Isotropic-Heisenberg to isotropic-dipolar crossover in amorphous ferromagnets with composition near the percolation threshold

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ac (''zero-field'') susceptibility of the amorphous ferromagnetic alloys $(Fe_pNi_{1-p})_{80}B_{16}Si_4$ $(0.0540 \leqslant p \leqslant 0.1375)$ and $(Co_{p'}Ni_{1-p'})_{80}B_{16}Si_4$ $(0.1125 \leqslant p' \leqslant 0.2375)$ with Fe or Co concentration above the critical concentration p_c or p'_c for the onset of long-range ferromagnetic order has been measured to very high precision in the absence and presence of dc (static) magnetic field over a wide temperature range embracing the critical region near the ferromagnetic-paramagnetic phase transition. Elaborate data analyses permit the *accurate* determination of the *asymptotic* critical exponents β_D, γ_D, and δ_D for spontaneous magnetization, susceptibility and the critical isotherm as well as the leading ''correction-to-scaling'' exponent for susceptibility (these exponents characterize the *isotropic dipolar* fixed point) and hence assert that the asymptotic critical behavior of amorphous ferromagnets with $p_c \lesssim p < 0.1$ or $p'_c \lesssim p' < 0.2$ (that exhibit reentrant behavior at low temperatures) is that of a $d=3$ *isotropic dipolar* ferromagnet. The presently determined values for the exponents β_D, γ_D, and δ_D do satisfy the Widom scaling relation $\beta+\gamma=\beta\delta$ accurately. In addition, the temperature dependence of the *effective* critical exponent for susceptibility observed in the amorphous ferromagnetic alloys with $p<0.1$ or $p'<0.2$ for temperatures above the Curie point displays features *characteristic* of the isotropic dipolar-to-isotropic Heisenberg crossover. A quantitative comparison between theory and experiment exposes certain limitations of the existing theories. By contrast, such a crossover is not observed even at reduced temperatures as close to Curie point T_C as $\epsilon=(T-T_C)/T_C \simeq 10^{-5}$ in the alloys with $p>0.1$ or $p'>0.2$, which behave as normal ferromagnets down to 3.8 K. A sharp contrast in the critical behavior of ferromagnets that either do or do not exhibit reentrant behavior at low temperatures is shown to reflect the decisive role played by the isotropic dipolar long-range interactions in establishing long-range ferromagnetic order in dilute magnetic systems exhibiting reentrant behavior.

I. INTRODUCTION

Recognizing that a complete theoretical description of the thermal critical behavior of *real* $d=3$ (d is the *space* dimensionality) ferromagnets demands that long-range dipole-dipole interactions are considered in addition to the isotropic short-range (nearest-neighbor) Heisenberg exchange interactions, Aharony and Fisher[1] used the renormalization-group (RG) approach to demonstrate that dipolar perturbations make the isotropic short-range (nearest-neighbor) Heisenberg fixed point of RG *unstable* and give rise to a new *stable* ''dipolar'' fixed point, whose nature depends on the *type* (Ising, XY, cubic) of anisotropy present. In an isotropic $d=3$, $n=3$ (n is the spin dimensionality) spin system, *isotropic dipolar* fixed point is characterized by critical exponents whose values differ only slightly[1,2] (by less than 0.5%) from those associated with $d=3$ *pure* isotropic Heisenberg ferromagnet. Subsequent RG treatments[3–6] of ferromagnets with both short-range exchange and long-range dipolar interactions revealed that despite the fact that the *asymptotic* critical behavior of an isotropic dipolar ferromagnets is practically indistinguishable from that of a $d=3$ ferromagnet with isotropic short-range Heisenberg exchange interactions only, their behavior in the intermediate *crossover* region displays certain striking features. Most notably, the *effective* critical exponent for susceptibility as a function of temperature, $\gamma_{eff}(\epsilon)$ (where $\epsilon=(T-T_C)/T_C$ and T_C is the Curie temperature of isotropic dipolar ferromagnet), goes through a deep minimum (a *dip*) in the crossover region for $\epsilon>0$. However, theoretical opinion is divided on the issue of whether or not the dip in $\gamma_{eff}(\epsilon)$ is a *universal* feature of $d=3$ isotropic dipolar ferromagnets. The RG theories that make use of epsilon expansion[3,4] and parquet graph[5] techniques assert that a pronounced dip in $\gamma_{eff}(\epsilon)$ in the crossover region is a *universal* feature of $d=3$ isotropic dipolar systems. On the contrary, the RG calculations employing field-theoretic method[6] reveal that this is true for *weak* isotropic dipolar systems only and that the occurrence of such a minimum in $\gamma_{eff}(\epsilon)$ in systems with *strong* dipolar interactions depends sensitively on the initial value ''u_i'' of the dimensionless renormalized coupling constant ''u,'' which essentially controls the flow of RG trajectories. To elucidate the latter point further, when dipolar interactions are *strong*, three distinctly different cases arise depending upon the value of u_i: (I) for $u_i=1/2$ (the value corresponding to the Heisenberg fixed point), the dip in $\gamma_{eff}(\epsilon)$, *characteristic* of $d=3$ weak dipolar systems, is *retained*, (II) for $u_i>1/2$, γ_{eff} starts with the $d=3$ isotropic dipolar value at $\epsilon=0$, goes through a minimum at $\epsilon=\epsilon_{min}$ and rises steeply to surpass the $d=3$ Heisenberg value for $\epsilon>\epsilon_{min}$, and (III) for $u_i<1/2$, $d=3$

Heisenberg fixed point is either partially or completely wiped out (depending upon the value of u_i) and a direct crossover from isotropic dipolar fixed point to *Gaussian* regime occurs.

