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High field magnetic behavior in *Boron* doped Fe_2VAl Heusler alloys

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ABSTRACT

We have investigated the magnetic behavior of $Fe_2VAl_{1-x}B_x$ (x=0, 0.03, 0.06 and 0.1) alloys under high temperature and high magnetic field conditions separately. Although, the low temperature DC magnetization data for the alloys above $x>0$ show clear magnetic transitions, the zero field cooled (ZFC) and field cooled (FC) curves indicate the presence of spin cluster like features. Further, critical exponent (γ) deduced from the initial susceptibility above the T_c, does not agree with standard models derived for 3 dimensional long range magnetic systems. The deviation in γ values are consistent with the short range magnetic nature of these alloys. We further extend the analysis of magnetic behavior by carrying the magnetization measurements at high temperatures and high magnetic fields distinctly. We mainly emphasize the following observations; (i) The magnetic hysteresis loops show sharp upturns at lower fields even at 900 K for all the alloys. (ii) High temperature inverse susceptibility do not overlap until T=900 K, indicating the persistent short range magnetic correlations even at high temperatures. (iii) The Arrott's plot of magnetization data shows spontaneous moment (M_S) for the x=0 alloy at higher magnetic fields which is absent at lower fields (<50 kOe), while the *Boron* doped samples show feeble M_S at lower fields. The origin of this short range correlation is due to presence of dilute magnetic heterogeneous phases which are not detected from the X-ray diffraction method.
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1. Introduction

It is now well established that Fe_2VAl Heusler alloy exhibits unusual transport and magnetic properties which has drawn more attention of the scientific community in the recent past [1,2]. A comparative study between experimental results and ab-initio calculations with various anti-site disorders on Fe_2VAl alloy aided in understanding the role of anti-site disorder [3] and chemical doping [4] on its ground state. In addition, band structure calculations reveal unmodified ground state character due to isoelectronic substitution at the *Al*-site [5]. On contrary, the experimental observations show a weak static and dynamic magnetic behavior due to the doping of isoelectronic elements (*B*, *Ga* and *In*) at the *Al*-site [4,6,7]. The origin of this contradictory nature is understood due to the presence of magnetic inhomogenities inhibited by the anti-site disorder [8,9] and chemical inhomogeneity [10]. Although, the anti-disorder was quantified by the analysis of X-ray as well as neutron diffraction pattern, no information was given in the past about the chemical inhomogeneity which was not identified by these diffraction techniques. On the other hand, the chemical inhomogeneity leads to soft magnetic-like hysteresis loops and higher magnetoresistance [10]. Hence, it is interesting to study the doping of isoelectronic elements at the *Al*-site in Fe_2VAl compound. Motivated by the earlier reports and to understand the influence of lower atomic size on the ground state properties of Fe_2VAl due to chemical compression, we have investigated series of $Fe_2VAl_{1-x}B_x$ (x= 0, 0.03, 0.06 and 0.1) alloys by measuring temperature variation of their magnetic and magneto-transport studies. The low temperature magnetization data on the same series reported elsewhere [10], reveal the presence of short range like magnetic characteristics of these alloys and motivate us to further explore the same by investigating magnetization of these materials at higher magnetic fields and high temperature conditions. The measurements here will address the questions like, (i) Is there any modification of these short range corrections towards the long range magnetic ordering upon application of high

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magnetic fields? (ii) Is there second magnetic transition that results in peculiar magnetic characteristics observed at 300 K. Thus, in this article, we report the magnetic properties of $Fe_2VAl_{1-x}B_x$ (x=0, 0.03, 0.06 and 0.1) alloys to verify the size of the non-magnetic sp-element effect on the magnetic and electric transport behaviors under higher fields (160 kOe) and high temperature (900 K) conditions.

2. Experimental methods

The alloy ingots were synthesized from high purity elemental constituents using conventional arc melting technique. Subsequently, the ingots were annealed at 1000 °C for 3 days followed by the quenching in a liquid nitrogen bath. The obtained ingot was again annealed at 600 °C for 24 h to relax the quenched disorder [8,10]. The magnetic measurements are taken by EverCool SQUID VSM (Quantum Design, USA) operated with the help of cryogen free cooling procedure using high purity He-gas. The samples were heated to paramagnetic regime in each time while taking the isothermal initial magnetization curves, which are used to analyze the magnetic critical exponents. Electrical transport properties have been carried out with conventional four probe technique under the magnetic fields upto 160 kOe.

3. Results

This article is started by presenting the evidence of magnetic short range order from the magnetic critical analysis of low field (50 kOe) magnetic isotherms taken around their respective transition temperatures followed by a discussion on the results obtained from the high field magnetization and high temperature data. In order to identify the phase transition, one can analyze the magnetization data in the vicinity of magnetic transition using conventional theories. In the vicinity of a second-order magnetic phase transition with Curie temperature T_c, the divergence of correlation length $\xi=\xi_0|1-T/T_c|^{-\gamma}$ leads to universal scaling laws for spontaneous magnetization M_{Sp} and initial susceptibility χ. The γ is a critical exponent. We have analyzed the magnetization data and obtained the critical exponents as described in the following.

