Critical behaviour of nanocrystalline gadolinium: evidence for random uniaxial dipolar universality class

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Abstract

We report on how nanocrystal size affects the critical behaviour of the rare-earth metal Gd near the ferromagnetic-to-paramagnetic phase transition. The asymptotic critical behaviour of the coarse-grained polycrystalline sample (with an average crystallite size of \(L \approx 100 \mu m\)) is that of a (pure) uniaxial dipolar ferromagnet, as is the case with single crystal Gd, albeit the width of the asymptotic critical region (ACR) is reduced. As the grain size approaches \(\sim 30 \text{ nm}\), the ACR is so narrow that it could not be accessed in the present experiments. Inaccessibly narrow ACR for \(L \sim 30 \text{ nm}\) and continuous increase in the width of the ACR as \(L\) decreases from 16 to 9.5 nm basically reflect a crossover to the random uniaxial dipolar fixed point caused by the quenched random exchange disorder prevalent at the internal interfaces (grain boundaries).

Keywords: critical behaviour, phase transitions, gadolinium, magnetism

(Some figures may appear in colour only in the online journal)

1. Introduction

For a long time, like the insulating EuS and EuO, the rare-earth metal Gd was considered to be an archetypal isotropic three-dimensional (\(d = 3\)) Heisenberg ferromagnet for two reasons. First, magnetocrystalline anisotropy is expected to be extremely weak [1] since Gd is made up of pure S-state Gd\(^{3+}\) ions with an orbital angular momentum \(\vec{L} = 0\). Second, isotropic Rudermann–Kittel–Kasuya–Yosida (RKKY) interactions, which mimic the Heisenberg form, couple the magnetic moments localized at the sites of the hexagonal close-packed (hcp) lattice of Gd. During the past decade, theoretical efforts to understand the physical origin of the observed magnetocrystalline anisotropy [2, 3], ferromagnetic ground state [4, 5] and unusually large volume magnetostriction near the Curie temperature, \(T_C\) [6], have resulted in substantial modifications to the long-held simplistic picture (\(d = 3\) Heisenberg ferromagnet) of Gd. Consequently, (i) the significant contribution of long-range dipole–dipole interactions to magnetocrystalline anisotropy as well as to polarizing the localized 4f spins and (ii) the polarization of the 5spd and 6s conduction-band electron spins due to RKKY coupling to the localized 4f spins, are now well established. Another important recent development is the resolution [7] of the basic issue [7, 8] of whether Gd is a ferromagnet with a collinear spin structure or an antiferromagnet with a helical spin arrangement, akin to other heavy rare-earth metals at temperatures ranging between the spin-reorientation temperature \(T_{SR} = 230 \text{ K}\) and \(T_C = 292.77 \text{ K}\).

The long-standing (spanning nearly four decades) controversy [9] about the asymptotic critical behaviour of Gd near the ferromagnetic-to-paramagnetic phase transition has finally been put to rest by demonstrating [10, 11] that single power laws alone cannot adequately describe the observed temperature variations of spontaneous magnetization, \(M(T, 0)\), and intrinsic susceptibility, \(\chi(T)\), in the asymptotic critical region (ACR), but do so only when the multiplicative logarithmic correctionsto these power laws, predicted by the
renormalization group (RG) calculations for a $d = 3$ uniaxial dipolar ferromagnet [12, 13], are taken into account. To be more specific, zero-field electrical resistivity/specific heat $C_{H=0}$ [14, 15], $M(T, 0)$ and $\chi(T)$, taken along the $c$ axis (easy direction of magnetization) of a high-purity Gd single crystal, respectively follow the RG-predicted temperature variations, $C_{H=0} \sim |\epsilon - \epsilon| \ln |\epsilon|^{1/3}$, with $\epsilon = (T - T_C)/|T_C|$ and $\alpha = 0$, $M(T, 0) \sim (\epsilon)^\beta |\ln |\epsilon|^{1/3}$, with $\beta = 0.5$, and

$$\chi^{-1}(T) = \Gamma_p^{-1} \epsilon \gamma |\ln |\epsilon|^{1-\gamma},$$

with $\gamma = 1$ and $x = 1/3$ over two decades in reduced temperature. For instance, equation (1) is obeyed in the ACR 5.1 $\times 10^{-5} \leq \epsilon \leq 2.05 \times 10^{-3}$ for $e > 0$, with $T_C = 292.77(1)$ K, $\gamma = 1.0008(5)$ and $x = 0.329(1)$ [10, 11]. Thus, Gd (single crystal) belongs to the $d = 3$ uniaxial dipolar universality class and will henceforth be referred to as a pure uniaxial dipolar (PUD) ferromagnet. In essence, theory has so far dealt with idealized imperfection-free model systems while the experiments have been mostly carried out on single crystal Gd.