Existing magnetic susceptibility $\chi(T)$ data[7–11] on the thermal critical behavior of well-known $d=3$ dipolar ferromagnets EuO and EuS fail to resolve this issue, as is evident from the following remarks. The neutron-scattering data[7] (which yield $\gamma \simeq 1.39$, in agreement with the theoretical estimate[1,2] for either a pure $d=3$ Heisenberg or a pure $d=3$ isotropic dipolar system) do not facilitate an unambiguous comparison with the theoretical predictions due to the uncertainty[2] introduced by the extrapolation method (which assumes the Ornstein-Zernike form for the correlation function) used to obtain zero wave-vector ($q=0$) static susceptibility from q-dependent susceptibility measured in such experiments. On the other hand, although static susceptibility, when measured directly, does not suffer from this drawback, such measurements[8–11] have persistently yielded as low values of the critical exponent γ as $\gamma \simeq 1.29$ for[8–10] EuO and $\gamma = 1.06 \pm 0.05$ for[11] EuS. Considering that the static susceptibility measurements[8–11] cover temperature ranges that fall within the crossover region (hence none of them unravels the true asymptotic critical behavior), it has been argued[3,4] that the reported[8–10] γ values for EuO correspond to the value of γ_{eff} at $\epsilon = \epsilon_{min}$ (i.e., $\gamma_{eff} \simeq 1.28$) predicted[4] by the theory. In complete disagreement with this interpretation, a reanalysis[12] of the existing[8–10] $\chi(T)$ data on EuO that takes into account the corrections to scaling yielded values of γ similar to the one theoretically expected for a pure $d=3$ isotropic Heisenberg ferromagnet. Moreover, the mean-field-like value[11] of γ in EuS has found no explanation so far. Additional complications arise from the theoretical prediction that the cubic anisotropy prevalent in EuO and EuS *destablizes*[2] the isotropic dipolar fixed point and results in a crossover[2] to the cubic-dipolar fixed point as $T \to T_C$. Thus the theoretical prediction concerning the behavior of $d=3$ isotropic dipolar ferromagnets in the crossover region still await experimental confirmation and the existence of a *stable* isotropic dipolar fixed point in such systems has not been experimentally demonstrated so far.

Detailed investigations of the thermal critical behavior of amorphous ($a-$)$Fe_xNi_{80-x}M$ (M stands for the metalloids) alloys with concentration *x well above* the critical concentration x_c for the onset of long-range ferromagnetic order previously undertaken by us[13–15] revealed that *weak isotropic* long-range dipole-dipole interactions *coexist* with relatively strong isotropic ''Heisenberg-like'' exchange interactions in these systems. In view of this finding, *weak* isotropic ''short-range'' exchange and weak isotropic dipolar interactions both are expected to be present in amorphous ferromagnetic alloys with *x just above* x_c. Hence such systems are ideally suited for rigorously testing the theoretical predictions. That this is indeed the case is amply demonstrated in this paper. The results reported here assert that the asymptotic critical behavior of amorphous ferromagnets with $x \gtrsim x_c$ is that of a $d=3$ isotropic dipolar ferromagnet, provide a conclusive evidence for the occurrence of isotropic Heisenberg-to-isotropic dipolar crossover as the temperature is lowered towards T_C and expose certain inadequacies of the existing theories.

II. THEORETICAL CONSIDERATIONS

Before embarking upon a detailed data analysis, a brief account of the theoretical considerations that form the basis for such an analysis is given below. According to the general theory[3,4,17,18] of crossover, ''zero-field'' susceptibility of a $d=3$ isotropic short-range Heisenberg ferromagnet with *weak* isotropic dipolar interactions of normalized strength g, for temperatures close to criticality, behaves as[3,4]

$$\chi = \Gamma \epsilon_H^{-\gamma_H} X(y), \tag{1}$$

where Γ is a nonuniversal constant, $\epsilon_H = [T - T_C(0)]/T_C(0)$, $T_C(0) \equiv T_C(g=0)$ is the transition temperature, and γ_H is the susceptibility critical exponent of pure ($g=0$) isotropic short-range Heisenberg ($d=3$, $n=3$) spin system, $y = x/x_g$, $x \equiv g/\epsilon_H^{\phi}$, $x_g \equiv g/\epsilon_g^{\phi}$, $\epsilon_g = [T_C(g) - T_C(0)]/T_C(0)$ is the shift in the transition temperature caused by long-range isotropic dipolar interactions, ϕ is the crossover exponent which, in the case under consideration, *equals*[1] γ_H, and $X(y)$ is the crossover scaling function. The asymptotic critical behavior is determined by the singularity in $X(y)$ occurring at $y=1$ (or equivalently, at $\epsilon = [T - T_C(g)]/T_C(g) = 0$):

$$X(y \approx 1) \sim (1-y)^{-\gamma_D}, \tag{2}$$

where γ_D is the susceptibility critical exponent and $T_C \equiv T_C(g)$ is the true transition temperature of isotropic dipolar ferromagnet. According to Eqs. (1) and (2), dipole-dipole interactions are expected to manifest themselves in the vicinity of a crossover temperature $\epsilon_{co} \equiv g^{1/\phi}$ such that for $\epsilon \gg \epsilon_{co}$ the spin system behaves as a *pure* $d=3$, $n=3$ system whereas for $\epsilon \ll \epsilon_{co}$ the asymptotic critical behavior is that of a $d=3$ isotropic dipolar ferromagnet. A detailed RG calculation[3,4] of the crossover scaling function yields the final expression for susceptibility and its *effective* critical exponent, defined as[18] $\gamma_{eff}(\epsilon) = \partial[\ln \chi^{-1}(\epsilon)]/\partial(\ln \epsilon)$, as

$$\chi(y) = \tilde{\Gamma} y^{\gamma_H/\phi}(1-y)^{-\gamma_D} p(y) \tag{3}$$

and

$$\gamma_{eff}(y) = (1 - y^{1/\phi})\left[\gamma_H + \phi\gamma_D\left(\frac{y}{1-y}\right) + \phi\left(\frac{y p'(y)}{p(y)}\right)\right], \tag{4}$$

where $p(y)$ is the ''correction-to-scaling'' function and $y^{-1/\phi} - 1 = \hat{\epsilon} = [(1+\epsilon_g)/\epsilon_g]\epsilon$. The explicit forms of $p(y)$ and its y derivative $p'(y)$ are given in Ref. 4. Equations (3) and (4) are valid across the entire crossover region and yield the values γ_D and γ_H for the susceptibility exponent γ in the isotropic long-range dipolar ($y \to 1$) and isotropic short-range Heisenberg ($y \ll 1$) limits. Moreover, in the asymptotic critical regime ($0 < \epsilon \ll \epsilon_{co}$) $y \simeq 1$, the function $p(y)$ in Eqs. (3) and (4) can be expanded to obtain[19]

$$\chi(\epsilon) = A_\chi \epsilon^{-\gamma_D}\,[1 + a_\chi \epsilon^{\Delta_D}], \tag{5}$$

$$\gamma_{eff}(\epsilon) = \gamma_D - a_\chi\,\Delta_D\,\epsilon^{\Delta_D}, \tag{6}$$

$$a_\chi \simeq 0.099\epsilon_g^{-\Delta_D}, \tag{7}$$

and

$$\epsilon_g \simeq 0.349\dot{\epsilon}, \tag{8}$$

where a_χ and Δ_D are the *leading* ''correction-to-scaling'' amplitude and exponent, respectively, and $\dot{\epsilon}$ is the reduced temperature ϵ at which the inverse initial (*intrinsic*) susceptibility χ^{-1} *equals* 4π.

