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Is gadolinium a helical antiferromagnet or a collinear ferromagnet?*

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Abstract. Controversial issues concerning the nature of magnetic ordering in gadolinium are briefly reviewed. The recent experimental results are shown to resolve most of such issues in that they rule out the possibility of a helical spin structure in Gd and clearly bring out the role of long-range dipolar interactions in stabilising collinear ferromagnetic order for temperatures between the spin-reorientation temperature and the Curie point.

Keywords. Magnetic order; ac susceptibility; gadolinium; critical phenomena; dipolar interactions; magnetic anisotropy.

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1. Introduction

Four decades ago, Belov and Ped'ko [1] observed that the magnetisation (M) of *polycrystalline* gadolinium (Gd) exhibits (i) a steep rise (in thermomagnetic curves taken at external magnetic fields $H_{\rm ext} \leq 15\,{\rm Oe}$) not at $T_{\rm C} \sim 290$ K (as expected for a ferromagnet) but at a lower temperature $T_{\rm C} \sim 210$ K as the temperature is lowered from $T > T_{\rm C}$ and (ii) a 'jump' (in M vs. $H_{\rm ext}$ isotherms taken in the range $T_1 \leq T \leq T_{\rm C}$) at a field $H^j_{\rm ext} (\leq 15$ Oe) which shifts to higher fields as the temperature is raised from T_1 to $T_{\rm C}$. Since these anomalies are reminiscent of those previously found to occur in dysprosium at the critical fields that mark the disappearance of 'helical' antiferromagnetism, Belov and Ped'ko [1] concluded that a *helical* spin structure similar to that prevalent in other heavy rare-earth metals also exists in Gd in the temperature range $T_1 \leq T \leq T_{\rm C}$, with the only difference that fields as low as 15 Oe suffice to transform the helical antiferromagnetism into collinear ferromagnetism. This picture of the spin structure in Gd had to be discarded after subsequent

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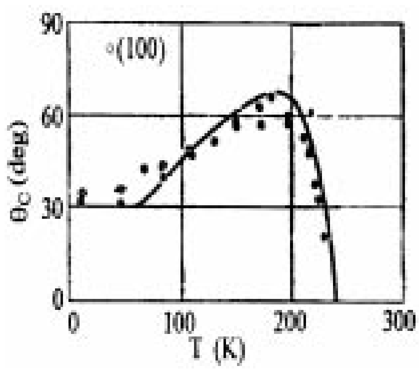

Figure 1. Temperature dependence of the tilt angle (θ_C) of easy direction of magnetisation of Gd with respect to *c*-axis (ref. [10]).

magnetic investigations [2–5] on Gd *single crystals* failed to reproduce such anomalies or kinks in low-field magnetisation, and neutron diffraction experiments [6,7] did not reveal any satellite reflections characteristic of helical spin structures. Magnetocrystalline anisotropy [8–10] and neutron diffraction [6,7] studies clearly demonstrated that (figure 1) the easy direction of magnetisation is *parallel* to the hexagonal *c*-axis from $T_C \sim 293$ K down to the spin re-orientation (SR) temperature T_{SR} of 230 K (where the anisotropy constant K_1 changes sign [8,9] and K_2 is vanishingly small, figure 2, [8,9]), moves away from the *c*-axis for $T < T_{SR}$ to a maximum tilt angle of $\theta_C \sim 60°$ near $T^* = 180$ K, and then tilts back to within 30° of the *c*-axis at low temperatures. The widely accepted experimental view that Gd is a normal ferromagnet with a rather complex (figure 1) temperature dependence of the spontaneous moment alignment has been challenged [11] recently. Based on the observation that the initial susceptibility $\chi_{ext}(T)$ of the needle-shaped single crystals of Gd is *not demagnetisation-limited* at T_C but at T_{SR}, it has been claimed [11] that the magnetic order in Gd in the temperature range $T_{SR} \leq T \leq T_C$ is akin to the helical spin structure previously found in erbium. This situation is further complicated by a sharply divided theoretical opinion [12,13] on the issue of whether the ground state of Gd is ferromagnetic or antiferromagnetic. Moreover, no theoretical consensus [14] regarding the nature of magnetic structure near T_C has been arrived at so far.

