

## Magnetism in gallium-doped CeFe<sub>2</sub>: Martensitic scenario

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Ce(Fe<sub>1-x</sub>Ga<sub>x</sub>)<sub>2</sub> compounds with  $x=0, 0.01, 0.025,$  and  $0.05$  have been investigated to unravel the effect of Ga on the magnetic state of CeFe<sub>2</sub>. We find that the dynamic antiferromagnetic phase present in CeFe<sub>2</sub> gets stabilized with Ga substitution. The hysteresis loops show that while the compounds with  $x=0$  and  $0.01$  show normal ferromagnetic behavior, the other two show sharp multiple magnetization steps across the antiferromagnetic-ferromagnetic transition region. Temperature and time dependences of magnetization show that the compounds with  $x \geq 0.025$  possess glassy behavior at low temperatures. The experimental findings clearly establish the fact that among the intermetallics, the Ga-doped CeFe<sub>2</sub> shows the closest resemblance with the martensitic scenario seen in the phase-separated magnetic oxides.

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### I. INTRODUCTION

Rare earth (*R*)-iron phase with the composition  $RFe_2$  generally crystallizes in the fcc Laves-phase structure and are simple ferromagnets or ferrimagnets.<sup>1-3</sup> Many members of this series have their Curie temperatures ( $T_C$ ) well above the room temperature and many of them show large magnetostriction. The *R*-Fe coupling is known to be ferromagnetic (FM) in the case of light rare earths and antiferromagnetic (AFM) in the case of heavy rare earths. The strong hybridization between Ce *4f* and Fe *3d* electrons makes CeFe<sub>2</sub> very special in the  $RFe_2$  series. Although Ce is a light rare earth, *4f* electrons couple antiferromagnetically with the *3d* electrons, which is due to the quenching of orbital *4f* moment by the band formation.<sup>1</sup> The lattice parameter of CeFe<sub>2</sub> does not follow the regular trend seen in the  $RFe_2$  series, but is close to that of HoFe<sub>2</sub>. The  $T_C=230$  K is almost lower by a factor of 3 as compared to that of LuFe<sub>2</sub> ( $T_C=610$  K) with its full *4f* shell and YFe<sub>2</sub> ( $T_C=545$  K), which has no *4f* electrons.<sup>2,4</sup> The saturation magnetic moment is also anomalously low ( $M_s=2.4\mu_B/\text{f.u.}$ ) as compared to that of LuFe<sub>2</sub> ( $M_s=2.9\mu_B/\text{f.u.}$ ).<sup>2,4</sup>

Another interesting observation in CeFe<sub>2</sub> is the occurrence of antiferromagnetic fluctuations.<sup>1</sup> It has been reported that substitution of small amounts of elements such as Co, Al, Ru, Ir, Os, and Re stabilizes the low-temperature dynamic AFM phase in CeFe<sub>2</sub>.<sup>3,5-7</sup> The coexistence of FM and AFM phases across the AFM-FM transition in the case of Ru-doped CeFe<sub>2</sub> has been shown by Hall probe imaging, and this transition bears distinct signatures of first-order phase transition, namely, supercooling, superheating, and time relaxation.<sup>8</sup> Very sharp magnetization step across the field-induced AFM-FM transition has been reported at temperatures less than 5 K when CeFe<sub>2</sub> is doped with Ru and Re.<sup>9</sup> Disorder-influenced first-order magnetostructural phase transition is reported to be the main reason for this anomalous behavior.<sup>9</sup> It has also been suggested that the single step may give way to multisteps in the case of single crystalline samples. Among the intermetallic compounds, this type of behavior is not very common and Gd<sub>5</sub>Ge<sub>4</sub> is the notable exception to this general observation.<sup>10</sup> The martensitic na-

ture of the AFM-FM transition is found to be responsible for the jumps in Gd<sub>5</sub>Ge<sub>4</sub>.<sup>11</sup>

Interestingly, single-step and multistep magnetization behaviors are observed in some mixed-valent manganites showing colossal magnetoresistance (CMR), even in polycrystalline form.<sup>12-20</sup> Basically, these materials are phase-separated systems, and the transformation between the two phases has a martensitic character. In addition to the AFM-FM transition, it has been found that the breakdown of charge and orbital ordering also leads to the magnetization jumps in the case of manganites, in general.<sup>15,17</sup> Martensitic accommodation of strain associated with the AFM-FM transition is found to affect the magnetization behavior.<sup>11</sup> Detailed studies have been performed on various manganites using the field sweep rate and time dependence of magnetization.<sup>11,21,22</sup> Wu *et al.*<sup>22</sup> suggested that an induction period exists for these steps to appear, implying that the dynamics of the strain field organization plays a crucial role behind this phenomenon. A few attempts have also been reported wherein the similarities between the intermetallic and oxide scenarios are compared to establish the universality of the phenomena responsible for the step behavior.<sup>11,23</sup>

Results on the Ru- and Re-doped CeFe<sub>2</sub> alloys show that they also possess a phase-separated magnetic structure while varying the field and (or) temperature.<sup>9</sup> In fact, among the substituted CeFe<sub>2</sub> compounds, until date, Ru and Re are the only two substituents that are known to result in sharp magnetization step behavior. In order to study the possibility of stabilizing the AFM state with other elements, we have substituted Ga and found that it is possible to induce the FM-AFM transition at low temperatures with Ga concentration above a critical value. To examine the deeper interconnection between the phase separation and magnetism in these materials, we have made a detailed magnetization study, and the results are presented in this paper. We have also used the sweep rate dependence and time delay measurements to probe the magnetic response.