III. EXPERIMENTAL DETAILS

Amorphous $(a-)(Fe_pNi_{1-p})_{80}B_{16}Si_4$ ($p=0.0540$, 0.0875, 0.1125, 0.1375) and $(Co_{p'}Ni_{1-p'})_{80}B_{16}Si_4$ ($p'=0.1125$, 0.1375, 0.2375) alloys were prepared in the form of long ribbons of cross section $0.04\times 2\,\mathrm{mm}^2$ by the single-roller melt-quenching technique under high-purity (99.999%) argon atmosphere using 99.998% (99.98%) pure Fe, Co, Ni, (B, Si) as starting materials. Amorphous nature of the ribbons so produced was first verified by x-ray-diffraction method and later confirmed by high-resolution scanning electron microscopic examination which revealed no crystalline regions. Actual concentrations of Fe, Co, Ni in the alloys in question were determined by the inductively coupled plasma method and optical emission spectroscopy and found to agree with the nominal ones (p or p' given above) to within ± 0.0007. More details about sample preparation and characterization have been furnished in our earlier reports[13–15] on similar alloy systems.

High-resolution (relative accuracy better than 10 ppm) ac susceptibility (χ_{ext}) measurements were performed on 30-mm-long alloy ribbons in the absence and presence of dc (static) magnetic fields H_{dc} at various[16] *fixed* frequencies ($18.7\leqslant\nu\leqslant 870$ Hz) and rms amplitudes ($1\leqslant H_{ac}\leqslant 300$ mOe) of ac driving field H_{ac}. Considerations such as optimum signal-to-noise ratio and *linear* response to H_{ac} when $H_{dc}=0$ (i.e., χ_{ext} is *independent* of the magnitude of H_{ac}, which in the present case is true for $H_{ac}\leqslant 25$ mOe) at a given temperature restrict the choice of H_{ac} and ν to 10 mOe and 87 Hz, respectively. Both H_{ac} and H_{dc} were applied along the length within the ribbon plane so as to minimize the demagnetizing effects. Zero-field ($H_{dc}=0$) ac susceptibility was measured after compensating for the Earth's magnetic field. Each data set was taken in two[16] runs. In the first run, χ_{ext} measurements were carried out at fixed (to within ± 5 mK) temperatures, 0.1 K apart, over a wide temperature range embracing the critical region near the ferromagnetic (FM)-paramagnetic (PM) phase transition and T_C was roughly identified with the temperature at which the peak occurs in $\chi_{ext}(T)$. In the second run, $\chi_{ext}(T)$ was measured in steps of $\simeq 25$ mK in the range $-0.05\lesssim\epsilon=(T-T_C)/T_C\lesssim 0.05$. Sharp cusps in the real part of the measured zero-field ac susceptibility, $\chi'_{ext}(T)$, at $T\simeq T_C$ (Fig. 1) and transition widths (defined as the interval bounded by the temperatures corresponding to the (Hopkinson) ''peak'' and ''knee'' in $\chi'_{ext}(T)$ curve) <1 K are indicative of very good chemical homogeneity of the samples. The Hopsinkon peak is a result of competing claims made by the critical order parameter fluctuations, which tend to diverge the susceptibility at T_C, and the formation of magnetic domains for temperatures just below T_C, which restricts the spin-spin correlation length to the size of domains and hence prevents the susceptibility from diverging. By comparison, the temperature at which the knee occurs in $\chi'_{ext}(T)$ curve marks roughly

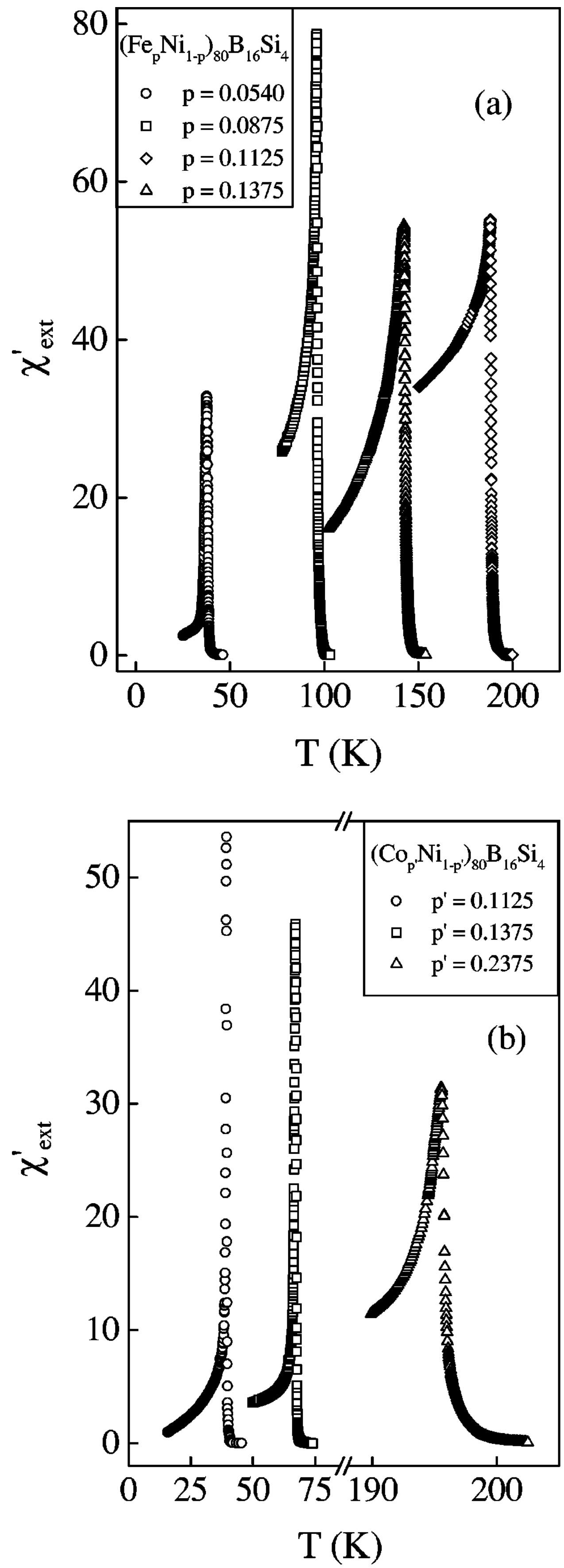

FIG. 1. Real component of zero-field ac susceptibility as a function of temperature for amorphous $(Fe_pNi_{1-p})_{80}B_{16}Si_4$ and $(Co_{p'}Ni_{1-p'})_{80}B_{16}Si_4$ alloys.

the temperature beyond which the critical fluctuations of the order parameter become less important.

