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2009 J. Phys.: Condens. Matter 21 216004

(http://iopscience.iop.org/0953-8984/21/21/216004)

J. Phys.: Condens. Matter **21** (2009) 216004 (9pp) doi:10.1088/0953-8984/21/21/216004

Observation of a Griffiths-like phase in the paramagnetic regime of $ErCo_2$

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Received 31 December 2008, in final form 6 April 2009
Published 29 April 2009
Online at stacks.iop.org/JPhysCM/21/216004

Abstract

A systematic x-ray magnetic circular dichroism study of the paramagnetic phase of $ErCo_2$ has recently allowed us to identify the inversion of the net magnetization of the Co net moment with respect to the applied field well above the ferrimagnetic ordering temperature, T_c. The study of small-angle neutron scattering measurements has also shown the presence of short range order correlations in the same temperature region. This phenomenon, which we have denoted *parimagnetism*, may be related to the onset of a Griffiths-like phase in paramagnetic $ErCo_2$. We have measured ac susceptibility on $ErCo_2$ as a function of temperature, applied field and excitation frequency. Several characteristics shared by systems showing a Griffiths phase are present in $ErCo_2$, namely the formation of ferromagnetic clusters in the disordered phase, the loss of analyticity of the magnetic susceptibility and its extreme sensitivity to an applied magnetic field. The paramagnetic susceptibility allows us to establish that the magnetic clusters are only formed by Co moments as well as the intrinsic nature of those Co moments.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The study of Griffiths phases [1] in intermetallic compounds has created much interest in recent years. The extended Griffiths model [2, 3] accounts for systems in which the magnetic correlations do not vanish completely at the first-order phase transition, but are present at higher temperatures. The Griffiths-like phase is then the region between the completely ordered state and the conventional disordered paramagnetic state. Experimentally a Griffiths-like phase has usually been identified as the paramagnetic susceptibility shows deviations from a Curie–Weiss law and extreme sensitivity to applied magnetic fields [4–9]. An increasing number of very interesting systems are being reported to show that kind of behavior. Among those, compounds that show a giant magneto-caloric effect like $Tb_5Si_{2+x}Ge_{2-x}$ [8, 10], colossal magneto-resistance like $La_{1-x}Sr_xMnO_3$ [11], $La_{1-x}Ca_xMnO_3$ [12, 13], $La_{1-x}Ba_xMnO_3$ [14], $Sm_{1-x}Ca_xMnO_3$ [15] or $La_{0.7-x}Dy_xCa_{0.3}MnO_3$ [16], strongly correlated electron systems like $CeNi_{1-x}Cu_x$ [17] or itinerant magnetic semiconductors like $Fe_{1-x}Co_xS_2$ [18] are remarkable examples. In those systems, the presence of a Griffiths-like phase is related to the existence of short range order magnetic correlations, which is shown from the enhancement of the magnetic signal in small-angle neutron scattering (SANS) measurements. It is very interesting therefore to revisit well-studied materials in which there is experimental evidence of important short range correlations well above T_c in order to determine whether or not they also show a Griffiths-like phase behavior. What is more, a general question should be open: are Griffiths-like phases a common phenomena in a wide range of intermetallic compounds and what role do they play in the magnetic properties of those compounds? In that sense, we present in this paper a study of the paramagnetic ac susceptibility of the intermetallic compound $ErCo_2$ in the search for the fingerprint of Griffiths-like phases.

Among the intermetallic compounds, the family of RCo_2 is particularly interesting as they have a fairly simple structure, which facilitates the understanding of their magnetic

properties. In fact, they have been widely studied for decades as a model system of the magnetism of itinerant electron systems and metamagnetic processes. However, it was not until recently that short range order correlations were shown to exist well within the paramagnetic phase of one of the members of the series. A SANS study in the compound $ErCo_2$ showed the presence of magnetic clusters of around 8 Å at temperatures as high as twice the ordering temperature when a magnetic field of 1 T is applied [19]. Indeed, this behavior is related to the existence of the new magnetic phase reported in [20], where a new magnetic phase diagram for $ErCo_2$ was proposed. We have denoted that new magnetic phase as the *parimagnetic* phase as—although no long range order exist in the compound—the Co moments are found to be, on average, oriented antiparallel to the Er moments.

The molecular field created by Er moments on the Co sublattice in $ErCo_2$ is just above the critical value to induce the metamagnetic transition in the Co moments. Therefore, the Co sublattice is especially sensitive to variations of the external parameters. Below T_c, the magnetic behavior is dominated by the Er sublattice, with an essentially temperature-independent magnetic moment of around 8.8 μ_B/atom, while the Co sublattice is ordered antiparallel to the Er one through a first-order magnetostructural transition [21, 22]. In the paramagnetic phase, the nature of the cobalt magnetic moment is an especially interesting open question. A recent work by Liu and Altounian [23] predicted theoretically a transition from a low-spin state for Co (∼0.1 μ_B) to the well-known high-spin state of the ferrimagnetic phase (∼1 μ_B) at the onset of the ordering temperature of $ErCo_2$. Moreover, whether or not an intrinsic magnetic moment in the Co sublattice exists has been a continuous matter of debate since the first susceptibility studies were carried out in the RCo_2 series [24, 25]. The present study of the paramagnetic ac susceptibility in $ErCo_2$ will also deal with the existence and nature of the Co moment in the paramagnetic phase.