### II. EXPERIMENTAL DETAILS

Polycrystalline samples of Ce(Fe<sub>1-x</sub>Ga<sub>x</sub>)<sub>2</sub> [ $x=0, 0.01, 0.025,$  and  $0.05$ ] were prepared by arc melting the stoichio-

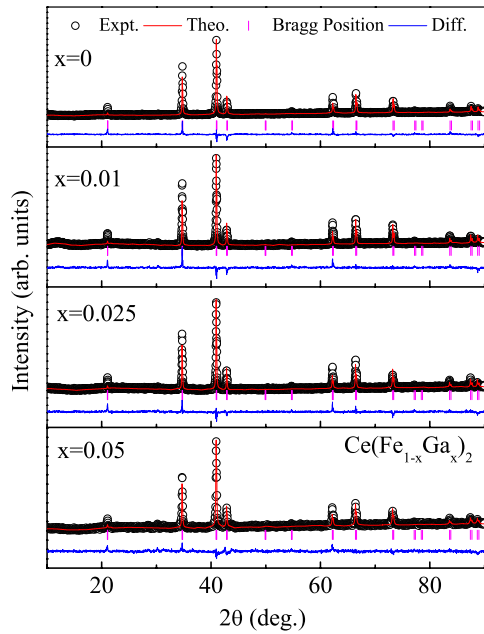


FIG. 1. (Color online) Powder x-ray diffraction patterns, along with the Rietveld refinement of  $\text{Ce}(\text{Fe}_{1-x}\text{Ga}_x)_2$  compounds. The plots at the bottom show the difference between the theoretical and the experimental data.

metric proportion of the constituent elements of at least 99.9% purity, in a water-cooled copper hearth in purified argon atmosphere. The resulting ingots were turned upside down and remelted several times to ensure homogeneity. The weight loss was monitored at the end of the melting process, and the characterization was performed only on samples whose final weight loss was less than 0.5%. The as-cast samples were annealed at 600 °C for two days, 700 °C for five days, 800 °C for two days, and 850 °C for a day.<sup>3</sup> The structural analysis of the samples was performed by collecting the room-temperature powder x-ray diffractograms (XRD) using  $\text{Cu-K}\alpha$  radiation. The refinement of the diffractograms was done by the Rietveld analysis using FULLPROF suite program. The lattice parameters were calculated from the refinement. The dc magnetization measurements in the temperature range of 1.8–300 K and in fields up to 90 kOe were performed with the help of physical property measurement system (PPMS, Quantum Design Model 6500), which has a vibrating sample magnetometer (VSM) attachment. Some measurements were done using Oxford Maglab VSM.

### III. RESULTS

Figure 1 shows the room-temperature powder x-ray diffraction pattern of  $\text{Ce}(\text{Fe}_{1-x}\text{Ga}_x)_2$  compounds along with the Rietveld refinement. The difference plot between the theoretical and the experimentally observed patterns is shown at the bottom of each plot. The refinement confirms that all these compounds are single phase, crystallizing in the  $\text{MgCu}_2$ -type cubic structure, with the space group of  $Fd\bar{3}m$ . The lattice parameters obtained from refinement are 7.3018(3) for  $x=0$ , 7.3059(3) for  $x=0.01$ , 7.3090(3) for  $x=0.025$ , and 7.3097(5) Å for  $x=0.05$ .

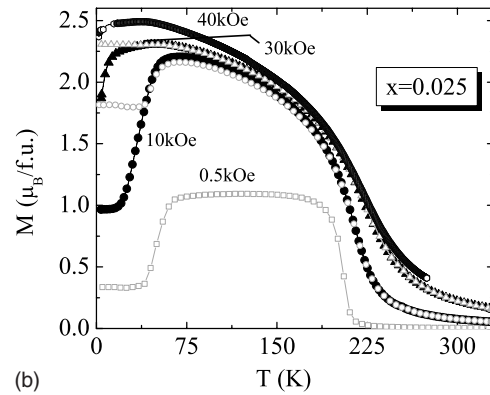
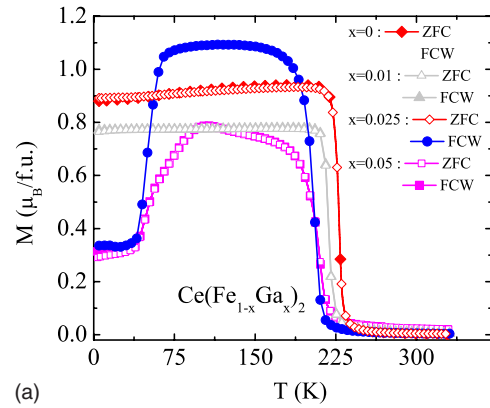


FIG. 2. (Color online) Temperature dependence of the magnetization of (a)  $\text{Ce}(\text{Fe}_{1-x}\text{Ga}_x)_2$  compounds at 500 Oe and (b)  $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$  in various fields. Closed symbols denote the ZFC data and the open symbols denote the FCW data.

The temperature ( $T$ ) variation in magnetization ( $M$ ) of  $\text{Ce}(\text{Fe}_{1-x}\text{Ga}_x)_2$  compounds has been studied in an applied field of 500 Oe both under “zero-field-cooled” (ZFC) and “field-cooled warming” (FCW) conditions. In both the modes, the data were collected during the warming cycle. In all the compounds, at low fields, both ZFC and FCW data follow almost the same path as the temperature is varied. Figure 2(a) shows the  $M$  vs  $T$  plots of  $\text{Ce}(\text{Fe}_{1-x}\text{Ga}_x)_2$  samples. In all the compounds, the transition around 225 K corresponds to the FM-paramagnetic (PM) transition. It can be seen that for  $x \geq 0.025$ , the unstable antiferromagnetic state of  $\text{CeFe}_2$  gets stabilized, as indicated by the low-temperature transition. Stabilization of the AFM phase in  $\text{CeFe}_2$  has been reported earlier with Co, Al, Ru, Ir, Os, and Re doping.<sup>3,5–7</sup> Another effect of Ga substitution is the gradual decrease in  $T_C$ . Figure 2(b) shows the typical field variation in the  $M$ - $T$  plots for the compound with  $x=0.025$ . The antiferromagnetic transition gets gradually suppressed by the field, and at 40 kOe it is almost completely suppressed. Analysis of the temperature variation in magnetization has shown that the magnetization at low temperature is dictated predominantly by the spin wave excitations than the Stoner excitation, which is in agreement with the observations in the Co-doped  $\text{CeFe}_2$ .<sup>24</sup>