A field of growing research activity relates to the influence of quenched randomness, i.e. compositional, topological, bond and/or site disorder, as well as confinement, proximity and translational symmetry breaking on magnetic properties [16–23]. Remarkable effects are routinely observed in real magnetic materials where free surfaces, internal interfaces, line and point defects, local composition fluctuations and preparation history form integral parts of the material's microstructure. For instance, the recent observation of an enhancement (reduction) in the magnetic moment per Eu ion and Curie temperature in EuTe nanoparticles [27]. The effects of randomness on second-order phase transitions have been a subject of sustained interest [28–35]. In this context, the celebrated ‘Harris criterion’ [36] provides general guidelines, i.e. (i) the addition of short-range disorder to an ordered (pure) system, which undergoes a second-order phase transition, should not affect the sharpness of the transition and hence leave the critical exponents unaltered, if the specific-heat critical exponent of the pure system is $\alpha_p < 0$ and (ii) a crossover to a new type of (random) critical behaviour could occur, if $\alpha_p > 0$. Since the specific heat of a $d = 3$ pure uniaxial dipolar ferromagnet (e.g. Gd) diverges asymptotically as $|\ln |\epsilon|^{1/3}$, according to the Harris criterion (ii), adding quenched randomness/disorder to a pure uniaxial dipolar ferromagnet should result in a new type of asymptotic critical behaviour. Indeed, RG calculations [37, 38], based on the quenched random exchange Ising model (which includes both quenched site- and bond-diluted models) with dipolar interactions, yield the multiplicative corrections to the leading singular behaviour in susceptibility and specific heat at $T_C$ that are drastically different from their pure uniaxial dipolar counterparts. For instance, they predict the intrinsic susceptibility for $e > 0$ as

$$\chi^{-1}(T) = \Gamma_i^{-1} \epsilon \exp \left( -\sqrt{D} |\ln |\epsilon| \right),$$

with $\gamma = 1$ and a universal constant $D \cong 0.113$ [38]. The asymptotic critical behaviour, now characterized by the new correction term $\exp(-\sqrt{D} |\ln |\epsilon|)$, corresponds to the $d = 3$ random uniaxial dipolar (RUD) universality class.

The main objective of the present study is to ascertain whether or not the quenched randomness/disorder present in nanocrystalline Gd metal gives rise to deviations from the pure uniaxial dipolar behaviour and if so, can the asymptotic leading singularity be identified and a universality class assigned to a possible new fixed point?

Before presenting the experimental details, we briefly specify the type of randomness/disorder prevalent in nanocrystalline Gd. It is a polycrystalline aggregate made up of randomly oriented nanometre-sized grains embedded in a manifold of grain boundaries. The core region of such grain boundaries accommodates the atomic mismatch between adjacent but differently oriented nanocrystallites. The atomic site mismatch in the grain boundaries gives rise to random site disorder that translates into random exchange interactions between localized atomic magnetic moments whereas the uniaxial anisotropy axis, oriented along the $c$ axis of a hexagonal close-packed crystal structure in each individual nanocrystal, varies randomly from grain to grain leading to a random distribution of easy axes in the grain ensemble.

The structural correlation length of such nanocrystallites, i.e. the grain size, $L$, can be varied depending on preparation and subsequent annealing conditions. Nanocrystalline Gd may, thus, be considered as a model system to study the influence of quenched random exchange disorder and random (intra-nanocrystal uniaxial) anisotropy on the ferromagnetic-to-paramagnetic phase transition. The control parameter $L$ permits manipulation of the strength of quenched random exchange disorder as well as the degree of random anisotropy. Actually, these quantities are coupled since both of them scale as $L^{-1,4}$.