This paper is organized as follows; we describe the synthesis and characterization of the samples in section 2. The experimental results and discussion will be presented in section 3: the ac susceptibility measurements as a function of temperature and as a function of the excitation frequency will be shown in sections 3.1 and 3.2, respectively. The determination of the Co magnetic moment from those data will be presented in section 3.3. Finally, in section 4, we summarize the main results obtained.

2. Sample characterization

The $ErCo_2$ samples are polycrystalline ingots and were prepared by melting the pure elements in an induction furnace under Ar atmosphere. The resulting ingots were further annealed under Ar atmosphere at 850 °C for a week, wrapped in tantalum foil. Several ingots were synthesized in order to check the reproducibility of the phenomena studied. X-ray diffraction analysis at room temperature was performed on powdered samples to check their quality. Rietveld analysis of the diffractograms was performed using FullProf software [26, 27], ensuring single-phase samples with good crystallization and the expected cubic $Fd\bar{3}m$. No impurities were found within the 1% accuracy of powder diffraction methods. Neutron diffraction measurements were also performed at low temperatures, confirming the ferrimagnetic coupling of Er and Co with magnetic moments for Er and Co of 8.84 ± 0.06 μ_B and 0.95 ± 0.03 μ_B, respectively, and the space group $R\bar{3}M$. The room temperature x-ray and the low temperature neutron diffractograms, together with their Rietveld refinements, can be found respectively in [28] and [20].

A complete magnetic characterization has been performed on the $ErCo_2$ samples. Magnetization measurements, $M(T, H)$, from 10 to 300 K and up to 5 T were performed in a SQUID Quantum Design magnetometer and up to 9 T in a commercial Quantum Design extraction magnetometer. Selected $M(H)$ and $M(T)$ curves are shown in figure 1, where the abrupt change in magnetization at the magnetostructural transition (e.g. $T_c = 34$ K at $H = 1$ T) can be observed. The data are fully consistent with those previously reported [29–37].

We have measured ac magnetic susceptibility as a function of temperature ($\chi_{ac}(T)$) and as a function of excitation frequency ω ($\chi_{ac}(\omega)$) in a variety of polycrystalline $ErCo_2$ samples. The measurements were performed in a commercial SQUID Quantum Design magnetometer under zero dc applied magnetic field, from 5 to 300 K, with an excitation ac field of 4.5 Oe and at ω ranging from 0.1 to 1000 Hz. We have also measured $\chi_{ac}(T)$ under applied magnetic fields up to 2500 Oe in a single crystal, courtesy of Professor Ernst Bauer from Vienna University of Technology. The ac susceptibility measurements will be presented along with this paper.

3. Experimental results and discussion

3.1. Susceptibility as a function of temperature

The real (χ'_{ac}) and imaginary (χ''_{ac}) contributions to the $ErCo_2$ ac susceptibility as a function of temperature under zero dc applied magnetic field are shown in figure 2. A sharp peak can be observed at the ordering temperature (32 K) both in the real and imaginary parts, followed by an abrupt drop of the signal above T_c. This drop corresponds to the expected Curie–Weiss dependence of χ'_{ac} in the paramagnetic phase:

$$\chi'_{ac}(T) = \frac{C}{T-\theta} \qquad \text{where } C = \frac{N\mu_B^2}{3k_B}\mu_{eff}^2. \tag{1}$$

However, together with that contribution to the paramagnetic ac susceptibility, a small anomaly near $T_f = 90$ K can be observed. This anomaly is more evident both in the inverse of χ'_{ac} and in χ''_{ac}, where a peak 20 K wide is present in the proximity of $T_f = 90$ K. If the region of the anomaly is excluded, the inverse of χ'_{ac} has a linear dependence with temperature and therefore can be fitted to a Curie–Weiss law using $\mu_{eff} = 8.8$ μ_B, the value of the atomic Er magnetic moment. The fit, shown in the inset in the upper panel in figure 2, is valid from high temperatures (∼300 K) down to 30 K above the magnetostructural transition and indicates that the Curie–Weiss contribution is due to independent Er

