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# Magnetic and thermal properties of $\text{Er}_{75}\text{Dy}_{25}$ single crystal

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We report on an experimental study of the thermal and magnetic properties of  $\text{Er}_{75}\text{Dy}_{25}$  single crystals with magnetic fields applied parallel to the  $[0001]$  and  $[10\bar{1}0]$  directions. The  $[0001]$  direction is the easy axis of magnetization while the  $[10\bar{1}0]$  direction is the hard axis of magnetization. Three major transitions are observed in the heat capacity, magnetization, and ac magnetic susceptibility data. A first order transition is observed at  $\sim 30$  K, and two second order transitions at  $\sim 45$  K and  $\sim 110$  K, respectively. The  $H$ - $T$  phase diagrams constructed from the experimental data reveal that several magnetic phases, including ferromagnetic cone, ferromagnetic fan, and  $c$  axis modulated phases, exist in  $\text{Er}_{75}\text{Dy}_{25}$  single crystal. Both similarities and dissimilarities are observed in the  $H$ - $T$  phase diagrams of  $\text{Er}_{75}\text{Dy}_{25}$  when compared to the  $H$ - $T$  phase diagrams of pure Er single crystal.

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## INTRODUCTION

The lanthanide metals erbium (Er) and dysprosium (Dy) exhibit complex yet fascinating thermal and magnetic properties.<sup>1,2,3,4,5,6,7</sup> Both metals adopt hexagonal closed packed (hcp) crystal structure at room temperature,<sup>6,8,9</sup> and upon cooling down from room temperature, they both undergo a series of phase transitions where either or both magnetic and crystallographic sublattices are affected.<sup>8</sup>

Earlier investigations of Er performed using neutron and x-ray diffraction<sup>1,10,11</sup> show that an antiferromagnetic (AFM) transition occurs at  $T_{N\parallel} \cong 84$  K (magnetization is parallel to the  $c$  axis), where paramagnetic (PM) compound orders in a  $c$  axis modulated (CAM) structure. At  $T_{N\perp} \cong 53$  K (magnetization is perpendicular to the  $c$  axis), another AFM transition takes place during which the CAM structure changes to an antiphase domain (APD) cone plus helix structure. Upon further cooling, at  $\sim 25$  K Er transforms into an alternating cone structure. The APD structure changes to a ferromagnetic (FM) cone along the  $c$  axis plus helix structure at  $T_C \cong 19$  K. This FM transition is of the first order and is accompanied by a discontinuous 0.38% change in the lattice parameter  $c$ .<sup>12</sup>

Synchrotron x-ray scattering study showed that Er also exhibits a sequence of lock-in transitions, which were explained by a spin slip model.<sup>13</sup> These spin slip transitions were also observed in the dc magnetometry and ac susceptometry studies of single-crystal Er.<sup>6</sup>

Elemental Dy, which is paramagnetic at room temperature, also exhibits several magnetic field and temperature driven magnetic transitions.<sup>7,14,15,16</sup> Upon cooling down to the Neel

temperature,  $T_N \cong 180$  K, in zero magnetic field, the PM Dy transforms into a helix AFM ordered state. The AFM order changes over to the FM one below  $T_C \cong 90$  K. When a magnetic field is applied anywhere between  $T_C$  and 135 K, the magnetic structure of Dy changes from a helix AFM to FM.<sup>7, 17</sup> Between 135 K and 180 K, the AFM-FM transition is separated by an intermediate fan magnetic phase, which is stable between  $\sim 7$  and  $\sim 25$  kOe.<sup>7, 17</sup> Low temperature x-ray diffraction studies of single crystal Dy showed that the FM transition at 90 K is accompanied by a structural transition from hcp to a distorted orthorhombic structure along with significant discontinuities in lattice parameters.<sup>18,19,20</sup> Below  $T_N$ , only a strong expansion of the  $c$  parameter is reported.

When two lanthanide metals are mixed, the magnetic and thermal properties of the resulting alloys may become very interesting. Some such alloy systems that have been investigated earlier include Tb-Er,<sup>21,22</sup> Nd-Dy,<sup>23</sup> and Er-Pr.<sup>24,25,26</sup> In the Er-Pr system it was found that the addition of Pr significantly increases the heat capacity of Er, making the Er-Pr system suitable for cryocooler regenerator applications.<sup>24,25</sup> Multiple magnetic ordering transitions were recently discovered in pseudo-binary  $R_{1-x}R'_xAl_2$  alloys, where R and R' are rare earth metals with second order Steven's factors of opposite sign.<sup>27,28,29,30</sup> Hence, mixing of two lanthanide metals can result in unusual behaviors not only in pseudo-binary lanthanide metals but also in rare earth based intermetallic compounds.

Among the  $R_{1-x}R'_xAl_2$  systems studied to date, the  $Er_{1-x}Dy_xAl_2$  alloys exhibit the most pronounced first order heat capacity peaks,<sup>Error! Bookmark not defined.</sup> which is the key characteristic of the multiple magnetic ordering phenomenon mentioned above. Among

the  $\text{Er}_{1-x}\text{Dy}_x\text{Al}_2$  alloys, the alloy with  $x = 0.25$  reveals the sharpest heat capacity anomaly. It is therefore interesting to explore the magnetic and thermal properties of pseudo-binary Er-Dy lanthanide alloy, especially with 25% Dy. Here we report an experimental study of  $\text{Er}_{75}\text{Dy}_{25}$  single crystals.

## EXPERIMENTAL DETAILS

Single crystal samples of  $\text{Er}_{75}\text{Dy}_{25}$  were prepared from 99.8+ at. % (99.98 wt. %) pure (with respect to all other elements in the periodic table) Er and Dy, by the Materials Preparation Center of the Ames Laboratory.<sup>31</sup> Appropriate quantities of the rare earth metals were arc melted together under an argon atmosphere several times, and then dropcast into a copper chilled mold. The ingot was cold worked by hitting it with a ball-peen hammer and then heat treated for 10 days at 1400°C. The resulting crystal was oriented by back reflection Laue technique, and samples were cut from the oriented ingot by spark erosion. The faces were ground flat and parallel by standard metallographic techniques. Three different samples (two for heat capacity measurements and one for magnetization measurements) were thus prepared. The specimen for the magnetization measurements was shaped as a parallelepiped,  $1.37 \times 1.4 \times 0.99 \text{ mm}^3$  in size. The specimens for the heat capacity measurements were bigger and measured  $5 \times 5 \times 2 \text{ mm}^3$ , with the *b*- or *c*- crystallographic axis parallel to the shortest dimension.