Other puzzling issues that have a direct bearing on the nature of magnetic ordering in Gd include the following. Considering that Gd metal is made up of *spherically symmetric* $^8S_{7/2}$ Gd^{3+} ions and *isotropic* Ruderman–Kittel–Kasuya–Yosida (RKKY) interactions between *localised* 4f magnetic moments give rise to ferromagnetism in this metal, Gd is expected to have a vanishing magnetocrystalline anisotropy and thus behave as an ideal *isotropic* three-dimensional (3D) *Heisenberg* ferromagnet. Contrary to this expectation, overwhelming experimental evidence in favour of a small uniaxial magnetocrystalline anisotropy, which ensures that the *c*-axis of the hexagonal-close-packed lattice is the preferred orientation of magnetisation in Gd at temperatures in the range $T_{SR} \leq T \leq T_C$, asserts that the critical behaviour of Gd is that of a 3D Ising ferromagnet. Numerous experimental investigations of the critical behaviour of Gd carried out till recently have failed to resolve the issue of whether Gd behaves as a 3D Heisenberg or as a 3D Ising ferromagnet in the critical region.

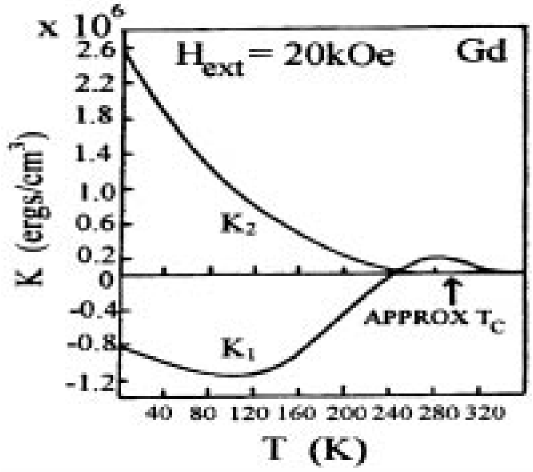

Figure 2. Temperature dependence of magnetocrystalline anisotropy constants K_1 and K_2 of Gd (ref. [9]).

2. Recent developments

Contrary to the recent claim [11] that Gd behaves as an antiferromagnet with a helical spin structure for temperatures between T_{SR} and the Néel point, high-resolution ac susceptibility and low-field bulk magnetisation data taken along the [0001] and [10$\bar{1}$0] hexagonal directions of high-purity Gd single crystals over a wide range of temperatures, when interpreted properly by taking into account both shape as well as magnetocrystalline anisotropies [15], provide ample experimental evidence for the existence of *collinear* ferromagnetism in Gd in the temperature range $T_{SR} \leq T \leq T_C$. A brief summary of the arguments that lead to this conclusion is presented here.

One of the characteristic properties of ferromagnets is the divergence of intrinsic magnetic susceptibility χ_{int} along the easy direction of magnetisation at $T = T_C$. When both shape as well as magnetocrystalline anisotropies are present, $\chi_{int}(T)$ is related to the measured initial susceptibility $\chi'_{ext}(T)$ as

