Probing a ferromagnetic critical regime using nonlinear susceptibility

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The second-order paramagnetic-ferromagnetic phase transition in a series of amorphous alloys ($Fe_5Co_{50}Ni_{17-x}Cr_xB_{16}Si_{12}$) is investigated using nonlinear susceptibility. A simple molecular-field treatment for the critical region shows that the third order suceptibility χ_3 diverges on both sides of the transition temperature and changes sign at T_C. This critical behavior is observed experimentally in this series of amorphous ferromagnets and the related asymptotic critical exponents are calculated. It is shown that using the proper scaling equations, all the exponents necessary for a complete characterization of the phase transition can be determined using linear and nonlinear susceptiblity measurements alone. Using meticulous nonlinear susceptibility measurements, it is shown that at times χ_3 can be more sensitive than the linear susceptibility χ_1 in unraveling the magnetism of ferromagnetic spin systems. A technique for accurately determining T_C is discussed, which makes use of the functional form of χ_3 in the critical region.

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I. INTRODUCTION

Nonlinear effects become more pronounced in the vicinity of a phase transition and the magnetization (M) measured in the presence of an exciting field H can be written as

$$M = M_0 + \chi_1 H + \chi_2 H^2 + \chi_3 H^3 + \cdots, \quad (1)$$

where χ_1, χ_2, and χ_3 are the first-, second- and third-order susceptibilities, respectively. The higher-order susceptibilities contain a wealth of information, but owing to the fact that they are usually a couple of orders smaller in magnitude than the first-order susceptibility, they are correspondingly more difficult to measure.

The second-order susceptibility (χ_2) arises due to the presence of a symmetry-breaking field and is observed only in the presence of a dc field or in materials with a large permanent magnetization. Though relatively unexplored, χ_2 has been used to show the coexistence of spin-glass phase and ferromagnetic order in a reentrant spin-glass system.[1]

The third-order susceptibility (χ_3) has been commonly employed to characterize the spin glass transition, where a negative cusp in χ_3 was used to identify the onset of a spin-glass transition and was used as a direct probe of the divergence of the Edward-Anderson order parameter.[2] χ_3 has also been used to distinguish a spin glass from a superparamagnet.[3–5] Among long-range-order systems, though χ_3 deduced from dc magnetization measurements has been used for studying the effect of quadrupolar interactions in rare-earth compounds,[6] it is interesting to note that nonlinear susceptibility measurements on ferromagnetic[7,8] and antiferromagnetic[9] materials are not common, and very few reports exist in literature. Moreover, most of the previous experimental works on nonlinear susceptibility in long-range-order systems are predominantly qualitative and not quantitative in nature. As far as the study of phase transitions is concerned, though the second-order paramagnetic to ferromagnetic phase transition has been extensively studied using linear susceptibility,[10] the behavior of χ_3 across the transition has been relatively less explored.

To bring out the advantages as well as the limitations of nonlinear susceptibility measurements in the investigation of a paramagnetic-ferromagnetic phase transition, we present a comprehensive study on a set of Fe-Co-Ni-Cr-B-Si amorphous alloys. This set of alloys is chosen because they were well studied in the past, and it was found through linear ac susceptibility and high-field dc magnetization measurements that they are reasonably well behaved three-dimensional (3D) Heisenberg ferromagnets.[11–13] As such, amorphous ferromagnets have been extensively studied using linear susceptibility to understand the influence of quenched disorder on the critical behavior of spin systems. The effect of disorder, as seen in the values of the critical exponents and the width of the asymptotic critical regime, have been studied and reviewed extensively.[14] Nonlinear susceptibility as measured in an amorphous ferromagnet, has been reported before,[15] but to the best of our knowledge, this is the first study on at least this class of materials, where linear and nonlinear susceptibility alone is used for a complete characterization of the critical regime.

At the outset, we present a simple molecular-field treatment to show that χ_3 diverges on both sides of the transition, and changes sign at the transition temperature. Thus the criticality is observed experimentally, and the related asymptotic critical exponents are calculated. It has been shown that using the relevant scaling equations, all the necessary exponents required for a complete characterization of the phase transition can be determined using linear and nonlinear susceptibility measurements alone. This can be considered as a major advantage of measurement of nonlinear susceptibility. Also, owing to the functional form of χ_3 across the phase transition, the determination of the true (zero-field) transition temperature T_C can be made to a better accuracy. It is shown that subtle features not discernable using normal linear susceptibility measurements can be picked up using nonlinear susceptibility, making it a very useful tool for studies of spin systems in the critical region. Hence, in this paper, we show the following advantages of χ_3: (i) determination of all the relevant critical exponents (ii) accurate determination of T_C, and (iii) detecting subtle features not seen in linear susceptibility measurements.