A Superconducting Quantum Interference Device (SQUID) magnetometer MPMS XL-7 made by Quantum Design Inc. was used to collect the dc and ac magnetic data. The

measurements were performed in the temperature range of 2 – 300 K and in applied magnetic fields of up to 7 T. The ac susceptibility measurements were performed in an ac field of  $H_{ac} = 5$  Oe and at frequencies ranging from 1 Hz to 1000 Hz in zero and applied dc magnetic fields of 0 to 40 kOe. A homemade adiabatic heat-pulse calorimeter was used to conduct the heat capacity measurements.<sup>32</sup> The measurements were performed in the temperature range from ~2 to 350 K in zero magnetic fields and in applied magnetic fields up to 15 kOe.

## RESULTS AND DISCUSSION

### Properties along the [0001] direction (or *c* axis)

The heat capacity,  $C_P$ , of single crystalline  $\text{Er}_{75}\text{Dy}_{25}$  measured in different magnetic fields applied parallel to the *c* axis is shown in Fig. 1a. Three transitions are observed: at ~ 30 K, ~ 45 K, and ~ 110 K. The sharp peak at  $T_C \cong 30$  K corresponds to the first order AFM-FM transition. A similar transition (sharp peak) is observed in the  $C_P$  data of pure Er at  $T_C \cong 19$  K.<sup>33,34</sup> In pure Dy, the first order  $C_P$  peak is observed at  $T_C \cong 90$  K.<sup>3,7</sup> The transition observed at ~45 K resembles the anomaly observed in the  $C_P$  of pure Er at  $T_{N\perp} \cong 53$  K, where the CAM structure transforms into the APD magnetic structure.<sup>3,33</sup> No such anomaly is observed in pure Dy. The anomaly at ~110 K corresponds to the second order AFM-PM transition and is similar to the anomalies observed in the heat capacities of pure Er and Dy at  $T_{N\parallel} \cong 84$  K and  $T_N \cong 180$  K, respectively. The spin slip transitions observed in pure Er<sup>34</sup> are not observed in the  $C_P$  of  $\text{Er}_{75}\text{Dy}_{25}$  single crystal.

The first order peak at  $\sim 30$  K is strongly suppressed by the magnetic field and it completely disappears when a magnetic field of 15 kOe is applied. A similar behavior is observed in pure Er where application of magnetic field suppresses the first order peak.<sup>34</sup> The 45 K anomaly shifts to lower temperatures and vanishes as the magnetic field is increased. The anomaly at 110 K remains practically unaffected by these magnetic fields. As Dy is mixed with Er, the  $T_C$  and  $T_{N\parallel}$  increase nearly linearly (see Fig. 1b, where the data points for pure Er and Dy are taken from References 3 and 33). This increase correlates with the magnitude of the de Gennes factor of  $Dy^{3+}$  (7.08), which is higher than that of  $Er^{3+}$  (2.55). On the contrary,  $T_{N\perp}$  decreases upon adding Dy.

The temperature dependencies of dc magnetization,  $M(T)$ , measured under zero field cooled (ZFC) warming, field cooled cooling (FCC), and field cooled warming (FCW) conditions in magnetic field of 100 Oe applied along the  $c$  axis are shown in Fig. 2. The inset of Fig. 2 shows the derivative of the  $M(T)$  data, and from these data the temperatures of different transitions observed in the  $M(T)$  data are obtained. The first order transition is observed at  $\sim 31$  K on heating and  $\sim 28$  K on cooling. A second order APD-CAM transition is observed at  $\sim 45$  K on both heating and cooling. The second order CAM-PM transition observed at  $\sim 110$  K in the  $C_P$  of  $Er_{75}Dy_{25}$  is barely visible in the  $M(T)$  data of Fig. 2; as shown in the inset of Fig. 2, a weak anomaly is observed in the derivative of the  $M(T)$  data at  $\sim 110$  K. It should be noted that the APD-CAM and CAM-PM transitions mentioned above, are assumed based on the similarities between the experimental observations for pure Er (e.g., see Refs. 5 and 6) and  $Er_{75}Dy_{25}$  (current

work). The 84 K transition in pure Er is also barely visible in the  $M(T)$  data.<sup>6</sup> The spin slip transitions that are observed in the  $M(T)$  data along the  $c$  axis of Er single crystal (see reference 6) are not observed in the  $M(T)$  data along the  $c$  axis of  $\text{Er}_{75}\text{Dy}_{25}$  single crystal.

Figure 3 shows the  $M(T)$  data of  $\text{Er}_{75}\text{Dy}_{25}$  measured along the  $c$  axis on heating at different magnetic fields ranging from 10 kOe to 50 kOe in 10 kOe increments. The only transition observed in each dataset is the first order transition at  $T_C$ , temperature of which increases linearly with increasing magnetic field. The APD-CAM transition at  $\sim 45$  K observed in the 100 Oe  $M(T)$  data is not observed in any of the  $M(T)$  data shown in Fig. 3. The anomaly at  $\sim 110$  K (see inset of Fig. 3) is more pronounced in the high magnetic field  $M(T)$  data.

The isothermal magnetization,  $M(H)$ , data of  $\text{Er}_{75}\text{Dy}_{25}$  along the  $c$  axis measured at 2 K and 5 K are shown in Fig. 4. The 2 K and 5 K data show insignificant differences, and a typical ferromagnetic behavior is observed at both temperatures suggesting that the easy magnetization direction of  $\text{Er}_{75}\text{Dy}_{25}$  is along the  $c$  axis, which is consistent with the easy axis of magnetization of pure Er being the  $c$  axis.<sup>35</sup> The magnetic moment at 70 kOe is 243 emu/g ( $7.23 \mu_B / \text{f.u.}$ ), which is less than the expected theoretical value of  $9.25 \mu_B$ . This suggests that similar to pure Er, the  $\text{Er}_{75}\text{Dy}_{25}$  alloy is a cone with a non-zero angle between the magnetic moment and the  $c$  axis. The upper inset of Fig. 4 shows the  $M(H)$  data at temperatures ranging from 23 K to 65 K along the  $c$  axis. The data show the transformation of the magnetic structure from FM to PM type with increasing temperature. The lower inset of Fig. 4 points to a weak metamagnetic behavior around 5

kOe in the vicinity 40 K. This correlates well with the transition observed at  $\sim 42$  K in the 100 Oe  $M(T)$ , and with the disappearance of this transition at higher magnetic fields (see Figs. 2 and 3).

