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Weak arrest-like and field-driven first order magnetic phase transitions of itinerant Fe_3Ga_4 revealed by magnetization and magnetoresistance isotherms

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ABSTRACT

The detailed magnetic study of complex 3*d*-electron based Fe_3Ga_4 is reported. It undergoes paramagnetic to antiferromagnetic (T_N) and antiferromagnetic to ferromagnetic (T_C) transitions respectively around 380 and 70 K. The thermal hysteresis of field-cooled cooling (FCC) and field-cooled warming (FCW) hints at first order phase transition below Curie temperature. A weak phase coexistence of ferro and antiferromagnetic phases is suggested by exploring the arrest-like first-order phenomenon. In the intermediate temperature range, field-driven metamagnetic transition from antiferro to ferromagnetic phase is confirmed. Further bringing the system very near to T_N, field-induced transitions disappear and above T_N predominant paramagnetic contribution is evident. The magnetic *H–T* phase diagram distinguishing different magnetic phases of Fe_3Ga_4 is obtained.

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1. Introduction

Materials which show first order magnetic transition by virtue of strong magnetostructural coupling are in the limelight. One of the reasons for this is the giant magnetocaloric effect associated with such transitions, which enable them to be practical magnetic refrigerant materials. Some rare earth-transition metal intermetallic compounds such as Gd_5Ge_4 belong to this class of materials. Furthermore, such materials show many exotic properties. Therefore, it is of interest to identify such materials, both from the point of view of fundamental interest as well as application potential. Certain 3*d*-transition metal alloys with other metals and semiconductors exhibit various exotic phenomena. For example, doped Fe i.e., $Fe_{1-x}A_x$ acquires different structural, magnetic and electrical ground states depending on the chemical nature of dopant A and its conduction electron bonding with 3*d*-band of Fe. The percentage of doping also plays a significant role in determining the structure and ground state properties. For example, when Fe is alloyed with 50% Si, formation of a correlated narrow band-gap semiconductor is reported [1] with cubic B20 structure. Ga doping in elemental Fe greatly affects the electrical and magnetic ground states of Fe. The quarter doped compound Fe_3Ga is a metal with shape memory characteristics [2,3] whereas $FeGa_3$ (75% doped) is a non-magnetic narrow-gap semiconductor [4,5]. Recently thin films of $Fe_{1-x}Ga_x$ are reported to be useful for magnetostrictive applications [6].

In the past few decades, the materials that show ferromagnetic (FM) and antiferromagnetic (AFM) orderings at different temperatures (*T*) are of great interest in the prospect of fundamental research as well as technological applications. Such materials are interesting because of the magnetic first order phase transitions like field-induced metamagnetic and phase coexistence properties. From the view point of technological applications, such materials could serve as magnetic refrigerants. These materials can be broadly divided into two classes; (i) with high-*T* FM and low-*T* AFM phases, (ii) with high-*T* AFM and low-*T* FM phases. Some of the well studied compounds that belong to the first class are Gd_5Ge_4 [7,8], doped $CeFe_2$ [9–12], $La(Fe, Si)_{12}$ [13], $Gd_5Ge_2Si_2$ [14], Nd_5Ge_3 [15] and $SmMn_2Ge_2$ [16,17] which are mainly the rare-earth based. Doped Mn_2Sb with Co and Cr for Mn [18] and Sn for Sb [19], also undergo high-*T* ferrimagnetic to low-*T* AFM phase transition. On the other hand, a few rare-earth rich compounds viz., R_3Co belonging to the latter class show metamagnetic nature (in high-*T* AFM phase) with low-*T* non-collinear ferromagnetic ground state [20,21]. Fe rich $HfFe_2$ for certain doping of Ta at Hf site [22,23] undergoes AFM to FM transition with decreasing temperature. Nevertheless, systems with temperature-driven dual magnetic transitions, phase coexistence and field-induced metamagnetic behavior with itinerant character are scarce in the literature.