### 2. Experimental details and data analysis

In order to unravel the asymptotic critical behaviour of nanocrystalline Gd, ac susceptibility, $\chi_{ac}$, was measured as a function of temperature in steps of 20–40 mK, particularly in the critical region, at the ac driving field of typical amplitude $1 \text{ Oe}$ and frequency $100 \text{ Hz}$ on several nanocrystalline Gd samples of different average grain sizes. Details of sample preparation [18] and characterization of structure, local magnetization and surface morphology by x-ray diffraction [39], Mössbauer spectroscopy [21], small-angle neutron scattering [23] and atomic force microscopy [39] are furnished elsewhere [18, 21, 23, 39]. Figure 1 serves to highlight the effect of grain size on the real part of $\chi_{ac}(T)$, $\chi'(T)$. After correcting $\chi'(T)$ for demagnetization to arrive at the intrinsic susceptibility, $\chi(T)$, the ‘range-of-fit’ (ROF) analysis, detailed in [10, 11, 40–42], is used to determine the effective and asymptotic amplitudes and critical exponents, appearing respectively in the single power law (SPL), i.e. $\chi^{-1}(T) = \Gamma_{eff}^{-1} \epsilon^{\gamma_{eff}}$, and in equations (1) and (2). In the ROF analysis, as a first step, changes in the values of the free fitting parameters, e.g. $T_C$, the effective or asymptotic critical amplitudes and exponents, as well as $\Gamma_i^{-1} \epsilon \gamma |\ln |\epsilon|^{1-\gamma}

Note that the values for $L$ quoted in this work represent the volume-weighted average grain diameters assuming a spherical crystallite shape [48].
the sum of deviation squares, are monitored as the fit range $\varepsilon_{\text{min}} \leq \varepsilon \leq \varepsilon_{\text{max}}$ is varied by keeping $\varepsilon_{\text{min}}$ ($\varepsilon_{\text{max}}$) fixed at a certain value and varying $\varepsilon_{\text{max}}$ ($\varepsilon_{\text{min}}$). In the next step, $T_C$ is fixed at the value that yields the optimum least-squares fit to the data in the temperature range in the immediate vicinity of $T_C$ where the remaining fitting parameters are insensitive to the variation in $\varepsilon_{\text{max}}$ for a given $\varepsilon_{\text{min}}$, and the variations of the effective or asymptotic critical amplitudes and exponents are obtained as a function of $\varepsilon_{\text{max}}$.

3. Results and discussion

Figure 2(a) displays $\gamma_{\text{eff}}(\varepsilon)$ and $\gamma(\varepsilon)$, obtained from the ROF analysis [10, 11, 40–42], based on SPL and PUD expressions, for the coarse-grained ($L = 100 \ \mu$m) Gd sample. Note that the logarithmic correction exponent $x$ in equation (1) is kept constant at the RG value $x = 1/3$ in the ROF analysis, which yields the same value $T_C = 291.917(3)$ K for $T_C$, within the uncertainty limits, in both SPL and PUD cases. From the data presented in figure 2(a) it is evident that $\gamma_{\text{eff}}$ (SPL) as well as $\gamma$ (PUD) are very close to the mean-field value of 1 for temperatures up to a well-defined crossover (‘co’) temperature, $\varepsilon_{\text{co}} = 1.5 \times 10^{-3}$, which marks the onset of crossover. Beyond $\varepsilon_{\text{co}}$, the effective and asymptotic susceptibility critical exponents increase steeply. The temperature range $0 < \varepsilon < \varepsilon_{\text{co}}$ equals the width of the ACR. The observation that $\gamma_{\text{eff}} \rightarrow 1$ as $\varepsilon \rightarrow 0$ is a strong indication of the PUD behaviour in the ACR. This inference is further supported by the observation that the PUD value $\gamma = 1.0001(7)$ is in much closer agreement with the RG value of $\gamma = 1.0$ (figure 3) than the SPL estimate $\gamma_{\text{eff}} = 0.999(3)$, within the ACR.