Figure 3(a)–3(c) shows the isothermal magnetization curves below 3 K for various concentrations of Ga-doped compounds. All the measurements have been made with a

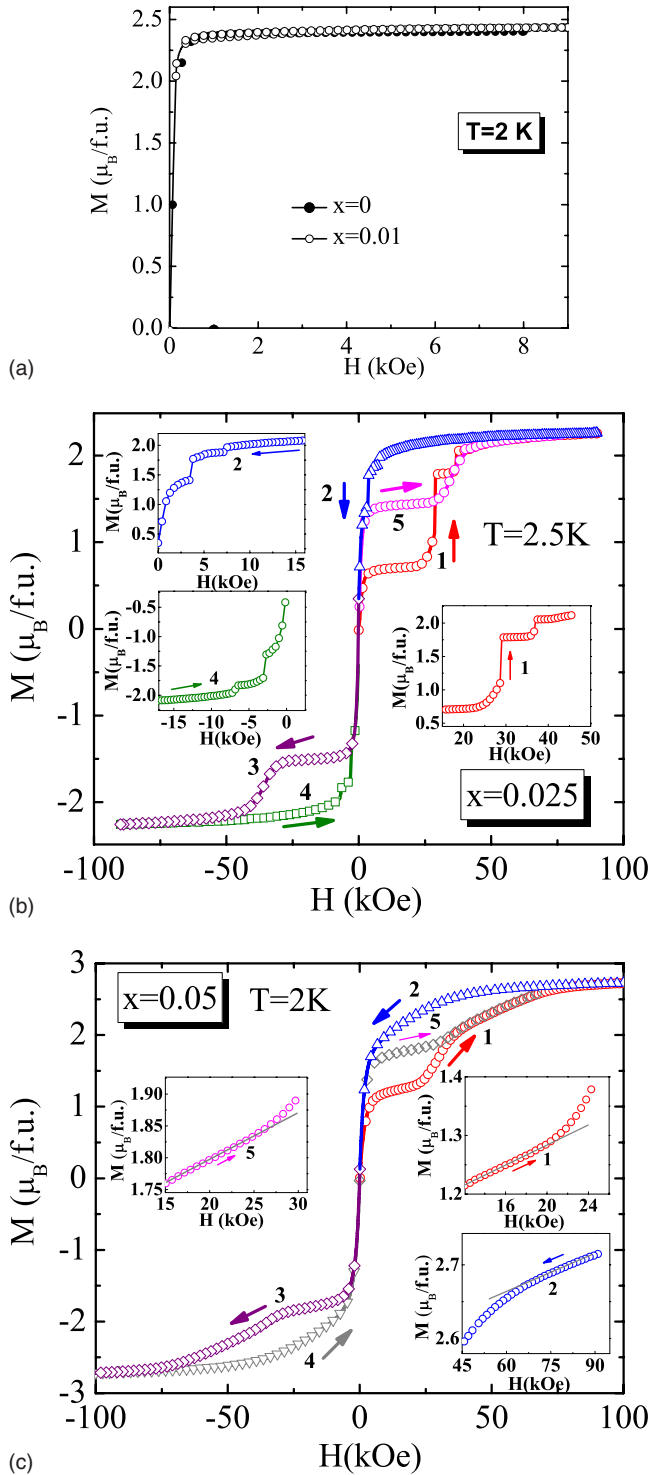


FIG. 3. (Color online)  $M$ - $H$  isotherms of (a)  $\text{CeFe}_2$  and  $\text{Ce}(\text{Fe}_{0.99}\text{Ga}_{0.01})_2$  at 2 K, (b)  $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$  at 2.5 K, and (c)  $\text{Ce}(\text{Fe}_{0.95}\text{Ga}_{0.05})_2$  at 2 K. The samples have been cooled in zero field to the measurement temperature. The arrows indicate the directions of field change. The insets in (b) and (c) highlight the expanded regions of the magnetization curve.

field sweep rate of 100 Oe/s. The samples were zero-field cooled to the measurement temperature. The set temperature was almost constant throughout the measurement time. It can be seen from Fig. 3(a) that the compounds with  $x=0$  and

0.01 show the normal ferromagnetic behavior. Both these compounds show saturation even in very low fields. The saturation moment observed ( $2.4\mu_B/\text{f.u.}$ ) in the case of  $\text{CeFe}_2$  matches very well with the theoretical value.<sup>2</sup> In contrast to these two compounds, the ones with  $x=0.025$  and 0.05 show distinct jumps in the magnetization curves, as can be seen from Fig. 3(b) and 3(c). We define a critical field ( $H_c$ ) at which  $M$ - $H$  curve changes the slope abruptly. It is important to note that the value of  $H_c$  depends on the details of measurement procedure and the thermal and magnetic history of the sample.

We have already seen that the fluctuating AFM state becomes stable in the compounds with  $x=0.025$  sample and 0.05 below 50 K [Fig. 2(a)]. Five quadrant  $M$ - $H$  isotherms have been taken on these compounds at temperatures below 3 K. The AFM phase gets converted to FM phase during the field increment from 0 to 90 kOe, giving rise to sharp multiple transitions in this path [path 1, inset of Fig. 3(b)] in the  $x=0.025$  sample, whereas comparatively smooth and single transition (at  $H_{c1}=20$  kOe) is observed in  $x=0.05$  sample [path 1, inset of Fig. 3(c)]. As mentioned earlier, it has been reported that Ru- and Re-doped  $\text{CeFe}_2$  show a single sharp magnetization step across the field-induced AFM-FM transition when the measurement is performed below 5 K.<sup>9</sup>

When the field is reduced, the system again goes to AFM state but in a relatively gradual way. In the case of  $x=0.025$  a few small steps are observed in path 2 [path 2, inset of Fig. 3(b)], which is absent in  $x=0.05$  sample. The growth of AFM phase with the decrease in  $H$  occurs at fields which are larger than the fields at which the AFM to FM transition occurs in the increasing field cycle, in both the compounds. The asymmetry between the increasing and decreasing field cycles is due to the supercooling associated with a first-order phase transition.<sup>25</sup> It is to be noted that this kind of asymmetry has not been reported in CMR manganites and  $\text{Gd}_5\text{Ge}_4$ .<sup>9</sup> Furthermore, the envelope curve lies inside the virgin curve. Overall, it is found that there is an excellent similarity between the magnetization behavior shown by the present system and Ru- or Re-doped  $\text{CeFe}_2$ . But, it should be noted that while Ru- or Re-substituted polycrystalline samples did not show the multiple magnetization jumps, Ga-substituted compounds in the present case show this behavior very prominently. Such an observation, though common in manganites, is very rare in the case of intermetallics. In view of the fact that  $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$  presents the best scenario among the compounds in this series with regard to the anomalous behavior, we have subjected this compound to some additional measurements.