The real component of the ac susceptibility,  $\chi'(T)$ , of  $\text{Er}_{75}\text{Dy}_{25}$  measured along the  $c$  axis in zero dc magnetic fields is shown in Fig. 5a. The measurements were performed on heating in an ac field ( $H_{ac}$ ) of 5 Oe at different frequencies. The  $\chi'(T)$  data exhibit a sharp peak at  $\sim 30$  K, due to the first order transition from FM to APD state, and a broader peak at  $\sim 45$  K due to the second order APD-CAM transition. As shown in the upper inset of Fig. 5a, below  $\sim 31$  K the  $\chi'(T)$  data are frequency dependent: with increasing frequency, the magnitude of  $\chi'$  below  $T_C$  decreases showing that the alloy becomes less responsive to the ac field. The sharp peak at  $T_C$  also tends to diminish with increasing frequency. The derivative of the  $\chi'$  data shows a third transition at  $\sim 110$  K, which is similar to the behaviors observed in Figs. 2 and 3, where the transition at 110 K is only observed in the derivative of the  $M(T)$  data.

Figure 5b shows the  $\chi'(T)$  data of  $\text{Er}_{75}\text{Dy}_{25}$  measured in zero and 5 kOe magnetic fields applied along the  $c$  axis of the crystal. Consistent with the behavior observed in the heat capacity data (see Fig. 1a), the peak at  $\sim 30$  K seen in the zero magnetic field data disappears as 5 kOe magnetic field is applied, while the peak at  $\sim 45$  K shifts to lower temperature. The magnitude of the 45 K is also enhanced when the 5 kOe field is applied.

The  $H$ - $T$  phase diagram of the  $\text{Er}_{75}\text{Dy}_{25}$  single crystal with the magnetic field applied along the  $c$  axis constructed using from Figs. 1-5 is shown in Fig. 6. Above  $T_{N\parallel}$ , which remains unchanged with varying magnetic field, the  $c$  axis moments are PM.  $T_{N\perp}$ , on the other hand, shows complex behavior as a function of magnetic field, while  $T_C$  increases with increasing magnetic field. Below  $T_C$  the  $\text{Er}_{75}\text{Dy}_{25}$  single crystal is FM along the  $c$  axis, and between  $T_C$  and  $T_{N\parallel}$  it belongs to the CAM phase. A fourth phase is observed below  $T_{N\perp}$ , but it only exists as a small region in low magnetic fields.

### **Properties along the $[10\bar{1}0]$ (or $a$ axis)**

Figure 7 shows the  $C_P$  of  $\text{Er}_{75}\text{Dy}_{25}$  measured at different magnetic fields applied parallel to the  $a$  axis. The sharp, first order peak is still observed in a 15 kOe field applied along the  $a$  axis, while it was completely suppressed by the same field applied along the  $c$  axis (see Fig. 1b). The insets (a) and (b) of Fig. 7 show the  $C_P$  data in the vicinity of the  $\sim 45$  K and  $\sim 110$  K transitions, respectively. It is interesting to note that the 45 K anomaly shifts to higher temperature region with increasing magnetic field applied along the  $a$  axis, while it shifts to lower temperature when the field is applied along the  $c$  axis (see Fig. 1b). As shown in the inset (b) of Fig. 7, the  $\sim 110$  K transition shifts to lower temperatures as the magnetic field increases.

All of the transitions observed in the  $C_P$  and  $c$  axis  $M(T)$  data are clearly observed in the  $a$  axis  $M(T)$  data measured in 100 Oe magnetic field, shown in Fig. 8. However, the

nature of the  $M(T)$  data along the  $a$  axis is quite different than the  $M(T)$  data along the  $c$  axis. This difference is due to the  $a$  axis being the hard magnetization direction. The 110 K anomaly that is weak and only observed in the derivative of the  $M(T)$  data along the  $c$  axis, is clearly seen as antiferromagnetic ordering in the  $M(T)$  along the  $a$  axis. The APD-CAM transition is also observed in the derivative of the  $M(T)$  data at  $T_{N\perp} \cong 42$  K, as shown in the inset of Fig. 8.

Figure 9 shows the  $M(T)$  data along the  $a$  axis of  $\text{Er}_{75}\text{Dy}_{25}$  measured in magnetic fields ranging from 10 kOe to 50 kOe. A sharp increase in magnetization is observed when the magnetic field is increased from 20 kOe to 30 kOe. Similar to the magnetization of pure Er,<sup>5</sup> this is most likely due to the field induced transition from an FM cone to an FM fan structure. A fourth transition is observed below  $T_C$  that shifts to a lower temperature when the magnetic field is increased and eventually disappears when the field reaches 50 kOe. This transition is more clearly observed in the derivative of the  $M(T)$  data as shown in the inset of Fig. 9, where the transition temperature is marked as  $T_4$ .

The 2 K and 5 K isothermal magnetization data of  $\text{Er}_{75}\text{Dy}_{25}$  measured with the magnetic field applied along the  $a$  axis are shown in Fig. 10. The metamagnetic nature of the magnetization data is clear in the figure. A sharp increase of magnetization starts at a critical magnetic field,  $H_C \cong 22$  kOe, which is consistent with the  $M(T)$  data shown in Fig. 9. The metamagnetic transition represents the field induced transformation from an FM cone phase to FM fan phase, which is observed in pure Er when field is applied along the  $a$  and  $b$  axes. Above  $\sim 28$  kOe, several step-like increases in magnetization are most

probably related to the fourth transition that is observed below  $T_C$  in Fig. 9. Hysteresis is observed in the magnetization data in the vicinity of 22, 28, and 60 kOe. At a magnetic field of 70 kOe the moment is  $\sim 267$  emu/g ( $7.93 \mu_B$ ), and does not saturate.

As shown in Fig. 11, magnetic field induced metamagnetic transitions from an FM cone to an FM fan are observed below 100 K. Above 100 K, the magnetization data exhibit PM behavior. Additional steps of magnetization observed in the 2 K and 5 K data (see Fig. 10) are not seen at 15 K and higher temperatures.

