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Observation of re-entrant spin glass behavior in $(\text{Ce}_{1-x}\text{Er}_x)\text{Fe}_2$ compounds

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Abstract – Experimental evidence of re-entrant spin glass (RSG) state has been revealed in Er-doped CeFe_2 compounds. The zero-field-cooled - field-cooled bifurcation in dc magnetization, frequency dependence of freezing temperature, relaxation in zero-field-cooled magnetization and presence of large remanence have been discussed in detail. Frequency dependence is found to follow the critical slowing-down mechanism of the type: $\tau = \tau_0(T_f/T_{SG} - 1)^{-z\nu}$. The RSG state is found to be associated with the randomly magnetized clusters instead of atomic level randomness.

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Introduction. – Among the RFe_2 (R = rare-earth element) series of compounds, CeFe_2 has attracted a lot of attention from various researchers in view of its anomalous magnetic properties such as low Curie temperature ($T_C = 230\text{ K}$), low saturation magnetic moment ($M_S = 2.4\ \mu_B/\text{f.u.}$) as compared to that of LuFe_2 ($T_C = 610\text{ K}$, $M_S = 2.9\ \mu_B/\text{f.u.}$) [1,2]. These anomalies are explained in terms of the $4f$ band magnetism scenario [1]. The fact that the light rare-earth Ce couples antiferromagnetically with the $3d$ moment is well understood in this scenario. CeFe_2 is known to be a ferromagnet (FM) with a fluctuating antiferromagnetic (AFM) ground state. This AFM state gets stabilized by certain substitutions such as Ru, Re, Ir, Al, Ga, Si etc. at the Fe site, thereby giving rise to a FM-AFM transition on cooling [3–6]. It is established that doped CeFe_2 compounds can be a model system to understand the physics of metamagnetic transitions, metastability and phase co-existence. This is important to understand the properties of certain functional materials which have drawn a lot of interest recently [6–8]. However, much less attention has been devoted to the substitution at the Ce site by other rare-earth elements [9–13]. Enhancement of Curie temperature has been found with R substitution, which enhances the trivalent behavior of Ce in them [9,12,13]. While

investigating the effect of various R (heavy) doping, we have found that Er substitution shows remarkable differences with regard to the magnetic properties, as compared to the other rare-earth dopants such as Gd or Ho. These include large magnetic hysteresis at low temperatures and the bifurcation of the zero-field-cooled (ZFC) and field-cooled (FC) magnetization data. In order to highlight these special features of Er in CeFe_2 , we focus on $(\text{Ce}_{1-x}\text{Er}_x)\text{Fe}_2$ compounds with $x = 0.08, 0.12, 0.15, 0.25$. We find that Er doping induces the re-entrant spin glass (RSG) state in this series for $x \leq 0.25$.

The physics of spin glasses is still to be resolved and there are a few well-known spin glass systems available in reality [14–16]. On the other hand, the re-entrant spin glass behavior is realized in a variety of systems such as AuFe , $(\text{Eu}, \text{Sr})\text{S}$, FeCr , NiMn , AlFe , $(\text{Pd}, \text{Fe})_{1-x}\text{Mn}_x$, $(\text{Eu}, \text{Sr})\text{Te}$, $(\text{Eu}, \text{Sr})\text{As}$, amorphous (a-) FeNi , a- FeMn , a- FeCr and a- ZrFe , a- $(\text{Fe}_{1-x}\text{Mn}_x)_{75}\text{P}_{16}\text{B}_6\text{Al}$, a- $(\text{Fe}_{1-x}\text{Ni}_x)_{75}\text{P}_{16}\text{BAI}_3$ [17–26]. Recently RSG has been seen in some oxides [27–32], shape memory alloys [33,34] and rare-earth-transition metal intermetallics [35] as well. RSG behavior occurs when the material shows spin glass behavior at temperatures lower than the ferromagnetic/antiferromagnetic ordering temperature. The observation of RSG behavior is sometimes controversial and ambiguous as similar experimental features appear due to the deviation from perfect ferromagnetic

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or antiferromagnetic state and/or competition between ferromagnetic and antiferromagnetic phases. So it is necessary to confirm the RSG with proper experimental tools. It is indeed possible to confirm RSG behavior as there are some unique experimental outcomes in the case of RSG [14,15].