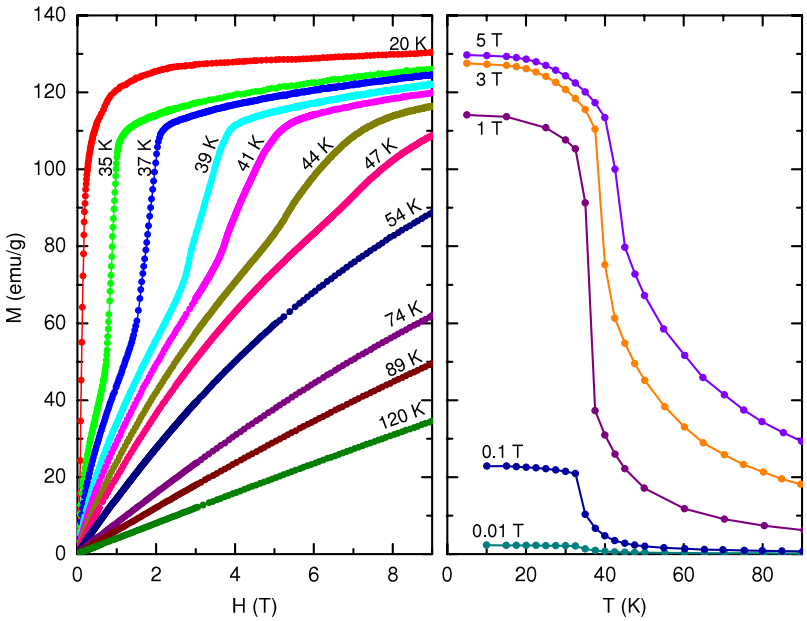

Figure 1. Selected $ErCo_2$ isothermal (left) and isofield (right) magnetization curves measured in SQUID and PPMS magnetometers.

moments. Accordingly, the difference of χ'_{ac} and the Er contribution (χ_{Er}) is the susceptibility due to the Co sublattice (χ_{Co}). The detachment of the different contributions to the paramagnetic susceptibility can be found in [20].

The existence of a Co contribution to the ac susceptibility, independent from that of Er, proves that the Co atoms have an intrinsic moment in the paramagnetic phase independent of the Er sublattice, in good agreement with the fact that temperature-dependent XMCD at the Co $L_{2,3}$ edges shows the presence of Co magnetic moment at temperatures above T_c [45].

Similar deviations of $1/\chi'_{ac}$ from a linear dependence with the temperature well above the ordering transition has been found in other intermetallic compounds like $CeNi_{1-x}Cu_x$ [17, 38, 39], $La_{1-x}Sr_xMnO_3$ [11], $Tb_5Si_2Ge_2$ [8], $Fe_xCo_{1-x}S_2$ [18] $La_{0.73}Ba_{0.27}MnO_3$ [5, 7], $Nd_{0.55}Sr_{0.45}Mn_{1-x}Ga_xO_3$ [9] and Gd_5Ge_4 [6] and have been related to the occurrence of a Griffiths-like phase well above their ordering temperatures. The appearance of a Griffiths phase is usually associated with the existence of disorder and competing interactions, which leads to percolative phenomena and clustering. Indeed, SANS experiments carried out in some of those compounds have demonstrated the occurrence of short range order correlations [8, 16, 17].

In the paramagnetic phase of $ErCo_2$, the existence of short range order correlations has been evidenced by a previous SANS study [19, 20]. Those SANS measurements revealed the existence of magnetic clusters in a wide temperature range well above T_c. The fits of the SANS data to Lorentzian and Lorentzian-squared functions—corresponding respectively to spin waves and static regions of spin ordering—show that in $ErCo_2$ the only significant contribution is Lorentzian, i.e. the short range correlations in its paramagnetic phase are due to spin waves. The correlation length, ξ, obtained from those fits experiences a continuous increase from high temperatures (>200 K) down to ≈60 K (at 1 T), where it reaches a plateau at an almost constant value of 7–8 Å (at lower temperatures, just above T_c, ξ diverges due to the establishment of long range order). The existence of such a plateau in ξ implies that the percolation does not occur due to the growing of clusters but by the increasing of the density of clusters with sizes around 7–8 Å. This process differs from the standard normal percolation process and has been predicted and observed for Griffiths-like phases [2–4]. Therefore, from what has been shown above, it is reasonable to establish that both the deviation from a Curie–Weiss law and the existence of clusters in the paramagnetic phase of $ErCo_2$ come from the presence of a Griffiths-like phase.

The characteristic features of the ac susceptibility in a Griffiths-like phase are usually suppressed under small magnetic fields, an effect that has been referred to as *extreme sensitivity* [5]. That is indeed the case in polycrystalline $ErCo_2$, where the observed peak in χ'_{ac} near $T_f = 90$ K at $H = 0$ T disappears when a magnetic field is applied. In the $ErCo_2$ single crystal, the same anomaly near $T_f = 90$ K can also be observed under moderately small applied magnetic fields. Figure 3 shows the real (upper panel) and the imaginary part (lower panel) of the susceptibility at zero applied magnetic field (left) and under applied magnetic fields up to 2500 Oe (right). A peak at T_c can be observed, although it is not as sharp as in the polycrystalline samples, probably due to the twinning originated in the high pressures this sample suffered in the past, as we confirmed by x-ray Laue diffraction experiments. Nevertheless, the anomaly at T_f is clearly visible both in the real and in the imaginary part, and its evolution with the applied magnetic field can be followed: the intensity of the anomaly decreases as soon as a few Oe are applied, at the same time as it moves to lower temperatures. At applied magnetic fields higher than 1000 Oe, the anomaly disappears and the linear dependence of $1/\chi'_{ac}$ with temperature is recovered. This evolution of $\chi_{ac}(T)$ with the applied magnetic field