Figure 4 shows the magnetization isotherms taken at four different temperatures after subjecting the sample to thermal cycling. At 3 K, a smooth transition is observed. When the temperature is reduced to 2.6 K, a step-type transition is observed. With further reduction in temperature, the  $M$ - $H$  isotherms consist of a number of ultrasharp steps before it transforms to the fully ferromagnetic phase. It has been reported that many manganites show multiple steps with reduction in temperature.<sup>14-17,26-28</sup> The  $M$ - $H$  curves were also recorded without thermal cycling. Figure 5 shows the two loop magnetization isotherms for  $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$  without thermal cycling. The data was taken as the field was varied in

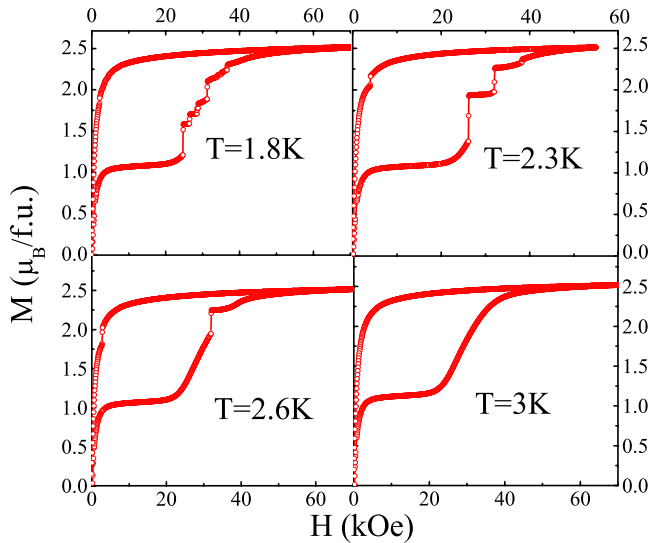


FIG. 4. (Color online) Two-loop magnetization isotherms for  $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$  at various temperatures.

the sequence 0-90-0-65-0 kOe. It can be seen that there is a large difference between the first and the second runs. Though the magnetization goes to zero at the end of the first cycle, during the field increase in the second cycle, the magnetization attains near saturation value in relatively lower fields. In addition, there is only one step which is quite broad, in contrast to the sharp multiple steps seen in the first run. These observations suggest that the thermal and magnetic history play an important role in determining the step behavior in this compound. Effect of thermomagnetic history has been demonstrated in Ru-doped  $\text{CeFe}_2$  as well.<sup>7,29</sup>

The dependence of the magnetization on the cooling field was also studied in  $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ . Figure 6 shows the field dependence of the ZFC and FCW magnetization at  $T = 1.8$  K. The sample was heated above 240 K ( $>T_C$ ) and then cooled to 1.8 K for each measurement. In the FCW mode, the cooling field was reduced to zero at the measurement temperature, and the magnetization was measured subsequently by increasing the field. The low-field magnetization is found to increase with increase in the cooling field, which implies that the FM fraction increases with cooling

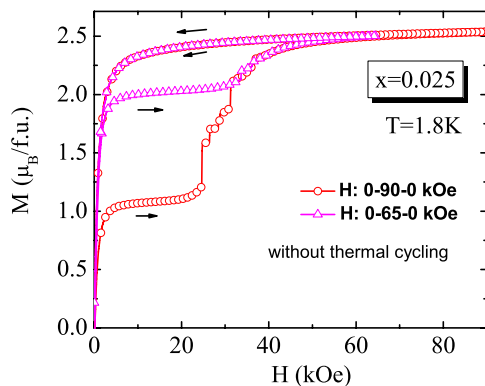


FIG. 5. (Color online) Two-loop magnetization isotherms for  $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$  without thermal cycling. The data was taken in the sequence 0-90-0-65-0 kOe.

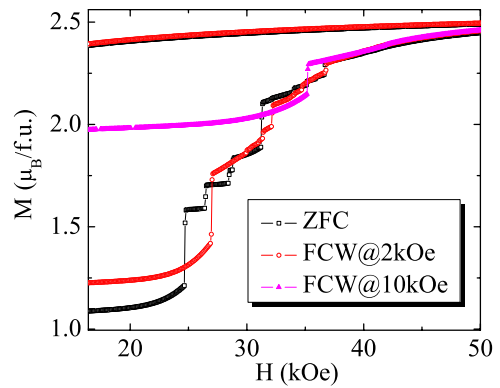


FIG. 6. (Color online) Field dependence of ZFC and FCW magnetization at  $T = 1.8$  K in  $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ . During FCW measurement, the cooling field was made zero at the measurement temperature, then data were taken for the increasing and decreasing field cycles.

