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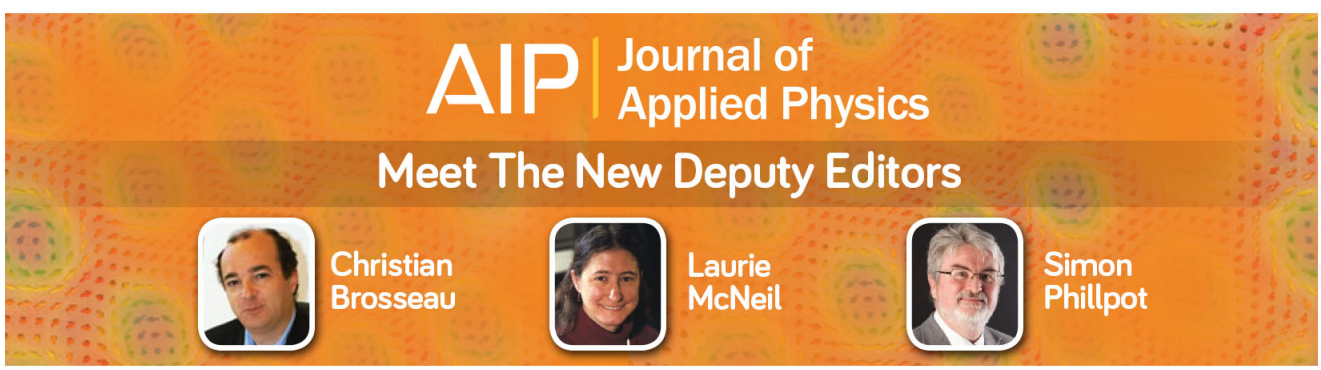

High field neutron diffraction study in $Ce(Fe_{0.95}Si_{0.05})_2$ compound

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Si substitution at Fe site causes the ferromagnetic to antiferromagnetic (AFM) transition at low temperatures in $CeFe_2$. We have carried out temperature and field variation of neutron diffraction measurements on $Ce(Fe_{0.95}Si_{0.05})_2$ to unravel its magnetic structure along with its lattice structure. The Curie temperature (T_C) and the Neel temperature (T_N) are 184 and 82 K, respectively. Above T_N, it exhibits a cubic ($Fd\bar{3}m$) structure. On lowering the temperature below T_N, weak superlattice reflections are observed indicating the onset of AFM ordering accompanied by a rhombohedral distortion. The crystal structure in the AFM state is refined in $R\bar{3}m$ space group. © *2011 American Institute of Physics*. [doi:10.1063/1.3556917]

I. INTRODUCTION

$CeFe_2$ series of compounds have attracted a lot of attention from a long time because of its rich phase diagram and the observation of anomalous features.[1–5] In the RFe_2 series of compounds $CeFe_2$ has a special position, as Ce couples antiferromagnetically with Fe, which is in contradiction with the general coupling trend seen between the light rare earth and transition metal. As a result, the Curie temperature and the saturation magnetization are comparatively low. Delocalized nature and the mixed valency of Ce 4*f* electrons have been found to be responsible for the anomalous features in $CeFe_2$.[1] Furthermore, substitution with certain elements at Fe site causes an additional phase transformation, namely ferromagnetic (FM) to antiferromagnetic (AFM) phase, on cooling. From neutron diffraction analysis it was shown that the system undergoes a structural transformation also along with the magnetic phase transition.[6]

It was observed that with Si substitution at Fe site causes the FM-AFM transition at low temperatures.[3,7] Temperature and field variations of x-ray diffraction (XRD) analysis on this system clearly establish the fact that antiferromagnetic phase transition is associated with a structural phase transformation from cubic to rhombohedral phase.[8] By comparing various dopants that stabilize the low temperature AFM state, we have found that Si doping causes better stabilization of the AFM state. Though there are a few reports on the magnetic structures of other doped $CeFe_2$ compounds,[6] no such reports are available on Si doped compounds. Therefore, we have carried out temperature and field variation of neutron diffraction measurements on $Ce(Fe_{0.95}Si_{0.05})_2$ to unravel its magnetic structure.