$$\chi_{int}^{-1}(T) = \chi_{ext}'^{-1}(T) - 4\pi N(T) \tag{1}$$

where $N(T) = N_d + N_K(T)$, the demagnetising factor N_d depends only on the sample shape, $H_d = -4\pi N_d M$ is the demagnetising field, $N_K(T) = H_K(T)/4\pi M_S(T)$ is the 'so-called' magnetocrystalline anisotropy factor, H_K is the *uniaxial* anisotropy field and M_S is the spontaneous magnetisation. According to eq. (1), χ_{int} diverges at a temperature T_0 where $\chi'_{ext}(T_0) = 1/4\pi\, N(T_0)$; T_0 can be significantly different from T_C if $N_K(T_C) \neq 0$. Figures 3 and 4 display the temperature variations [15] of the real, χ'_{ext}, and imaginary, χ''_{ext}, components of the ac susceptibility measured when $H_{dc} = 0$ and $H_{ac}(\nu = 87\ \text{Hz}) \equiv H_{ext} = 10$ mOe is applied along the cylindrical axis in *as-grown* crystal rod of diameter 1.55 mm and length 26.8 mm, sample 1 (figure 3) and along the directions *parallel* (*c*-axis or [0001] direction) and perpendicular (the [10$\bar{1}$0] direction) to the cylindrical axis in an *oriented* single crystal rod of diameter 1.5 mm and length 1.7 mm, sample 2 (figure 4) in which

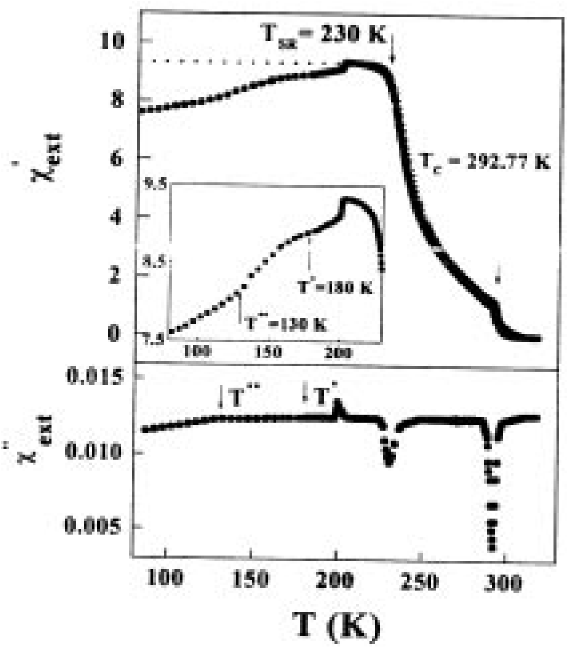

Figure 3. Temperature (T) dependence of the real ($\chi'_{\rm ext}$) and imaginary ($\chi''_{\rm ext}$) components of ac susceptibility of single crystal sample 1 of Gd at 87 Hz frequency for ac field amplitudes of 10 mOe (closed circles) and 1 Oe (crosses) applied along the c-axis (see text) (ref. [15]). The inset shows expanded plot for the region $T = 80$–230 K. The horizontal dashed line indicates the demagnetisation limited value $\chi'_{\rm ext} = 1/4\pi N_{\rm d}$ (see text).

the cylindrical axis coincides with the c-axis. The horizontal dashed lines in these figures indicate the demagnetization-limited values. The observed temperature variations of $\chi'_{\rm ext}$ (figures 3 and 4) can be understood in terms of eq. (1) by considering the following cases.

Case I: $H_{\rm ext}$ is applied along the sample dimension for which $N_{\rm d}$ has the smallest value (e.g., $N_{\rm d} = 0.0085$ when $H_{\rm ext}$ is along the cylindrical axis of sample 1), but this direction is not favoured by magnetocrystalline anisotropy, i.e., when $N_{\rm d} \ll N_{\rm K}$. With decreasing temperature, $\chi'_{\rm ext}$ rises steeply from a small value $\sim 1/4\pi N_{\rm K}$ at $T_{\rm C}$ (since $N_{\rm K}$ is large) to a large value $= 1/4\pi N_{\rm d}$ at $T_{\rm SR}$, where $K_1 = K_2 = 0$ (figure 2) and hence $N_{\rm K} = 0$, since $N_{\rm d}$ is extremely small.