field. A cooling field of 10 kOe almost completely converts the AFM phase into FM phase, without any steps. However, comparing the ZFC and FCW data, it may be noted that the steps are shifted to higher fields, as the cooling field is increased. Furthermore, it can be seen that the number of steps in the  $M$ - $H$  curve has decreased in the FCW curves, as compared to the ZFC curve. These observations are very similar to the scenario reported in some phase-separated manganites.<sup>14</sup> It is quite surprising that the increase in the cooling field leads to an increase in the step fields, even while increasing the FM fraction. This behavior can be explained on the basis of nucleation and growth of FM clusters in the AFM matrix. Due to the structural differences between the FM and AFM phases, there is elastic-misfit energy present between the two phases. According to the theory of nucleation, when clusters form in solids, elastic-misfit strain energy is generally present because of volume and/or shape incompatibilities between the cluster and the matrix. The strain energy term, which is positive, acts along with the interfacial energy term and acts as a barrier to the nucleation. The magnitude of the elastic energy term generally depends upon factors such as cluster shape, the mismatch between the cluster and the matrix, etc.<sup>30</sup> Increment in the FM fraction in the field-cooled (FC) case indicates increased growth of FM phase in AFM matrix compared to ZFC case. This increases the positive term of the free-energy change during nucleation, compared to the ZFC case. As a result of this it is hard to grow to a completely new phase (FM) because it acts a barrier to nucleation, resulting in an upward shift in critical field under the FC mode.

In view of the strong similarities between the oxides and the present compound, the effect of changing the sweep rate of the field on the magnetization behavior was also investigated. Figure 7 shows the  $M$ - $H$  plots of  $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$  with sweep rates of 0.5 kOe/min and 4 kOe/min. Here also, the sample was heated above the  $T_C$  before each measurement. It is quite evident from this figure that the sweep rate has a significant role in determining the number of steps in the magnetization isotherms. Also, there seems to be a general trend that as the sweep rate increases, the steps occur at

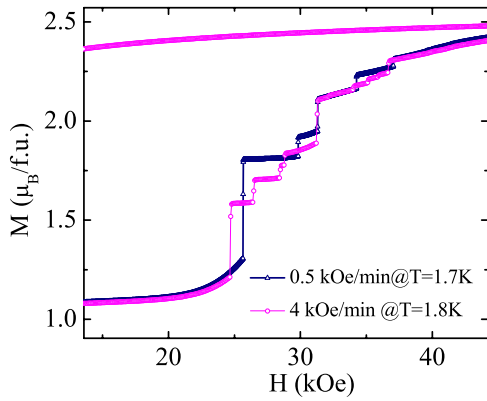


FIG. 7. (Color online) Two loop  $M$ - $H$  isotherms measured for  $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$  at  $T=1.8$  K with a field sweep rates of 0.5 kOe/min and 4 kOe/min. The sample was zero-field cooled from 240 K before each measurement.

lower fields. These observations are also identical to the behavior of phase-separated manganites and  $\text{Gd}_5\text{Ge}_4$ .<sup>11,15,17,20</sup> In the case of manganites, the different crystal structures of the FM and AFM phases in the phase-separated state is responsible for the sweep rate dependence. The sweep rate dependence of the step behavior is a signature of martensitic strains in the system.<sup>11,26</sup>

The effect of sweep rate was also investigated in another manner as shown in Fig. 8, which compares the  $M$ - $H$  curves obtained in the normal sweep and in the interrupted sweep. In the normal sweep, the data have been taken continuously in time as the field was increased from 0 to 70 kOe. A step at about 32 kOe was found in this curve. In the interrupted sweep, 20 kOe was maintained for 1.5 h and 32 kOe was maintained for 1 h. A step is observed at 20 kOe when the field was held for 1.5 h, which was not observed in the normal sweep. At 32 kOe, when it was held for 1 h, it gave rise to an additional step. Similar observations have been made in certain manganites as well.<sup>21,22</sup> These observations also lend credence to the martensitic scenario expected for the present compound. This also suggests that the occurrence and number of steps in the magnetization isotherms are determined by the dynamics of the martensitic strain distribution.

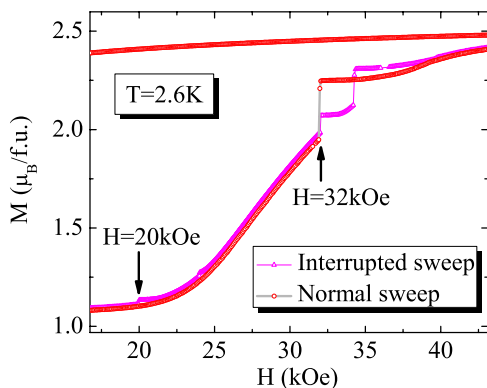


FIG. 8. (Color online) Isothermal ZFC magnetization at  $T=2.6$  K of  $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$  in a typical sweep and in an interrupted sweep. In interrupted sweep, fields of 20 kOe and 32 kOe were held constant for 1.5 h and 1 h.

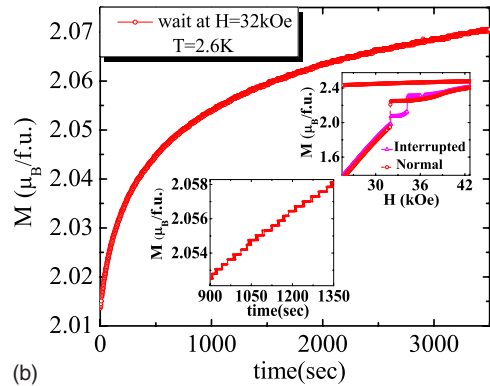
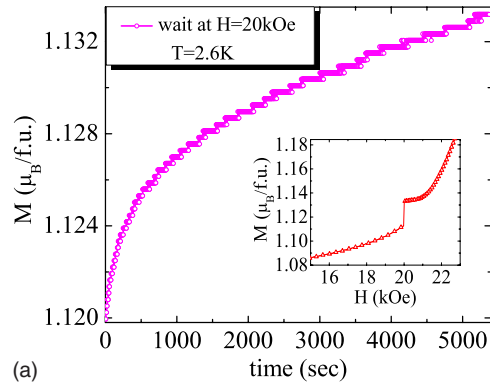


FIG. 9. (Color online) Time evolution of isothermal (ZFC) magnetization at 2.6 K for  $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$  during the holding time at (a) 20 kOe and (b) 32 kOe. Insets show the magnetization jumps during the holding time.

