Induced spin polarization in ferromagnetic $Gd_{62.4}Y_{37.6}$

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Evidence of a spin moment, induced through a Ruderman-Kittel-Kasuya-Yosida-type interaction in $Gd_{62.4}Y_{37.6}$, is presented. The additional moment, of $0.16\pm0.03\mu_B$, arises from polarization of Y-like electrons in the alloy. The moment was detected in a Compton scattering experiment via the measurement of the one-dimensional projection of the momentum space electron-spin density in Gd and in the alloy. The result is consistent with theoretical predictions calculated using the linear muffin-tin orbital method within the local spin-density approximation.

Oscillatory exchange coupling[1] was first observed in superstructures of Gd and Y. This was quickly followed by observations of giant magnetoresistance in other similar multilayer systems. The coupling between successive ferromagnetic Gd layers is thought to rely upon the spin-polarization of the Y layers,[2] although this polarization has never been observed. Given that yttrium plays a similar important role in bulk Gd-Y alloys, we have studied the latter in order to search for evidence of such an induced moment. The bulk alloy system exhibits interesting magnetic behavior, having three ordered phases,[3] and is an excellent system for studying the magnetic interactions. At high Gd concentrations (above 70% Gd) the alloy has different ferromagnetic phases at low and high temperature. In compositions containing less than 60% Gd the alloy exhibits a helical antiferromagnetic phase. In the intermediate compositions there is a delicate balance between the three phases. For a large composition range, the total magnetic moment is greater than would be expected simply from the dilution of the Gd.[4,5] There has been considerable effort made towards an understanding of the magnetic structures of the Gd-Y alloys, but the nature of the excess moment has not hitherto been fully resolved. Moreover, the physics of the magnetic ordering of the Gd-Y alloy system is also considered to arise from an indirect exchange interaction involving the Y electrons. In this paper we present experimental evidence for the polarization of the Y electrons, in the form of an excess *spin* moment in ferromagnetic $Gd_{62.4}Y_{37.6}$.

Gadolinium metal is a $4f$ ferromagnet, with a magnetic moment of $7.63\mu_B$ (Ref. 6) and a Curie temperature of 294 K. The moment comprises $7\mu_B$ from the half filled $4f$ shell, plus an induced conduction electron moment of $0.63\mu_B$. As predicted by Hund's rules, there is no orbital component ($L=0$). Gadolinium has the hcp structure, and the moment aligns along the c axis down to 235 K, below which it becomes canted.[7] Yttrium has the same structure and a similar atomic volume to Gd, and hence the alloys readily form a continuous solid solution with only small changes in their lattice parameters. The nonmagnetic Y ''impurities'' might be thought to have very little effect on the magnetic properties, acting simply as a diluent. However, in studies of the paramagnetic moments, Thoburn *et al.*,[4] in early work, and Foldeaki *et al.*, more recently,[5] showed that the addition of Y does not simply monotonically reduce the total moment; an extra contribution is apparent. Thoburn *et al.* had also found a large additional moment in the ferromagnetic phase, although its size may be ascribable to the lower value they quoted for the pure Gd moment [$7.12\mu_B$ compared to the now accepted value of $7.63\mu_B$ (Ref. 6)]. The behavior of the total moment could be explained in two ways: either the presence of yttrium modifies the crystal field, resulting in an *orbital* contribution to the moment, or the hybridized conduction bands in the alloy enable a larger *spin* moment to be induced.

