

Magnetostructural transition in Ce (Fe 0.975 Ga 0.025) 2 compound

Arabinda Haldar, Niraj K. Singh, Ya. Mudryk, Ajaya K. Nayak, K. G. Suresh, A. K. Nigam, and V. K. Pecharsky

Citation: *Journal of Applied Physics* **107**, 09E133 (2010); doi: 10.1063/1.3365064

View online: <http://dx.doi.org/10.1063/1.3365064>

View Table of Contents: <http://scitation.aip.org/content/aip/journal/jap/107/9?ver=pdfcov>

Published by the [AIP Publishing](#)

Articles you may be interested in

[Pressure induced magneto-structural phase transitions in layered RMn 2 X 2 compounds \(invited\)](#)

J. Appl. Phys. **115**, 172617 (2014); 10.1063/1.4870582

[Elastically controlled magnetic phase transition in Ga-FeRh/BaTiO3\(001\) heterostructure](#)

Appl. Phys. Lett. **104**, 022401 (2014); 10.1063/1.4861455

[Structural and magnetic properties of the Heusler compound Fe2MnGa](#)

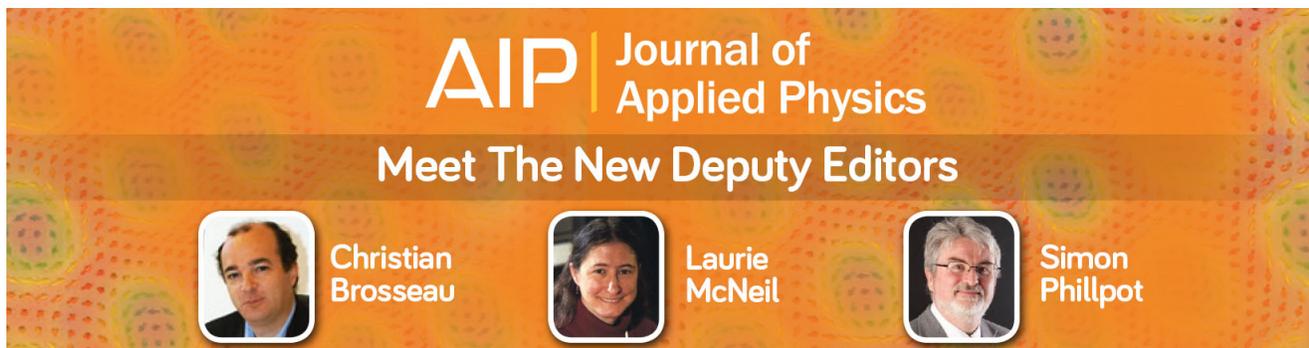
J. Appl. Phys. **113**, 17E301 (2013); 10.1063/1.4794979

[Effect of fast neutron irradiation induced defects on the metamagnetic transition In Ce\(Fe0.96Ru0.04\)2](#)

J. Appl. Phys. **112**, 063922 (2012); 10.1063/1.4754577

[Giant exchange bias based on magnetic transition in \$\gamma\$ -Fe 2 MnGa melt-spun ribbons](#)

Appl. Phys. Lett. **97**, 242513 (2010); 10.1063/1.3526377



AIP | Journal of Applied Physics

Meet The New Deputy Editors

	Christian Brosseau		Laurie McNeil		Simon Phillpot
---	---------------------------	---	----------------------	---	-----------------------

Magnetostructural transition in $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ compound

Arabinda Haldar,^{1,a)} Niraj K. Singh,² Ya. Mudryk,² Ajaya K. Nayak,¹ K. G. Suresh,¹ A. K. Nigam,³ and V. K. Pecharsky^{2,4}

¹Department of Physics, Indian Institute of Technology Bombay, Mumbai 400076, India

²The Ames Laboratory U. S. Department of Energy, Iowa State University, Ames, Iowa 50011-3020, USA

