Temperature dependence of the spin and orbital magnetization density in $Sm_{1-x}Gd_xAl_2$ around the spin-orbital compensation point.

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Non-resonant ferromagnetic x-ray diffraction has been used to separate the spin and orbital contribution to the magnetization density of the proposed zero-moment ferromagnet $Sm_{0.985}Gd_{0.015}Al_2$. The alignment of the spin and orbital moments relative to the net magnetization shows a sign reversal at 84K, the compensation temperature. Below this temperature the orbital moment is larger than the spin moment, and vice versa above it. This result implies that the compensation mechanism is driven by the different temperature dependencies of the 4f spin and orbital moments.

Specific heat data indicate that the system remains ferromagnetically ordered throughout.

Recently, it was proposed that the Laves phase compound $Sm_{1-x}Gd_xAl_2$ exhibits a spin-orbital compensation point at $\approx 85K$ when $x = 0.0185$. Magnetometry showed that the net moment dipped to zero at this temperature, but was finite either side in the magnetically ordered phase (the Curie temperature is 128K). At the compensation temperature, magnetic Compton scattering shows a net spin moment, indicating that the system consists of a ferromagnetically ordered spin sublattice. For the net moment to be zero, this spin moment must be exactly compensated by the orbital moment. Although it is thought that this may be driven by the different temperature dependencies of the Sm 4f spin and orbital moments, this had not yet been investigated. An understanding of the compensation mechanism may be gained by studying the temperature dependence of the spin and orbital moments near the compensation point, which requires direct measurement of the spin and orbital magnetization. Such an x-ray diffraction study is reported here for the first time. Our result conclusively proves that the compensation point is driven by the different temperature dependence of the spin and orbital moments. Our specific heat data indicate that the system remains magnetically ordered.

The magnetism of Sm and its compounds has been the focus of many investigations as a result of the importance of the conduction electron polarization and the complex crystalline electric field (CEF) in the material. The spin and orbital contribution to the $Sm^{3+}$ 4f moment are of similar size and aligned antiparallel, and the polarized conduction electron spin moment is thought to align parallel with the 4f spin moment. The three components to the site magnetization almost cancel, leaving a small net local moment. Interestingly, the temperature dependencies of the spin and orbital components are not identical due to a complex thermal admixture of nearly degenerate J multiplets in which the $Sm^{3+}$ ion exists in (the ground state 5/2 multiplet is 1500K from first excited state 7/2). The admixture arises from the CEF effect on the degeneracy of the J states and has long been an explanation of the magnetism in Sm compounds.

A solid solution of $Gd^{3+}$ introduces a large ($7.6 \mu_B$) spin moment onto the Sm site in $Sm_{1-x}Gd_xAl_2$ and the small induced lattice distortion alters the CEF deformation potential. It also critically affects the RKKY exchange interaction due to the increase in conduction electron polarization. These factors have a considerable influence on the temperature dependence of the Sm site moment, as the thermal admixture of J states is renormalized. The result is to change the temperature dependencies of the 4f spin and orbital moments. In the undoped compound the moments are $M_t \approx 4.3\mu_B$ and $M_s \approx -3.8\mu_B$ respectively. The change in the CEF allows the Sm orbital and Gd/Sm spin contributions to cancel each other completely at a distinct temperature below $T_C$: at this point the material has no net moment, and is referred to as compensated. This effect in itself is not unusual in some ferrimagnetic systems, where two sublattice magnetizations become equal and opposite at a particular temperature. However in this case the magnetism exists only on the rare earth site (a solid solution of Sm / Gd ions). A naive picture of the temperature dependence has three order parameters, 4f orbital magnetism, 4f spin magnetism, and conduction electron spin polarization with the latter probably having the same temperature dependence as that of the 4f spin. If the order parameters are of opposite sign, with non-identical temperature dependence the system can become compensated. Previous work has concentrated on the bulk magnetization and the type of magnetic ordering at the compensation point. However the mechanism of compensation in $Sm_{1-x}Gd_xAl_2$ has not been investigated.