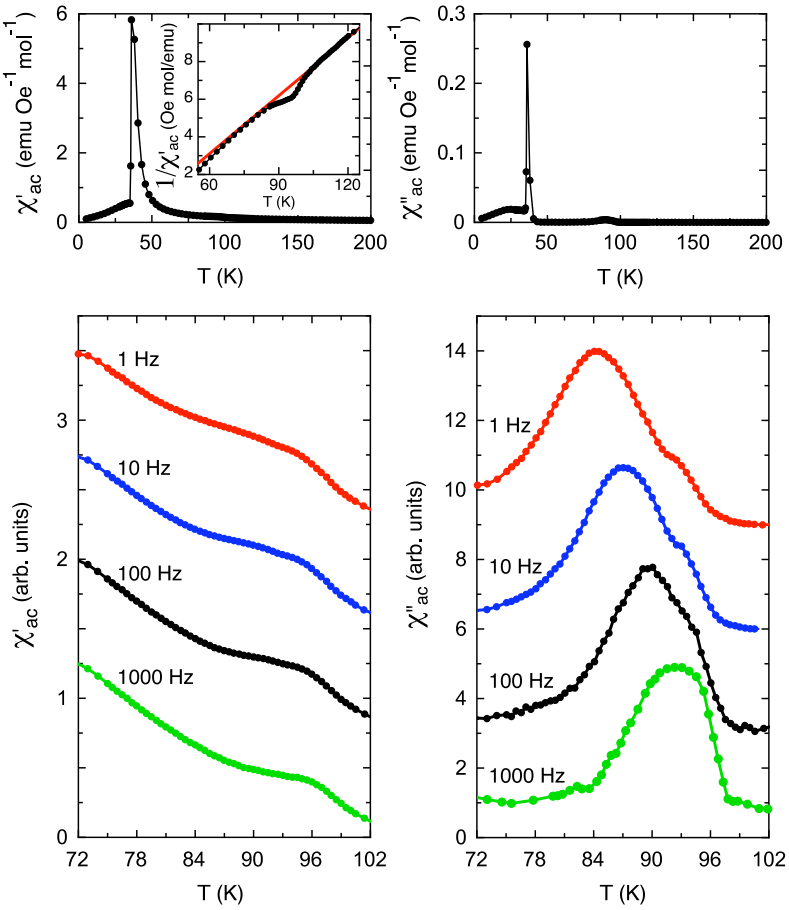

Figure 2. $ErCo_2$ paramagnetic ac susceptibility as a function of temperature at $H = 0$ T. Upper panels: real (χ'_{ac}) and imaginary (χ''_{ac}) components of the ac susceptibility at $\omega = 100$ Hz are shown on the left and right, respectively. Lower panels: real and imaginary components of the ac susceptibility at $\omega = 1$, 10, 100 and 1000 Hz in the vicinity of T_f. In this case the signals have been vertically displaced for clarity. Inset in upper left panel: inverse of χ'_{ac} (full dots) and its fit to a Curie–Weiss law (continuous line) in the paramagnetic region (see text).

again supports the existence of a Griffiths-like phase above the ferrimagnetic order in $ErCo_2$.

3.2. Frequency dependence of the paramagnetic susceptibility

To further investigate the fingerprint of this Griffiths-like phase in the paramagnetic susceptibility, we have also studied the dependence of the ac susceptibility with the excitation frequency ω. Figure 2 shows $\chi_{ac}(T)$ measurements in the vicinity of T_f for $\omega = 1$, 10, 100 and 1000 Hz. The anomaly already shown for 100 Hz in figure 2 is present for all the frequencies, but a shift to higher temperatures (of 9 K when going from 1 to 1000 Hz) can be observed as the frequency is increased both in the real and imaginary parts of the susceptibility. This fact confirms that the origin of the anomaly at T_f is not in the magnetic ordering of any impurity which may be present in the sample occurring at that temperature. Excluding the temperature region where the anomaly is present, no other frequency dependence can be observed, as the Er contribution to the paramagnetic susceptibility is ω-independent for that frequency range. The frequency dependence of the Co contribution, together with the recovery of the Curie–Weiss dependence below T_f (i.e. the Co contribution going to zero below T_f), allows us to identify the origin of the anomaly as a relaxation process in the Co sublattice in the proximities of T_f [40].

Therefore, the next step is to study the dynamic response of this Co contribution to the paramagnetic susceptibility. To do so, we have performed ac susceptibility measurements as a function of the excitation frequency ($\chi_{ac}(\omega)$) at zero applied magnetic field, excitation frequencies between 0.05 and 1500 Hz and in the temperature region between 80 and 100 K.

Owing to the fact that Er moments do not suffer major changes in that temperature range, the Er contribution to the susceptibility can be considered as constant at fixed temperatures. This offers an advantage with respect to the study presented in the previous subsection, as it facilitates the identification of the Co contribution. i.e. the Co contribution is the only contribution varying as a function of ω in an isothermal measurement.