³Tata Institute of Fundamental Research, Homi Bhabha Road, Mumbai 400005, India

⁴Department of Materials Science and Engineering, Iowa State University, Ames, Iowa 50011-2300, USA

(Presented 19 January 2010; received 21 October 2009; accepted 17 November 2009; published online 5 May 2010)

The magnetic and magnetostructural properties of the polycrystalline $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ have been investigated as a function of temperature and magnetic field. In $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ the magnetic transition from antiferromagnetic (AFM) to ferromagnetic (FM) state is accompanied by a structural transformation from rhombohedral to cubic structure. Phase coexistence is present during both the temperature and field driven transformations from AFM to FM phase. © 2010 American Institute of Physics. [doi:10.1063/1.3365064]

I. INTRODUCTION

Different classes of magnetic materials exhibiting characteristic features of the first order phase transition, such as steps in magnetization isotherms, phase coexistence, field sweep rate dependence, superheating/supercooling, unusual relaxation, etc., have attracted considerable interest recently.^{1–12} Among various materials, doped CeFe_2 compounds have evinced the above features of the first order transition rather convincingly.^{11–14} The occurrence of these exotic properties is believed to be a consequence of the magnetostructural transition.

Recently, we have reported the magnetic properties of $\text{Ce}(\text{Fe}_{1-x}\text{Ga}_x)_2$.¹⁵ It was shown that in $\text{Ce}(\text{Fe}_{1-x}\text{Ga}_x)_2$, the low temperature antiferromagnetic (AFM) state can be stabilized only for $x \geq 0.025$. Metamagnetic transition from the AFM to the ferromagnetic (FM) state with multiple steps was observed in compounds with the AFM ground state. The AFM-FM transition was shown to exhibit many distinct features of first order phase transition and the martensitic scenario was invoked to explain the observed anomalous properties.¹⁵ Therefore, in order to further improve our understanding of the Ga doped CeFe_2 compounds, we have carried out investigations of the magnetostructural properties of $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ using magnetization measurements and the temperature and field dependent x-ray powder diffraction (XRD) experiments. The latter technique is a unique tool to probe the structural part microscopically as a function of temperature as well as magnetic field.^{16–18} Our in-field XRD investigations reveal that in $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ the magnetic and structural degrees of freedom are intimately coupled.

II. EXPERIMENTAL DETAILS

The polycrystalline $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ compound was prepared as described in Ref. 15. Magnetization measurements were carried using a commercial Physical Property Measurement System (Quantum Design model 6500), which

has a vibrating sample magnetometer attachment. The temperature (10–295 K) and field (0–35 kOe) dependent XRD data were collected using a Rigaku TTRAX powder diffractometer with Mo $K\alpha$ radiation.¹⁹ Multiple sets of diffraction data were collected in step scanning mode (0.5–2 s/step) with a 0.01° step of 2θ over the range of $9^\circ \leq 2\theta \leq 45^\circ$. Each data set was analyzed by the Rietveld refinement to determine the unit cell parameters and the phase contents, when two different crystallographic phases coexisted in certain field and temperature regimes.

III. RESULTS AND DISCUSSION

Figure 1 shows the temperature variation of magnetization (M) data of $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ measured in field cooled warming (FCW) mode under an applied field (H) of 500 Oe. We note that owing to the presence of AFM state, the magnetization at low temperatures is quite small. Upon warming, the compound undergoes two magnetic transitions: AFM to FM state and then from FM state to paramagnetic (PM) state. These transitions occur at 63 K (T_N) and 206 K (T_C).