In this letter we report the use of non-resonant x-ray diffraction to investigate the magnetization density of $Sm_{0.98}Gd_{0.012}Al_2$ as a function of temperature through the spin-orbital compensation point by monitoring a Bragg reflection. The technique has the advantage of allowing the separation of the spin and orbital form factors by changing the experimental geometry. At the wavevector sampled, the conduction electron moment makes...
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The technique of non-resonant x-ray diffraction has re-
cently been developed as a convenient method of studying
the spin density in ferromagnetic materials using ellipti-
cally polarized synchrotron radiation. It is particularly
useful for materials where the neutron technique is not
viable due to the high neutron absorption cross section.
The technique also gives a convenient method of separ-
ating the spin and the orbital contribution to the total
magnetic form factor by changing the experimental geo-
metry (see fig 1). Essentially the technique makes use of
the suppression of Thompson charge scattering at a
scattering angle of 90\degree
for radiation linearly polarized in
the scattering plane. When elliptically polarized photons
are incident, the charge and magnetic Bragg intensities
interfere. This leads to modulation of the signal with
reversal of the sign of magnetic component, this can be
achieved by either flipping the sample magnetization vec-
tor (in the scattering plane), or by flipping the helicity
of the incident beam polarization. When the pure charge
scattering is a minimum, the signal modulation, result-
ning from the magnetic scattering cross section, tends to
a maximum, which facilitates the measurement of a flip-
ning ratio.

The fractional change in intensity upon reversal of the
sample magnetization or the photon helicity is related to
the orbital ($F_L$), spin ($F_S$) and charge($F_C$) form factors
as:

$$ R(\alpha) = \frac{I_\alpha - I_{\bar{\alpha}}}{I_\alpha + I_{\bar{\alpha}}} = g f_p \frac{2F_S(k)\sin\alpha + F_L(k)(\sin\alpha + \cos\alpha)}{F_C(k)} $$

In the past most experiments have made use of a poly-
chromatic incident beam of x-rays, in order to collect
data on a number of Bragg peaks simultaneously, using
energy dispersive Ge detectors. However, the white beam
method suffers from multiple diffraction which corrupts
the signal and is difficult to model. In this investigation
a monochromatic beam was used to avoid these uncer-
tainties. However the principle remains identical to that
described previously for white beam experiments[7][8].

A single crystal sample of $Sm_{1-x}GdxAl_2$ with $x = 0.018$ was produced by the Bridgemann method, with
the polycrystalline boule sealed in a Ta can to maintain
stoichiometry. The structure of the resulting crystal was
verified as the $C_{15}$ Laves phase using Laue photography.
The non resonant magnetic diffraction experiment was
performed on the XMaS beamline[9] at ESRF. Ellipti-
cally polarized radiation was extracted by viewing the
bending magnet source at a angle of $\approx 0.3$ mrad from
the plane of the synchrotron: the optimum position in
terms of signal to noise. The sample was mounted in a
Be shrouded closed cycle He cryostat. An incident en-
ergy of 5.736keV was selected using the double bounce
Si monochromator, such that the 333 reflection was in
the Bragg condition with a scattering angle of 90\degree
in the plane of the synchrotron (see fig 1). The calculated po-
larizion of the beam at the incident energy used was
$P_l = 0.99470$ and $P_c = -0.02937$, yielding a polariza-
tion factor, $f_p$, of -5.5. The diffracted beam was de-
tected using a fast NaI scintillator with an average count

FIG. 1: Schematic layout of a generic non-resonant ferromag-
netic diffraction experiment. Left hand figure shows experi-
mental configuration for measurement of the orbital compo-
nent of the form factor. Right hand figure shows configuration
for measurement of the total (L+S) form factor.
rate of \( \approx 85000 \text{cps} \) at the diffraction peak. The magnetic field was applied using a 1T electromagnet, which was flipped at intervals of 20s in order to average over the beam position fluctuations inherent with the bending magnet source. In this configuration a single flipping ratio was acquired over an integration time of 2 hours. The flipping ratios of the 333 reflection were measured as a function of temperature, in the total (eq 3) moment configuration and the orbital only (eq 2) configuration, in both heating and cooling cycles to ensure reproducibility of the data. At \( \sin \theta / \lambda = 0.32 \AA^{-1} \) on the form factor curve only the 4f moments contribute to the magnetic signal.

A comprehensive investigation of the magnetic properties of the sample was performed at Warwick University using SQUID and VSM magnetometry, specific heat measurements and AC susceptibility, in order to check sample quality and to investigate the complex magnetic properties of the sample comprehensively.