Case II: $H_{\rm ext}$ is applied along the easy direction of magnetisation (e.g., the cylindrical axis of sample 2 ($N_{\rm d} = 0.31(1)$) which is also the [0001] direction) so that $N_{\rm K} = 0$ (as the magnetocrystalline energy, $E_{\rm K}$, is minimum). Consequently, $\chi'_{\rm ext}$ gets *limited* at the value $1/4\pi N = 1/4\pi N_{\rm d}$ from $T_{\rm C}$ (where $\chi_{\rm int}^{-1} = 0$) down to $T_{\rm SR}$.

Case III: $H_{\rm ext}$ points in the *hard* direction (e.g. the $[10\bar{1}0]$ direction in sample 2 ($N_{\rm d} = 0.345$)), for which $E_{\rm K}$ is *maximum* and $4\pi N_{\rm K} = 2K_1/M_{\rm S}^2$ is sizable since K_1 is large. $\chi'_{\rm ext}(= 1/4\pi N)$ attains a value at $T_{\rm C}$ which lies well below the demagnetisation limit $= 1/4\pi N_{\rm d}$ as $N_{\rm K} > N_{\rm d}$, increases with decreasing temperature because K_1 (and hence $N_{\rm K}$) decreases

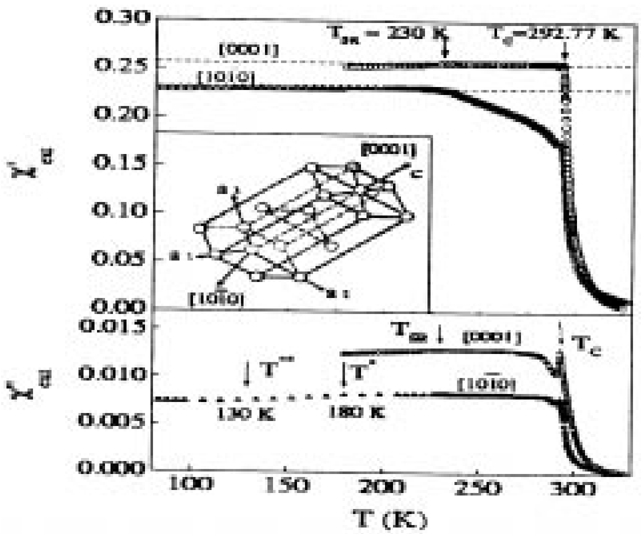

Figure 4. Temperature dependence of the real (χ'_{ext}) and imaginary (χ''_{ext}) components of ac susceptibility of single crystal sample 2 of Gd at 87 Hz frequency for ac field amplitude of 10 mOe applied along the crystallographic directions [0001] (open circles) and $[10\bar{1}0]$ (open triangles) (ref. [15]). The inset shows the crystal structure of Gd indicating the directions [0001] and $[10\bar{1}0]$. The horizontal dashed lines indicate the demagnetisation limited value $\chi'_{\text{ext}} = 1/4\pi N_{\text{d}}$ (see text).

(figure 2), and reaches the demagnetisation limit at T_{SR} where $K_1 = 0$ (as such $N_{\text{K}} = 0$) while $K_2 = 0$ in the range $T_{\text{SR}} \leq T \leq T_{\text{C}}$ (figure 2). The cases I, II and III correspond to the data presented in figures 3 and 4. The structure observed in $\chi'_{\text{ext}}(T)$ and $\chi''_{\text{ext}}(T)$ curves at T^* and T^{**} is a manifestation of the peak at $T^* = 180$ K and the crossover from rapid to slow variation at $T^{**} = 130$ K in the $\theta_{\text{C}}(T)$ curve shown in figure 1. For details, the reader is referred to [15].