Selected magnetization isotherms of $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ are shown in Fig. 2. We note that in the AFM regime, the isotherms show multiple steps, which are attributed to the field induced AFM-FM transition. The $M(H)$ data also show

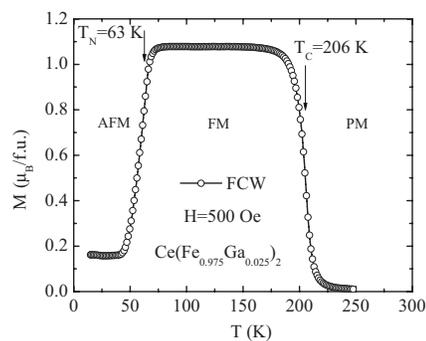


FIG. 1. Temperature variation of magnetization of $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ compound in FCW mode under an applied field (H) of 500 Oe.

^{a)}Electronic mail: arabinda.haldar@gmail.com.

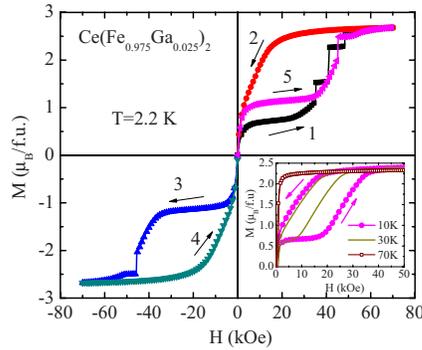


FIG. 2. (Color online) Magnetization isotherm of $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ at $T=2.2$ K. The inset shows the isotherms at higher temperatures.

large hysteresis and the virgin curve (labeled as 1) lies outside the envelope curve (labeled as 5). These features are attributed to the supercooling/superheating effect and the kinetic arrest of first order phase transition.¹⁵ A similar step behavior in magnetization isotherms has been reported in manganites¹⁻⁸ and in a few intermetallic compounds.^{10,11} Similarities in the magnetization behavior in different classes of materials and a universal picture arising from their study have been reported by Roy *et al.*²⁰ We believe that a martensitic scenario arising from the structural mismatch between the FM and the AFM phases leads to the observed step behavior in this system.¹⁵

It was mentioned above that a martensitic scenario may be responsible for the anomalous magnetic properties of $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$. Thus, in order to further understand the $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$, we have carried out the temperature dependent XRD measurements and the results of Rietveld refinement of XRD results are shown in Fig. 3. Here, the

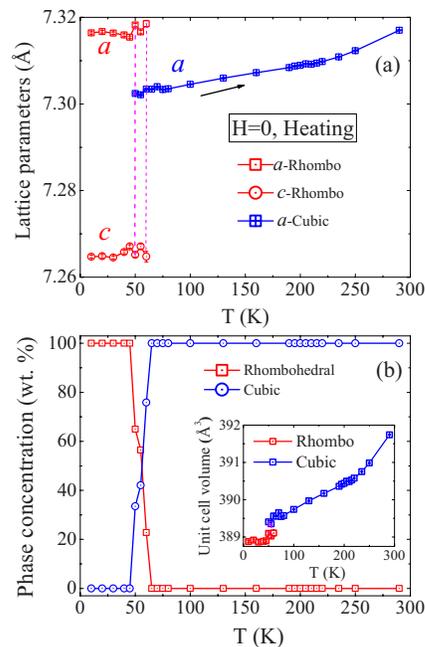


FIG. 3. (Color online) (a) Temperature variation of the normalized lattice parameters of $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ during heating in zero field. (b) Temperature dependencies of the concentrations of the rhombohedral and cubic phases obtained from zero field XRD data. The inset shows the variation in unit cell volume with temperature.