II. EXPERIMENTAL DETAILS

Polycrystalline $Ce(Fe_{0.95}Si_{0.05})_2$ compound was prepared by arc melting of the constituent elements Ce (99.9%), Fe (99.999%), and Si (99.999%) in a water cooled copper hearth in argon atmosphere. The alloy button was remelted several times. As cast alloy was then 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. Then it was quenched in normal water flow. The formation of the compound has been confirmed by analyzing the room temperature x-ray diffraction patterns obtained using Cu Kα radiation. Neutron diffraction studies on this compound have been carried out on the E6 diffractometer ($\lambda = 0.24$ nm) at Berlin Neutron Scattering Center (BENSC), Germany. The Rietveld refinement of diffraction data was carried out using FullProf Suite program.[9] Magnetization measurement has been performed in a Physical Property Measurement System (Quantum Design).

III. RESULTS AND DISCUSSION

Temperature variation of zero field cooled (ZFC) magnetization is shown in Fig. 1(a). This variation indicates that the compound undergoes paramagnetic (PM) to ferromagnetic transition at 184 K, and on further cooling it undergoes ferromagnetic to antiferromagnetic phase transition at 82 K which is in agreement with our earlier report.[7] Curie temperature (T_C) and the Neel temperature (T_N) are estimated from the dM/dT versus T plot. Figure 1(b) shows the $M(H)$ isotherm at $T = 30$ K of $Ce(Fe_{0.95}Si_{0.05})_2$. For this measurement the sample was zero field cooled to the measurement temperature and the data was taken by varying the magnetic field. A clear hysteresis between the increasing and the decreasing field cycles can be seen from this plot, which is typical for first order type transition.[5]

From the magnetization measurements, we have obtained the critical temperatures (function of H) and fields (function of T) across AFM-FM and FM-PM transitions. With this knowledge of plausible magnetic phases in this compound we have chosen the measurement temperatures and fields for the neutron diffraction experiment. Figure 2(a) shows the neutron diffraction patterns at selected temperatures. This compound exhibits a cubic structure at 250 K (not shown in the figure).

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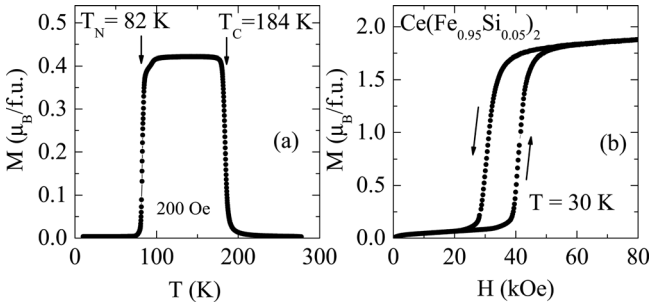

FIG. 1. (a) Temperature variation of magnetization of $Ce(Fe_{0.95}Si_{0.05})_2$ in ZFC mode at $H = 200$ Oe. (b) Two quadrant $M(H)$ isotherm of $Ce(Fe_{0.95}Si_{0.05})_2$ after cooling the sample in zero field down to the measurement temperature ($T = 30$ K).

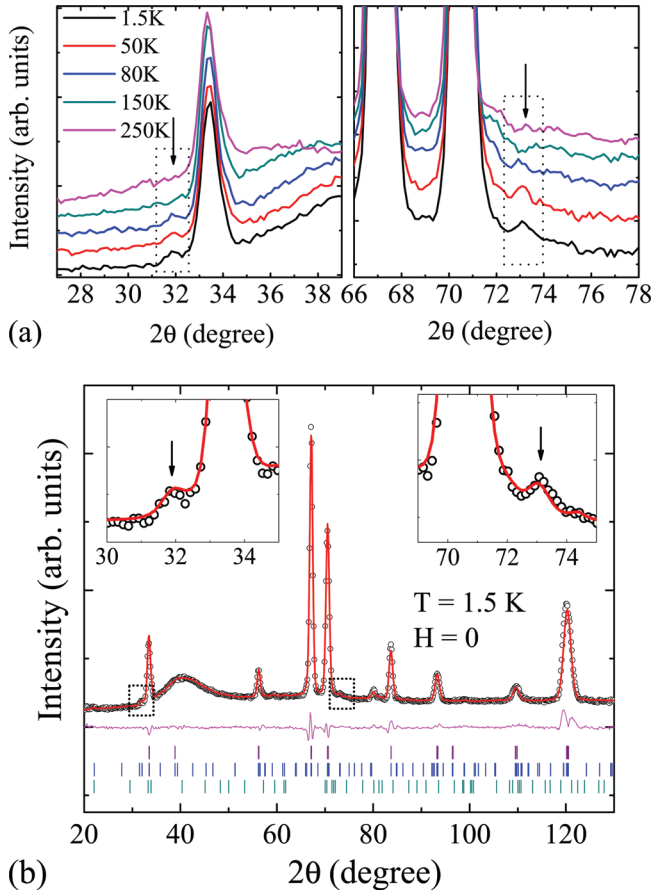

FIG. 2. (Color online) (a) Neutron diffraction pattern at various temperatures in zero magnetic field. Arrows indicate the low temperature magnetic peak. (b) Rietveld refinement of 1.5 K pattern along the difference plot. The tick marks give the positions of reflections of the chemical, magnetic and secondary (Ce_2Fe_{17}) phases (from top to bottom).