3.1. Critical exponents

According to the scaling hypothesis, the second order magnetic phase transitions are described by the critical exponents such as β (exponent just below T_c), δ (exponent of critical isotherm at T_c) and γ (exponent just above T_c). These critical exponents are associated with the spontaneous magnetization (M_S), initial susceptibility ($\chi_0 = \lim_{H\to 0} M/H$), and magnetization isotherm (M vs H) near the critical region by simple power laws (SPL) as given below.

$$M_s \propto \eta^{\beta} \quad (H=0) \quad \eta<0 \tag{1}$$

$$M(H, T_c) \propto H^{(1/\delta)} \quad (H\to 0) \quad \eta=0 \tag{2}$$

$$\lim_{H\to 0}\left(\frac{M}{H}\right) = \chi_0(T) \propto \eta^{-\gamma} \quad \eta>0 \tag{3}$$

where $\eta=\pm[T-T_c]/T_c$ is the reduced temperature, +/- indicates the temperature above and below T_c.

The values of M_{sp}, χ_0 and T_c are often determined from the isothermal magnetization curves, presented as M^2 *vs* H/σ known as Arrott-Plots (AP). Extrapolation of AP to M^2=0 and H/M=0 gives intercepts on M^2 axis and H/M axis, which are equal to the M^2 for $T<T_c$ and χ_0^{-1} for $T>T_c$. The AP plots for $Fe_2VAl_{1-x}B_x$ with x=0.03, 0.06 and 0.1 alloys show a nonlinear behavior (not shown here) that does not allow one to determine the values of M_{sp}, χ_0^{-1} and T_c accurately by extrapolation method. Therefore, a modified Arrot-Plot (MAP) method [11,12] has been suggested by the following empirical expression to the M^2 vs H/σ plot,

$$\left(\frac{H}{M}\right)^{1/\gamma} = \frac{(T-T_c)}{T_x} + \left(\frac{\sigma}{M_x}\right)^{1/\beta} \tag{4}$$

where T_x and M_x are constants that depend on the materials. In this method, the M–H isotherms at different temperatures near the critical region are used to construct $M^{1/\beta}$ vs $(H/M)^{1/\gamma}$ plots. The values of critical exponents β and γ are varied so as to make these isotherms as straight lines over a wide range of H/M values as much as possible (ie. low χ^2 value of fitting) and parallel to each other in a narrow temperature range around T_c. In addition to this, the isotherm data taken above and below the T_c must have negative and positive intercepts respectively on Y-axis very nearer to the origin. This analysis was repeated self consistently until the values of critical exponents are converged within the tolerance.

Fig. 1(a) and (b) show the M_{SP}, $1/\chi_0$ data derived from the linear fittings of $M^{1/\beta}$ vs $(H/M)^{1/\gamma}$ plots obtained from the MAP method along with the fits to the (Eqs. 1 and 3). In order to verify their numerical consistency, we further determined the critical exponents from the *Kouvel–Fisher* method [13] (KF) as shown in Fig. 1(c) and (d), using expressions as given below.

$$X(T) = \left(M_s(T)\left(\frac{dM_s(T)}{dT}\right)^{-1}\right) = \left(\frac{(T-T_c)}{\beta_{eff}}\right) = \left(\frac{\eta T_c}{\beta_{eff}}\right) \tag{5}$$

and

$$Y(T) = \left(\chi_0^{-1}\left(\frac{d\chi_0^{-1}}{dT}\right)^{-1}\right) = \left(\frac{(T-T_c)}{\gamma_{eff}}\right) = \left(\frac{\eta T_c}{\gamma_{eff}}\right) \tag{6}$$

According to this method, $X(T)$ vs T and $Y(T)$ vs T have been plotted in the asymptotic critical region, where σ_s and χ_0^{-1} can be approximated by power laws of the form (Eqs. 5 and 6) with straight lines whose slopes are $(-1/\beta_{eff})$ and $(1/\gamma_{eff})$, respectively. These straight lines yield intercepts on their η-axis upon extrapolation to the abscissa, which gives the T_c. The obtained β, γ and T_c values are listed in the Table 1.

Typically, the long range magnetic ordering is characterized by the standard theoretical models such as mean field theory (β=0.5, γ=1), Heisenberg model (β=0.365, γ=1.386), Ising like (β=0.325, γ=1.241) and XY-like (β=0.345, γ= 1.316) behavior for the 3D systems [14]. It may be noticed that the values of β obtained for the present compositions are close to these conventional models, however, γ values could not be explained on the basis of these models. It is interesting to point out that Salamon et al. [15], reported a crossover from short range to long range magnetic ordering in La-based manganites by calculating the critical exponents. Their study clearly indicated that short range order has unusual critical exponent values which are not satisfied by the standard theoretical models. Such deviations from the standard models have also been reported for the short range magnetic system [16–18]. As the presently obtained γ values are similar to that reported in literature, we can contemplate the presence of short-range type magnetic ordering in the present compositions (x=0.03, 0.06 and 0.1). Moreover, the studies on magnetic properties at low temperatures as mentioned in our earlier reports [10], show an existence of inhomogeneous FM-like phase even at room temperatures, which are far above from their Tc's. Hence, it is very

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