IV. DATA ANALYSIS AND RESULTS

Previous study[20] of magnetic phase diagrams for the amorphous alloy systems in question revealed that (i) the critical concentrations for the onset of long-range ferromagnetic order in $a-(Fe_pNi_{1-p})_{80}B_{16}Si_4$ and $a-(Co_{p'}Ni_{1-p'})_{80}B_{16}Si_4$ alloys are $p_c = 0.0285(5)$ and $p'_c = 0.0688(5)$, respectively, and (ii) the alloys with composition in the range $p_c \lesssim p \lesssim 0.1$ or $p'_c \lesssim p' \lesssim 0.2$ exhibit reentrant[20] behavior at temperatures well below the Curie temperature whereas the alloys with $p > 0.1$ or $p' > 0.2$ behave as normal ferromagnets for temperatures down to 1.6 K. The present results have to be viewed against this background.

A. Zero-field susceptibility

The *intrinsic* (volume) susceptibility $\chi(T)$ is obtained from the real part of the measured zero-field ac susceptibility $\acute{\chi}_{ext}(T)$ (Fig. 1) using the relation

$$\chi^{-1}(T) = \acute{\chi}_{ext}^{-1}(T) - 4\pi N, \tag{9}$$

where N is the demagnetizing factor. Since in the present case N ($\simeq 5\times 10^{-4}$) is negligibly small, $\chi^{-1}(T)$ essentially equals $\acute{\chi}_{ext}^{-1}(T)$. The $\chi^{-1}(T)$ data for different compositions, obtained by correcting $\chi^{-1}{}_{ext}(T)$ for demagnetization, are displayed in Fig. 2 and analyzed in terms of the single power law,

$$\chi^{-1}(T) = A_{eff}\epsilon^{\gamma_{eff}}, \quad \epsilon > 0, \tag{10}$$

with the aid of ''range-of-fit'' analysis.[13–16] In this type of analysis, changes in the values of free fitting parameters [e.g., A_{eff}, T_C and γ_{eff} in Eq. (10)] and the sum of deviation squares are monitored as the fit range $\epsilon_{min} \leqslant \epsilon \leqslant \epsilon_{max}$ is varied by keeping ϵ_{min} *fixed* at its lowest value (which in the present case is $\simeq 10^{-5}$) and progressively raising ϵ_{max} from a value not too far above ϵ_{min} to the upper limit of the temperature range covered in the experiment. The range-of-fit analysis,[13–16] based on Eq. (10), is performed in two steps. In the first step, T_C is accurately determined from the $\chi^{-1}(T)$ data spanning temperatures in the immediate vicinity of T_C by varying all the three parameters A_{eff}, T_C, and γ_{eff}. In the second step, T_C is kept constant at the value obtained in step one and the temperature dependence of γ_{eff}, depicted in Fig. 3, is arrived at by optimizing the values of A_{eff} and γ_{eff} in different temperature ranges. The salient features of the $\gamma_{eff}(\epsilon)$ data, presented in Fig. 3, are as follows. For the alloys that exhibit *reentrant* behavior at low temperatures, (I) a *dip* in γ_{eff} occurs at ϵ_{min} and ϵ_{min} shifts rapidly towards $\epsilon = 0$ as the Fe or Co concentrations increases, (II) *irrespective* of the alloy composition, γ_{eff} has the value $\gamma_{eff}^{min} = 1.280(2)$ at ϵ_{min}, and (III) γ_{eff} assumes the values $\gamma_D \simeq 1.40$ and $\gamma_{eff}(\epsilon^{**}) = \gamma_H \simeq 1.38$ for $\epsilon \simeq 0$ and $\epsilon \gg \epsilon_{min}$, respectively. By contrast, no such dip in $\gamma_{eff}(\epsilon)$ is observed at any temperature above T_C for the alloys with $p > 0.1$ or $p' > 0.2$, which behave as *normal ferromagnets* down to the lowest temperature; instead (I') γ_{eff} possesses