As evidenced from the results of the SPL-ROF analysis displayed in figure 2(b) (cf figure 2(a)), $\gamma_{\text{eff}}(\varepsilon)$ exhibits a completely different behaviour in the nanocrystalline Gd sample with $L = 25.6$ nm as compared to coarse-grained Gd in that there is no clear indication of the asymptotic critical behaviour. Instead, $\gamma_{\text{eff}}$ tends to approach 1 when $\varepsilon < 4.9 \times 10^{-4}$ (the temperature closest to $T_C = 285.63$ K in the experiment) and attains the value $\gamma_{\text{eff}} = 1.35(5)$ on either side (i.e. in the reduced temperature ranges $7.4 \times 10^{-4} \leq \varepsilon \leq 8.7 \times 10^{-4}$ and $1.8 \times 10^{-3} \leq \varepsilon \leq 3.5 \times 10^{-3}$) of the minimum ($\gamma_{\text{eff}}(\varepsilon_{\text{min}}) = 1.25(5)$ occurring at $\varepsilon_{\text{min}} = 1.17 \times 10^{-3}$. That such a non-monotonic temperature variation of $\gamma_{\text{eff}}$, indicative of a series of crossovers in the critical region (which result from an interplay between the different types of interactions), is completely reproducible, has been established by repeated experimental runs on this sample.

For a PUD ferromagnet, the RG calculations [12, 13] predict the sequence of crossovers uniaxial dipolar (UD) $\rightarrow$ isotropic dipolar (ID) $\rightarrow$ isotropic short-range Heisenberg (IH) $\rightarrow$ Gaussian regime, as the temperature increases from $T_C$. According to the RG treatment [43, 44] of ferromagnets with isotropic short-range Heisenberg and long-range dipolar interactions, the characteristic experimental signature [45] for the ID–IH crossover is a well-defined minimum in $\gamma_{\text{eff}}(\varepsilon)$ (at $\varepsilon_{\text{dip}}$ with $\gamma_{\text{eff}}(\varepsilon_{\text{dip}})$ $\approx 1.28$) that separates the asymptotic UD regime (characterized by the critical exponent $\gamma_{\text{ID}} = 1.372$) from the IH regime (with $\gamma_{\text{IH}} = 1.365$). A direct comparison between theory and experiment thus reveals that in the present

Figure 2. Temperature variations of the effective, $\gamma_{\text{eff}}$, and asymptotic, $\gamma$, susceptibility critical exponents, deduced from the ROF analysis based on the PUD (equation (1)) and/or SPL expressions (figures (a) and (b)), or the SPL and RUD (equation (2)) expressions (c) for (a) coarse-grained Gd ($L = 100 \ \mu$m) with $T_C = 291.917$ K, (b) nanocrystalline Gd ($L = 25.6$ nm) with $T_C = 285.63$ K and (c) nanocrystalline Gd ($L = 9.5$ nm) with $T_C = 252.335$ K.
experiments on the sample with \( L = 25.6 \text{ nm} \), the asymptotic regime could not be accessed as it is extremely narrow and lies well below \( \epsilon = 4.9 \times 10^{-4} \); outside the ACR, the observed temperature variation of \( \gamma_{\text{eff}} \) conforms well with the RG predictions. The behaviour of \( \gamma_{\text{eff}}(\epsilon) \) similar to that found in the sample \( L = 25.6 \text{ nm} \) is also observed in \( L = 33.7 \text{ nm} \) and even in the latter case, the ACR remained inaccessible although the \( T_C = 287.22 \text{ K} \) was approached as closely as \( \epsilon = 4.7 \times 10^{-4} \).