The  $\chi'(T)$  data measured along the  $a$  axis of  $\text{Er}_{75}\text{Dy}_{25}$  single crystal at several frequencies are shown in Fig. 12. As seen here, there is no frequency dependence of the  $\chi'(T)$ . The behavior of the  $\chi'(T)$  data is similar to the 100 Oe  $M(T)$  data shown in Fig. 8. All of the transitions observed in the 100 Oe  $M(T)$  dc data are observed in the  $\chi'(T)$  ac susceptibility data. The derivative of the  $\chi'(T)$  data, as shown in the inset of Fig. 12, also compares very well with the derivative of the  $M(T)$  data as shown in Fig. 10. This was not the case with the measurements along the  $c$  axis of  $\text{Er}_{75}\text{Dy}_{25}$ , where the behavior of the  $M(T)$  and  $\chi'(T)$  data were quite different (see Figs. 2 and 5).

As shown in Fig. 13, the behavior of  $\chi'(T)$  with the field applied along the  $a$  axis of  $\text{Er}_{75}\text{Dy}_{25}$  changes significantly when bias dc magnetic fields are applied in the same direction. The peak at  $\sim 30$  K (shown by a vertical arrow in Fig. 13a) is strongly enhanced as the magnetic field increases. Up to 20 kOe, the enhancement of the 30 K peak is

proportional to the applied magnetic field. The peak enhancement is most dramatic when a magnetic field of 25 kOe is applied (see the inset of Fig. 13a). When the magnetic field exceeds 25 kOe, the peak begins to diminish. The peak at 110 K (shown by a tilted arrow in Fig. 13a) both sharpens and shifts to lower temperature with increasing magnetic field up to 30 kOe. At 35 kOe the peak value of  $\chi'$  diminishes and at 40 kOe, the 110 K anomaly is no longer visible (Fig. 13b).

The  $H$ - $T$  Phase diagram of  $\text{Er}_{75}\text{Dy}_{25}$  single crystal along the  $a$  axis is shown in Fig. 14. The phase diagram along the  $a$  axis is significantly different and more complex than the phase diagram along the  $c$  axis (see Fig. 6). The transition temperatures show strong dependence on the applied magnetic field.  $T_C$  remains at  $\sim 30$  K until the field reaches  $\sim 20$  kOe, above which  $T_C$  increases with increasing magnetic field. The  $H_C$  obtained from the  $M(H)$  data initially remains same up to  $\sim 30$  K and then increases with increasing temperature. The  $\sim 45$  K anomaly representing the APD –CAM transition shifts to higher temperatures as the field is increased while the  $\sim 110$  K anomaly that represents the CAM-PM transformation shifts to lower temperatures with increasing field. The two transitions merge at  $\sim 70$  K and  $\sim 40$  kOe. As labeled in Fig. 14, below  $H_C$  the FM cone phase is dominant along the  $a$  axis of  $\text{Er}_{75}\text{Dy}_{25}$  single crystal. Between  $\sim 30$  K and  $\sim 50$  K and below  $H_C$  the APD phase is dominant. The CAM phase dominates between  $\sim 50$  K and  $\sim 110$  K and below  $H_C$ . Above 110 K the PM phase is dominant. Above  $H_C$  and between 10 K and 60 K FM fan phase is stable. Below 20 K and above  $H_C$  (region labeled as FM ?? in Fig. 14) the phase is still FM-like but its nature is unclear.

The  $H$ - $T$  phase diagram of  $\text{Er}_{75}\text{Dy}_{25}$  single crystal along the  $a$  axis has some similarities with the  $a$  axis phase diagram of Er single crystal.<sup>5</sup> Magnetic phases similar to the FM, FM fan, APD, and CAM phases observed in pure Er are also observed in  $\text{Er}_{75}\text{Dy}_{25}$  single crystal. However, in pure Er,  $T_{N\parallel}$  does not change with the magnetic field while along the  $a$  axis of  $\text{Er}_{75}\text{Dy}_{25}$   $T_{N\parallel}$  decreases with increasing magnetic field. Also  $T_{N\perp}$  decreases with increasing magnetic field along the  $a$  axis of Er single crystal, while in  $\text{Er}_{75}\text{Dy}_{25}$  single crystal the behavior of  $T_{N\perp}$  is opposite. These differences in the  $H$ - $T$  phase diagram of  $\text{Er}_{75}\text{Dy}_{25}$  and Er single crystals can be attributed to the addition of Dy in  $\text{Er}_{75}\text{Dy}_{25}$  single crystal. Since the easy magnetization directions and the  $4f$  charge densities of Dy and Er are different from one another, mixing these two elements result in the observed magnetic structures shown in Fig. 14.

Finally, the sharp peak in the heat capacity data and the corresponding thermal hystereses observed in the  $M(T)$  data of  $\text{Er}_{75}\text{Dy}_{25}$  single crystal strongly suggest that the 30 K anomaly represents a first order transition. It is unclear if a structural transition is associated with this first order transition. Since Dy undergoes a structural transition at  $T_C$ , it is possible that a structural change may be associated with the first order transition in of  $\text{Er}_{75}\text{Dy}_{25}$  single crystal. In order to confirm the existence of a structural transition in  $\text{Er}_{75}\text{Dy}_{25}$ , we plan to perform temperature dependent x-ray diffraction measurements in the future.

## CONCLUSIONS

Experimental investigations on  $\text{Er}_{75}\text{Dy}_{25}$  single crystal show that the  $c$  axis is the easy magnetization direction. Along the  $c$  axis, three temperature driven phase transitions are observed. A first order FM transition is observed at  $T_C \cong 30$  K, followed by two AFM transitions at  $\sim 45$  K, and  $\sim 110$  K, respectively. The first order peak is revealed as sharp peak in the heat capacity data. The application of magnetic fields along the  $c$  axis suppresses the sharp peak, which disappears when the field reaches 15 kOe. The 45 K transition also disappears as magnetic field is applied along the  $c$  axis. The transitions shift either to lower or higher temperatures when magnetic fields are applied along the  $a$  axis of the  $\text{Er}_{75}\text{Dy}_{25}$  single crystal. Field induced metamagnetic transition from FM cone to FM fan phase is observed also along the  $a$  axis. The multiple spin slip transitions that are observed in pure Er single crystals are not observed in the  $\text{Er}_{75}\text{Dy}_{25}$  single crystal. The  $H$ - $T$  phase diagrams along the  $c$  and  $a$  axis are significantly different. The phase diagram along the  $c$  axis shows the existence of FM, APD, CAM and PM phases, while the phase diagram along the  $a$  axis shows that the  $\text{Er}_{75}\text{Dy}_{25}$  single crystal exhibits the FM cone, FM fan, an unknown FM, APD, CAM and PM phases.