It is important to note that though the present study deals with amorphous ferromagnets, criticality in χ_3 can be used equally effectively in the study of phase transitions in their crystalline counterparts.

II. EXPERIMENTAL DETAILS

A well characterized and extensively studied series of amorphous ferromagnets ($Fe_5Co_{50}Ni_{17-x}Cr_xB_{16}Si_{12}$) with $x=5$, 10, 15 referred to as A2, A3, and A4, respectively, have been used in this study. These samples prepared by melt quenching are in the form of ribbons of approximate dimensions 10 mm$\times$1 mm$\times$0.03 mm).[11]

In one of the earlier works,[6] χ_3 was determined by fitting the measured isothermal magnetization using Eq. (1), or alternatively by plotting M/H versus H^2, and calculating the slope in the linear low-field range.

Another technique involves using the Arrots plots M^2 versus H/M, where the slope in the low-field range can be related to χ_3 through the relation

$$\chi_3 = -\chi_1^4[dM^2/d(H/M)]. \tag{2}$$

However, these methods are unsuitable for studying the critical behavior across the ferromagnetic transition primarily due to two reasons.

(i) The measuring fields are relatively higher and this can at times mask the true critical behavior of the system.

(ii) χ_3 is determined after fitting the dc magnetization data. This method is not only time consuming but also introduces significant errors in the value of χ_3.

In the present study the linear and nonlinear susceptibilities are measured by monitoring the change in the induced voltage across two oppositely wound secondary coils in a home made susceptometer.[16] Both the linear and nonlinear susceptibilities were determined by using a lock-in amplifier to measure the signal seen at ω and 3ω, respectively, where ω is the exciting frequency. Neglecting the higher-order terms, the voltage measured at 3ω is given by

$$V_{3\omega} = \frac{3}{4}\omega\chi_3 H^3,$$

where H is the magnitude of the applied field.

It is normal practice to bundle a few ribbons together to increase the measured signal but in this case a solitary ribbon was stuck on to a sapphire single crystal, which made up the sample holder, with the temperature sensor and a nonmagnetic heater on the same crystal. This was done to avoid temperature gradients across the samples. A calibrated Platinum resistance thermometer was used for measuring the temperature and control to an accuracy of 0.01 K was achieved using a Lakeshore temperature controller DRC-93A. Exciting fields from about 100 mOe to a few Oe and frequencies ranging from 73 Hz to 1.33 kHz were used for different experimental runs. All the measurements were performed with the applied field parallel to the long edges of the samples to minimize the effect of the demagnetization fields.

III. RESULTS AND DISCUSSIONS

A. Linear susceptibility

ac susceptibility is amongst the finest tools for the study of critical phenomenon, not only due to the relative ease with which measurements can be done but also due to the fact that very low-field measurements can be performed. The most important advantage is that it directly gives the true initial susceptibility χ_0, which otherwise has to be calculated from data taken at comparatively higher measuring fields. Considering the fact that the ideal second-order paramagnetic-ferromagnetic phase transition is defined in zero external field, ac susceptibility becomes a very good tool for studying criticality, as larger exciting fields used during the course of measurements are likely to mask the true critical behavior of the system under study. Also, the Kouvel-Fisher (K-F) analysis[17] of the ac-susceptibility data is a good means of accurately determining both the transition temperature T_C as well as the susceptibility exponent γ. Figures 1(a), 1(b), and 1(c) show the Kouvel-Fisher plots $[1/\chi_0 d/dT(\chi_0^{-1})$ versus $T]$ for the three samples. The inverse of the slopes directly gives us the value of the susceptibility exponent γ and the intercepts of the straight line provides us with an independent estimate of the transition temperature T_C. This series has been well studied in the past[11] and the values of T_C and γ reported are in reasonable agreement with the values we have obtained, as is shown in Table I.