In the present case of Er-doped CeFe_2 compounds, we discuss the observation of RSG behavior as revealed by dc and ac susceptibility measurements. The distinct features of spin glass behavior below the Curie temperature is established by examining the frequency dependence of susceptibility, relaxation in dc magnetization and remanence present in these compounds. To the best of our knowledge, RSG state has not been observed in any of the doped CeFe_2 compounds. Though certain substitutions at the Fe site are known to give rise to a magnetic glass due to the first-order transition, the observation of spin glass state is quite unexpected in the CeFe_2 compounds.

Experimental details. – Polycrystalline compounds, $(\text{Ce}_{1-x}\text{Er}_x)\text{Fe}_2$ ($x = 0.08, 0.12, 0.15, 0.25$) were prepared by the arc melting method in a water-cooled copper hearth under argon atmosphere. The constituent elements, of at least 99.9% purity, were melted by taking their stoichiometric proportion. The alloys buttons were remelted several times to ensure homogeneity. The arc melted samples were annealed for 10 days in the following way: 600 °C for 2 days, 700 °C for 5 days, 800 °C for 2 days and 850 °C for 1 day [5]. The structural analysis was performed by the Rietveld refinement of room temperature X-ray diffraction (XRD) patterns. The ac magnetic susceptibility data has been carried out in PPMS (quantum design) in the frequency range 33–9997 Hz. The ac measurements have also been performed in various ac amplitudes and various dc bias fields. All the ac measurements have been taken during heating after cooling the sample in zero field. The dc magnetization and heat capacity measurements were also performed in the PPMS. DC magnetization has been measured during heating after zero-field cooling (ZFC) and field cooling (FC) the sample.

Results. – All the compounds have formed in single phase which is confirmed by analyzing room temperature X-ray diffraction patterns. At room temperature, these compounds possess the MgCu_2 type cubic structure with the space group $Fd\bar{3}m$. The lattice parameter is found to increase from 7.3029(2) Å for $x = 0.08$ to 7.3047(2) Å for $x = 0.25$. This variation is in good agreement with the report by Tang *et al.* [9].

Figure 1 shows the temperature variation of dc magnetization, $M(T)$, for all the compounds at $H = 100$ Oe. It can be noticed that all the compounds undergo paramagnetic (PM) to ferromagnetic transition and that the T_C monotonically increases with Er content. Therefore, it is clear that there is a net enhancement of the ferromagnetic ordering with Er. It can be seen that the FC curves reflect the ferromagnetic behavior in all the cases, except in $x = 0.25$ (see fig. 1(d)). This latter compound does not

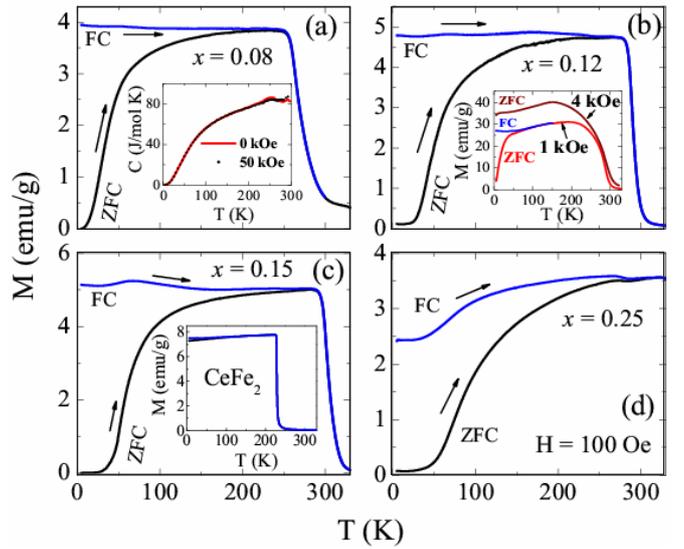


Fig. 1: (Colour on-line) Temperature variation of dc magnetization of $(\text{Ce}_{1-x}\text{Er}_x)\text{Fe}_2$ ($x = 0.08, 0.12, 0.15$ and 0.25) compounds during heating in ZFC and FC modes at $H = 100$ Oe. The inset in (a) shows the variation of heat capacity as a function of temperature (at $H = 0$ and 50 kOe) in the case of $x = 0.08$. The inset in (b) shows ZFC and FC magnetization data in 1 kOe and 4 kOe in $x = 0.12$. The ZFC and FC M - T data in the undoped CeFe_2 in 100 Oe is shown in the inset of (c).