The evolution with temperature of the real and imaginary parts of $\chi_{ac}(\omega)$ can be observed in figure 4. As expected, the real part decreases as the frequency is increased. A small shoulder is present in the vicinity of T_f, which moves to higher frequencies as the temperature is raised (in agreement with the shift observed in figure 2). In the imaginary contribution to

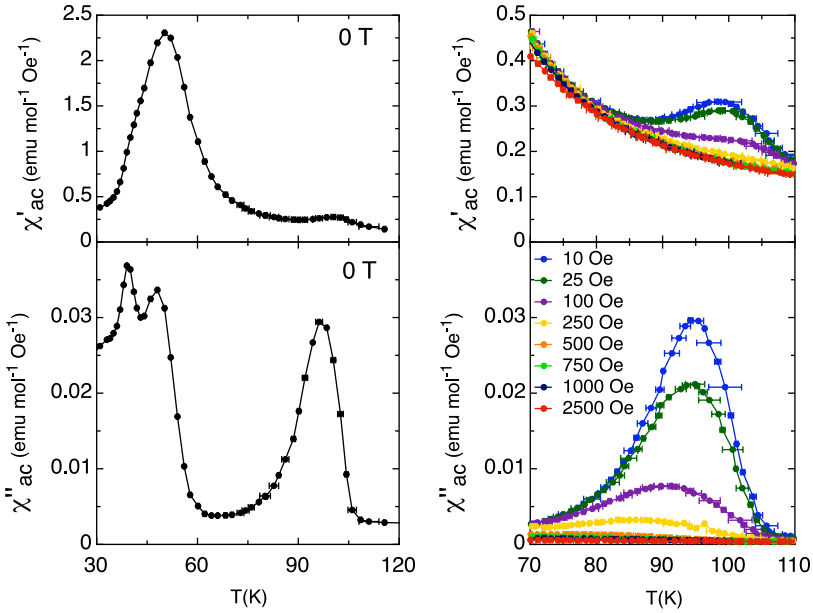

Figure 3. AC susceptibility as a function of temperature in an $ErCo_2$ single crystal. Left graph: real (upper panel) and imaginary (lower panel) parts at $H = 0$ T. Right graph: evolution of the real (upper panel) and imaginary (lower panel) parts in the paramagnetic region at selected applied magnetic fields up to 2500 Oe.

$\boldsymbol{\chi}_{ac}(\omega)$, two maxima can be observed, which also move to higher frequencies as the temperature is raised.

Figure 5 shows isothermal χ''_{ac} versus χ'_{ac} curves for selected temperatures. This kind of representation—where the excitation frequency is an implicit parameter—is the so-called Cole–Cole representation. For a system of independent spins undergoing a relaxation process (i.e. a Debye process with only one relaxation time) χ''_{ac} versus χ'_{ac} is a perfect semicircle, represented in figure 5 by a discontinuous line (note that the horizontal and vertical axes have different scales, and therefore the circle appears to be an ellipse). This representation allows us to determine, just by mere visual inspection, deviations in the susceptibility from a Debye process: the Cole–Cole representation for $ErCo_2$ shows stretched and asymmetric curves and, in the frequency range studied, two maxima can be observed. This shows that the observed anomaly in $\boldsymbol{\chi}_{ac}(\omega)$ and $\boldsymbol{\chi}_{ac}(T)$ is not due to a single relaxation time but to a bimodal distribution of relaxation times.

The ac susceptibility in a Debye process with only one relaxation time can usually be fitted to a Debye law [41]:

$$\boldsymbol{\chi} = \chi_0 + \frac{\chi_{\mathrm{eq}} - \chi_0}{1 - \mathrm{i}\omega\tau} \tag{2}$$

where τ is the relaxation time, χ_0 is the value at infinite ω and χ_{eq} is the value at $\omega = 0$, which follows a Curie–Weiss law as a function of temperature. However, in systems with broad distributions of relaxation times, as in $ErCo_2$, $\boldsymbol{\chi}(\omega)$ does not obey equation (2) and there are numerous empirical approximations to parameterize the ac susceptibility. Many of those approximations are modifications of the Debye law. In particular, the Davidson–Cole model [42, 43] introduces the factor γ ($0 < \gamma \leqslant 1$), which accounts for asymmetry and a smoother dependence of the susceptibility with $\omega\tau$:

$$\boldsymbol{\chi} = \chi' + \mathrm{i}\chi'' = \chi_0 + \frac{\chi_{\mathrm{eq}} - \chi_0}{(1 - \mathrm{i}\omega\tau)^{\gamma}}. \tag{3}$$

Figure 6 shows the fit of the measured ac susceptibility at 90 K to the sum of two Davidson–Cole functions, i.e.