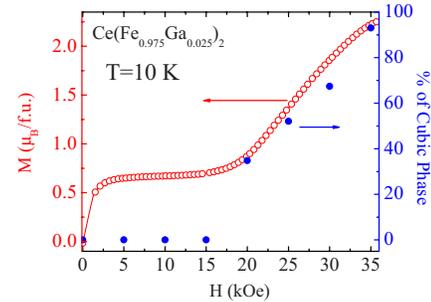


FIG. 4. (Color online) Field (H) dependence of magnetization (M) and the percentage of cubic phase in $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ at $T=10$ K.

sample was zero field cooled (ZFC) to 10 K, and XRD data were collected during warming. At 10 K, the compound possesses a rhombohedral structure, which is preserved up to 45 K. With an increase in temperature, the compound transforms from the rhombohedral to the cubic structure and, between 45 and 65 K, both polymorphs coexist. At 65 K, the compound completely adopts the cubic structure and this structure is retained up to room temperature. We note that the lattice parameters of the rhombohedral phase shown in Fig. 3(a) were calculated by the Rietveld refinement using hexagonal setting of the $R\bar{3}m$ space group and the calculated a_{rh} and c_{rh} parameters were modified (a_{rh} was multiplied by $\sqrt{2}$, and c_{rh} was divided by $\sqrt{3}$) in order to be directly comparable with the high-temperature cubic lattice parameter. The unit cell volume [inset in Fig. 3(b)] of the rhombohedral structure was also normalized ($V=V_{\text{rh}}\times 4/3$).

Therefore, the XRD data reveal that in the low temperature AFM phase, $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ possesses the rhombohedral structure, whereas in the high temperature FM and PM phases, it adopts the cubic structure. The same is clearly illustrated in Fig. 3(b), where the phase fractions of different phases as a function of temperature were calculated from x-ray data. We note that the temperature range over which the rhombohedral to cubic transformation occurs [45–65 K (see Fig. 3)] matches well with the broad AFM-FM transition seen in the $M(T)$ data of Fig. 1.

Figure 4 shows the results of the field dependent XRD examination carried out at 10 K along with the $M(H)$ isotherm of $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ measured at the same temperature. Here the sample was ZFC to 10 K and then the field dependent XRD patterns were recorded with a field step of 5 kOe up to a maximum field of 35 kOe. The field dependent XRD data reveal that in zero field the $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ possesses a rhombohedral structure and this structure is preserved up to $H=15$ kOe. However, at $H=20$ kOe, about 35% of the rhombohedral phase is converted into the cubic phase. It is interesting to note that the ZFC $M(H)$ of $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ also shows the metamagnetic phase transition at the same field. With further increase in the field, the concentration of the cubic phase increases and at 35 kOe field, about 93% of the rhombohedral phase is converted into the cubic phase. Thus, the field dependent XRD data reveal that in $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ the metamagnetic transition from AFM to FM state is associated with the field induced structural transformation from rhombohedral to cubic phase. We find that the growth of the cubic phase follows the evolution

of the FM phase (see Fig. 4). Thus, both the temperature and field dependent XRD results indicate that in $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ the magnetic and structural degrees of freedom are intimately coupled.

There are many similarities between the properties of $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ and other materials, which exhibit first order magnetostructural transition.^{16–18} Dependence of magnetic properties on measurement protocols, such as magnetic field sweep rate, time delay, etc., was reported earlier for the title material.¹⁵ The underlying physics of these phenomena can be explained with this evidence of structural transition along with the magnetic transition. The slow relaxation observed in this compound arises due to the mismatch between the experimental time scale and the transformation time scale of the lattice structure.²¹ So the experimental time scale produces metastable states, which relax to equilibrium state when the specimen is allowed to relax. As the moment and the lattice structure are strongly coupled with each other, jumps are expected in magnetization isotherms as well as in magnetoresistance.

IV. CONCLUSION

Temperature and field variation of structural properties of $\text{Ce}(\text{Fe}_{0.975}\text{Ga}_{0.025})_2$ have been studied using XRD technique. The strong magnetostructural coupling seen from the combination of the XRD and magnetization data lends direct experimental evidence to the martensitic scenario predicted in this compound. The observation of the first order nature of the magnetostructural transition between the AFM and the FM states and the phase coexistence of structural and the magnetic phases over a certain regime of temperature and field are the highlights of this study. Comparison with the different classes of materials (magnetocaloric materials and magnetoresistive oxides) brings to light the universality of phase separated systems.