follow the FM behavior, possibly due to the fact that the T_C in this case is more than 330 K and that the field cooling is not started from the paramagnetic state. Figure 1 also shows that at temperatures below T_C , the ZFC and FC data follow different paths, resulting in a large bifurcation between them. This behavior roughly indicates the magnetic frustration and glassy behavior at low temperatures ($T < T_C$). It is of interest to note that undoped CeFe_2 does not exhibit this bifurcation as can be seen from the inset of fig. 1(c). In the presence of a higher field (1 kOe), the ZFC-FC difference decreases and at $H = 4$ kOe, the ZFC data shows almost a normal ferromagnetic behavior (see inset of fig. 1(b)), in the case of $x = 0.12$. Therefore the M - T data is indicative of a possible spin glass phase below the Curie temperature in these compounds. As is clear from the inset of fig. 1(a), the heat capacity does not show any anomaly close to the region where the ZFC magnetization decreases considerably.

Notably, a spin glass state shows characteristic frequency dependence in the ac susceptibility. Figure 2 shows the temperature variation of the in-phase (χ'_{ac}) and the out-of-phase (χ''_{ac}) components of ac magnetic susceptibility for the $x = 0.15$ compound as a typical example, at different frequencies over a wide temperature range from 5 K to the Curie temperature, at fixed $H_{ac} = 5$ Oe and $H_{dc} = 0$. It is to be noted here that when the temperature is reduced below T_C , the in-phase component decreases giving a high value at T_C (fig. 2(a)). On the other hand,

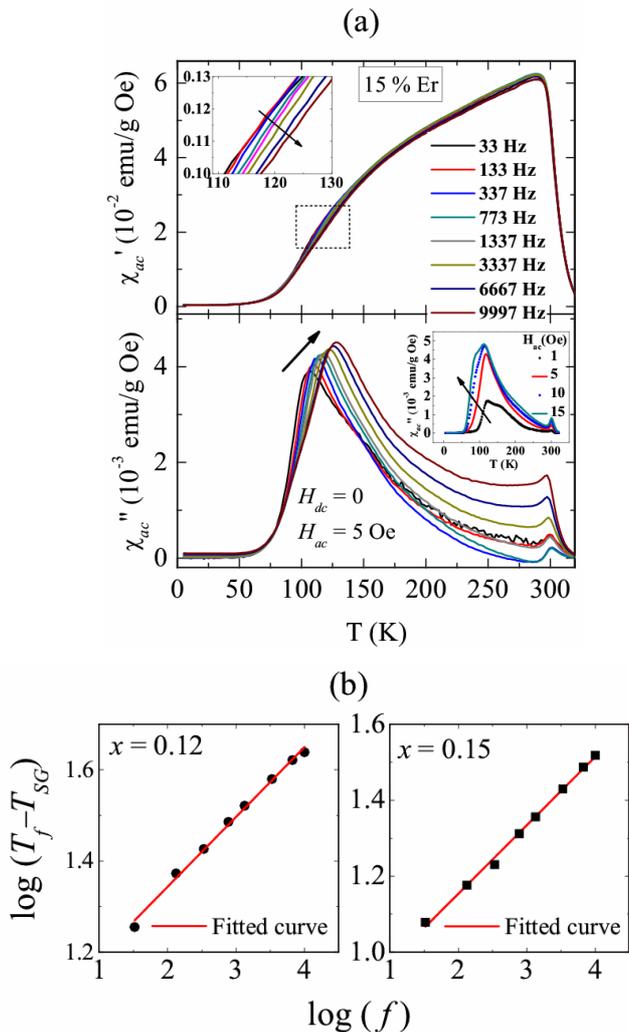


Fig. 2: (Colour on-line) (a) Temperature variation of in-phase (upper panel) and out-of-phase (lower panel) of ac susceptibility at $H_{ac} = 5$ Oe for $(\text{Ce}_{0.85}\text{Er}_{0.15})\text{Fe}_2$ at different frequencies. Inset of upper panel shows the expanded square region. Inset of lower panel shows the ac amplitude dependence of out-of-phase ac susceptibility. (b) Frequency dependence of the freezing temperature in $x = 0.12$ and 0.15 along with the fit to the critical slowing-down formula, $\tau = \tau_0(T_f/T_{SG} - 1)^{-zv}$.

the χ''_{ac} data shows a weak peak at T_C , but a pronounced peak is observed close to the temperature at which the ZFC dc magnetization changes considerably. The decrease of χ'_{ac} usually indicates the reduction in the ability of the material to respond to the low ac magnetic field. The behavior of $\chi'_{ac}(T)$ is determined by the change of domain wall motion and domain magnetization reorientation in the alternating magnetic field [36]. The peak in $\chi''_{ac}-T$ plot indicates energy absorption associated with the domain wall motion and domain rotation, which implies that such losses are quite large at temperatures below T_C in the present case.