$$\chi' = \chi_0 + \mathrm{Re}\left(\frac{\chi_{\mathrm{A}}}{(1 - \mathrm{i}\omega\tau_{\mathrm{A}})^{\gamma}} + \frac{\chi_{\mathrm{B}}}{(1 - \mathrm{i}\omega\tau_{\mathrm{B}})^{\gamma}}\right)$$
$$\chi'' = \mathrm{Im}\left(\frac{\chi_{\mathrm{A}}}{(1 - \mathrm{i}\omega\tau_{\mathrm{A}})^{\gamma}} + \frac{\chi_{\mathrm{B}}}{(1 - \mathrm{i}\omega\tau_{\mathrm{B}})^{\gamma}}\right) \tag{4}$$

where χ_{A} and χ_{B} are the values of the jump in χ' for each relaxation process at τ_{A} and τ_{B}. The same value of $\gamma = 0.1$ for both χ'' maxima has been used, giving an idea of how broad the distribution is ($\gamma = 1$ is the Debye law limit).

In systems with broad distributions of relaxation times, the relaxation time frequently follows a Vogel–Fulcher law [44]:

$$\tau = \tau_0 \exp\left(\frac{Q}{k_{\mathrm{B}}(T - T_{\mathrm{o}})}\right) \tag{5}$$

where T_{o} is the so-called Vogel–Fulcher temperature and corresponds to the temperature at which the relaxation process occurs at zero frequency. From the χ''_{ac} maxima at $\omega\tau = 1$, the relaxation time as a function of the temperature can be obtained and from the fit of $\tau(T)$ to equation (5) $T_{\mathrm{o}} = 70$ K is obtained, which is a reasonable temperature for the zero frequency relaxation process to occur.

3.3. Determination of the Co paramagnetic moment from susceptibility measurements

As mentioned earlier, χ_{eq} follows a Curie–Weiss law with temperature and, using equation (1), the value of the effective

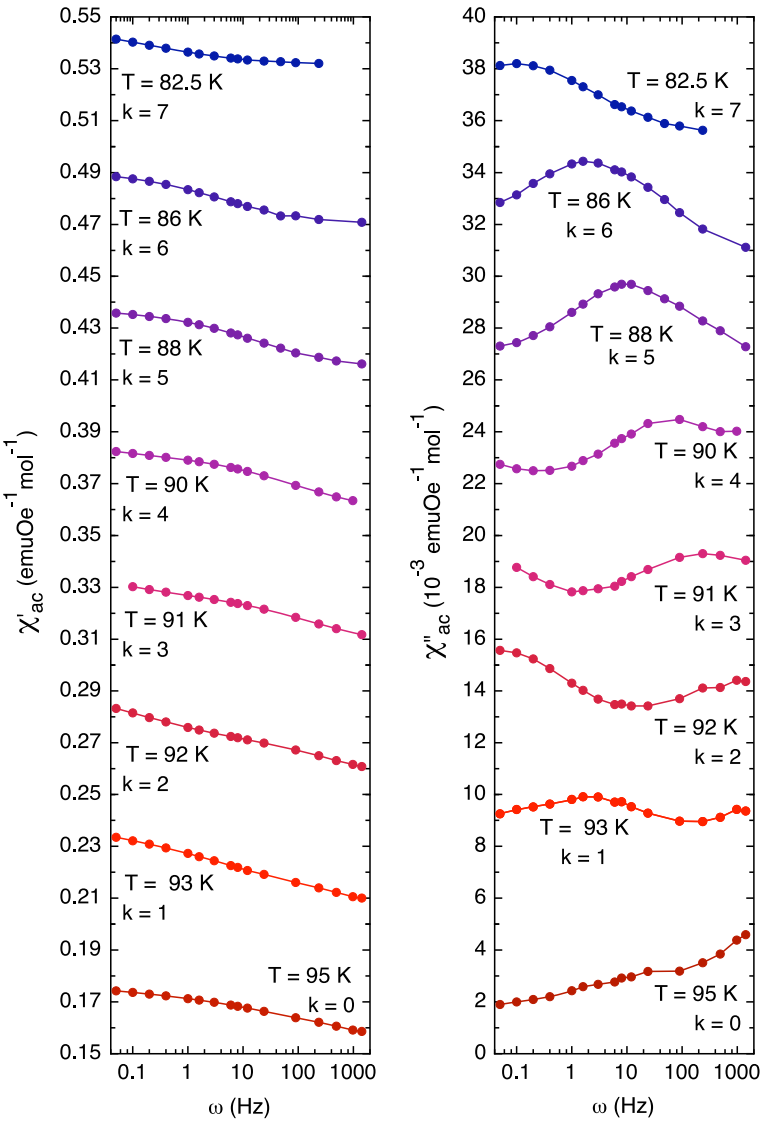

Figure 4. χ'_{ac} (left) and χ''_{ac} (right) as a function of the excitation frequency at selected temperatures between 82.5 and 95 K. The signals have been vertically displaced $k \times 0.05$ for the real part and $k \times 0.005$ for the imaginary part, where k is indicated in each curve below the corresponding temperature.