The $4f$ electrons of Gd are highly localized, and the magnetic ordering in both the Gd metal and in the Gd-Y alloy

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arises from an indirect exchange interaction mediated via the conduction electrons. Hence the ordering mechanism in the Gd-Y alloys is quite different from that observed in the transition-metal alloys such as YFe_2,[8] where significant hybridization is expected with the $3d$ Fe electrons. In the rare earths, a Ruderman-Kittel-Kasuya-Yosida (RKKY)-type interaction is required, which is explained in terms of the wave-vector dependent susceptibility $\chi(\mathbf{q})$.[9] In pure Gd, $\chi(\mathbf{q})$ has a maximum at $\mathbf{q}=0$, leading to the observed ferromagnetic ordering.[10] If this maximum is at a nonzero $\mathbf{q}$, because of yttrium induced Fermi surface nesting in the alloys,[11,12] then a more complex arrangement of spins may form,[3,13] such as seen by x-ray resonant magnetic scattering in a DyLu thin film.[14] The RKKY-type interaction relies on the polarizability of the Y conduction bands. However, it has recently been proposed that the additional moment can be accounted for purely by considering orbital contributions originating from the modified crystal field.[5] This was reasoned from the experimental behavior of the effective Landé g factor and total angular momentum J, which is compatible with the assumption that the crystal field is changed by the presence of Y, permitting spin-orbit coupling to induce the orbital moment. The presence of a spin moment in the low-temperature ferromagnetic (''ferro-II'') phase would indicate that the exchange-splitting persists and that the Y bands are polarized, obviating the need for a large orbital contribution in the alloy. The goal of this experiment was to determine whether there is indeed an extra spin moment contribution to the magnetization in the low-temperature ferro-II phase.

The experiment was performed using magnetic Compton scattering, a uniquely sensitive probe of the spin component of the magnetization. The Compton profile $J(p_z)$ is defined as the one-dimensional projection of the electron momentum distribution $n(\mathbf{p})$,

$$J(p_z)=\int\int n(\mathbf{p})\mathrm{d}p_x\mathrm{d}p_y, \tag{1}$$

and the integral of $J(p_z)$ is simply the total number of electrons per unit cell. The profile can be obtained experimentally from the energy spectrum of the inelastically scattered photons. This is achieved by exploiting the Compton effect, in which monochromatic photons scattered through a given angle by stationary electrons would have a single energy determined purely by the scattering angle. However, because bound electrons must have some distribution of momenta, the photon energy is Doppler broadened into an energy distribution. This is related to the Compton profile, defined above, via the scattering cross section,[15] within the impulse approximation.[16] If the photons impinging on a sample have a component of circular polarization, then a small spin dependence appears in the scattering cross section.[17] Reversing either the photon polarization or the magnetization of the sample changes the sign of the spin-dependent signal, which enables the spin part to be isolated. The resultant profile, known as the magnetic Compton profile (MCP), is a projection of the momentum density of only those electrons with unpaired spins,

$$J_{\mathrm{mag}}(p_z)=\int\int [n\uparrow(\mathbf{p})-n\downarrow(\mathbf{p})]dp_xdp_y. \tag{2}$$

Here, $n\uparrow(\mathbf{p})$ and $n\downarrow(\mathbf{p})$ are the momentum dependent spin densities. The area under the MCP is equal to the number of unpaired electrons, that is, the total spin moment per formula unit:

$$\int_{-\infty}^{\infty} J_{\mathrm{mag}}(p_z)\mathrm{d}p_z=\mu_{\mathrm{spin}}. \tag{3}$$

Magnetic Compton scattering is now an established technique for probing momentum space spin densities and band structures in magnetic materials.[18,19] Within the impulse approximation, the method is solely sensitive to *spin* magnetic moments;[18,20,21] that is to say, the orbital moment is not measured.[22] The value of magnetic Compton scattering lies in its uniform sensitivity to the whole of the spin-resolved electron momentum distribution. Since the MCP is a difference between Compton profiles, the contributions from the nonmagnetic electrons and from unwanted systematic sources disappear. Spin-polarized positron angular correlation experiments also probe the spin density,[23,24] but are subject to both positron-electron correlation effects and repulsion of the positron by the positive ion cores, so that the positron does not sample electrons in all states equally.[25] Furthermore, the incoherent nature of the Compton scattering process means that the electron-density distribution can be sampled at all momenta, notably at the low momenta where the conduction electrons contribute.