Fe_3Ga_4 is one such itinerant-electron system which is reported to crystallize in monoclinic structure bearing C/2 m space group

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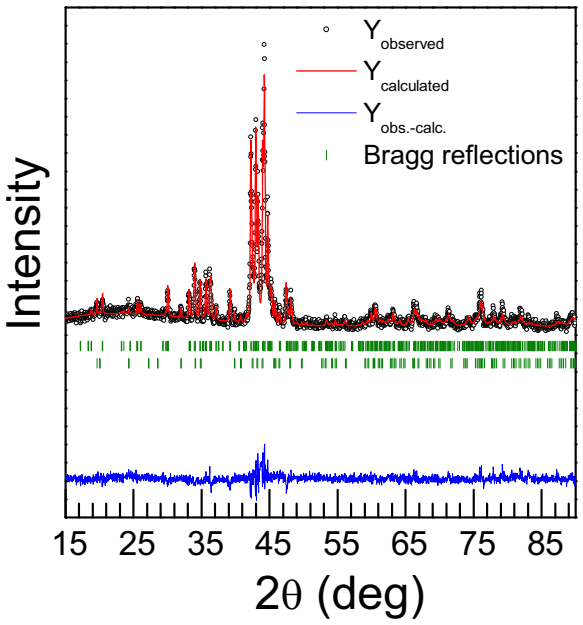

Fig. 1. The refinement of x-ray diffraction pattern of Fe_3Ga_4 taken at room temperature. The lattice parameters are about $a=10.08$ Å, $b=7.66$ Å and $c=7.86$ Å with $\beta \simeq 106.3°$. The black circles and red solid line represent experimental and calculated data. Bragg reflections are given in green solid lines. The blue line is the difference pattern. The refinement reveals a minor secondary phase of non-magnetic $FeGa_3$ which is about 6.17%. (For interpretation of the references to color in this figure caption, the reader is referred to the web version of this paper.)

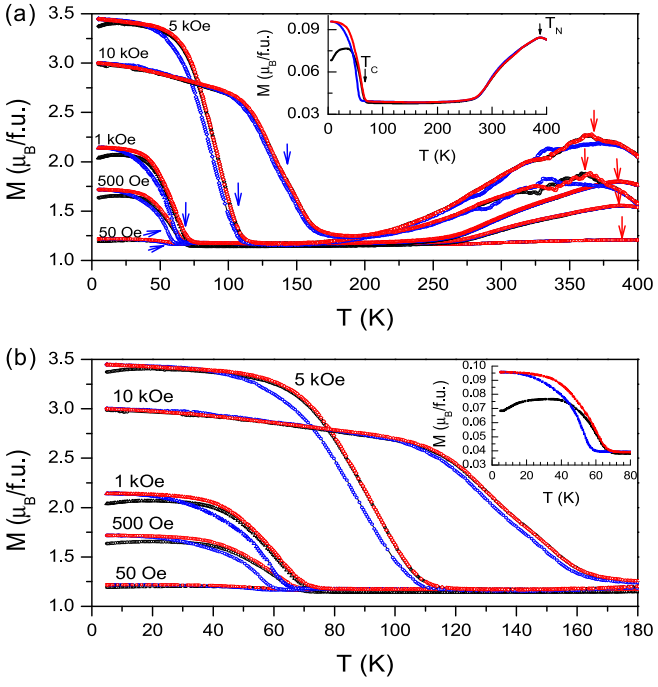

Fig. 2. (a) Temperature dependence of magnetization in 50 Oe, 500 Oe, 1 kOe, 5 kOe and 10 kOe. For clarity and comparison, the data in 50 Oe, 500 Oe, 1 kOe and 10 kOe is shifted along magnetization axis by 1.123, 0.98, 0.84 and −0.952 μ_B. Inset shows the data in 50 Oe. The antiferromagnetic (T_N) and ferromagnetic (T_C) transition temperature are indicated by arrows. ZFC, FCC and FCW are shown in the black, blue and red curves respectively. (b) The zoomed portion (from 0 to 180 K) of the thermal hysteresis region of FCC and FCW at $T \leq T_C$ along with the bifurcation of ZFC and FCC is presented. Inset represents $M(T)$ in 50 Oe. (For interpretation of the references to color in this figure caption, the reader is referred to the web version of this paper.)

