

## Exploration of Domain Nucleation Mechanisms in the Mean Field System $\text{Fe}_{1.5}\text{Mn}_{1.5}\text{Si}$

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### ABSTRACT

$\text{Fe}_{1.5}\text{Mn}_{1.5}\text{Si}$  undergoes a paramagnetic to a ferromagnetic transition with true long range ordering. The critical behavior of the system near the transition temperature has been studied by analyzing dc magnetization data with the help of the Arrot plot. The present work is focused to answer an important question, whether the nucleation of domains starts at a temperature close to  $T_c$  or different from it. The enhancement technique in ac susceptibility helps us to determine the domain nucleation temperature. The domain nucleation temperature is found to be close to  $T_c$  obtained from scaling analysis. At the same temperature, the imaginary component of the ac susceptibility ( $\chi''$ ) also shows a peak.

**KEYWORDS:** Alloys, Critical analysis, Domain nucleation

### I. INTRODUCTION

AC susceptibility (ACS) is a useful experimental technique for examining the nature of magnetic phase transitions. ACS diverges at the critical temperature ( $T_c$ ) [1]. Critical exponents also give us important information regarding the interaction mechanism near the paramagnetic-ferromagnetic transition. The critical temperature and the critical exponents can also be obtained from the analysis of isothermal magnetization data. The enhancement technique of ACS is the study of ACS in the presence of a second biasing AC field ( $h_2 \sin \omega_2 t$ ) in addition to the measuring one ( $h_1 \sin \omega_1 t$ ) [2]. According to Jiles and Atherton, [3] below the critical temperature in ferromagnetic materials, the magnetization lags behind the low AC field due to the impedance caused to the domain wall motion by the pinning centres. A second AC field causes an extra perturbation to the domain walls and makes their motion more free. Therefore, we get an enhancement in the measured in-phase component of ACS ( $\chi_1'$ ) below the critical temperature in materials having a domain structure. When we plot the in-phase component,  $\chi_1'$  as a function of  $T$ , the data in the absence of enhancing field becomes separated from the data in the presence of enhancing field below a temperature at which the domain nucleation starts [2].

The present work focuses on, whether the domain nucleation in a ferromagnetic system starts exactly at the  $T_c$  associated with the paramagnetic to ferromagnetic transition, or at a temperature different from it.

$\text{Fe}_3\text{Si}$  is a ferromagnetic system with  $T_c = 830$  K.  $\text{Fe}_3\text{Si}$  and  $\text{Mn}_3\text{Si}$  combine to form a complete solid solution with the  $DO_3$  structure. This may be understood in terms of a unit cell which consists of four interpenetrating sublattices A, B, C, D, with origins at the points (0,0,0), ( $\frac{1}{4}, \frac{1}{4}, \frac{1}{4}$ ), ( $\frac{1}{2}, \frac{1}{2}, \frac{1}{2}$ ), ( $\frac{3}{4}, \frac{3}{4}, \frac{3}{4}$ ). Three of the four sites (A, B, C) are occupied by Fe, and the remaining D site by Si. The A and C sites are magnetically different from the B site. The A and C sites are surrounded by a first near-neighbour shell of 4 Fe and 4 Si. On the other hand, the B sites have 8 Fe nearest neighbours. For  $\text{Fe}_3\text{Si}$ , the Fe atom on a B site has a magnetic moment of 2.3  $\mu_B$ , whereas that on either A or C site has a magnetic moment of 1.2  $\mu_B$  [4]. For the system  $\text{Fe}_{3-x}\text{Mn}_x\text{Si}$ , (for  $x < 0.75$ ) the Mn atoms occupy B sites only. The substitution of Mn for Fe(B) reduces the Fe(A, C) moment equally. The moment on the B site does not deviate significantly from the 2.3  $\mu_B$  observed for  $x < 0.75$ . This implies that the Mn atoms carry a magnetic moment approximately equal to that of the Fe atoms they replace [4]. On the other hand, the moment on the Fe atoms in A and C sites have an environment in which number of Mn atoms increases as  $x$  increases [4]. This causes the moment in A and C sites to fall from the value of 1.2  $\mu_B$  (at  $x = 0$ ) to 0.4  $\mu_B$  (at  $x = 0.75$ ). However, for  $x \geq 0.75$ , Mn atoms start to occupy A and C sites also. The moment on the B site then drops from 2.3  $\mu_B$ . At  $x = 1.5$  the moment on the B site is  $\sim 0.5 \mu_B$ , and that on the A/C site is 0.4  $\mu_B$  [4]. Therefore, the low value of the moments at A, B and C sites is inherent in the system and depends only on the number as well as type of the nearest neighbours. All moments of the system are coupled ferromagnetically. It may be mentioned that the work of Yoon and Booth [5] suggests that  $\text{Fe}_{1.5}\text{Mn}_{1.5}\text{Si}$  is a normal ferromagnet below  $T_c = 150$  K and down to 65 K, around which the drop in ACS occurs due to canting [6].