The [0001] MCP for $Gd_{62.4}Y_{37.6}$ was measured on the high-energy x-ray beamline at the ESRF. The experiment was performed in reflection geometry[26] with an incident beam energy of 200 keV, selected by the {311} reflection of a Si monochromator, and a scattering angle of 168°. The samples were 5 mm diameter $\times 1.3$ mm thick disks and were oriented so that the resolved direction was within $\pm 2°$ of [0001]. The temperature of the samples was maintained at 70 ± 2 K. At present, it is difficult to reverse the polarization of the synchrotron x-ray beam, but in soft ferromagnets like these, the sample's magnetization can be easily reversed. Here, the magnetization was kept alternately parallel and antiparallel to [0001] with a 0.96 T rotating permanent magnet. The energy spectrum of the scattered x rays was measured by a solid-state Ge detector. The momentum resolution obtained was 0.44 atomic units (a.u., where 1 a.u.$=1.99\times 10^{-24}$ kg m s^{-2}). The total number of counts in each of the charge profiles was 1.5×10^8, resulting in 3.7×10^6 in the MCP with a statistical precision of $\pm 3\%$ at the magnetic Compton peak in a bin of width 0.09 a.u. The usual correction procedures for the energy dependence of the detector efficiency, for absorption, relativistic scattering cross section and magnetic multiple scattering were applied and after checking that the profiles were symmetric about zero momentum, the MCP's were folded about this point to increase the effective statistical precision of the data.

The experiments were complemented by LMTO band-structure calculations[27] performed within the local spin-density approximation (LSDA).[28] The authors recently demonstrated the ability of this technique to predict the magnetic Compton profiles of ferromagnetic Gd metal and the details of the calculations are described more fully there.[26] In order to predict the MCP of the disordered $Gd_{62.4}Y_{37.6}$ alloy, a

TABLE I. Calculated partial spins in Bohr magnetons per atom for pure Gd and for Gd and Y in $Gd_{62.5}Y_{37.5}$. Also shown are the total moments per formula unit of the alloy.

	s	*p*	*d*	*f*	Total
Gd (pure)	0.025	0.141	0.580	6.94	7.64
Gd (alloy)	0.032	0.153	0.525	6.85	7.56
Y (alloy)	0.000	0.124	0.205	0.022	0.350
Total	0.020	0.142	0.405	4.29	4.91

16-atom supercell approach was adopted. By taking ten Gd and six Y atoms, a effective composition of $Gd_{62.5}Y_{37.5}$ was obtained, very close to that of the measured sample. The results were essentially unchanged for different configurations of the 16 atoms. In order to provide a suitable comparison, the pure Gd MCP was also calculated in the supercell and it was found to be essentially identical to that from the standard calculation. The lattice parameter and c/a ratio were, respectively, 6.8710 a.u. and 1.590 for Gd, and 6.8758 a.u. and 1.584 for the alloy. The predicted moments and their associated characters for Gd and $Gd_{62.5}Y_{37.5}$ are presented in Table I. It can be seen that an extra spin moment of $0.35\mu_B$ is expected per Y atom, mainly on the *p*- and *d*-like itinerant electrons deriving from Y orbitals, and this corresponds to $0.13\mu_B$ per formula unit. It is important to note that, while this moment is not actually on the Y site, it is nevertheless associated with Y electrons and no increase in the Gd itinerant moment is observed. The predicted [0001] MCP's are presented in Fig. 1, where the $4f$ moments have been normalized to account for the Gd dilution in the alloy. This clearly shows that the conduction electron contribution increases relative to the $4f$ moment in the alloy.

In Fig. 2, we present the experimental MCP's for Gd and $Gd_{62.4}Y_{37.6}$, normalized according to their $4f$ moments. These are presented together with the theoretical predictions after convolution with a Gaussian of FWHM 0.44 a.u. to simulate the experimental resolution. The Gd profile is analyzed in more detail in Ref. 26. The salient features here are the narrow conduction electron contributions at low momentum superimposed on the broader $4f$ profile, as expected.