To further probe the ac response of these compounds, the frequency dependence of the ac magnetic susceptibility

was measured, as shown in fig. 2(a). It is evident from the inset of fig. 2(a) that the χ'_{ac} data shows strong frequency dependence, with an upward shift of the curve, at around 90–130 K. Similarly, the peak in the χ''_{ac} data (in the same region) also shows a significant upward shift (lower panel of fig. 2(a)). These observations underline the frustrated magnetic state associated with the spin glasses. This kind of large frequency dependence in χ''_{ac} is also observed in shape memory alloys [33]. Furthermore, the crossing of the χ''_{ac} curves as seen here was also reported in spin glass systems [37]. On the other hand, the frequency dependence of the peak at T_C is found to be quite negligible, for both the in-phase and the out-of-phase components. The low-frequency (33 Hz) ac susceptibility anomaly coincides with the temperature at which the dc (low-field) magnetization falls rapidly. We define this temperature as T_f , the freezing temperature from the position of the maximum of $\chi''_{ac}(T)$ and sudden drop at ZFC dc $M(T)$ data. The frequency shift $Q = \Delta T_f/[T_f \log_{10}(f)]$ [15], is calculated to be 0.09 and 0.07 for $x = 0.12$ and 0.15 compounds, respectively. It is of interest to note that the present values compare well with the values of 0.06 seen in certain shape memory alloys showing RSG state [33], 0.037 seen in metallic glasses [38] and 0.095 reported in $\text{LaCo}_{0.5}\text{Ni}_{0.5}\text{O}_3$ [39]. The T_f is found to obey the critical slowing-down dynamics (see fig. 2(b)) governed by the relation $\tau = \tau_0(T_f/T_{SG} - 1)^{-zv}$, where τ is relaxation time and zv is known as dynamic exponent [29]. The maximum frequency used here is 10 kHz, which is good enough to investigate the critical behavior. We found the best fit with $T_{SG} = 82$ K, $\tau_0 = 2.7 \times 10^{-7}$ s, $zv = 5.55$ for the $x = 0.12$ and $T_{SG} = 95$ K, $\tau_0 = 1.4 \times 10^{-6}$ s, $zv = 6.67$ for $x = 0.15$. For a conventional spin glass, τ_0 is $\sim 10^{-10}$ – 10^{-13} s and zv lies in the range of 4–13 [38]. It is important to note here that the order of τ_0 value remains in the range 10^{-6} – 10^{-8} s while varying the T_f value by ± 5 K. The fact that the present τ_0 values are higher implies that the relaxation is slower and that the RSG phase is constituted by randomly magnetized clusters, instead of atomic level randomness. Such higher τ_0 values have also been found in other RSG systems such as Heusler alloys, $\text{LaCo}_{0.5}\text{Ni}_{0.5}\text{O}_3$, pyrochlore molybdates etc. [33,39,40]. It should be mentioned that though we tried $\log_{10}(f)$ vs. $1/T_f$ fitting, the fit was not as good as the critical slowing-down fitting shown in fig. 2(b). It is also found that the magnitude of the peak in χ''_{ac} increases with increase in frequency. This is again a signature of conventional spin glasses, though some known RSG systems show the opposite trend [33]. It is of importance to mention here that the strong frequency dependence that we have seen in $x = 0.08, 0.12$ and 0.15 is absent in the case of $x = 0.25$.

By applying a dc bias field (H_{dc}), the response of the ac susceptibility is found to show significant modification in its behavior. The magnitude of χ'_{ac} is strongly suppressed by the dc bias field. Furthermore, it shows a well-defined double-hump behavior for $H_{dc} \geq 500$ Oe. The variation of χ''_{ac} is also more or less similar to that of χ'_{ac} .