B. Nonlinear susceptibility

1. Theory

The critical behavior of χ_3 has been investigated in the past using the Sherrington-Kirkpatric model[18] and the Bethe approximation.[19] The behavior of χ_3 was shown by a simple molecular-field theory[8] by Sato and Miyako. Here we present a detailed calculation based on the molecular-field approach.

Consider a collection of n particles, each with spin 1/2. When an external field H is applied, the net magnetic moment is given by

$$m = n\mu \tanh\left(\frac{\mu H}{kT}\right). \tag{3}$$

where μ is the Bohr's Magneton and k is the Boltzman constant. The net applied field which the dipole sees is given by

$$H = H + \lambda M,$$

where λ is the molecular-field constant. Thus, the magnetization can now be written as

$$M = N\mu \tanh\frac{\mu}{kT}(H+\lambda M), \tag{4}$$

where N is the particle density ($N=n/V$).

Case A: $T>T_C$. In the paramagnetic region, as the magnetization M has inversion symmetry with respect to the applied field H, the magnetization can be written as

$$M = \chi_1 H + \chi_3 H^3 + \chi_5 H^5 + \cdots. \tag{5}$$

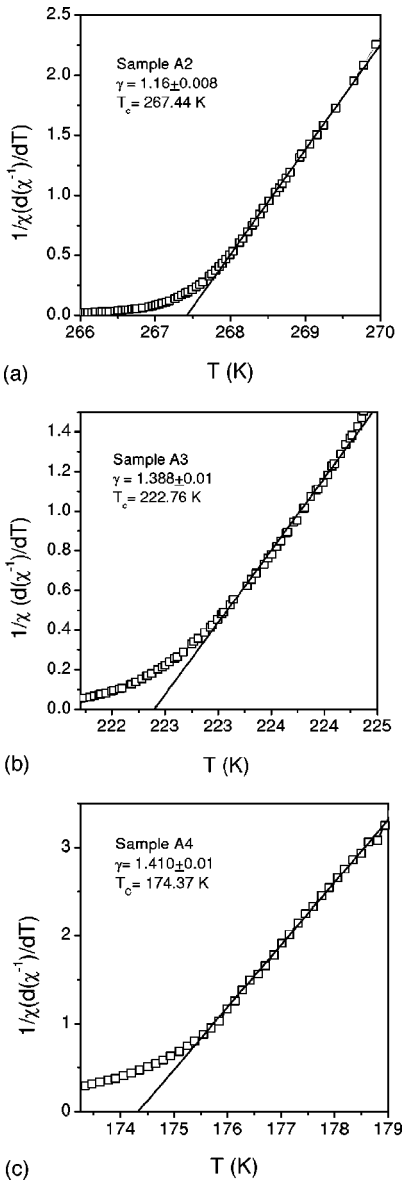

FIG. 1. Kouvel-Fischer plots of the real part of the first-order susceptibility for the samples A2, A3, and A4. The inverse of the slope gives the value of the susceptibility exponent γ, and the intercept on the temperature axis provides the transition temperature T_C.

Substituting this in Eq. (4), we get

$$\chi_1 H+\chi_3 H^3 = N\mu \tanh\frac{\mu}{kT}(H+\lambda[\chi_1 H+\chi_3 H^3+\cdots]). \tag{6}$$

Now, using the expansion

$$\tanh(x)=x-\frac{1}{3}x^3;$$

using the expression for T_C from the mean-field theory, $kT_C=N\mu^2\lambda$; and comparing the coefficients of H, H^3, etc. we get

TABLE I. Values of the susceptibility exponent γ and the transition temperature T_C as determined from the Kouvel-Fischer analysis of the first-order susceptibility. The values determined by the earlier workers is given for the sake of comparison.

	A2	A3	A4
T_C	267.44	222.76	174.37
	267[a]	222.2[a]	174[b]
γ	1.16±0.008	1.388±0.01	1.41±0.01
	1.19[a]	1.38[a]	1.73[b]

[a]Values determined in Ref. 11 using ac-susceptibility measurements.
[b]Values determined in Ref. 11 using high-field dc magnetization.

$$\chi_1=\frac{N\mu^2}{kT_C}\frac{1}{\left(\frac{T}{T_C}-1\right)}, \tag{7}$$

$$\chi_3=-\frac{N\mu^4}{3k^3T^3}\frac{T}{T_C}\frac{1}{\left(\frac{T}{T_C}-1\right)^4}. \tag{8}$$

It is clear that in the limit $T\Rightarrow T_C^+$, χ_1 diverges positively, whereas χ_3 shows a negative divergence.