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## FIGURE CAPTIONS

Fig. 1 (Color online) (a) The heat capacities of the Er<sub>75</sub>Dy<sub>25</sub> single crystal measured in different magnetic fields applied along the *c* axis. The inset clarifies the behavior around

the 45 K anomaly. (b) The different transition temperatures, observed in pure Er,  $\text{Er}_{75}\text{Dy}_{25}$ , and pure Dy as a function of Dy concentration.

FIG. 2 (Color online). The temperature dependencies of the dc magnetization of  $\text{Er}_{75}\text{Dy}_{25}$  measured in 100 Oe magnetic field applied along the  $c$  axis under ZFC, FCC, and FCW conditions. The inset shows the derivatives of the ZFC and FCC data.

FIG. 3 (Color online). The temperature dependencies of dc magnetization of  $\text{Er}_{75}\text{Dy}_{25}$  measured in applied magnetic fields from 10 kOe to 50 kOe in 10 kOe increments applied along the  $c$  axis. The inset shows the derivatives of the data obtained at each field.

FIG. 4 (Color online). The field dependence of magnetization along the  $c$  axis of  $\text{Er}_{75}\text{Dy}_{25}$  measured at 2 and 5 K. The upper inset shows the field dependence of magnetization of the same measured at higher temperatures. The lower inset shows the low field behavior of the magnetization data shown in the upper inset.

FIG. 5 (Color online). (a) The temperature dependence of real component of the ac magnetic susceptibility ( $\chi'$ ) of  $\text{Er}_{75}\text{Dy}_{25}$  measured along the  $c$  axis in an ac field of 5 Oe and frequencies from 1 Hz to 1000 Hz. The upper inset shows the  $\chi'$  data in the vicinity of the transition at 30 K. The lower inset shows the derivative of the 1 Hz frequency  $\chi'$  data. (b) The temperature dependence of real component of the ac magnetic susceptibility

( $\chi'$ ) of  $\text{Er}_{75}\text{Dy}_{25}$  measured in dc magnetic fields of 0 and 5 kOe applied along the  $c$  axis in an ac field of 5 Oe and frequency of 100 Hz.

FIG. 6 (Color online). The  $H$ - $T$  phase diagram of  $\text{Er}_{75}\text{Dy}_{25}$  single crystal for applied magnetic fields along the  $c$  axis constructed from the data points obtained from magnetization, ac susceptibility, and heat capacity measurements.

Fig. 7 (Color online) The heat capacity of the  $\text{Er}_{75}\text{Dy}_{25}$  single crystal as a function of temperature measured at different magnetic fields applied along the  $a$  axis. Inset (a) shows the region near the 45 K anomaly. Inset (b) shows the region near the 110 K anomaly.

FIG. 8 (Color online). The temperature dependencies of dc magnetization of  $\text{Er}_{75}\text{Dy}_{25}$  measured in an applied field of 100 Oe applied along the  $a$  axis under ZFC, FCC, and FCW conditions. Inset shows the derivatives of the ZFC and FCC data.

FIG. 9 (Color online). The temperature dependencies of dc magnetization of  $\text{Er}_{75}\text{Dy}_{25}$  measured in different magnetic fields from 10 kOe to 50 kOe in 10 kOe increments applied along the  $a$  axis. The inset shows the derivative of the  $M(T)$  data obtained at 20 kOe.

FIG. 10 (Color online). The field dependence of magnetization along the  $a$  axis of  $\text{Er}_{75}\text{Dy}_{25}$  measured at 2 and 5 K.

FIG. 11 (Color online). The field dependence of magnetization along the  $a$  axis of  $\text{Er}_{75}\text{Dy}_{25}$  measured at different temperatures.

FIG. 12 (Color online). The temperature dependence of real component of the ac magnetic susceptibility ( $\chi'$ ) of  $\text{Er}_{75}\text{Dy}_{25}$  measured along the  $a$  axis in an ac field of 5 Oe and frequencies from 1 Hz to 1000 Hz. The inset shows the derivative of the 1 Hz frequency  $\chi'$  data.

FIG. 13 (Color online). The temperature dependence of real component of the ac magnetic susceptibility ( $\chi'$ ) of  $\text{Er}_{75}\text{Dy}_{25}$  measured in dc magnetic fields of (a) 0, 10, 20, and 25 kOe (the inset shows the complete  $\chi'$  data measured at 25 kOe); (b) 30, 35, and 40 kOe, applied along the  $a$  axis in an ac field of 5 Oe and frequency of 1 Hz.

FIG. 14 (Color online). The  $H$ - $T$  phase diagram of  $\text{Er}_{75}\text{Dy}_{25}$  single crystal for applied magnetic fields applied along the  $a$  axis constructed from the data points obtained from heat capacity, magnetization and ac susceptibility measurements.