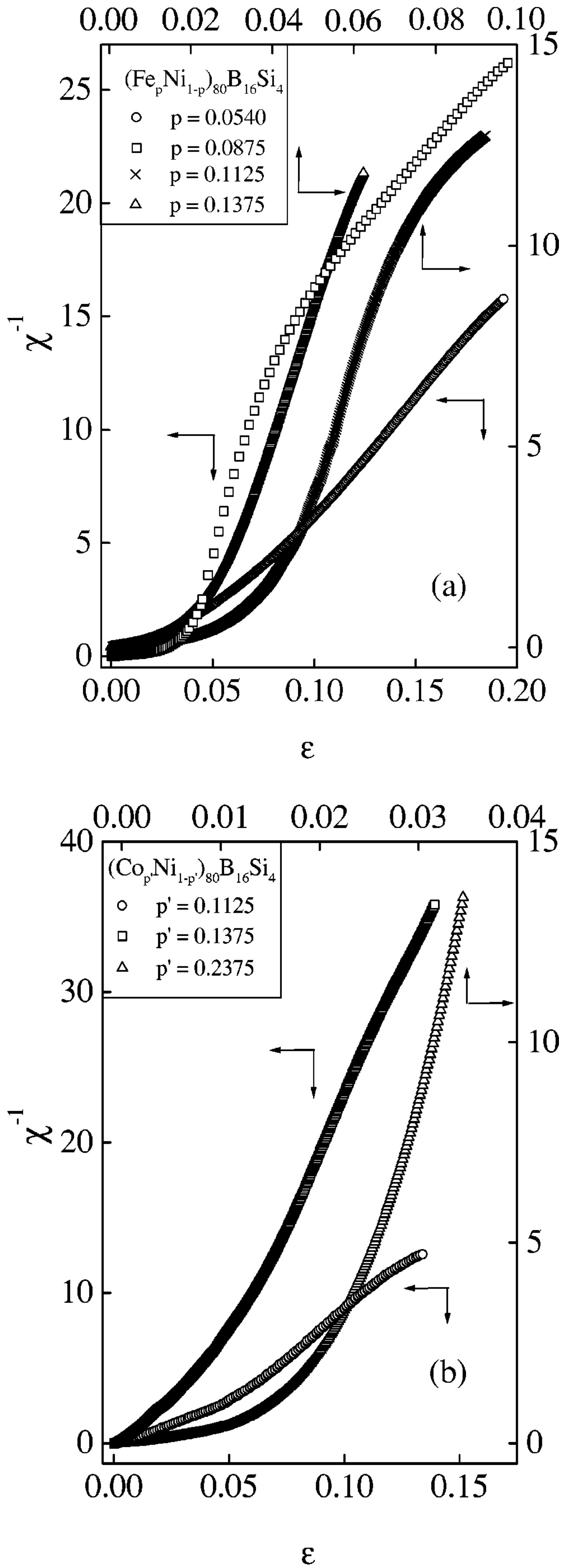

FIG. 2. Inverse intrinsic susceptibility versus reduced temperature. Arrows indicate the relevant ordinate and abscissa scales.

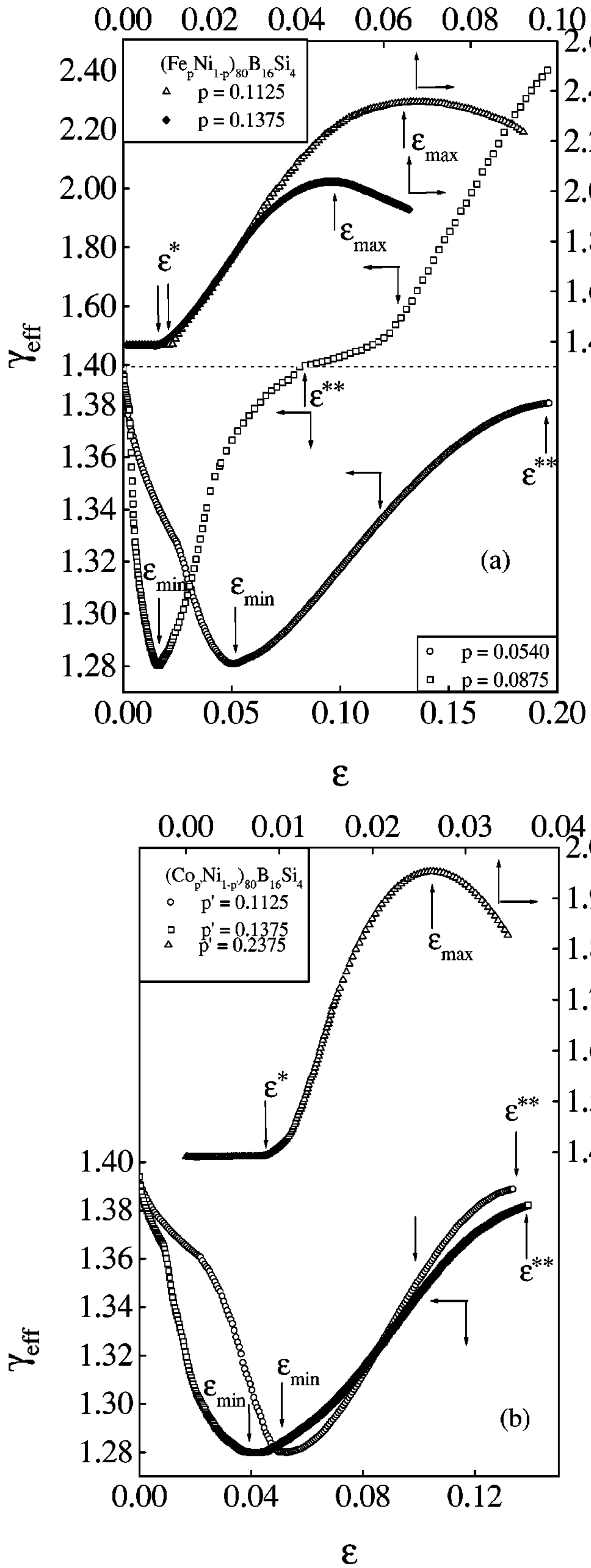

FIG. 3. Temperature variation of the effective critical exponent for susceptibility γ_{eff}. $\epsilon_{min}, \epsilon_{max}, \epsilon^*$, and ϵ^{**} mark, respectively, the temperatures corresponding to the minimum, maximum, crossover, and limiting values of γ_{eff} at high temperatures in a given $\gamma_{eff}(\epsilon)$ curve. Note the change in the sensitivity of the ordinate scale at $\epsilon^{**} \simeq 0.08$ for the $\gamma_{eff}(\epsilon)$ data (open squares) of the alloy with $p = 0.0875$ in the series $(Fe_pNi_{1-p})_{80}B_{16}Si_4$.

the $d=3$ isotropic Heisenberg value of 1.386(4) for temperatures in the close proximity to T_C ($\epsilon \leqslant \epsilon^*$), ($II'$) for $\epsilon > \epsilon^*$, γ_{eff} increases steeply to go through a *peak* at ϵ_{max}, and (III') the peak value γ_{eff}^p reduces drastically while ϵ^* and ϵ_{max} get displaced towards T_C as p (or p') increases. Now that the composition $p=0.0875$ is close to the boundary[20] between reentrant and ferromagnetic phases in the magnetic phase diagram of the $a-(Fe_pNi_{1-p})_{80}B_{16}Si_4$ alloy series, $\gamma_{eff}(\epsilon)$ exhibits a *dip* as well as a *peak* (not shown in Fig. 3) for this alloy.

With a view to ascertain the asymptotic critical behavior of zero-field susceptibility, $\chi(T)$ data have been analyzed in terms of the single power law, i.e., Eq. (10), and the expression, Eq. (5), that includes the leading correction-to-scaling term, using the range-of-fit analysis. Such an exercise reveals that in a narrow temperature range $T \geqslant T_C$ (Table I), Eq. (5) describes the $\chi(T)$ data *far better* than Eq. (10) in the case of amorphous reentrants ($p_c \lesssim p \lesssim 0.1$ or $p_c' \lesssim p' \lesssim 0.2$) whereas the reverse is true for the amorphous ferromagnets with $p > 0.1$ or $p' > 0.2$. The values for the *asymptotic* critical exponent γ_D (or γ_H), correction-to-scaling amplitude a_χ and correction-to-scaling exponent Δ_D, that characterize the *isotropic dipolar* (or isotropic Heisenberg) fixed point, obtained in this way and listed in Table I, are cross checked by least-squares fitting Eq. (6) to the $\gamma_{eff}(\epsilon)$ data in the temperature ranges specified in Table I. The continuous curves and straight lines through the $\chi^{-1}(\epsilon)$ and $\gamma_{eff}(\epsilon^{\Delta_D})$ data points, symbols, in Fig. 4 (Fig. 5) testify to the validity of Eqs. (5) and (6), respectively, [Eq. (10) with $\gamma_{eff}=\gamma_H$], and to the accuracy of the presently determined values of $\gamma_D(\gamma_H), \Delta_D$, and a_χ displayed in Table I.