[24]. The parent [25] and Al-doped [26] compounds of Fe_3Ga_4 exhibit temperature driven first order and field induced metamagnetic transition from AFM to FM. The magnetic nature of these compounds is described based on the itinerant electron models of Moriya-Usami [27] and Isoda [28]. The detailed Mössbauer studies of $(Fe_{1-x}M_x)_3Ga_4$ (M=Ti, Cr and Mn) along with parent compound [29,30] are also reported. Recently Mendez et al. [31] have explored the competition between ferromagnetic and antiferromagnetic states and disordered effects (at different annealing conditions) and stable FM state is predicted by supercell calculations with Fe anti-site defects. Surprisingly, specific heat data does not exhibit any pronounced anomaly at the magnetic ordering temperatures. However, the detailed temperature and field dependent magnetization studies on Fe_3Ga_4 are lacking in the literature.

In this study, we report magnetic properties of Fe_3Ga_4 based on magnetization and resistivity. The temperature range is broadly divided into three regions namely (I): $T < T_C$, (II): $T_C < T < T_N$; (i)-towards T_C, (ii) towards T_N and (III): $T > T_N$. At low temperatures, the phase coexistence of FM and AFM and arrest-like features are demonstrated phenomenologically. In the mid temperature range, field-induced AFM to FM metamagnetic phase change is illustrated. Towards the end, a probable magnetic phase ($H - T$) diagram is proposed.

2. Experimental details

Polycrystalline Fe_3Ga_4 is prepared by arc melting the constituent elements of purity better than 99.99% in Argon atmosphere. For homogeneity the ingot is melted three times by flipping each time. The refinement of the x-ray diffraction pattern taken at room temperature is shown in Fig. 1. The obtained parameters (Rietveld refined using FullProf suite) are approximately $a=10.08$ Å, $b=7.66$ Å and $c=7.86$ Å with $\beta=106.3°$. A minor secondary phase of non-magnetic $FeGa_3$ of about 6.17% is detected as estimated from refinement. However, heat treatment did not improve the homogeneity [26]. $FeGa_3$ being non-magnetic [5], its effect on the magnetization of Fe_3Ga_4 is neglected. The temperature (5–400 K) and magnetic field (0–50 kOe) dependent magnetization is measured using 9 T VSM and 7 T SQUID-VSM (Quantum Design, USA). The electrical resistivity measurements are performed by linear four-probe method using 9 T Physical Property Measurement System (QD, USA).

3. Results

Fig. 2(a) shows the temperature dependence of magnetization, M, in 0.05, 0.5, 1, 5 and 10 kOe. The specimen is initially cooled down to the lowest temperature (5 K) at which the required field is applied. Then the data is acquired during warming (ZFC) up to 400 K and cooling (FCC) down to 5 K. Subsequently, without switching off the magnetic field, $M(T)$ is taken during warming (FCW) up to 400 K. The same protocol is employed in each field. $M(T)$ for the full measured range of temperatures is given in the insets. Fe_3Ga_4 undergoes paramagnetic (PM) to AFM transition around 370 K with a Néel temperature (T_N = 370 K). Upon further cooling, it undergoes a first order transition from AFM to FM with a Curie temperature, T_C, of about 70 K. The data is analyzed by separating it into different temperature regimes mentioned earlier.

The increase of fields resulted in the enhancement of T_C and a suppression of T_N. T_{cross} at which FCC crosses ZFC is approximately 44.5, 31.8 and 29.8 K at $H=0.05$, 0.5 and 1 kOe respectively. A difference in the base values of ZFC and FCC for $T \in$ [70 K, 260 K] may be due to the fact that the compound is not completely

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