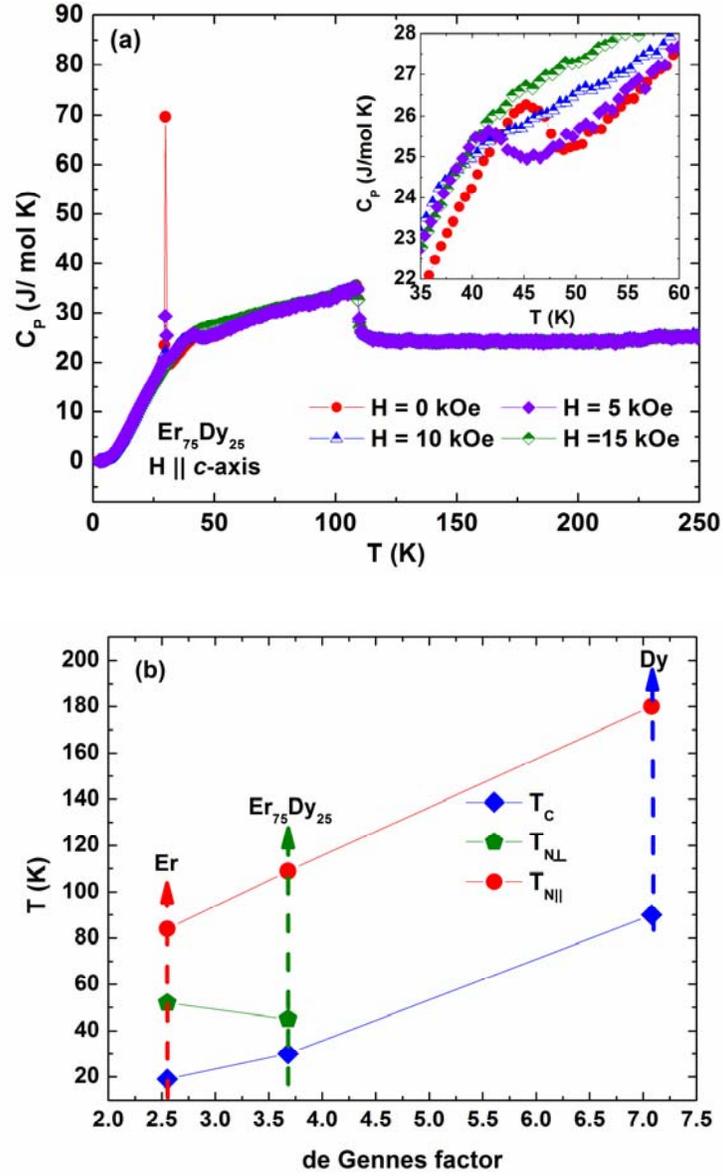


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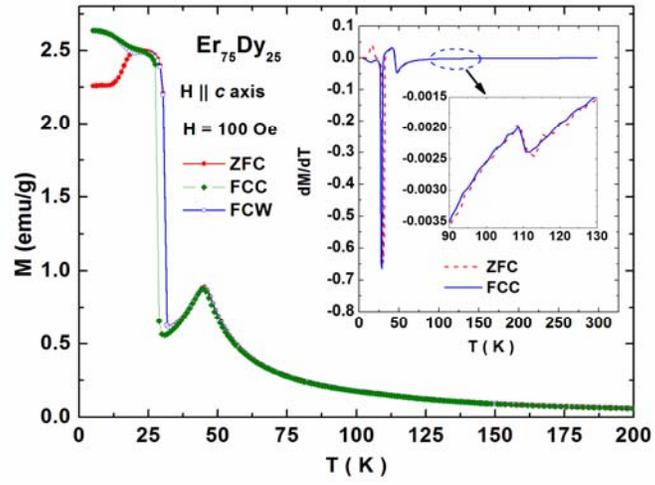


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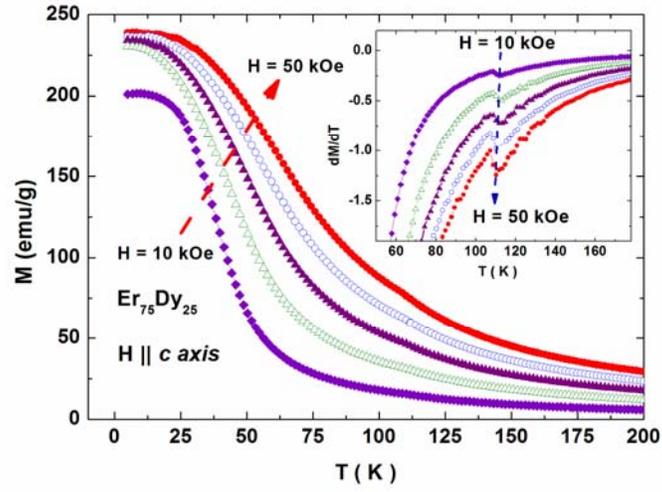


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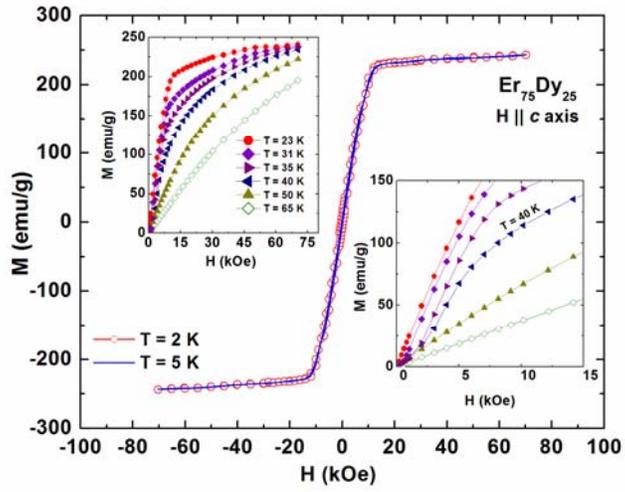


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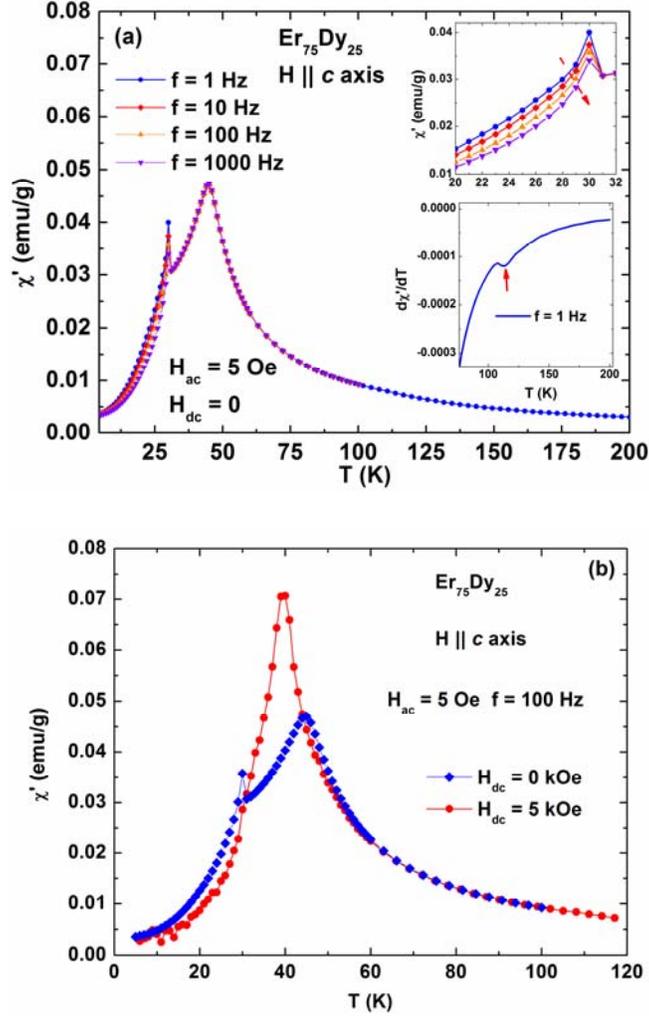