Rietveld structure refinement was carried out in the $Fd\bar{3}m$ space group at this temperature to obtain the structural parameters and occupancies. The refined parameters are summarized in Table I. The refined lattice parameters are in good agreement with that obtained from x-ray diffraction analysis. On lowering the temperature below 100 K, weak superlattice reflections are observed [indicated by arrows in Fig. 2(a)]. The diffraction pattern is similar to that reported in the case of $Ce(Fe_{0.8}Co_{0.2})_2$. The appearance of the superlattice reflections coincides with the rapid decrease observed in $M(T)$ and clearly establishes the antiferromagnetic nature of the sample. Due to the relatively poor resolution of the instrument we have not observed the expected changes in the diffraction pattern accompanying the cubic (FM)-rhombohedral (AFM) transition. Therefore, we used the structural parameters from previously reported x-ray diffraction studies. The refined neutron diffraction pattern at 1.5 K is shown in Fig. 2(b). The hump near 40° in all the patterns originates from the deuterated alcohol which was used during the sample preparation to prevent the rotation of individual particles by the magnetic field. The chemical structure at 1.5 K has been refined in $R\bar{3}m$ space group (hexagonal setting). The magnetic cell used is $a \times 2b \times c$ and is refined in the P1 space group. In this model the moment is assumed to be on Fe alone and the AFM structure is noncentrosymmetric. The contribution of Ce in the AFM phase is absent and has been observed in similar pseudobinary alloys.[6] The AFM structure is similar to that reported for $CeFe_2$.[10] The moment on Fe is found to be $1.0(1)\mu_B$. The weak nature of the superlattice reflections restricts the accuracy of the moment estimation. The value of the moment on Fe is close to that reported for Fe moment in $CeFe_2$.

In Fig. 1 it is observed that, at $T = 30$ K, the compound is antiferromagnetic in the absence of a magnetic field. On application of high field (> 48 kOe at $T = 30$ K), the antiferromagnetic phase is suppressed and a transformation to a ferromagnetic phase is observed [Fig. 1(b)]. Field induced magneto-structural transition has been probed by high field neutron diffraction measurements and the results are shown in Fig. 3. Selected 2θ windows have been shown in the upper

TABLE I. Space group, lattice parameters, atomic co-ordinates of main phases at two selected temperatures for $Ce(Fe_{0.95}Si_{0.05})_2$ compound in zero magnetic field.

T	Space group	Lattice parameters (Å)	Atom	Site	x	y	z	Occ.	χ^2
1.5 K	$R\bar{3}m$	a = b = 5.1612	Ce	6c	0	0	0.12290	1	5.3
		c = 12.6149	Fe1	9e	0.5	0	0	0.95	
			Fe2	3b	0	0	0.5	0.95	
			Si1	9e	0.5	0	0	0.05	
			Si2	3b	0	0	0.5	0.05	
250 K	$Fd\bar{3}m$	a = b = c = 7.3051	Ce	8b	0.375	0.375	0.375	1	3.9
			Fe	16c	0	0	0	1.9	
			Si	16c	0	0	0	0.1	