We have studied the critical behavior in  $\text{Fe}_{1.5}\text{Mn}_{1.5}\text{Si}$  by analyzing our isothermal DC magnetization data near  $T_c$  with the help of Arrot plot. This will also help us to estimate  $T_c$  accurately. We have compared the  $T_c$  obtained from analyzing ACS and DC magnetization data with the domain nucleation temperature obtained from enhancement study.

## II. MATERIALS AND METHODS

The magnetization measurements were performed using superconducting quantum interference device magnetometer (Quantum Design). The data were collected at 3 K intervals over the temperature range from 120 K to 165 K, in fields from 0 to 60 kOe. The data for enhancement study has been taken from the reference [7]. In [7] a measuring field of 1 Oe and 137 Hz and an enhancing field of 8 Oe and 433 Hz, were used. The sample details and experimental techniques have been given elsewhere [6, 7].

## III. RESULTS AND DISCUSSION

The second order magnetic phase transition near the Curie point is characterized by a set of critical exponents,  $\beta$  (associated with the spontaneous magnetization),  $\gamma$  (associated with the initial susceptibility), and  $\delta$  (related to the critical magnetization isotherm). They are defined as [8]

$$M_s(T) = M_0 (-\varepsilon)^\beta, \sim \varepsilon < 0 \quad (1)$$

$$\chi^{-1}_0(T) = h_0 / M_0 (\varepsilon)^\gamma, \sim \varepsilon > 0 \quad (2)$$

$$M = A_0(H)^\delta \quad (3)$$

where  $\varepsilon = (T - T_c)/T_c$ ,  $T_c$  is the Curie temperature, and  $M_0$ ,  $h_0/M_0$ ,  $A_0$  are the critical amplitudes.

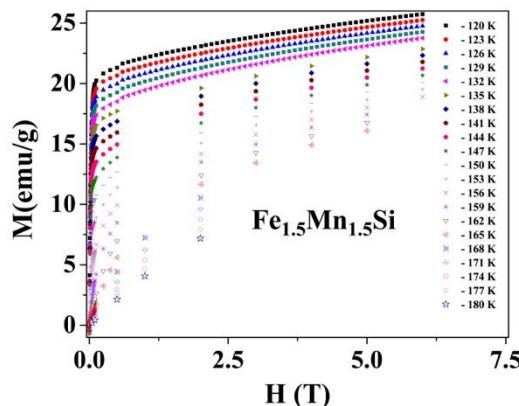


Figure 1 :  $M$  versus  $H$  Curve.

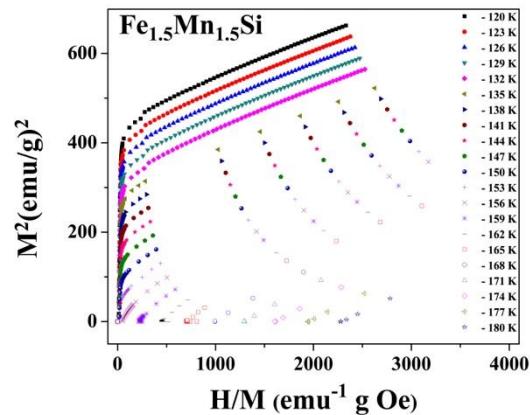


Figure 2 :  $M^2$  versus  $H/M$  plot

Figure 1 shows the  $M$  versus  $H$  curve for different temperatures (in the range 120 K to 165 K in steps of 3 K, up to 6 T). These curves show a gradual transition from a ferromagnetic state to a paramagnetic state. We have taken the data up to 6 T for the lower temperatures (120 K - 132 K). However, for higher temperatures we have performed experiment with limiting value of  $H$  as 0.5 T. The behavior of saturation is a characteristic of samples with true long-range-order ferromagnetism [9, 10]. We have used these data to determine the critical exponents  $\beta$ ,  $\gamma$  and  $\delta$ .