With reference to the coarse-grained specimen, the behaviour of \( L = 25.6 \text{ nm} \) seems to suggest that the reduction in the grain size by four orders of magnitude weakens the effective uniaxial anisotropy to some extent, but promotes the quenched random exchange disorder at grain surfaces/interfaces and in grain boundaries, such that, at such grain sizes, \( L \geq 25-34 \text{ nm} \), quenched random exchange disorder has a strength just sufficient to qualify as a relevant perturbation (or a relevant scaling field in the RG sense) which, in turn, renders the PUD fixed point unstable and causes a crossover to the RUD fixed point. The ACR is thus so narrow as to remain inaccessible to experiments. If this line of argument is pursued, the RUD ACR is expected to progressively increase in width as a result of increasing quenched random exchange disorder with decreasing \( L \). Consistent with this expectation, the width of the RUD ACR increases continuously as the grain size reduces from \( L = 16 \text{ nm} \) to \( L = 9.5 \text{ nm} \); the upper bound of the RUD ACR increases from \( \epsilon_{\text{co}} = 2.5 \times 10^{-3} \) with \( T_C = 285.481 \text{ K} \) for \( L = 16 \text{ nm} \) to \( \epsilon_{\text{co}} = 8.6 \times 10^{-3} \) with \( T_C = 252.335 \text{ K} \) for \( L = 9.5 \text{ nm} \).

Figure 2(c) displays \( \gamma_{\text{eff}}(\epsilon) \) and \( \gamma(\epsilon) \) that the ROF analysis, based on the SPL and the RUD (equation (2)) expressions, yields for the nanocrystalline Gd sample with \( L = 9.5 \text{ nm} \), when \( T_C \) is fixed at 252.335 K. Judging by the reduced sum of deviation squares, we find that the SPL does not describe \( \chi^{-1}(T) \) as accurately as the RUD expression (equation (2)) does in the ACR (figure 4, top panel). The middle and bottom panels of figure 4 show the results of the ROF analysis of the \( \chi^{-1}(T) \) data (the top panel), based on the SPL (\( \Gamma_{-1}^{\text{eff}} \)) and the RG RUD (\( \Gamma_{-1}^{\text{RUD}} \)) and the RG RUD (\( \Gamma_{-1}^{\text{RUD}} \)) expressions. Inclusion of the multiplicative logarithmic correction, besides the leading single power law, i.e. equation (2), vastly improves the robustness of the fitting parameters against the variation in the temperature range of the fit (ACR being the largest fit range); e.g. compare \( \Gamma_{-1}^{\text{RUD}} = 0.55(20) \text{ with } \Gamma_{-1}^{\text{RUD}} = 1.13(4) \text{ and } \gamma_{\text{eff}} = 1.035(35) \text{ with } \gamma = 1.0002(8) \). But for the change in the ACR width with \( L \), these results are representative of the samples with \( L = 12 \text{ and } 16 \text{ nm} \) as well. In stark contrast to the extremely narrow ACR in the \( L = 33.7 \text{ and } 25.6 \text{ nm} \) samples, the ACR widens at smaller grain sizes. A plausible explanation for this observation has already been provided.

Figure 3. Temperature variations of the effective, \( \gamma_{\text{eff}} \), and asymptotic, \( \gamma \), critical exponents for susceptibility (top panel) and those of the corresponding inverse critical amplitudes \( \Gamma_{-1}^{\text{eff}} \) and \( \Gamma_{-1}^{\text{RUD}} \) (bottom panel) for coarse-grained Gd (\( L = 100 \mu\text{m} \)) in the asymptotic critical regime (\( \epsilon \leq \epsilon_{\text{co}} \)).