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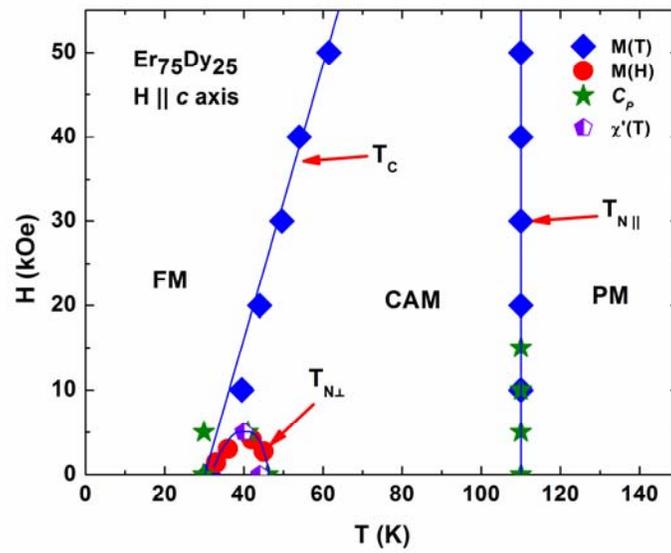


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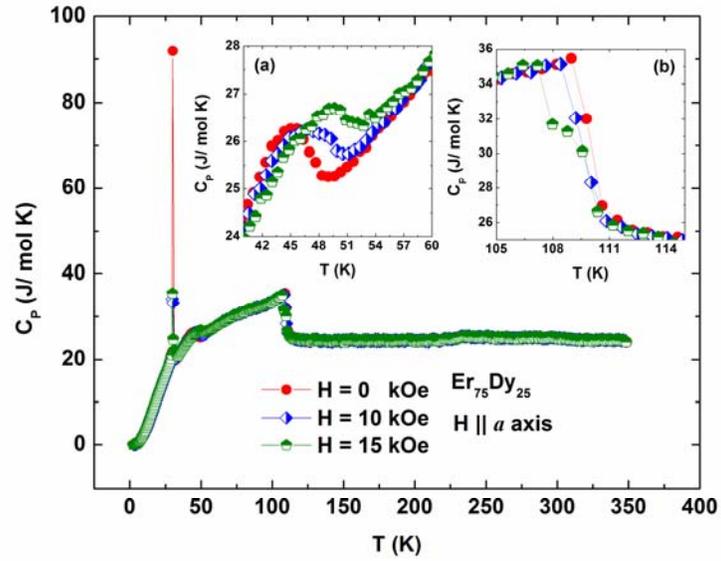


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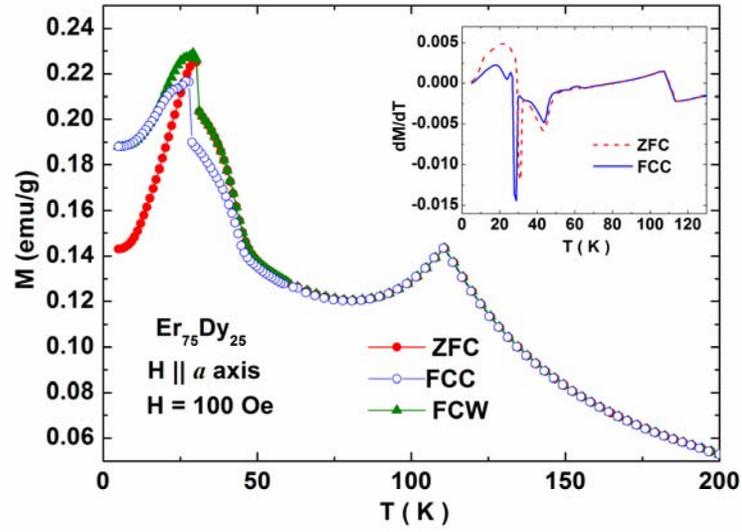


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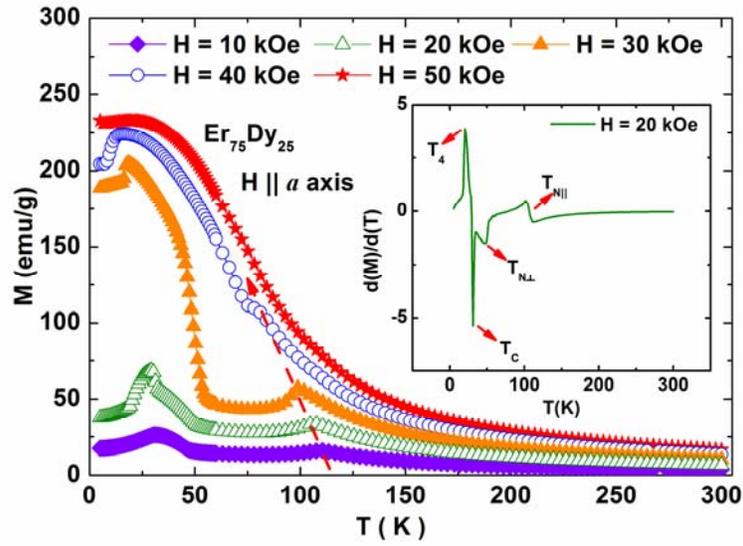


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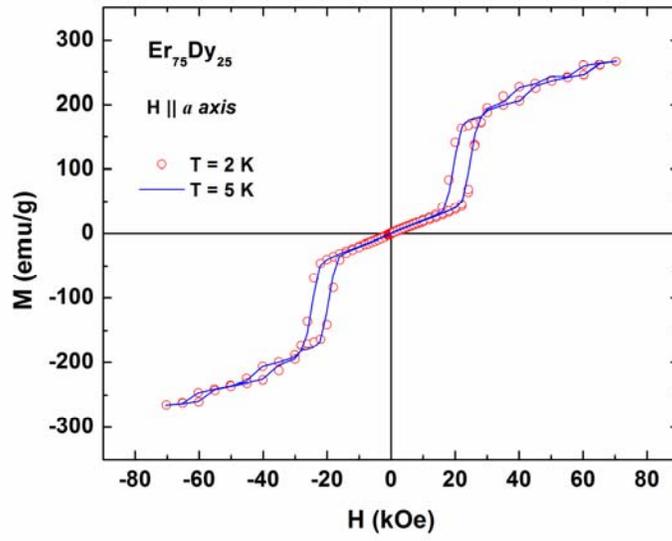