Figure 2 shows the  $M^2$  versus  $H/M$  plot or the Arrot plot. According to the mean field theory near  $T_c$ ,  $M^2$  vs  $H/M$  at various temperatures should show a series of parallel lines. The line at  $T = T_c$  should pass through the origin. For  $T > T_c$ , the intercept on the  $H/M$  axis gives  $1/\chi$  in the limit of zero field *i.e.*,  $1/\chi_0(T)$ . The slope of  $1/\chi_0(T)$  versus temperature gives a measure of the magnetic moment per atom of the material. For  $T < T_c$ , the intercept on the  $M^2$  axis gives a measure of  $M_s(0, T)$  *i.e.*, the spontaneous magnetization at zero field [9, 10]. Figure 2 shows that the isotherms are parallel straight lines at higher values of  $H/M$ . This suggests that this system can be well described by mean field theory. The high field straight line portions of the isotherms can be linearly extrapolated to obtain the spontaneous magnetization  $M_s(0, T)$ .

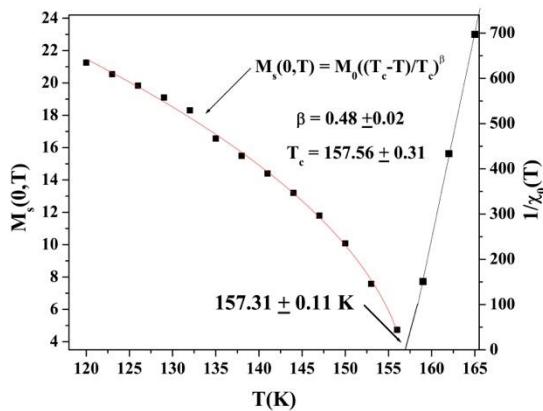


Figure 3 : Variation of  $M_s(0,T)$  as a function of temperature,  $1/\chi_0(T)$  as a function of temperature.

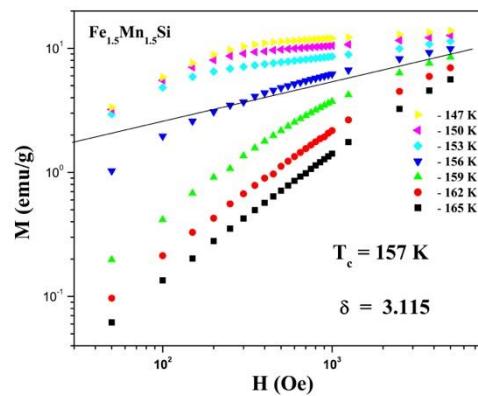


Figure 4 :  $M$  versus  $H$  plot on a log scale

Figure 3 shows the variation of  $M_s(0,T)$  as a function of temperature. The curve obtained can be fitted to the power law  $M_s(T) = M_0(-\varepsilon)^\beta$ ,  $\varepsilon < 0$  with  $\varepsilon = (T - T_c)/T_c$  and the exponent  $\beta = 0.48 \pm 0.02$ , and  $T_c = 157.56 \pm 0.31$  K. In the same figure we plotted  $1/\chi_0(T)$  as a function of temperature. The curve can be fitted to a straight line with intercept on the  $T$  axis as  $157.31 \pm 0.11$  K. According to the equation (2) the straight line fitting suggests  $\gamma = 1$ , and  $T_c = 157.31 \pm 0.11$  K. The value of  $\delta$  can be obtained from Widom scaling relation, *i.e.*,  $\delta = 1 + \gamma/\beta = 3.08 \pm 0.08$ . Figure 4 shows the  $M$  versus  $H$  plot on a log scale at few temperatures close to  $T_c$ . The straight line shows the fit for the interpolated data for  $T_c \sim 157$  K. This gives the value of  $\delta = 3.11$ . Our analysis of dc magnetization data suggests the system to be like a mean field one. The mean field exponents are  $\beta = 0.5$ ,  $\gamma = 1$ ,  $\delta = 3$ . These values are very close to our experimental values  $\beta = 0.48 \pm 0.02$ ,  $\gamma = 1$ ,  $\delta = 3.08 \pm 0.08$ . In this experiment our estimated  $T_c$  value is between 157.2 K and 157.87 K.