During the holding times, the magnetization grows in small steps to its final value (Fig. 9). The size of these steps is found to be become smaller at  $H=32$  kOe, as can be seen from Figs. 9(a) and 9(b). The growth of magnetization with time is found to show a stretched exponential behavior, implying that the low-temperature magnetic state is similar to that of glassy systems.<sup>22,31</sup> Insets in these figures show the magnetization jumps during the holding time.

Figure 10 demonstrates the nature of high-temperature  $M$ - $H$  isotherms of the compounds with  $x=0.025$  and  $x=0.05$ . The transition is rather continuous, and the critical field for the growth of FM phase in  $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$  is about 14 kOe, which is less compared to the value of 24 kOe obtained at 2.5 K. Further increase in temperature makes the transition broader, and the area of the hysteresis loop also becomes smaller. For  $\text{Ce}(\text{Fe}_{0.95}\text{Ga}_{0.05})_2$ , the transition is not as sharp as in  $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ , as we have seen earlier. The AFM-FM transition temperatures for these compounds are around 50 K, as found from the  $M$ - $T$  curve (Fig. 2).

In order to understand the nature of the AFM-FM and FM-PM transitions, the Arrott's plots have been recorded for the compounds with  $x=0.025$  and 0.05 and are shown in Fig. 11. S-shaped nature of the Arrott's plots clearly suggests that the AFM-FM transition is first order in nature.<sup>32</sup> However, the FM-PM transition is found to be of second order. Also, the strength of first order is more in the case of  $x=0.025$ , as compared to that of  $x=0.05$ , which is consistent with the difference in the anomalous behavior between these two

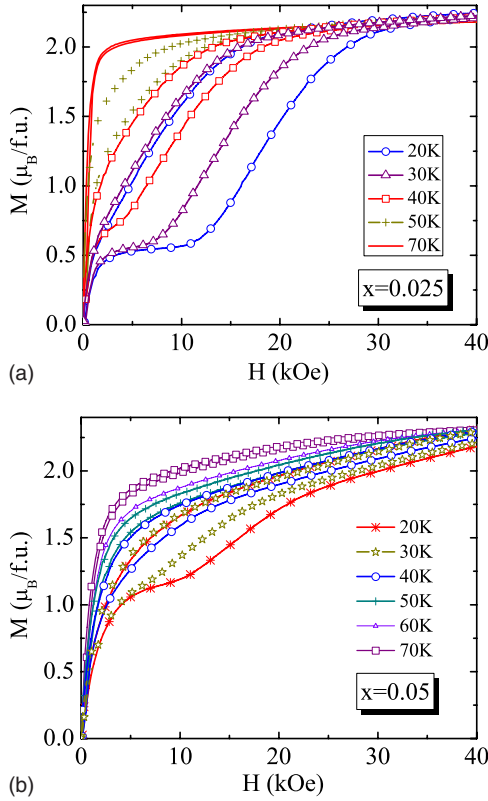


FIG. 10. (Color online) Isothermal magnetization at high temperatures ( $\geq 20$  K) in (a)  $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$  and (b)  $\text{Ce}(\text{Fe}_{0.95}\text{Ga}_{0.05})_2$ .

compounds. Therefore, the martensitic scenario presumed in this system earlier is supported by the order of phase transition.

Based on the  $M$ - $H$  curves obtained at various temperatures the magnetic phase diagram has been constructed for the compounds with  $x=0.025$  and  $0.05$  and is shown in Fig. 12. In this figure,  $H_{c1}$  and  $H_{c2}$  refer to the lower and upper critical fields.

IV. DISCUSSION

The major findings from the results presented in the above section are (i) stabilization of low-temperature antiferromag-

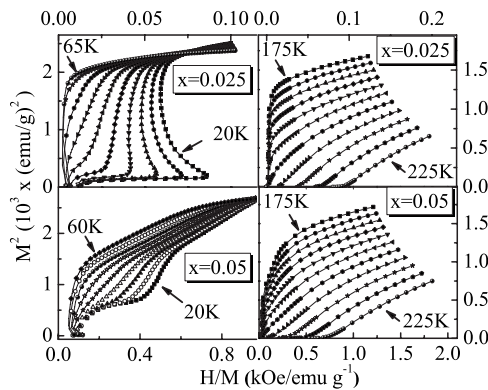


FIG. 11. Arrott's plots for  $x=0.025$  and  $x=0.05$  compounds at AFM-FM and FM-PM transition regions.

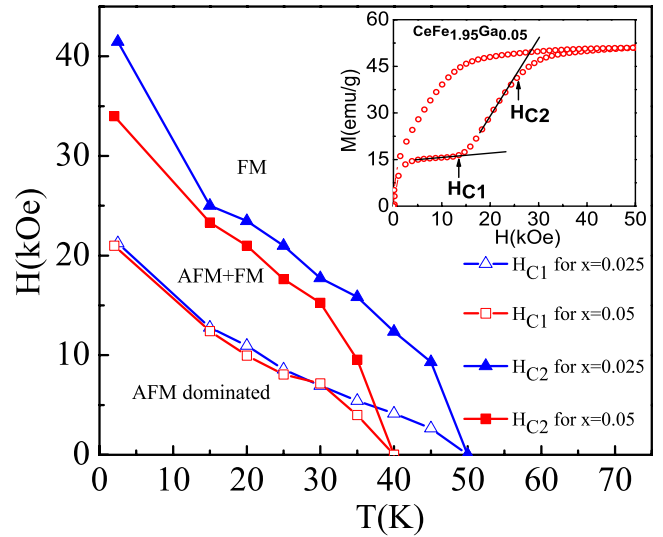


FIG. 12. (Color online) Magnetic phase diagram for  $\text{Ce}(\text{Fe}_{1-x}\text{Ga}_x)_2$  compounds with  $x=0.025$  and  $0.05$

netic phase with Ga substitution, (ii) “ultrasharp and multi-step” behavior in the magnetization isotherms in the Ga-substituted compounds across the AFM-FM transition, (iii) the fact that the virgin curve lies outside the envelope curve, (iv) change in the order of the AFM-FM transition and the anomalous behavior as a function of Ga concentration, (v) anomalous sweep rate and cooling field dependences of magnetization, and (vi) the similarity between these compounds and the martensitic oxides with regard to the magnetization relaxation behavior.