ACKNOWLEDGMENTS

K.G.S. and A.K.N. thank BRNS (DAE) for the financial assistance for carrying out this work. The work at Ames

Laboratory was supported by the Office of Basic Energy Sciences, Materials Sciences Division of the U.S. Department of Energy under Contract No. DE-AC02-07CH11358 with the Iowa State University. The authors thank Professor K. A. Gschneidner, Jr. for fruitful discussions.

- ¹R. Mahendiran, A. Maignan, S. Hebert, C. Martin, M. Hervieu, B. Raveau, J. F. Mitchell, and P. Schiffer, *Phys. Rev. Lett.* **89**, 286602 (2002).
- ²S. 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).
- ³D. S. Rana, D. G. Kuberkar, and S. K. Malik, *Phys. Rev. B* **73**, 064407 (2006).
- ⁴D. S. Rana and S. K. Malik, *Phys. Rev. B* **74**, 052407 (2006).
- ⁵S. Nair, A. K. Nigam, A. V. Narlikar, D. Prabhakaran, and A. Boothroyd, *Phys. Rev. B* **74**, 132407 (2006).
- ⁶D.-Q. Liao, Y. Sun, R.-F. Yang, Q.-A. Li, and Z.-H. Cheng, *Phys. Rev. B* **74**, 174434 (2006).
- ⁷V. Hardy, S. Majumdar, M. R. Lees, D. McK. Paul, C. Yaicle, and M. Hervieu, *Phys. Rev. B* **70**, 104423 (2004).
- ⁸F. 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).
- ⁹A. Maignan, S. Hébert, V. Hardy, C. Martin, M. Hervieu, and B. Raveau, *J. Phys.: Condens. Matter* **14**, 11809 (2002).
- ¹⁰E. M. Levin, K. A. Gschneidner, Jr., and V. K. Pecharsky, *Phys. Rev. B* **65**, 214427 (2002).
- ¹¹Z. W. Ouyang, V. K. Pecharsky, K. A. Gschneidner, Jr., D. L. Schlagel, and T. A. Lograsso, *Phys. Rev. B* **76**, 134406 (2007).
- ¹²S. B. Roy, M. K. Chattopadhyay, and P. Chaddah, *Phys. Rev. B* **71**, 174413 (2005).
- ¹³S. 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).
- ¹⁴S. B. Roy and B. R. Coles, *J. Phys.: Condens. Matter* **1**, 419 (1989).
- ¹⁵A. Haldar, K. G. Suresh, and A. K. Nigam, *Phys. Rev. B* **78**, 144429 (2008).
- ¹⁶V. K. Pecharsky, A. P. Holm, K. A. Gschneidner, Jr., and R. Rink, *Phys. Rev. Lett.* **91**, 197204 (2003).
- ¹⁷N. K. Singh, D. Paudyal, Ya. Mudryk, V. K. Pecharsky, and K. A. Gschneidner, Jr., *Phys. Rev. B* **79**, 094115 (2009).
- ¹⁸Ya. Mudryk, A. P. Holm, K. A. Gschneidner, Jr., and V. K. Pecharsky, *Phys. Rev. B* **72**, 064442 (2005).
- ¹⁹A. P. Holm, V. K. Pecharsky, K. A. Gschneidner, Jr., R. Rink, and M. N. Jirmanus, *Rev. Sci. Instrum.* **75**, 1081 (2004).
- ²⁰S. B. Roy, P. Chaddah, V. K. Pecharsky, Jr., and K. A. Gschneidner, *Acta Mater.* **56**, 5895 (2008).
- ²¹K. J. Singh, S. Chaudhary, M. K. Chattopadhyay, M. A. Manekar, S. B. Roy, and P. Chaddah, *Phys. Rev. B* **65**, 094419 (2002).