The low field response of a ferromagnetic system is due to the domain wall motion only. However this domain wall motion is impeded by various pinning centres. In the ACS enhancement technique, the presence of the second biasing ac field changes the sample state to one in which the domains are relatively free. So we get an enhancement in the measured value of  $\chi$  below  $T_c$ . However, so long as the ferromagnetic domains are not nucleated, the measured ACS in the presence of enhancing field is the same as that in the absence of the enhancing field. Moreover, these two are different, once the domains are formed. Therefore, we identify the temperature corresponding to the point of separation of the two ACS curve as the domain nucleation temperature.

Figure 5 shows, the ac susceptibility curve in the presence of enhancing field becomes separated from that in the absence of enhancing field at a temperature around 156 K. The data has been taken from reference 7. This temperature was identified as the domain nucleation temperature. Our estimated  $T_c$  ( $\sim 157$  K) from dc magnetization data is close to the domain nucleation temperature ( $\sim 156$  K). The plot of the imaginary component of ac susceptibility ( $\chi''$ ) obtained from our general ACS data (in the absence of enhancing field) as a function of temperature shows also a peak around the same temperature 156 K.

In summary, for a mean field system like  $\text{Fe}_{1.5}\text{Mn}_{1.5}\text{Si}$ ,  $T_C$  estimated from dc measurements is very close to the domain nucleation temperature. The difference is within 1 K. At the same temperature,  $\chi''$  also shows a peak. The slight discrepancy may be due to the presence of finite ac field. It may be noted that, the domain nucleation

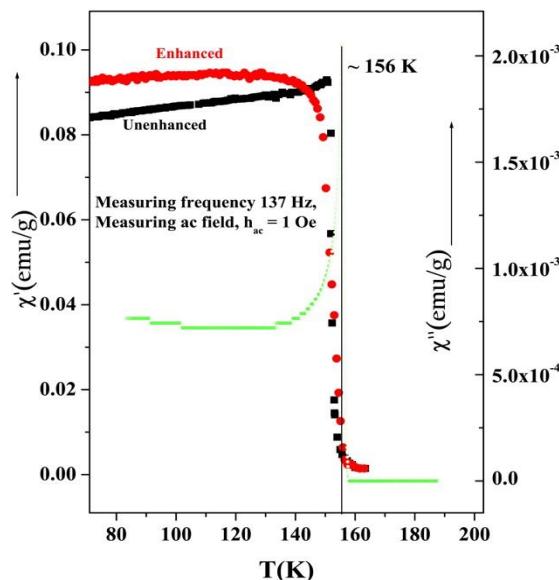


Figure 5:  $\chi'(T)$  in the presence of enhancing field, and that in the absence of enhancing field (Left y-axis) ;  $\chi''(T)$  in the absence of enhancing field (Right y-axis).

temperature is estimated in the presence of a finite measuring ac field, while  $T_C$  from the dc magnetization data is obtained in the absence of any field.

#### IV. CONCLUSION

In the absence of any field,  $\text{Fe}_{1.5}\text{Mn}_{1.5}\text{Si}$  undergoes a paramagnetic to a ferromagnetic transition near the critical temperature 157.5 K. Its behavior in the ferromagnetic state is mean field like suggesting a true long-range ferromagnetic order. Our enhancement study suggests that the domain nucleation for this system starts at a temperature close to the critical temperature estimated from isothermal dc magnetization data. The small discrepancy may be due to the presence of measuring ac field in the later case. Actually the domain nucleation starts near the critical temperature for this system. However, similar experiments in other systems are needed to generalize this conclusion.

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