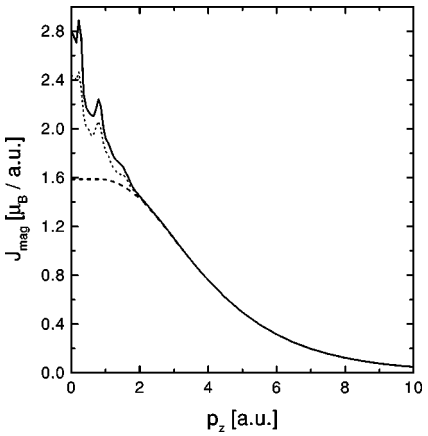

FIG. 1. Calculations of the magnetic Compton profile of Gd (dots) and $Gd_{62.5}Y_{37.5}$ (solid line) resolved along [0001], performed using a 16-atom supercell. Also shown is the equivalent free atom profile for Gd $4f$ electrons (dashes).

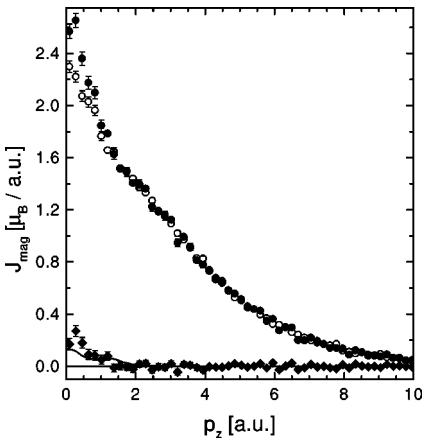

FIG. 2. The experimental magnetic Compton profiles of Gd, normalized to $7.62\mu_B$ (open circles) and $Gd_{62.4}Y_{37.6}$ (solid circles), scaled to $4f$ tails of the Gd profile. The diamonds represent the difference in Bohr magnetons per a.u. for a formula unit of the alloy. The difference in the theoretical profiles (see Fig. 1), convoluted with a Gaussian with full width at half maximum $=0.44$ a.u. to represent the experimental resolution, is presented as a solid line.

Comparison of the experimental profiles for the pure metal and the alloy shows that there is a small, but genuine difference for $p_z<2$ a.u., consistent with that predicted by theory. By integrating over p_z, the value of the induced moment can be calculated as

$$\Delta\mu_{\text{spin}}=0.16\pm0.03\mu_B. \tag{4}$$

In order to investigate the robustness of this small difference, we performed a number of checks during the data analysis. The difference is present in the raw, uncorrected data. The corrections applied to the Gd and $Gd_{62.4}Y_{37.6}$ data sets are essentially the same, since both measurements were performed with the same setup, and the detector efficiency and scattering cross sections are identical in both cases. The multiple-scattering contribution was calculated to be an order of magnitude smaller than the measured difference signal. The absorption correction was calculated to simulate possible misalignments in the setup, but the results remained unchanged. It should be noted that near $p_z=0$ a.u. this correction is almost a linear function of p_z and any error will be unlikely to affect the difference observed, especially when the data are folded about the origin. Hence the result is robust to any reasonable variations in the corrections applied.

In conclusion, we have presented evidence of an additional spin moment in ferromagnetic $Gd_{62.4}Y_{37.6}$, corresponding to a polarization of the Y-band electrons. It is unaffected by the corrections that need to be applied to the experimental data. Band-structure calculations performed within the LSDA are consistent with our result. It should be remembered that the presence of this spin moment does not rule out the existence of an additional small orbital moment. However, in contrast with the interpretation of Foldeaki *et al.*,[5] we conclude that, irrespective of any orbital contribu-

tion, there is indeed a substantial polarization-induced spin moment of $0.16 \pm 0.03 \mu_B$ in the alloy.

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