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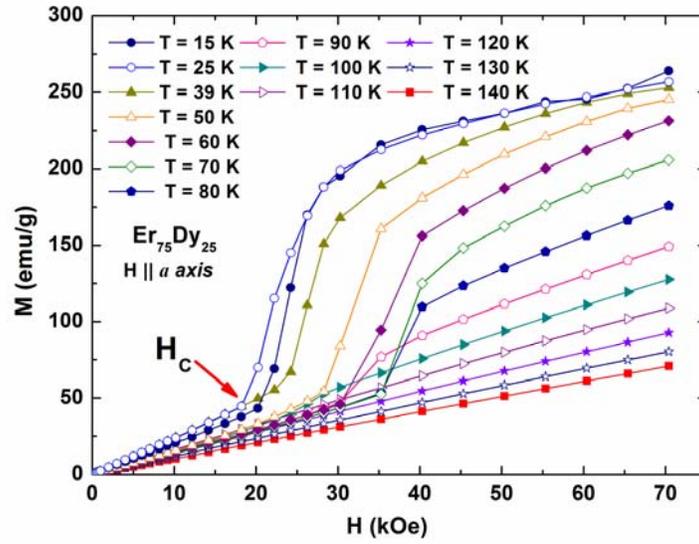


FIG. 11 (Color online). The field dependence of magnetization along the  $a$  axis of  $\text{Er}_{75}\text{Dy}_{25}$  measured at different temperatures.

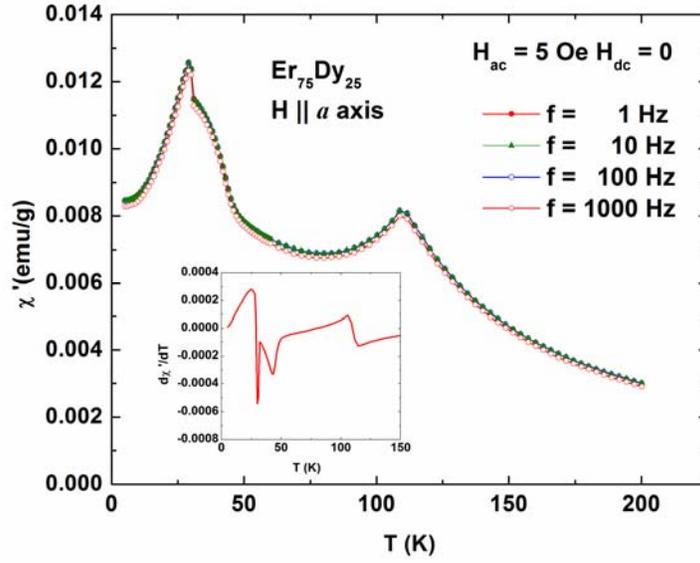


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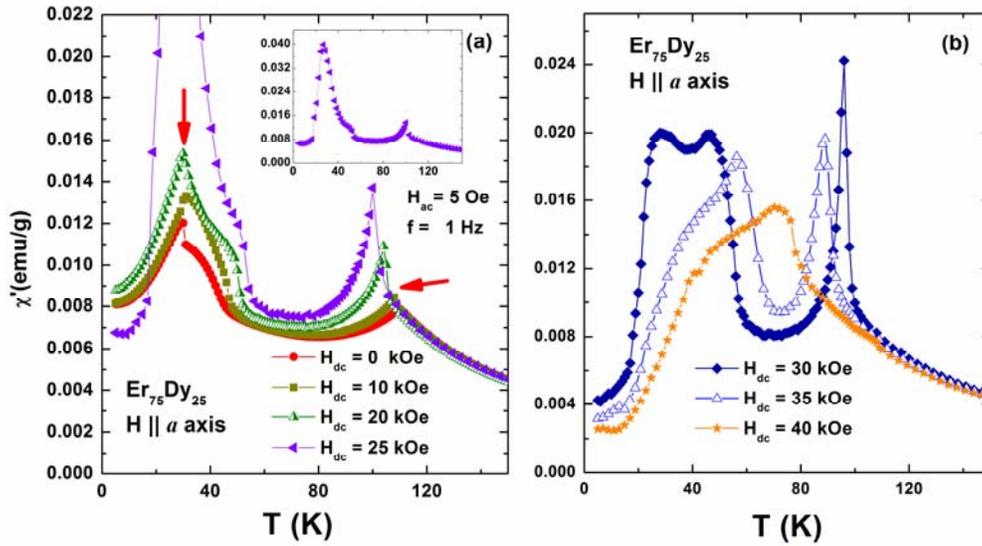


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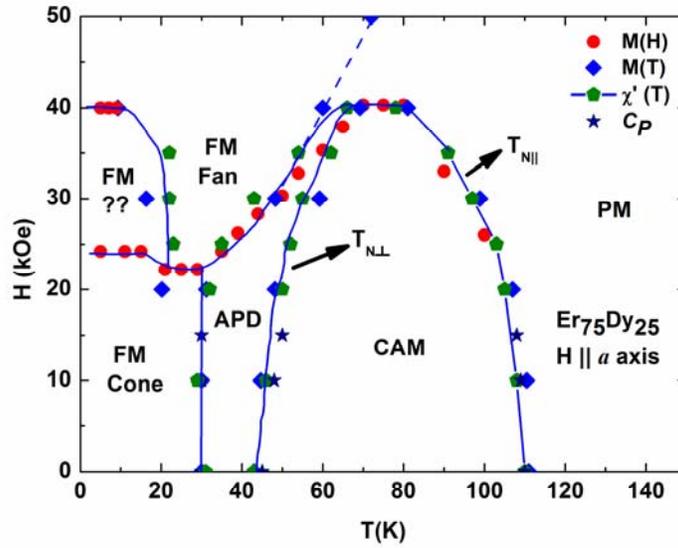


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