moment contributing the susceptibility can be calculated:

$$\mu_{\text{eff}} = \left(\chi_{\text{eq}}(T - T_{\text{o}})\frac{3k_{\text{B}}}{N\mu_{\text{B}}^2}\right)^{1/2} \tag{6}$$

where in this case N is the number of dynamic entities (i.e. number of clusters), T_{o} is the temperature calculated from equation (5) and χ_{eq} can be obtained from both the fits of $\chi_{ac}(\omega)$ to Davidson–Cole functions and from the $\chi_{ac}(T)$ measurements. The obtained value in both cases, $\mu_{\text{eff}} \sim 20\ \mu_{\text{B}}$, is greater than any individual moment present in the system and thus can only be explained as coming from the relaxation process of a group of moments or cluster. Indeed, this is in good agreement with the reported cluster sizes of 7–8 Å at T_{f} from SANS experiments, i.e. around 100 Co atoms and/or 50 Er atoms in the cluster [20]. However, the obtained effective moment is only compatible with the SANS results if the clusters exclusively consist of Co low-spin moments. Therefore, if c is the number of Co atoms in the clusters, the moment of the Co atoms in the paramagnetic phase of $ErCo_2$ is $m_{\text{Co}} = \mu_{\text{eff}}/c$. The results are presented in table 1, together with the value of the Co moment obtained from an independent set of x-ray magnetic circular dichroism (XMCD) experiments presented in [45].

Table 1. Co moment in paramagnetic phase calculated from susceptibility measurements in this work and from XMCD data in [45].

Experimental data	m_{Co} (μ_{B})
$\chi(\omega)$	0.24 ± 0.03
$\chi(T)$	0.22 ± 0.03
XMCD [45]	0.19 ± 0.02

The existence of magnetic moment in the Co sublattice in the paramagnetic phase of the RCo_2 compounds, as well as the intrinsic or induced nature of the moment, has long been

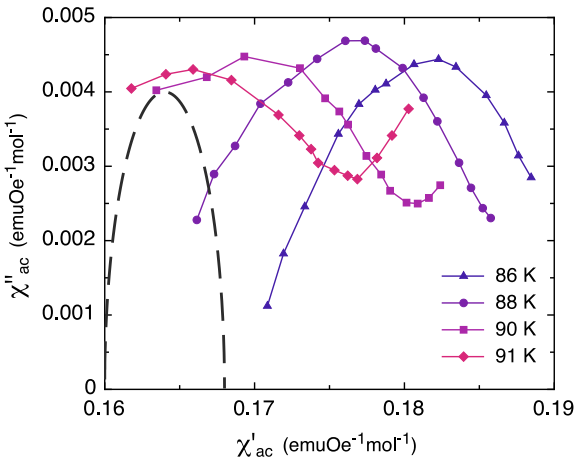

Figure 5. Cole–Cole representation (i.e. χ'_{ac} versus χ''_{ac}) of the paramagnetic ac susceptibility in $ErCo_2$ at selected temperatures (full symbols). The dashed line is the curve corresponding to a theoretical Debye process with only one relaxation time.

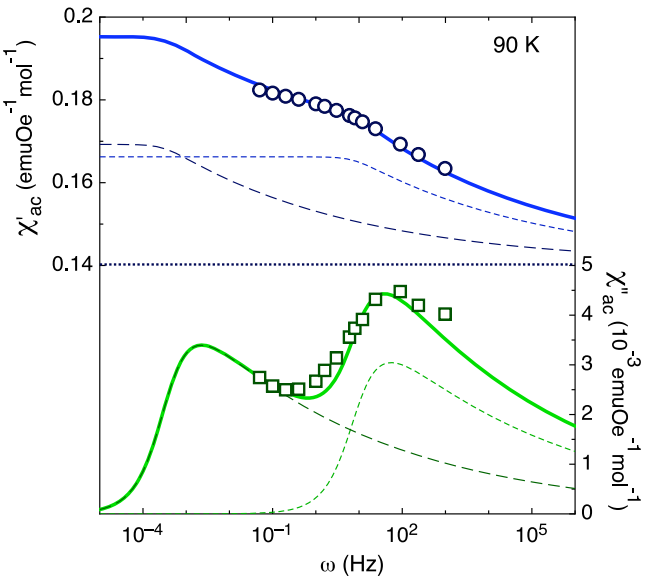

Figure 6. χ'_{ac} (open circles) and χ''_{ac} (open squares) as a function of ω at 90 K in $ErCo_2$. The continuous line is the fit of the data to a sum of two Davidson–Cole functions, the dashed lines are the two functions separately and the dotted line is the adiabatic value of the susceptibility, χ_0, at 90 K.

studied in the literature [23, 24, 29, 46, 47]. The $\chi_{ac}(T)$ and $\chi_{ac}(\omega)$ measurements presented in this work demonstrate its existence and quantifies the value of the Co magnetic moment in the paramagnetic phase of $ErCo_2$. What is more, the presence of an intrinsic Co component is needed to explain the relaxation process observed near T_f. These results are also in agreement with the results from XMCD experiments [45] as well as with recent first-principles calculations carried out on $ErCo_2$, which demonstrate the existence of a small but yet significant Co moment at high temperatures [48]. Moreover, the existence of a low-spin state for Co at the onset of the ordering temperature of $ErCo_2$ was also predicted theoretically by Liu and Altounian [23]. Our results show that the low-spin state is stable up to temperatures well above the ordering temperature T_c.