Figure 4. Top panel: the best RUD fit (continuous curve) to the inverse intrinsic susceptibility, \( \chi^{-1}(T) \), of nanocrystalline Gd with \( L = 9.5 \text{ nm} \) in the ACR (\( \epsilon \leq \epsilon_{\text{co}} \)). Temperature variations of the effective and asymptotic critical exponents \( \gamma_{\text{eff}}(\epsilon) \) and \( \gamma(\epsilon) \) (middle panel), and of the corresponding inverse critical amplitudes \( \Gamma_{-1}^{\text{eff}}(\epsilon) \) and \( \Gamma_{-1}^{\text{RUD}}(\epsilon) \) (bottom panel) in the ACR.
To discuss our results, we treat the single-crystalline Gd metal as the reference state with regard to structure and the ferromagnetic-to-paramagnetic phase transition. The change of microstructure from single-crystalline to coarse-grained polycrystalline Gd with a grain size of 100 μm leads to a slight narrowing of the ACR from $\varepsilon_{\text{co}} = 2.05 \times 10^{-3}$ to $\varepsilon_{\text{co}} = 1.5 \times 10^{-3}$, but leaves the PUD asymptotic critical behaviour unaltered. Assuming that the spin–spin correlation length $\xi$ (the distance over which the order-parameter fluctuations are correlated) grows well beyond $L$ due to sufficiently strong spin coupling across the grain boundaries but does not reach the system size at $T = T_C$ (as contrasted with single crystal Gd, wherein $\xi$ diverges at $T_C$), the effective uniaxial anisotropy weakens due to the averaging over the random crystal orientations within the spin-correlated volume ($\sim L^3$) with the result that the ACR of the uniaxial dipolar ferromagnet shrinks. However, as $L$ approaches the nanometre range, the number of atoms (and hence spins) at the grain surface increases rapidly at the cost of the atoms within the core. Consequently, quenched random exchange disorder picks up in strength and, beyond a threshold, causes a crossover from the PUD to RUD asymptotic critical behaviour. In nanocrystalline Gd, this threshold is reached at $L \geq 34$ nm and the RUD behaviour in the ACR is found in the samples with $L \approx 9.5$ to 16 nm (figure 2(c) and 4). A continuous reduction in the grain size from the bulk to 9.5 nm or, equivalently, the increase in the degree of quenched random exchange disorder thus results in a gradual crossover from the pure uniaxial dipolar critical behaviour in single crystal Gd to that of a random uniaxial dipolar ferromagnet in the asymptotic critical region in nanocrystalline Gd with grain sizes $\lesssim 35$ nm.

At this stage, it is interesting to note that traditionally the crystalline Gd$_{1-x}$Nd$_x$Cl$_3$ and LiTb$_{1-x}$Y$_x$F$_4$ systems have been regarded as random uniaxial dipolar ferromagnets with dipolar interactions [37, 38] but low Curie temperatures have precluded the observation of RUD asymptotic critical behaviour in them. Thus, nanocrystalline Gd with $L \leq 16$ nm happens to be the first experimental realization of the random uniaxial dipolar critical point behaviour.

Finally, the present results on the static critical phenomena in nanocrystalline Gd are compared with those reported [46, 47] previously for nanocrystalline-(intergran amorphous) Gd and Gd$_{0.36}$Fe$_{0.054}$ composite systems with average grain sizes of 24(3) nm and 68 nm, respectively. The most refined values quoted for the effective critical exponent for susceptibility, obtained from the Arrrott–Noakes scaling equation of state analysis of bulk magnetization, within the reduced temperature ranges $1.0 \times 10^{-3} \leq \varepsilon \leq 1.5 \times 10^{-2}$ and $8.9 \times 10^{-4} \leq \varepsilon \leq 1.5 \times 10^{-3}$ were $\gamma_{\text{eff}} = 1.300(14)$ and $\gamma_{\text{eff}} = 1.24(3)$ for the 24 and 68 nm nanocomposite samples. The estimate $\gamma_{\text{eff}} = 1.300(14)$ for the 24 nm sample [46] compares favourably with the average value $\gamma_{\text{eff}} = 1.30(5)$ determined in this work in the temperature range $7.4 \times 10^{-4} \leq \varepsilon \leq 3.5 \times 10^{-3}$ outside the ACR for $L = 25.6$ (figure 2(b)). The present work scores over the preceding ones in that true asymptotic critical exponents for susceptibility, determined for all the nanocrystalline Gd samples, except for those with $L = 25.6$ and 33.7 nm, enable fixing the universality class to which the systems in question belong.

4. Summary

In summary, an elaborate analysis of the intrinsic magnetic susceptibility reveals that, like in single crystal Gd, the asymptotic critical behaviour of polycrystalline coarse-grained Gd (grain size: $\approx 100$ μm) in the critical region near the ferromagnetic-to-paramagnetic phase transition is that of a (pure) uniaxial dipolar ferromagnet. For nanocrystalline Gd with $L \sim 34$ nm, asymptotic critical behaviour could not be discerned, which we believe is due to a crossover to the random uniaxial dipolar fixed point with an extremely narrow critical region. At grain sizes $\lesssim 16$ nm, nanocrystalline Gd behaves as a random uniaxial dipolar ferromagnet in the asymptotic critical region and hence it belongs to the random uniaxial dipolar universality class.

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