001]$ and $[100]$ directions in the temperature range from 9 to 593 K. Some of the raw data are shown in Figs. 1 and 2 which illustrate typical temperature dependences of the spin-wave spectra at small and large q .

Figure 1 shows high resolution data at temperatures near T_c for a wavevector of $0.15 \, 2\pi/c$ along the $[001]$ direction. The central peak consists of a temperature independent component, which is probably the incoherent scattering from the furnace, plus a component that increases with decreasing temperature and which may be domain wall scattering. The peaks

symmetrically displaced about the central peak correspond to spin waves propagating along the c-axis direction. These move continuously inward with increasing temperature and collapse into a single peak centered at $\omega = 0$ at T_c . This is the expected behavior at small q where the spin-wave energies should follow the magnetization and vanish at T_c . At these high temperatures, spin-wave interactions should produce a broadening of the spin-wave peaks. However, this broadening is strongly q dependent ($\propto q^4$ at small q) (5) and is barely detectable at this particular q value.

After correction for background and for the higher order wavelength contamination at the monitor, the neutron data must be folded with the instrumental resolution function for fitting. So far, this fitting has been done only for the small q data obtained under high resolution conditions. Fitted frequency values are given in Table I. The damping parameters are not tabulated but are less than 0.5 for all of these q and T values. If the spin-wave energies are represented by

$$\hbar\omega_0(q) = Dq^2 \quad (2)$$

where D is the stiffness constant, then the observed temperature dependence near T_c is given by

$$D(T) = D(0) \left(\frac{T_c - T}{T_c} \right)^{0.33} \quad (3)$$

where $D(0) = 35 \text{ meV-Å}^2$. This temperature dependence is consistent with dynamic scaling theory (5).

Typical large q behavior is illustrated in Fig. 2. These are medium resolution data for a wavevector of $0.9 \text{ 2}\pi/\text{c}$ along $[001]$. These spectra show a pronounced broadening of the spin-wave linewidths near T_c and an energy renormalization less rapid than that observed at small q . The peaks do not collapse into a single peak at T_c ; instead a distinct shoulder remains in the excitation spectrum above T_c . Other data, not included in Fig. 2, show that this shoulder persists up to about 350 K. Because of the the strong damping, it is not clear that these excitations above T_c should be characterized either as propagating or overdamped spin waves. Their shapes suggest that there are characteristic frequencies associated with the spin correlations and this feature remains up to about $1.2 T_c$.

The temperature dependence of the magnetic excitations in Gd is qualitatively similar to that reported for Fe and Ni in that the small wavevector behavior follows the predictions of hydrodynamic theory while strong departures from that behavior are observed at larger wavevectors. However, the spin wave peaks above

T_C in Fe and Ni are more pronounced and persist to higher temperatures ($1.4 T_C$ in Fe, $2 T_C$ in Ni) than in Gd. The presence of spin waves in the paramagnetic region is presumably associated with short range ordered regions which exist over large enough correlation lengths and times to support short-wavelength, high-frequency spin waves. It would seem then that short-range order is much more pronounced in Fe and Ni than in Gd.

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Table I.

Temperature Dependence of Small q Spin-Wave
Frequencies in Gd

$T(^{\circ}\text{K})$	$\nu_0(0.10)^*$	$\nu_0(0.15)^*$	$\nu_0(0.20)^*$	$\nu_0(0.25)^*$
9	0.126	0.252	0.42	-
200	0.110	0.184	0.329	-
250	0.057	0.122	0.216	0.349
273	0.041	0.084	0.155	0.271
282	0.034	0.068	0.132	0.242
287	0.030	0.061	0.118	0.214

* $\nu_0(q)$ in THz for q in $2\pi/c$

FIGURE CAPTIONS

- Fig. 1. Temperature dependence of the magnetic excitation spectrum of Gd at $q = 0.15$ in the $[001]$ direction (q is given in units of $2\pi/c$).
- Fig. 2. Temperature dependence of the magnetic excitation spectrum of Gd at $q = 0.9$ (in $2\pi/c$) along $[001]$.

Gd

$$q = 0.9 \left(\frac{2\pi}{c} \right)$$