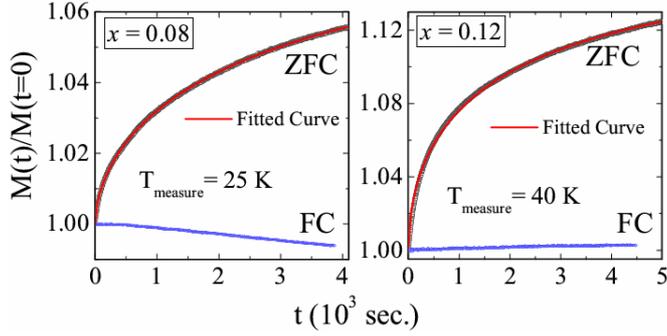


Fig. 3: (Colour on-line) Time relaxation of magnetization, $M(t)$ for $(\text{Ce}_{0.92}\text{Er}_{0.08})\text{Fe}_2$ and $(\text{Ce}_{0.88}\text{Er}_{0.12})\text{Fe}_2$ at $(T_{\text{measure}}, H_{\text{dc}}) = (25 \text{ K}, 500 \text{ Oe})$ and $(40 \text{ K}, 100 \text{ Oe})$, respectively. Data has been taken during ZFC and FC modes. The red line shows the typical relaxation fit of the type: $M_t(H) = M_0(H) + [M_\infty(H) - M_0(H)][1 - \exp\{-t/\tau\}^\alpha]$.

It is noteworthy that the χ''_{ac} peak broadens with dc bias field. At $H_{\text{dc}} = 4 \text{ kOe}$, both χ'_{ac} and χ''_{ac} become almost zero. This shows that the material is unable to respond at low H_{ac} value ($= 5 \text{ Oe}$) when the dc bias field is as large as 4 kOe . Similar findings have been reported in the case of re-entrant spin glass compound $\text{La}_{0.96-y}\text{Nd}_y\text{K}_{0.04}\text{MnO}_3$ [30].

Varying the amplitude of the ac field also causes a huge enhancement and modification of both the in-phase and the out-of-phase susceptibility near the freezing temperature (see inset of fig. 2(a)). With increase in the ac field both the in-phase and the out-of-phase peaks shift towards lower temperatures. This observation is consistent with the fact that a higher ac magnetic amplitude weakens the occurrence of spin glass state which in turn shifts the T_f towards lower temperature.

The metastability of the low-temperature glassy phase has been investigated by magnetization relaxation measurement. Comparison of the time dependent ZFC and FC magnetization data is shown in fig. 3. This is one of the measurement protocols used to investigate the glassy dynamics [29,41]. For this measurement, the sample was cooled in presence (FC) or absence (ZFC) of a field to the measurement temperature. Subsequently the measuring field was applied in the ZFC case, while the cooling field was retained in the FC case. The time variation of the growth of magnetization was recorded. The ZFC magnetization measured in this way shows a huge relaxation at $T = 25 \text{ K}$ (for $x = 0.08$) and 40 K (for $x = 0.12$), which indicates the metastability of the low-temperature magnetic state. However, there is no considerable relaxation observed in the FC magnetization. This is in sharp contradiction with a magnetic glass where FC magnetization relaxes and ZFC shows no relaxation [41]. This difference, shown in fig. 3, clearly shows that the Er-doped compounds are not magnetic glasses like $\text{Ce}(\text{Fe}_{0.96}\text{Ru}_{0.04})_2$, but re-entrant spin glasses. In this respect we would also