Case B: $T<T_C$. In the ferromagnetic region, the emergence of spontaneous magnetization causes M to lose its inversion symmetry with respect to H, and now the magnetization can be written as

$$M=M_0+\chi_1 H+\chi_2 H^2+\chi_3 H^3+\cdots, \tag{9}$$

where M_0 is the spontaneous magnetization. Substituting this in Eq. (4) we get

$$M=N\mu\tanh\left\{\frac{\mu}{kT}[\lambda M_0+(\lambda\chi_1+1)H+\lambda\chi_2 H^2+\lambda\chi_3 H^3]\right\}. \tag{10}$$

Expanding $\tanh(x)$ and comparing the coefficients of the different powers of H, we get

$$M_0=N\mu\left[\frac{\mu\lambda M_0}{kT}-\frac{1}{3}\frac{\mu^3\lambda^3 M_0^3}{k^3T^3}\right], \tag{11}$$

$$\chi_1=\frac{N\mu^2}{kT_C}\frac{\left[1-\left(\frac{T_C}{T}\right)^2\left(\frac{M_0}{N\mu}\right)^2\right]}{\left[\frac{T}{T_C}-\left(1-\left(\frac{T_C}{T}\right)^2\left(\frac{M_0}{N\mu}\right)^2\right)\right]}, \tag{12}$$

$$\chi_2=-\frac{kT_C M_0\chi_1^3}{N^3\mu^4}\frac{1}{\left[1-\left(\frac{T_C}{T}\right)^2\left(\frac{M_0}{N\mu}\right)^2\right]^3}, \tag{13}$$

$$\chi_3 = \frac{\chi_1^4}{\left[1-\left(\frac{T_C}{T}\right)^2\left(\frac{M_0}{N\mu}\right)^2\right]^4}\left(\frac{\lambda}{N^2\mu^2}\right)\left[2\frac{\lambda T_C}{T}\frac{M_0^2}{(N\mu)^2}\frac{\chi_1}{\left[1-\left(\frac{T_C}{T}\right)^2\left(\frac{M_0}{N\mu}\right)^2\right]}-\frac{1}{3}\frac{T}{T_C}\right]. \quad (14)$$

Equation (11) is solved for the spontaneous magnetization and yields for $T<T_C$

$$\left(\frac{M_0}{N\mu}\right)^2 = 3\left(\frac{T}{T_C}\right)^2\left(1-\frac{T}{T_C}\right). \quad (15)$$

Substituting this value of $(M_0/N\mu)^2$ in the value of χ_1 and simplifying, we get

$$\chi_1 = \frac{N\mu^2}{2kT_C}\frac{\left[3\frac{T}{T_C}-2\right]}{\left[1-\frac{T}{T_C}\right]}. \quad (16)$$

Hence it is clear that χ_1 has a positive divergence as $T \Rightarrow T_C^-$.

Now, substituting Eqs. (15) and (16) in the value of χ_3 and simplifying, we obtain

$$\chi_3 = \frac{8}{3}\frac{\left(\frac{N\mu^2}{2kT_C}\right)^4}{\left(1-\frac{T}{T_C}\right)^4}\frac{\lambda T}{(N\mu)^2 T_C}, \quad (17)$$

where it is obvious that like χ_1, χ_3 too diverges positively in the limit $T \Rightarrow T_C^-$.

2. Experiments

Normally, χ_3 is seen to show a sharp negative peak around the transition. It has been observed in an electrical analog (across the second-order paraelectric-ferroelectric transition in a triglycine sulfate monocrystal) that the nonlinearity parameter a_3 [$\approx \epsilon_3/(\epsilon_1)^4$, where ϵ_1 and ϵ_3 are the first- and third-order dielectric susceptibilities, respectively] was seen to minimize at T_C but had positive values both above and below the transition temperature.[20] However, as is clear from the preceding section, theoretically, χ_3 is expected to diverge on both sides of the phase transition and change sign exactly at T_C. However, this critical behavior is not easy to observe. The reason for it is that the expansion of M in terms of higher powers of H, which was shown in the preceding section is truly speaking only valid for small values of H. Hence, to observe the true critical behavior, measurements have to be done at very low measuring fields, and higher fields tend to smear off the transition. It is difficult to predict *a priori* the fields at which this critical behavior will be seen, as it depends on the extent of nonlinearity in the system, and thus differs from one sample to another. It must be kept in mind that at higher measuring fields, the contribution from domains can become large, thus masking the true critical behavior of the spin system.