B. In-field susceptibility

Figure 6 serves to illustrate the effect of the static biasing field H_{dc} ($\leqslant 14.5$ Oe) on the temperature variation of (ac) susceptibility. As H_{dc} grows in strength, the Hopkinson peak, evident in the zero-field susceptibility (Fig. 1), progressively broadens, gets suppressed, and shifts to *lower* temperatures, revealing, in the process, a secondary maximum in $\chi(T)$ at $T \approx T_C$. The data presented in Fig. 7 and 8 highlight the *typical* static field dependence of such secondary maxima observed in amorphous reentrants and normal ferromagnets, respectively. In both the cases, the secondary peak reduces in height (χ_m) and its position (T_m) gets displaced to *higher* temperatures as H_{dc} increases. It is widely accepted[21–25] that such peaks are a *characteristic signature* of critical fluctuations of the order parameter that drive the (second-order) magnetic phase transition at T_C since the field and temperature dependences of these peaks are consistent with the static scaling description of such a transition.

Magnetic behavior near the ferromagnetic (FM)-paramagnetic (PM) phase transition is described by the scaling equation of state[23,26,27]

$$m(h,t) = t^\beta F(h/t^{\beta+\gamma}), \quad (11)$$

where the (unknown) scaling function F is different for temperatures below and above T_C, $t=(T-T_C)/T_C$ stands for both ϵ_H and $\epsilon \equiv \epsilon_D$, h is H_{dc} corrected for demagnetization, h and t are the conventional linear relevant scaling fields,[27] the critical exponents β, γ, and δ, for spontaneous magneti-

TABLE I. Comparison between experiment and theory. Abbrevations: ID = isotropic dipolar, IH = isotropic Heisenberg, p and p' denote the Fe and Co concentrations in the amorphous alloy series $a-(\mathrm{Fe}_p\mathrm{Ni}_{1-p})_{80}\mathrm{B}_{16}\mathrm{Si}_4$ and $a-(\mathrm{Co}_{p'}\mathrm{Ni}_{1-p'})_{80}\mathrm{B}_{16}\mathrm{Si}_4$ while D (H) in the subscript/superscript refer to the quantities relevant to the isotropic dipolar (isotropic Heisenberg) fixed point.

Parameters	Experiment/alloy composition							RG theory	
	p				p'			$d=3$	$d=3$
	0.0540	0.0875	0.1125	0.1375	0.1125	0.1375	0.2375	ID [a]	IH [b]
$T_C(g)\equiv T_C^D(K)$	38.735(5)	96.040(5)			39.675(5)	67.595(5)			
γ_D	1.397(2)	1.396(2)			1.396(2)	1.396(2)		1.372	
a_χ	0.90(1)	0.83(1)			0.53(1)	0.76(1)			
Δ_D	0.53(5)	0.54(5)			0.54(5)	0.55(5)		0.425	
Fit range for above parameters									
$\epsilon(10^{-4})$	4.1-275	0.52-28.2			6.3-223	0.59-89.4			
ϵ_{min}	0.050(2)	0.016(1)			0.053(1)	0.041(2)			
γ_{eff}^{min}	1.281(1)	1.281(1)			1.280(1)	1.280(1)		1.28	
$\epsilon_g\simeq 0.349\dot{\epsilon}$	0.054	0.018			0.047	0.024			
$T_C(0)=\frac{T_C(g)}{1+\epsilon_g}(K)$	36.75(1)	94.38(1)			37.90(1)	66.00(1)			
$\gamma_H\simeq\gamma_{eff}(\epsilon^{**})$	1.382	1.390			1.388	1.384			
β_D	0.364(4)	0.366(4)			0.364(4)	0.367(3)		0.381	
δ_D	4.82(3)	4.82(3)			4.83(3)	4.80(3)		4.45	
$\beta_D+\gamma_D$	1.760(3)	1.762(2)			1.759(2)	1.764(3)		1.753	
$\beta_D\delta_D$	1.754(30)	1.764(30)			1.758(30)	1.761(25)		1.700	
$T_C^H(K)$			142.380(5)	187.950(5)			195.690(5)		
β_H			0.364(6)	0.366(6)			0.365(5)		0.365(3)
γ_H			1.386(2)	1.385(2)			1.387(2)		1.386(4)
Fit range for γ_H									
$\epsilon(10^{-4})$			0.63-106	0.63-79.5			0.2-81.9		
δ_H			4.79(4)	4.81(4)			4.81(2)		4.80(4) [c]
$\beta_H+\gamma_H$			1.750(4)	1.752(6)			1.751(5)		
$\beta_H\delta_H$			1.744(40)	1.760(40)			1.755(35)		

[a]References 2 and 4.
[b]Reference 28.
[c]Value obtained through the scaling relation $\delta = 1+(\gamma/\beta)$.

zation $m(0,t)$, zero-field susceptibility $\chi(0,t)$, and critical $m-h$ isotherm $m(h,0)$, that characterize the FM-PM phase transition, are defined as $m(0,t)\sim t^{\beta}(t<0)$, $\chi(0,t)\sim t^{-\gamma}(t>0)$, and $m(h,0)\sim h^{1/\delta}(t=0)$. Equation (11) permits determination of in-field susceptibility $\chi(h,t)$ as follows:

$$\chi(h,t)=\partial m/\partial h=h^{(1/\delta)-1}G(h/t^{\beta+\gamma}), \tag{12}$$

where $G(x)=x^{\gamma/(\beta+\gamma)}\dot{F}(x)$ and $\dot{F}(x)=\partial F/\partial h$. In arriving at Eq. (12), use has been made of the Widom scaling equality $\beta+\gamma=\beta\delta$. A maximum in susceptibility, measured in a *fixed* value of h (so that the prefactor $h^{1/\delta-1}$ is constant) as a function of temperature, occurs when $d\chi/dt=0$, which implies that G does not depend on t at $t=t_m=(T_m-T_C)/T_C$. This condition is satisfied if $h/t^{\beta+\gamma}=$const. It immediately follows that the peak position t_m and peak height χ_m depend on h as