As mentioned earlier, the features seen in the magnetization data of Ga-doped  $\text{CeFe}_2$  compounds are very similar to those seen in many phase-separated oxide materials.<sup>14–20</sup> Among the intermetallics,  $\text{Gd}_5\text{Ge}_4$  is known to show many similarities with the oxides.<sup>11,23</sup> Though there are several factors thought to be responsible for the multistep behavior, the most striking and accepted reason common to the oxide materials and the intermetallics seems to be the martensitic transition driven by the applied field. It has been proposed that the temperature<sup>33</sup> and the magnetic-field-induced<sup>12</sup> phase evolution in these systems resemble that of a martensite. According to this view, the mechanism behind the steps is attributed to the catastrophic relief of strain developed during the first-order martensitic phase transition from the AFM to the FM state.<sup>22</sup> It has been shown that avalanchelike behavior is expected in martensitic systems at low temperatures. Though Ru- and Re-substituted  $\text{CeFe}_2$  is known to show many of the anomalies shown by the present series of compounds, the sharp multistep magnetization behavior is found to be present only in the latter case. It is also important that Ga-substituted compounds show such a distinct behavior even in polycrystalline form.

In the present case, the FM phase grows above 50 K for  $x=0.025$  and  $x=0.05$  samples. It is well documented that the magnetic phase transition in these kinds of compounds is also associated with structural transition from rhombohedral AFM phase to cubic FM phase.<sup>25,29,34,35</sup> Consequently, this system appears to behave like a phase-separated system,

where FM phase can be induced by applying field externally. The sweep rate dependence also confirms the proposition of the martensitic nature of this system. The appearance of sharper steps under a fast sweep rate, compared to that with a slow sweep rate, is a characteristic of martensitic systems.<sup>11,26</sup> A slower sweep is supposed to transform the two phases smoothly, compared to the higher sweep rate. A gradual accommodation of martensitic strains during the field-induced order-order transition leads to higher critical field and fewer steps.<sup>11</sup> In addition, the relaxation behavior, similar to the one seen in the present case, has been observed in many phase-separated oxides and Gd<sub>5</sub>Ge<sub>4</sub>. The kind of relaxation seen in these systems suggests that the magnetic states are metastable. The various metastable states are separated by energy barriers associated with strain, magnetic field, or temperature. The competition among these contributions determines the nature of the response of the system to an applied field. Until now, only Ru- and Re-doped<sup>9</sup> CeFe<sub>2</sub> are reported to show the strain-induced first-order magnetostructural transition, and therefore, the present study gives yet another evidence for this anomalous behavior of the CeFe<sub>2</sub>-based systems.

Another point to be noted is that the sharp and multiple steps seen in  $x=0.025$  have given way to a broad single step in  $x=0.05$ . This can be attributed to the weakening of the first-order nature of the AFM-FM transition with increase in Ga. Considering the various substitutions carried out in CeFe<sub>2</sub>, it can be inferred that the magnetization steps and the related effects are seen prominently when the region of coexistence of FM-AFM phases is narrow.<sup>34</sup> Therefore, it is quite likely that though AFM state may be stabilized with a certain substituent, it need not necessarily give rise to the step behavior. One cannot rule out the role of grain-size variation (as a function of Ga content) and its effect in absorbing the martensitic strains to explain the difference seen between the compounds with  $x=0.025$  and  $0.05$ .<sup>33</sup> The varia-

tions in the antiferromagnetic domain size may also contribute to the difference in the magnetization behavior between various Ga concentrations.<sup>14</sup> Finally, we also feel that the difference in the step behavior seen between Ru- or Re-doped compounds and the Ga-doped compounds arises due to the differences in their microstructure, given the fact that the AFM-FM transition is of first order in both the cases.

## V. CONCLUSIONS

The present study shows that Ga doping stabilizes the dynamic AFM state in CeFe<sub>2</sub>. The FM-AFM transition is found to be of first order in the case of  $x=0.025$  and  $0.05$ . We find that phenomena such as strain-induced first-order jumps in the magnetization curves, asymmetry between the  $M$ - $H$  curves during the increasing and decreasing field cycles, the envelope curve being inside the virgin curve, occur in these compounds as well. Thermomagnetic history is found to influence the magnetization behavior. It is found that the magnetization steps can be induced by proper relaxation procedure. Experimental evidences clearly show that the system behaves like a glassy phase at low temperatures. The results, in general, show that Ga-doped CeFe<sub>2</sub> ( $\geq 0.025$ ) shows a martensiticlike behavior due to the strong magnetostructural coupling. The experimental finding from the present work clearly establishes the fact that among the intermetallics, the Ga-doped CeFe<sub>2</sub> shows the closest resemblance with the martensitic scenario seen in the phase-separated oxides.

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<sup>1</sup>L. Paolasini, B. Ouladdiaf, N. Bernhoeft, J.-P. Sanchez, P. Vulliet, G. H. Lander, and P. Canfield, Phys. Rev. Lett. **90**, 057201 (2003).

<sup>2</sup>O. Eriksson, L. Nordstrom, M. S. S. Brooks, and B. Johansson, Phys. Rev. Lett. **60**, 2523 (1988).

<sup>3</sup>S. B. Roy and B. R. Coles, J. Phys.: Condens. Matter **1**, 419 (1989).

<sup>4</sup>C. Giorgetti, S. Pizzini, E. Dartyge, A. Fontaine, F. Baudelet, C. Brouder, P. Bauer, G. Krill, S. Miraglia, D. Fruchart, and J. P. Kappler, Phys. Rev. B **48**, 12732 (1993).

<sup>5</sup>S. B. Roy and B. R. Coles, Phys. Rev. B **39**, 9360 (1989).

<sup>6</sup>A. K. Rajarajan, S. B. Roy, and P. Chaddah, Phys. Rev. B **56**, 7808 (1997).