Careful low-field measurements have shown this critical feature in this series of samples. Figures 2(a), 2(b), and 2(c) show the critical behavior in the samples A2, A3, and A4, respectively. As predicted in the preceding section, χ_3 changes sign across T_C and the exact T_C can be directly determined from the crossover of χ_3 on the temperature axis. These experimental T_C values are given in Table II. It is to

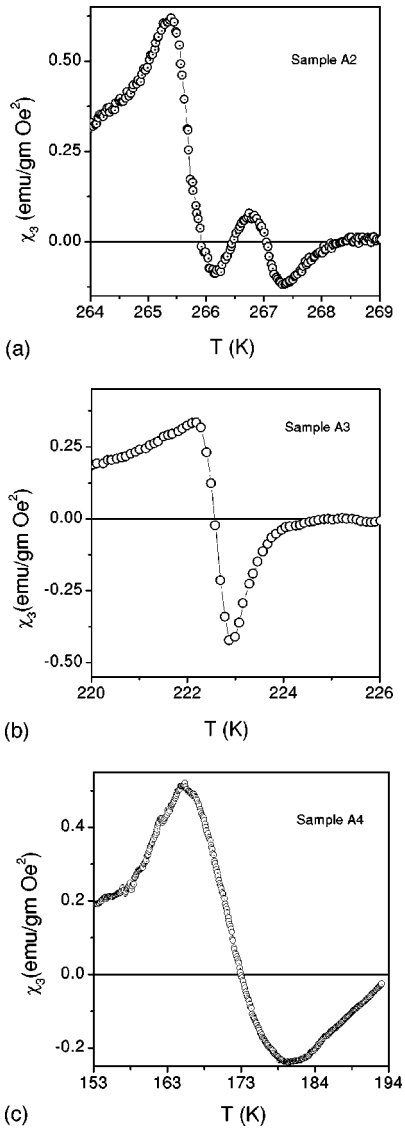

FIG. 2. The critical feature of χ_3 as observed in the real part of the third-order susceptibility in the samples A2, A3, and A4. The measurements were done at a frequency of 133.33 Hz and the exciting field is of the order of 200 mOe.

TABLE II. Values of the all the critical exponents as calculated using linear and nonlinear ac susceptibilities in this series of samples. $T_C(\chi_3)$ is the value of the transition temperature determined from the crossover of χ_3 in the temperature axis. The values quoted by the previous workers is also given for the sake of comparison.

	A2	A3	A4
γ	1.16 ± 0.008	1.388 ± 0.01	1.41 ± 0.01
	1.19 [a]	1.38 [a]	
γ_3	4.57 ± 0.08	4.805 ± 0.12	5.04 ± 0.09
$\triangle$	1.705	1.708	1.815
β	0.545	0.32	0.405
	0.35 [a]	0.41 [a]	0.52 [a]
α	-0.25	0.03	-0.22
	0.2 [a]	-0.2 [a]	-0.7 [a]
δ	3.12	5.33	4.48
	4.42 [a]	4.49 [a]	4.32 [a]
$T_C(\chi_3)$	267.05	222.55	173.45

[a]From Ref. 11.

be noted here that the T_C determined from the crossover of χ_3 matches well with that determined from the Kouvel-Fischer plot (Table I). This matching of T_C's from two different measurements not only substantiates the fact that the measured χ_3 is a genuine response of the spin system but also that the crossover of χ_3 can be used as a direct method to determine T_C. The other advantages of determination of T_C from the crossover of χ_3 will be discussed in the following section.

As is seen in Fig. 2(a), a double transition showing two crossovers is observed in the sample A2. As will be shown later, this sample gives an uncharacteriztically low value of the susceptibility exponent γ, a fact observed by the earlier workers as well. Considering the fact that this sample has the largest percentage of Ni and is closest to the critical concentration x_c, this double transition is not difficult to explain, as

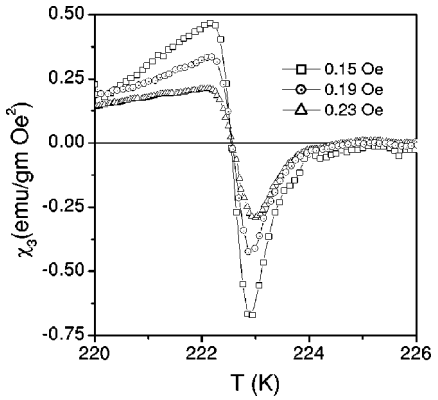

FIG. 3. The field dependence of the third-order susceptibility as seen in the sample A3 at a measuring frequency of 133.33 Hz. As is clearly seen, the critical behavior is supressed with increasing applied field.