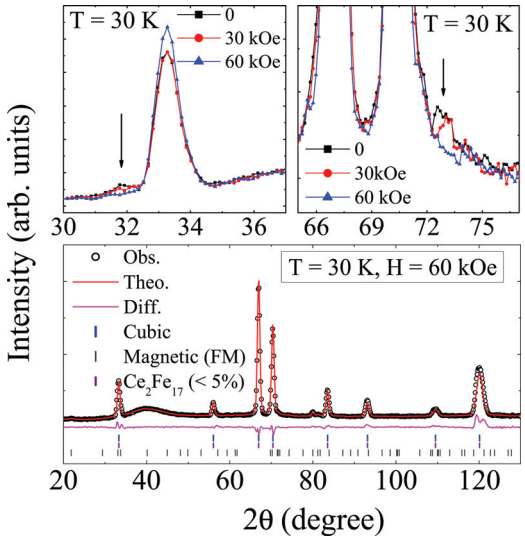

FIG. 3. (Color online) Neutron diffraction pattern at various magnetic fields at $T = 30$ K (upper panels). Lower panel shows the Rietveld refinement of the diffraction pattern obtained at $H = 60$ kOe along with difference plot. Bragg positions and the difference plot is also shown. The tick marks give the positions of reflections of the cubic, magnetic, and secondary (Ce_2Fe_{17}) phases (from top to bottom).

panels which clearly illustrate the disappearance of magnetic superlattice peaks at 60 kOe. Additionally, the fundamental (111) reflection gains in intensity. The suppression of the intensity of the superlattice reflection and gain in intensity of the fundamental reflection indicates the complete transformation of the AFM structure to FM structure at 60 kOe and is in agreement with *M(H)* data. Our previous XRD studies in magnetic field have shown that the sample completely transforms from rhombohedral to cubic on application of magnetic field.[8] Therefore, we have analyzed the data at $T = 30$ K in 60 kOe field using $Fd\bar{3}m$ space group. The ferromagnetic moment is confined to Fe alone. The absence of any enhancement in the intensity of the (220) reflection indicates the absence of moment on Ce.[4] Similar behavior in the ferromagnetic phase, albeit in the absence of magnetic field, has been observed with other dopants like Co and Al as well.[6] Figure 3 shows the Rietveld refinement of the data at 30 K in $H = 60$ kOe. The refined magnetic moment on Fe is 1.9(1) μ_B, which is in agreement with *M(H)* data. The field induced lattice distortion is consistent with our earlier investigation using the in-field x-ray diffraction analysis.[8]

IV. CONCLUSIONS

To conclude, we have probed magnetic and lattice structures of $Ce(Fe_{0.95}Si_{0.05})_2$ compound using neutron diffraction experiment. The observation of superlattice reflection below T_N establishes the antiferromagnetic nature of the sample. However, it is not clear from this experiment if the ferromagnetic state is completely suppressed in the AFM state or they coexist. The magnetic transition on lowering the temperature is accompanied by a structural distortion and the structure changes from cubic to rhombohedral. It is observed that by varying temperature (at fixed H) and magnetic field (at fixed T), magnetic and structural phase transformations occur in this compound. Moment is assumed to be on Fe alone, and the refined value of the moment in the AFM state is found to be $\sim 1\mu_B$. A field of $H = 60$ kOe is found to suppress the AFM ordering (at $T = 30$ K) and the rhombohedral distortion and induce FM ordering with cubic structure. The refined magnetic moment on Fe in the ferromagnetic state is 1.9(1) μ_B. This is consistent with *M(H)* data and the in-field x-ray diffraction data. It is found that there is a good agreement with the results obtained from the magnetization and the in-field XRD measurements. It also establishes that the structure and magnetic phases are strongly coupled with each other.

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[1]O. Eriksson, L. Nordström, M. S. S. Brooks, and B. Johansson, Phys. Rev. Lett. **60**, 2523 (1988).
[2]N. Pillmayr, G. Schaudy, T. Holubar, and G. Hilscher, J. Magn. Magn. Mater. 104–107, **881** (1992).
[3]A. K. Grover, R. G. Pillay, V. Balasubramanian, and P. N. Tandon, Solid State Commun. **67**, 1223 (1988).
[4]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).
[5]A. Haldar, K. G. Suresh, and A. K. Nigam, Phys. Rev. B **78**, 144429 (2008).
[6]S. J. Kennedy and B. R. Coles, J. Phys.: Condens. Matter **2**, 1213 (1990).
[7]A. Haldar, K. G. Suresh, and A. K. Nigam, J. Phys.: Conf. Ser. **200**, 032021 (2010).
[8]A. Haldar, N. K. Singh, Ya. Mudryk, K. G. Suresh, A. K. Nigam, and V. K. Pecharsky, Solid State Commun. **150**, 879 (2010).
[9]J. Rodríguez-Carvajal, Physica B **192**, 55 (1993).
[10]L. Paolasini, B. Ouladdiaf, N. Bernhoeft, J-P. Sanchez, P. Vulliet, G. H. Lander, and P. Canfield, Phys. Rev. Lett. **90**, 057201 (2003).