<sup>7</sup>K. J. Singh, S. Chaudhary, M. K. Chattopadhyay, M. A. Manekar, S. B. Roy, and P. Chaddah, Phys. Rev. B **65**, 094419 (2002).

<sup>8</sup>S. B. Roy, G. K. Perkins, M. K. Chattopadhyay, A. K. Nigam, K. J. S. Sokhey, P. Chaddah, A. D. Caplin, and L. F. Cohen, Phys. Rev. Lett. **92**, 147203 (2004).

<sup>9</sup>S. B. Roy, M. K. Chattopadhyay, P. Chaddah, and A. K. Nigam, Phys. Rev. B **71**, 174413 (2005).

<sup>10</sup>E. M. Levin, K. A. Gschneidner, Jr., and V. K. Pecharsky, Phys. Rev. B **65**, 214427 (2002).

<sup>11</sup>V. Hardy, S. Majumdar, S. J. Crowe, M. R. Lees, D. McK. Paul, L. Herve, A. Maignan, S. Hebert, C. Martin, C. Yaicle, M. Hervieu, and B. Raveau, Phys. Rev. B **69**, 020407(R) (2004).

<sup>12</sup>S. Hebert, V. Hardy, A. Maignan, R. Mahendiran, M. Hervieu, C. Martin, and B. Raveau, J. Solid State Chem. **165**, 6 (2002); Solid State Commun. **122**, 335 (2002).

<sup>13</sup>A. Maignan, S. Hebert, V. Hardy, C. Martin, M. Hervieu, and B. Raveau, J. Phys.: Condens. Matter **14**, 11809 (2002).

<sup>14</sup>R. Mahendiran, A. Maignan, S. Hebert, C. Martin, M. Hervieu, B. Raveau, J. F. Mitchell, and P. Schiffer, Phys. Rev. Lett. **89**, 286602 (2002).

<sup>15</sup>D. S. Rana and S. K. Malik, Phys. Rev. B **74**, 052407 (2006).

<sup>16</sup>S. Nair, A. K. Nigam, A. V. Narlikar, D. Prabhakaran, and A. Boothroyd, Phys. Rev. B **74**, 132407 (2006).

<sup>17</sup>D. S. Rana, D. G. Kuberkar, and S. K. Malik, Phys. Rev. B **73**, 064407 (2006).

- <sup>18</sup>F. M. Woodward, J. W. Lynn, M. B. Stone, R. Mahendiran, P. Schiffer, J. F. Mitchell, D. N. Argyriou, and L. C. Chapon, *Phys. Rev. B* **70**, 174433 (2004).
- <sup>19</sup>V. Hardy, S. Majumdar, M. R. Lees, D. McK. Paul, C. Yaicle, and M. Hervieu, *Phys. Rev. B* **70**, 104423 (2004).
- <sup>20</sup>V. Hardy, S. Hebert, A. Maignan, C. Martin, M. Hervieu, and B. Raveau, *J. Magn. Magn. Mater.* **264**, 183 (2003).
- <sup>21</sup>V. Hardy, A. Maignan, S. Hebert, C. Yaicle, C. Martin, M. Hervieu, M. R. Lees, G. Rowlands, D. M. Paul, and B. Raveau, *Phys. Rev. B* **68**, 220402(R) (2003).
- <sup>22</sup>T. Wu and J. F. Mitchell, *Phys. Rev. B* **69**, 100405(R) (2004).
- <sup>23</sup>K. Kumar, A. K. Pramanik, A. Banerjee, P. Chaddah, S. B. Roy, S. Park, C. L. Zhang, and S.-W. Cheong, *Phys. Rev. B* **73**, 184435 (2006).
- <sup>24</sup>H. Fukuda, H. Fujii, H. Kamura, Y. Hasegawa, T. Ekino, N. Kikugawa, T. Suzuki, and T. Fujita, *Phys. Rev. B* **63**, 054405 (2001).
- <sup>25</sup>M. K. Chattopadhyay, S. B. Roy, A. K. Nigam, K. J. S. Sokhey, and P. Chaddah, *Phys. Rev. B* **68**, 174404 (2003).
- <sup>26</sup>D. Q. Liao, Y. Sun, R. F. Yang, Q. A. Li, and Z. H. Cheng, *Phys. Rev. B* **74**, 174434 (2006).
- <sup>27</sup>Z. W. Ouyang, V. K. Pecharsky, K. A. Gschneidner, Jr., D. L. Schlager, and T. A. Lograsso, *Phys. Rev. B* **76**, 134406 (2007).
- <sup>28</sup>H. Tang, V. K. Pecharsky, K. A. Gschneidner, Jr., and A. O. Pecharsky, *Phys. Rev. B* **69**, 064410 (2004).
- <sup>29</sup>M. A. Manekar, S. Chaudhary, M. K. Chattopadhyay, K. J. Singh, S. B. Roy, and P. Chaddah, *Phys. Rev. B* **64**, 104416 (2001).
- <sup>30</sup>Robert W. Balluffi, Samuel M. Allen, and W. Craig Carter, *Kinetics of Materials* (Wiley, Hoboken, 2005).
- <sup>31</sup>R. Hoogerbeets, W.-L. Luo, and R. Orbach, *Phys. Rev. B* **34**, 1719 (1986).
- <sup>32</sup>N. K. Singh, K. G. Suresh, A. K. Nigam, S. K. Malik, A. A. Coelho, and S. Gama, *J. Magn. Magn. Mater.* **317**, 68 (2007).
- <sup>33</sup>V. Podzorov, B. G. Kim, V. Kiryukhin, M. E. Gershenson, and S. W. Cheong, *Phys. Rev. B* **64**, 140406(R) (2001).
- <sup>34</sup>S. J. Kennedy and B. R. Coles, *J. Phys.: Condens. Matter* **2**, 1213 (1990).
- <sup>35</sup>J. S. Sokhey, M. K. Chattopadhyay, A. K. Nigam, S. B. Roy, and P. Chaddah, *Solid State Commun.* **129**, 19 (2004).