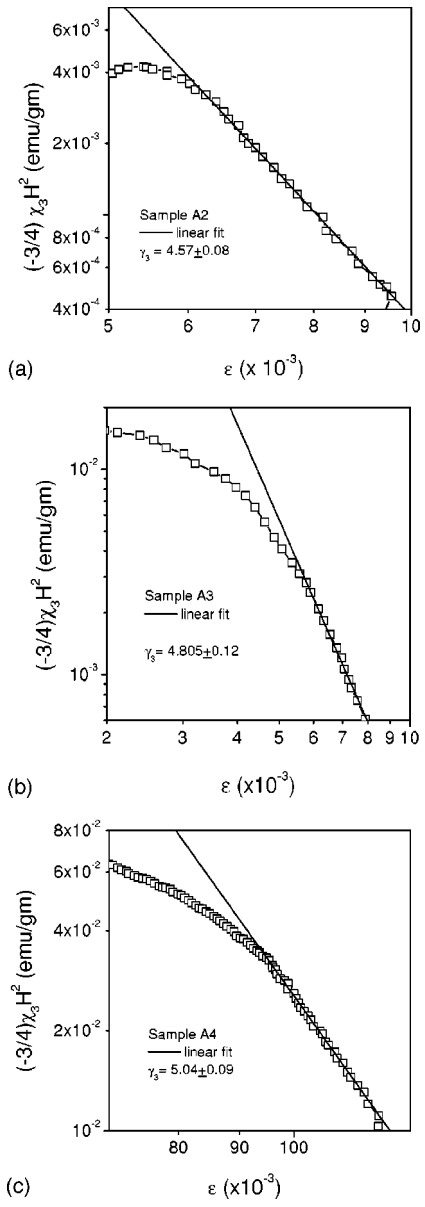

FIG. 4. Double-logarithmic plots of χ_3 vs the reduced temperature for the samples A2, A3, and A4. The slopes of the straight lines give the value of the exponent γ_3.

in this class of materials, the formation of clusters showing a distribution of T_C's have been speculated.[11,14] However, it is interesting to note that no direct evidence of such a distribution is evident from the linear susceptibility measurements. This only goes on to show that low-field nonlinear susceptibility is a more sensitive tool than the linear susceptibility, as far as studies of spin systems near the transition is concerned.

As mentioned earlier, the field in which the measurement is performed is an important consideration in the study of criticality in χ_3. This is clearly shown in Fig. 3, which shows the field dependence of χ_3 in the sample A3 at a measuring frequency of 133.33 Hz. It is clearly seen that the critical feature is sharpest at the lowest measured field, and increas-

ing the measuring field wipes off this critical behavior. It is interesting to note that we have observed a nontrivial frequency dependence in this series of samples . This could possibly be due to the presence of superparamagnetic clusters, as no frequency dependence would be expected in a long range ferromagnetically ordered system. These results will be dealt with in a later communication.

Large uncertainties in $(d/dT)(\chi_3^{-1})$ make it very difficult to determine the value of the critical exponent associated with the third-order susceptibility γ_3, using the Kouvel-Fischer formalism. Hence the value of γ_3 is determined by plotting a double-logarithmic plot of $(-\frac{3}{4})\chi_3 H^2$ versus ϵ, where $\epsilon=[(T-T_C/T_C)]$ is calculated by taking the crossover point of χ_3 as T_C. Figures 4(a), 4(b), and 4(c) shows the double-logarithmic plots for samples A2, A3, and A4, respectively. As shown in the figure, as $T\rightarrow T_C^+$ a curvature from the straight-line fit is clearly seen. This arises due to the presence of higher-order terms close to the transition temperature. Exponent calculations give the value of γ_3 to be 4.57, 4.80, and 5.04 for this series of samples, which matches well with the 3D Heisenberg value of 4.88.