The existence of Co intrinsic moment in the paramagnetic phase of $ErCo_2$ does not agree with the calorimetric study by Imai *et al* [31], where the Co moment shows little influence on the magnetic entropy. However, the existence of a Griffiths-like phase implies that the Co entropy should be released at temperatures much higher than the ordering temperature. On the other hand, the entropy as a function of temperature also discards Er moments taking part in the formation of the clusters. More detailed calorimetric studies in a larger temperature range should be performed on $ErCo_2$, and adequate references like $ErAl_2$ and YCo_2 should be measured to identify the influence of the formation of the Co clusters on the magnetic entropy at high temperatures.

4. Conclusions

Magnetic ac susceptibility measurements as a function of temperature and excitation frequency have been performed in order to study the dynamic properties of the $ErCo_2$ paramagnetic phase. The $\chi_{ac}(T)$ measurements show the expected Curie–Weiss dependence coming from the Er independent moments. However, a deviation from that Curie–Weiss law occurs in a narrow temperature region. We have studied the dependence of that anomaly with the temperature, the applied magnetic field and the excitation frequency, showing that a relaxation process occurs in the Co sublattice. The $\chi_{ac}(T)$ and $\chi_{ac}(\omega)$ measurements also confirm that the net Co magnetic moment in the $ErCo_2$ paramagnetic phase is not zero and that an intrinsic component is needed to explain the relaxation process.

From the analysis of the ac susceptibility measurement, together with the already reported existence of magnetic clusters in the paramagnetic phase of $ErCo_2$, it follows that a Griffiths-like phase is formed at temperatures well above the ordering temperature of $ErCo_2$, leading to a handful of new interesting phenomena in this compound. In particular, the presence of a Griffiths-like phase is related to two new phenomena observed in $ErCo_2$. On the one hand, the existence of a Griffiths-like phase is related to the anomaly in the paramagnetic ac susceptibility, which is due to a relaxation process of Co clusters. On the other hand, a previous work showed that the reversal of the Co magnetization occurs at a much higher temperature than the ferrimagnetic ordering and that the phenomena can only be observed if there exist Co clusters [20]. The new phenomenology observed in $ErCo_2$ is due to a competition of interactions, in which the strong Co–Co exchange interaction plays a very important role, being responsible for the existence of short range order correlations in the paramagnetic phase of $ErCo_2$.

The generalized concept of Griffiths-like phases involves an arbitrary distribution of exchange constants, which can be achieved when there are competing interactions in the system together with a source of randomness. However, in $ErCo_2$, the competing interactions acting in the Co sublattice are not accompanied by a disorder in the lattice. There are two possible sources of disorder that could percolate the

formation of the Co clusters, and therefore the formation of the Griffiths-like phase. On the one hand, the appearance of small regions slightly off-stoichiometric is inherent to the synthesis process of the RCo_2 compounds (see the discussion in [28] and references therein). A slighter richer region in Co—not detectable by x-ray diffraction—could be the seed to nucleate the clusters. Although this conjecture cannot be ruled out, one would expect that, if that were the case, then the phenomena would be sample-dependent and radically different in a single crystal. On the other hand, it is well known that spin fluctuations play a very important role in the Co Laves phases. This allows us to postulate that the source of random interactions in $ErCo_2$ comes from those spin fluctuations. Indeed, the fits of the SANS measurements to a Lorentzian function rather than to a Lorentzian-squared function indicate that the Co clusters in $ErCo_2$ are not inhomogeneous regions of spin ordering but dynamical clusters. Moreover, it is well known that the spin fluctuations are quenched when a magnetic field is applied [49, 50], in agreement with the already mentioned extreme sensibility of the Griffiths-like phase to the applied magnetic field.

However, although the occurrence of a Griffiths-like phase in $ErCo_2$ has some distinct characteristics, the scenario is very similar to what has been reported in other intermetallic compounds. Indeed, there is an increasing number of compounds in which the characteristic fingerprint of Griffiths-like phases is now being reported, such as manganites, rare-earth alloys, heavy-fermion systems, itinerant magnetic semiconductors and now the case of $ErCo_2$. It is therefore very likely that, if the paramagnetic phase of other well-studied intermetallic compounds are revisited, several of them would show Griffiths-like behavior. The characterization of those compounds would surely help in the understanding of both the magnetic properties of those particular systems and of the paramagnetic phase of intermetallic compounds in general.

Acknowledgments

We acknowledge the Spanish CICYT research project MAT 2008/01077/NAN, the FEDER program and the Aragonése DECRYPT and CAMRADS research groups. We thank F Luis and V Pecharsky for fruitful discussion, N Plugaru and M J Pastor for sample preparation and Professor E Bauer for the single crystal. JH acknowledges Fundación Ramón Areces for financial support.

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