$$t_m\sim h^{1/(\beta+\gamma)} \tag{13}$$

and

$$\chi_m(h,t_m)\sim h^{(1/\delta)-1}. \tag{14}$$

The $t_m(h)$ and $\chi_m(h,t_m)$ data, *characteristic* of amorphous reentrants (e.g., the alloy with $p=0.0875$) and ferromagnets (e.g., the alloy with $p'=0.2375$) investigated in this work, displayed in Fig. 9, demonstrate the validity of Eqs. (13) and (14). The values of the critical exponents $\beta+\gamma$ (hence of β as γ has already been accurately determined from the zero-field susceptibility data taken in the asymptotic critical region) and δ obtained in this way are listed in Table I. In sharp contrast with the customary approach of estimating critical exponents β and γ from spontaneous magnetization (for $T<T_C$) and zero-field susceptibility (for $T>T_C$) data obtained through *extrapolation* to zero field of the magnetization data taken in *finite* fields, the present method of determining these exponents completely avoids such extrapolations and hence uncertainties[23] inherent in them.

V. DISCUSSION

Table I compares the presently determined values of the asymptotic critical exponents as well as the leading correction-to-scaling amplitude and exponent with those predicted by the RG theories[2,4,18,28] for pure $d=3,n=3$ spin systems with or without isotropic dipolar long-range interac-

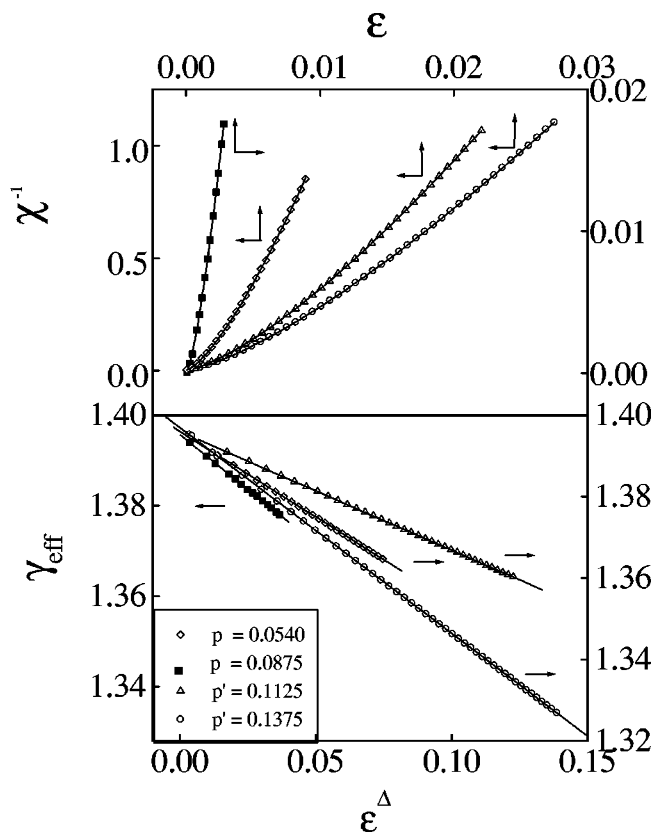

FIG. 4. Temperature variations of χ^{-1} and γ_{eff} in the asymptotic critical region of the ferromagnetic-paramagnetic phase transition for the alloys with $p=0.0540$, 0.0875, and $p'=0.1125$, 0.1375 in the series $(Fe_pNi_{1-p})_{80}B_{16}Si_4$ and $(Co_{p'}Ni_{1-p'})_{80}B_{16}Si_4$ that exhibit reentrant behavior at low temperatures. The continuous curves and straight lines through the $\chi^{-1}(\epsilon)$ and $\gamma_{eff}(\epsilon^{\Delta_D})$ data points depict the best least-squares fits based on Eqs. (5) and (6) of the text.

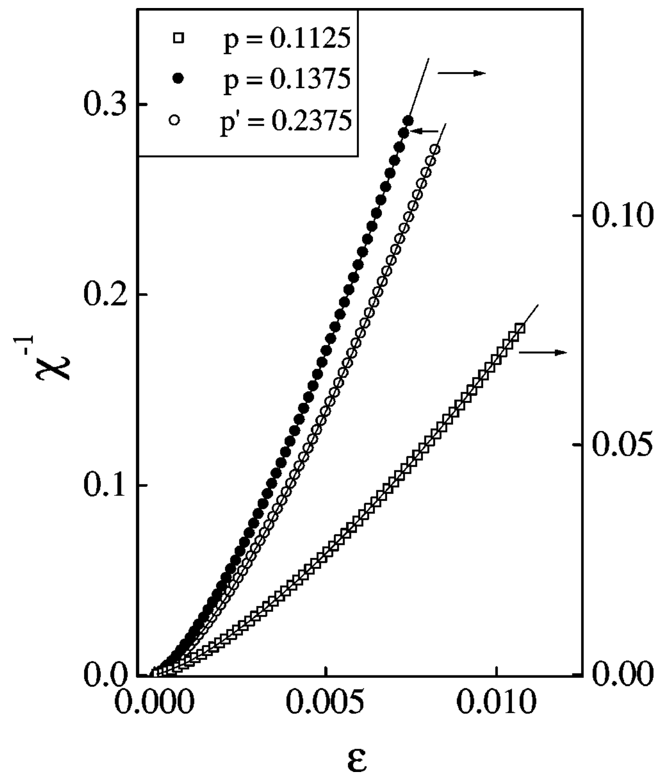

FIG. 5. Temperature dependence of χ^{-1} in the critical region ($\epsilon \leqslant \epsilon^*$) for the alloys with $p=0.1125$, 0.1375 and $p'=0.2375$ in the series $(Fe_pNi_{1-p})_{80}B_{16}Si_4$ and $(Co_{p'}Ni_{1-p'})_{80}B_{16}Si_4$ that behave as normal ferromagnets down to the lowest temperature. The curves through the $\chi^{-1}(\epsilon)$ data points represent the best least-squares fits based on Eq. (10) of the text.