Using the scaling relations $\gamma_3^+=\gamma+2\triangle$, $\triangle=\gamma+\beta$, $\delta=1+\gamma/\beta$, and $\alpha+2\beta+\gamma=2$ (where α, β, γ, and $\triangle$ are the exponents associated with the specific heat, magnetization, susceptibility, and the gap exponent,[21] respectively), all the exponents required for a complete characterization of the system have been determined and are given in Table II. As is clearly seen our values match reasonably with that given in Ref. 11. The only exponent in which a large difference is seen is α which is calculated by using the Rushbrooke equality $\alpha+2\beta+\gamma=2$. The ambiguity in the value of this exponent is probably because the values determined in Ref. 11 are calculated by using both high dc field and low ac field measurements, as is the general practice, whereas in our case α is determined by using low-field linear and nonlinear ac-susceptibility measurements alone.

C. Determination of T_C using nonlinear susceptibility

Accurate determination of T_C has always been an important consideration in the study of criticality across the ferromagnetic transition. The most popular techniques used till date have been the K-F formalism (discussed in Sec. III A) and the Arrots plots.[22]

Arrots plots work on the principle of plotting the isothermal experimental data as M^2 versus H/M at different temperatures in the transition region. They should be straight lines in the critical region with the intercepts of these lines with the H/M axis being positive if $T>T_C$ and negative when $T<T_C$. However, it is important to note that this is valid only when the domain alignment is complete, i.e., the low-field experimental data is completely excluded. This can at times be a nontrivial problem, specially in systems with large field induced effects, causing uncertainties in the accurate determination of T_C. Also, in inhomogeneous systems, such as amorphous ferromagnets, the Arrots plots tend to be curved even at large measuring fields, thus making the identification of T_C even more difficult.[23]

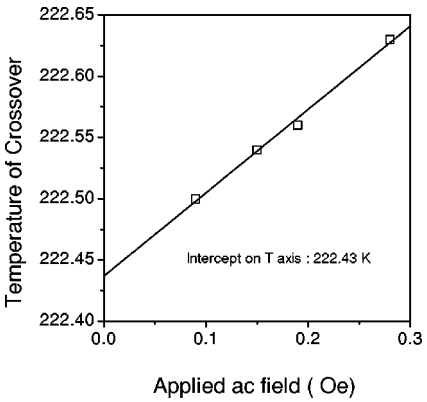

FIG. 5. The temperature of crossover T_C vs the measuring field H_{ac} for the sample A3. A linear behavior is observed and the interpolation to zero applied field gives us a unique way of determining the true transition temperature.

As has been described earlier, χ_3 diverges on both sides of the transition temperature, and changes sign exactly at T_C. Moreover, this critical behavior is extremely sensitive to the magnitude of the field in which the measurement is done, and this critical feature is seen to vanish with increasing field. This makes χ_3 a very promising candidate for an accurate determination of T_C, as even by theory the ferromagnetic transition is defined for the limit $H\Rightarrow 0$, and any large measurement field is likely to change the thermodynamic properties of the system under study. Plotting the temperature of crossover T_C versus the field used during the measurement (H_{ac}), it is clearly seen that T_C varies linearly with the applied field. Thus, extrapolating this to $H=0$, one will be able to determine T_C to a very high accuracy. This is shown in Fig. 5, where T_C versus H is plotted for the sample A3. The intercept at $H=0$ gives us the value of $T_C=222.43$ K.

IV. CONCLUSIONS

A simple molecular treatment of the second-order paramagnetic-ferromagnetic phase transition shows that χ_3 diverges on both sides of the transition and changes over sign at T_C. This criticality has been experimentally observed in a well studied series of amorphous alloys and the related assymptotic critical exponents have been determined. It is shown that sensitive nonlinear susceptibility measurements, along with linear susceptibility, can be used to fully characterize the phase transition, without the need for any other type of measurements, such as dc magnetization, specific heat, etc. Moreover, due to the functional form of χ_3 at the transition, the transition temperature T_C can be determined accurately. Using the field dependence of χ_3, an extrapolation to true zero field can be made, which gives us a unique way of determining the transition temperature. Subtle features not seen in the first-order susceptibility, such as the presence of magnetic clusters with a distribution of T_C's, is observed. This only goes on to show that at times nonlinear susceptibility can be a more sensitive tool than its linear counterpart in understanding the magnetism of spin systems in the critical region.

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