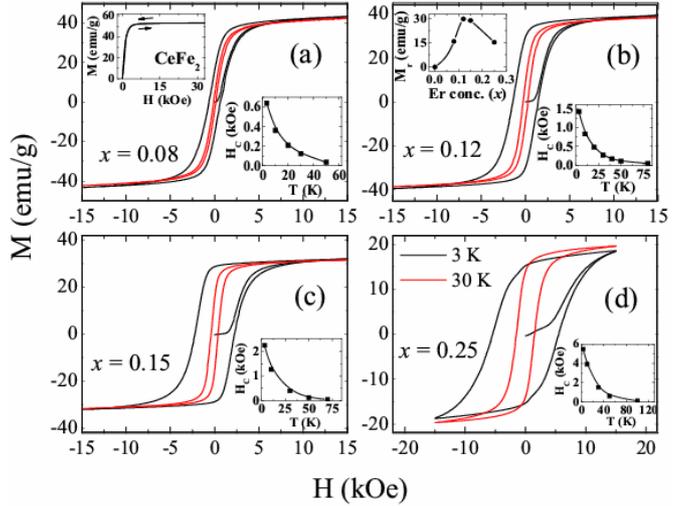


Fig. 4: (Colour on-line) M - H plots of $(\text{Ce}_{1-x}\text{Er}_x)\text{Fe}_2$ ($x = 0.08, 0.12, 0.15$ and 0.25) compounds at 3 K and 30 K . The insets in the lower right corners show the variation of coercive fields (H_c) with temperature. The inset in the upper left corner of (a) shows the two-loop $M(H)$ plot for CeFe_2 at 3 K . The inset in the upper left corner of (b) shows the variation of remanent magnetization (M_r) at 3 K with Er concentration.

mention here that we have performed measurement of $M(T)$ after cooling and heating the sample in unequal fields which was proposed recently by the authors of ref. [38]. Interestingly this system shows similar behavior as shown by a typical re-entrant spin glass material $\text{Au}_{82}\text{Fe}_{18}$ [41]. In the present compounds, the ZFC magnetization at a constant field and temperature grows as a function of time and the growth can be fitted well to a stretched exponential of the type: $M_t(H) = M_0(H) + [M_\infty(H) - M_0(H)][1 - \exp\{-t/\tau\}^\alpha]$, where τ is the characteristic relaxation time and α is called stretching parameter that ranges between 0 and 1. Best fit curve gives $\alpha = 0.58$ and 0.53 for the $x = 0.08$ and 0.12 compounds, respectively.

Another important characteristic of the RSG state is the presence of considerable remanence and coercivity [29]. To probe this, the M - H plots have been recorded in all the compounds both under ZFC and FC modes. It is found that there is no difference in the data between these two modes. Magnetization isotherms (ZFC only), which are presented in fig. 4, show clear hysteresis at $T = 3 \text{ K}$ and 30 K which was not observed in undoped CeFe_2 (see inset in the upper corner of fig. 4(a)). We would like to highlight here that no such hysteresis was observed with other rare earths such Gd or Ho even at the lowest temperature. Tang *et al.* have shown that even with Er, the hysteresis is visible only when the Er concentration is below a critical value ($x = 0.7$) [9]. Though Gd substitution does not alter the magnetocrystalline anisotropy considerably, replacing Ce with Ho should have increased the net rare-earth sublattice anisotropy. The absence of hysteresis in both Gd and Ho compounds positively indicates that the

hysteresis seen in the present (Er doping) case is not due to the increase in the magnetocrystalline anisotropy usually expected with Er addition. We would like to emphasize that even in the field-cooled mode, the M - H loops are all symmetric with respect to both the M and H axes, thereby indicating the absence of any exchange bias/exchange anisotropy.

The magnetization is found to be saturated in all the compounds at around 15 kOe and the saturation value is almost unchanged between 3 and 30 K, except in $x = 0.25$. In this compound ($x = 0.25$), the saturation magnetization is found to be more at 30 K as compared to that at 3 K, indicating the predominant ferrimagnetic coupling in this compound. It may be noted that the M - T curve in the FC mode (fig. 1) also indicated this behavior, though it is true that the field cooling did not start from the paramagnetic phase. But since the M - H plots in the other Er concentrations showed no difference between the FC and ZFC modes, we can take the anomalous M - H behavior of $x = 0.25$ as real. At this point, it may be recalled that the frequency dependence of ac susceptibility was almost negligible in this compound. With increase in temperature, the hysteresis in $M(H)$ isotherms is found to decrease as can be seen from the decrease in the coercive field (H_C) shown in the insets in the lower right corners of fig. 4. The demagnetization correction was ignored as only a qualitative analysis of the hysteresis curves is of importance in this context. The value of H_C is found to increase with x . The remanent magnetization (M_r) initially increases and then decrease at $x = 0.25$ (see the inset in the upper left corner of fig. 4(b)). We have also observed that at $x = 0.5$, the M - H curve at 3 K shows a metamagnetic transition (not shown), similar to that seen in other reports [9]. It has also been reported that $(\text{Ce}_{1-x}\text{Tb}_x)\text{Fe}_2$ as well as $(\text{Ce}_{1-x}\text{Dy}_x)\text{Fe}_2$ show metamagnetic transition around $x = 0.5$ [12,13]. We would also like to mention here that there was no frequency dependence in $\text{Ce}(\text{Fe}_{1-x}\text{M}_x)_2$ ($\text{M}=\text{Ga}, \text{Si}, \text{Al}$) compounds, which are known to be magnetic glasses [41].