For reflections of the type \( hh'h \) where \( h \) is odd, only the 4f site contributes to the phase factor. The temperature dependence of the orbital, spin and total form factor curve at \( \sin \theta / \lambda = 0.32 \AA^{-1} \) is shown in fig 2. It is clear that below the compensation temperature the orbital contribution is positive (fig 2 A), and thus the derived spin contribution is negative (fig 2 B), with a smaller magnitude as expected, since at this wave vector the conduction electron polarization is not measured. The spin-only form factor result is in good agreement with that measured previously for the un-doped sample[11]. Above the compensation temperature the spin and orbital contributions are reversed, with approximately equal magnitudes. The total form factor (fig 2 C) is positive below the compensation temperature, as expected, since the total magnetization will follow the large orbital contribution. Interestingly above \( T_{comp} \) the total form factor is negligible. It is clear that both the orbital and the spin component to the form factor have a complex temperature dependence, furthermore both components flip sign at \( T_{comp} \), with the total form factor tending to zero at \( T_{comp} \). This result implies that the systems exhibits no ferro-magnetic character at \( T_{comp} \). This does not mean that the system becomes paramagnetic however, or that the orbital, or spin magnetizations disappear at the compensation point, for the following reasons.

The orbital and spin components to the magnetization have different temperature dependence and an anti-parallel arrangement. The net moment in the system will always align with the field, (when the field is large i.e. 1T see next section). At low temperature this results in a positive contribution to the magnetization density arising from the orbital moment and a negative contribution arising from the spin moment, with a net positive magnetization density where \( L>S \). When the system becomes compensated the orbital and spin components are in effect antiferro-magnetically aligned. However the orbital and the spin components should still be finite. Our data show a definite sign reversal of the spin and the orbital components. The change of sign results from the spin component becoming dominant above \( T_{comp} \), hence the net moment is re-aligned with respect to the field.

The fact that our orbital data tend to zero smoothly at \( T_{comp} \) rather than exhibiting a sharp step-like transition, is a statistical artefact produced from a combination of unwanted beam movements, from the synchrotron bending magnet source, and temperature fluctuations in the cryostat due to reversing the applied field. If one assumes that the temperature is only stable to within \( \pm 0.5 K \) one may easily reach a point whereby the sample is driven from one side of \( T_{comp} \) to the other, by the eddy current heating effect, throughout the period of the measurement, thereby measuring zero.

Above \( T_{comp} \) the measured form factor is negligible. This means that the 4f components to the magnetization are of a similar size, which in turn implies that the conduction electron spin component (not measured by the diffraction experiment) is of critical importance. Our diffraction data provide clear evidence that the spin and the orbital contributions to the 4f magnetization density cancel at the compensation point, and that the compensation point occurs as a result of the different temperature dependence of the spin and orbital form factor.

This diffraction result will now be discussed in the context of the bulk properties of the system. Firstly the magnetization data observed for the sample as a function of temperature (fig 2 A). The bulk magnetization data clearly show that, at the compensation point, the net moment in the sample is zero. However the magnetic behav-
The closed hysteresis loop at MAE and hence realign the net moment in the system.

...field of 1T, which is large enough to overcome the large is clarified. The diffraction data were taken in an applied field of 1T, which is large enough to overcome the large size of the Gd moment. It is also clear that the 4f contributions above $T_{\text{comp}}$ appear, from our data to be equal. Thus it is reasonable to assume that the net magnetization observed above $T_{\text{comp}}$ (fig 3:A) results almost exclusively from the conduction electron moment in the system, the temperature dependence of which is unknown (although it is reasonable to assume it is similar to the 4f spin moment) Such a measurement is planned using the magnetic Compton scattering technique, which directly samples the polarization of all spin polarized electrons.

...below $T_{\text{comp}}$ drastically alters, depending on whether the sample is field cooled or zero field cooled. On cooling in a small $10^{-2}$T field the magnetization shows a large diamagnetic effect below $T_{\text{comp}}$ (fig 3:A, triangles). The size of the diamagnetic effect can be altered by changing the magnitude of the applied field (up to 10 T) at $T_{\text{comp}}$. D: Specific heat capacity.

FIG. 3: Low temperature properties of $\text{Sm}_{1-x}\text{Gd}_x\text{Al}_2$. A: Magnetization as a function of temperature. Triangles Field cooled in $10^{-2}$T. Circles field cooled in 0.1T. Diamonds zero field cooled. Squares field cooled in 1T. B: Temperature dependence of the coercive field. C: Magnetization as a function of field (up to 10 T) at $T_{\text{comp}}$. D: Specific heat capacity.

...compensation temperature. We have demonstrated that the compensation mechanism is driven by the temperature dependence of the spin and orbital moments in the system. We have shown that the unusual temperature dependence of the bulk magnetization is driven by the reversal of the dominant 4f component at the compensation temperature, i.e. $T < T_{\text{comp}}$:L>S and $T > T_{\text{comp}}$:S>L. The fact the bulk measurement of specific heat shows no anomaly at the $T_{\text{comp}}$ implies that magnetic system remains ordered, as one may expect due to the high magnetocrystalline anisotropy energy.

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