Renormalisation group treatment [16] of spin systems, such as Gd, in which uniaxial dipolar (UD) and isotropic dipolar (ID) interactions of normalised coupling strengths g_{UD} and g_{ID} (such that $g_{\text{UD}} \ll g_{\text{ID}}$) occur in association with isotropic Heisenberg (IH) interactions predicts the sequence of crossovers: Gaussian regime $\rightarrow$ short-range IH $\rightarrow$ long-range ID $\rightarrow$ UD fixed point when temperature is lowered from high temperatures to T_{C}. Recently, high resolution 'zero-field' ac susceptibility and bulk magnetisation data taken along the *c*-axis (easy direction of magnetisation) of high-purity Gd single crystals have unambiguously demonstrated the following (figure 5) [17,18]. (i) The asymptotic critical behaviour of Gd is that of a uniaxial dipolar ferromagnet. (ii) As the temperature is raised above T_{C}^{UD}, a crossover from UD to ID fixed point occurs at a sharply-defined temperature $\varepsilon_{\text{CO}}^{\text{UD}\rightarrow\text{ID}} = 2.05(10) \times 10^{-3}$, where $\varepsilon = (T - T_{\text{C}}^{\text{UD}})/T_{\text{C}}^{\text{UD}}$ and this crossover, at high temperatures, is followed by a very sluggish ID $\rightarrow$ Gaussian crossover which proceeds without the (theoretically predicted) intervening isotropic short-range Heisenberg regime. (iii) The lowering of temperature below T_{C}^{UD} results in a crossover from UD to isotropic short-range Heisenberg fixed point at a temperature $\varepsilon_{\text{CO}}^{\text{UD}\rightarrow\text{IH}} = -2.08(5) \times 10^{-3}$, which is close

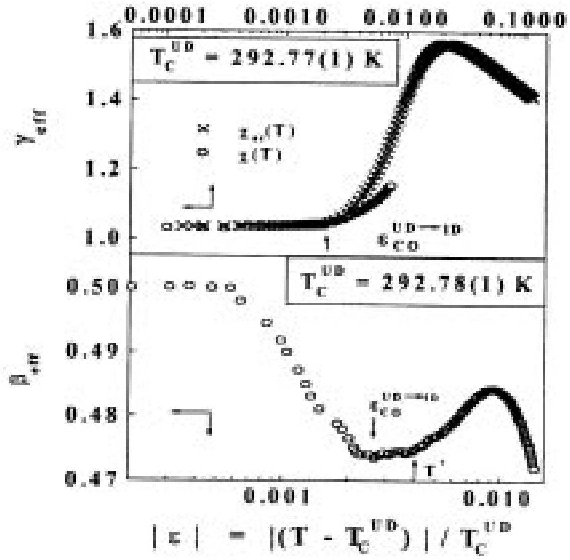

Figure 5. Variation of effective critical exponents, β_{eff} and γ_{eff}, with reduced temperature $|\varepsilon|$ (see text).

to the temperature $T^\dagger$ at which a transition from linear (uniaxial dipolar/Ising) to Bloch (Heisenberg) domain wall occurs. Details concerning the observed crossover scenario in the thermal critical behaviour of Gd are given in refs [17,18].

3. Concluding remarks

A detailed comparison [19] between the results of a mode coupling theory (which includes dipolar coupling and uniaxial anisotropic effects) and critical spin dynamics experiments lends firm support to the observation that the asymptotic critical behaviour of Gd is that of a uniaxial dipolar ferromagnet. This finding is consistent with (i) the theoretical prediction [20] that the long-range dipole–dipole interaction between magnetic moments localised at the sites of the hcp lattice favour the *c*-axis as the easy direction of magnetisation when the unit cell parameter ratio c/a falls below its ideal value of $c/a \approx 1.63$ (this is the case in Gd when $T > T^\dagger \approx 291$ K) and (ii) the result of early neutron diffraction [6,7] and magnetocrystalline anisotropy [8,9] investigations that Gd is a collinear ferromagnet for temperatures between T_{SR} and T_{C}.

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