Discussion. – Occurrence of RSG state in $(\text{Ce}_{1-x}\text{Er}_x)\text{Fe}_2$ is rather unexpected. The RSG state is found to be a result of randomly magnetized clusters, instead of random atomic moments. The fact that it is not achieved with rare earths such as Ho or Gd makes it even more interesting to probe its origin. An experimental observation that has been seen with various rare-earth dopings in CeFe_2 is that the T_C increases with the rare-earth content. This implies that there is an increase in the net ferromagnetic coupling with the addition of R, even when the low-temperature state shows spin glass signatures. It should also be noted that the moment on Ce in undoped CeFe_2 is quite small, due to the delocalized character of the $4f$ shell. In fact this is the main reason for the low T_C of CeFe_2 . Substitution of rare earths at the Ce site is found to change the valence state of Ce [9,12,13]. It has been reported that the Ce valency

in CeFe_2 is close to 3.29, which decreases to almost 3 on R substitution, resulting in an increase in the Ce moment. Because of the reduction in the delocalization of Ce, the Fe sublattice moment would also increase, resulting in the increase in T_C because of the increase in the $3d$ - $3d$ exchange. Since Er occupies random (Ce) positions in the unit cell, it is quite likely that the Ce moment is non-uniform in the Er-doped compounds. It is also known that the coupling of both Ce and heavy rare earths, with Fe is anti parallel. All these point towards the fact that the magnetic structure in doped compounds, especially with smaller x values, is non-collinear. One can attribute the magnetic glassiness and the frustration to this non-collinearity. But the interesting point is that though Er doping resulted in the RSG state, the Ho doping did not show any evidence of it, though the T_C and the lattice parameter variations are identical to those of Er. In this context, it is to be noted that ErFe_2 is the only member in the RFe_2 series which shows magnetic compensation (at 468 K) in the M - T data. The $(\text{Ce}_{1-x}\text{Er}_x)\text{Fe}_2$ shows compensation below 300 K [9]. However, there are no reports of such compensation in Gd or Ho doping. The absence of ZFC-FC bifurcation and magnetic hysteresis completely rule out the existence of RSG state in Gd- and Ho-doped CeFe_2 compounds [42]. Therefore, it seems that the formation of spin glass state depends on whether the Fe and (R + Ce) sublattice moments are comparable or not. If they are comparable, it is reasonable to assume that the direct FM coupling of the Fe sublattice competes with the indirect AFM coupling between Fe and (R + Ce) moments. The fact that above a certain concentration of Er, the compound behaves more or less like a normal ferrimagnet supports this proposition. Another noteworthy point is the absence of any exchange bias in the FC magnetization isotherms. This probably indicates that in the RSG state, there is no FM component, unlike in some materials in which the spin glass state is assumed to coexist with the FM state [33]. It should be mentioned here that, like Er substitution, a certain concentration of Tb and Dy doped in CeFe_2 (namely $\text{Ce}_{0.6}\text{Tb}_{0.4}\text{Fe}_2$ and $\text{Ce}_{0.6}\text{Dy}_{0.4}\text{Fe}_2$, respectively) is reported to show a compensation point, with the former showing the compensation below 300 K [13].

Conclusions. – In this report we have shown that $(\text{Ce}_{1-x}\text{Er}_x)\text{Fe}_2$ ($x = 0.08, 0.12, 0.15, 0.25$) compounds show re-entrant spin glass behavior. Frequency dependence of ac susceptibility, relaxation in ZFC dc magnetization below the freezing temperature and intrinsic remanence are shown to establish the re-entrant spin glass state. We show here that the freezing temperature follows the critical slowing-down mechanism and that the parameters obtained are comparable to those of typical spin glasses and other known RSG systems. The RSG state is attributed to the random distribution of Er and the modification of the Ce and Fe moments due to the band structural changes brought about by the lattice

expansion. We further conclude that the occurrence and the strength of the RSG state in this system are dependent on the Er concentration. With increase in Er concentration, the system is found to gradually change to a non-collinear ferrimagnet and finally to a normal ferrimagnet at $x = 1$. The fact that all rare earths do not give rise to these features makes this study interesting.

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