tions. From a perfect agreement between the experimental and theoretical values of the critical exponents β_H, γ_H, and δ_H, we infer that the critical behavior of the alloys with $p=0.1125$, 0.1375, and $p'=0.2375$ (which behave as *normal ferromagnets* down to the lowest temperature) is that of a $d=3$ isotropic Heisenberg (IH) ferromagnet. A *peak* in $\gamma_{eff}(\epsilon)$ (Fig. 3) concomitant with $d=3$ Heisenberg-like critical behavior is a feature that these alloys share with a host of other *disordered* $d=3$, $n=3$ spin systems.[13–15,21–23] This feature, *characteristic* of such systems, has been generally attributed to the *interplay*[13–15,22,23] between the length scales describing the *infinite* $d=3$ ferromagnetic (FM) matrix or network and *finite* FM spin clusters within the framework of the model originally proposed by Kaul.[23] In sharp contrast with the temperature dependence of γ_{eff} observed in normal ferromagnets with quenched disorder, the $\gamma_{eff}(\epsilon)$ in the alloys with $p=0.054$, 0.0875 and $p'=0.1125$, 0.1375 (that enter into the *reentrant* state at temperatures well below the Curie point) exhibits a pronounced *dip* at ϵ_{min} and attains different limiting values as $\epsilon \to 0$ and $\epsilon(\gg \epsilon_{min}) \to \epsilon^{**}$ (Fig. 3). In order to ascertain whether or not the observed variation of γ_{eff} with ϵ for the amorphous reentrants is the experimental signature of the isotropic short-range Heisenberg (IH) to isotropic dipolar (ID) crossover, which the RG theories[3,4,6] predict for a $d=3$ ferromagnet with both isotropic short-range Heisenberg exchange and isotropic long-range dipolar interactions, a detailed quantitative comparison between the experiment and theory is attempted below.

Having determined ϵ_g (the shift in the transition temperature caused by long-range isotropic dipolar interactions) from Eq. (8), and thereby $\hat{\epsilon}=(1+\epsilon_g)/(\epsilon/\epsilon_g)$, γ_{eff} is plotted against $\log_{10}\hat{\epsilon}$ in Fig. 10 for the alloys exhibiting reentrant behavior and the observed variation is compared with that predicted by the RG theories, which make use of two-loop matching technique[2,4] (continuous curve) or field-theoretic method[6] (dashed curve[29]). The more accurate[29] RG calculation of the two, leading to Eq. (4) and the continuous curve in Fig. 10, yields the *same* value for γ_{eff}^{min} as the experimentally determined one (Table I) and the variation of γ_{eff} with $\hat{\epsilon}$ that is in better agreement with the observed $\gamma_{eff}(\hat{\epsilon})$. However, even this RG theory[3,4] predicts a much *broader* crossover region and *lower* values for the susceptibility critical exponents γ_D and γ_H, as is evident from Table I. The entries in this table also reveal that the *composition independent*, and hence universal, magnitudes of the critical exponents $\beta_D=0.365(5)$, $\gamma_D=1.397(3)$ [or $\gamma_H=1.386(4)$], $\delta_D=4.82(5)$, and the leading correction-to-scaling exponent $\Delta_D=0.54(6)$ [characterizing the ID (or IH) fixed point], obtained in this work, substantially differ from the corresponding theoretical estimates.[2,4] Similar discrepancy is found between the experimental values of the leading correction-to-scaling amplitude a_χ and those calculated using Eqs. (7) and (8) (see Table I). Such a disparity exposes the inherent limi-

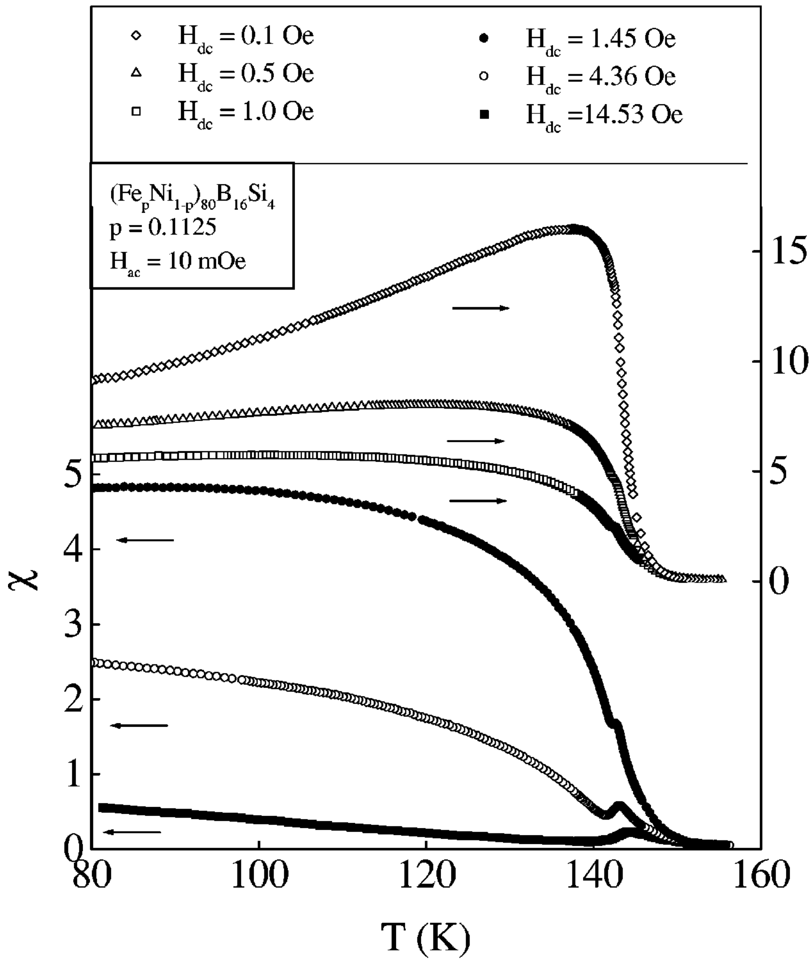

FIG. 6. ac susceptibility as a function of temperature for the $a-(Fe_pNi_{1-p})_{80}B_{16}Si_4$ alloy with $p=0.1125$ measured at various fixed static biasing field (H_{dc}) values. The data presented in this figure demonstrate the suppression and broadening of the Hopkinson peak observed [Fig. 1(a)] in zero-field susceptibility, as well as the emergence, evolution and shifting to higher temperatures of the critical peak at $T \approx T_C$ with increasing H_{dc}.

tation of the epsilon expansion method to yield accurate numerical estimates for the quantities just mentioned. Notwithstanding this numerical disagreement, there seems to be some internal consistency between the theoretical predictions and experimental observations when it comes to the relative magnitudes of the *asymptotic* critical exponents of pure isotropic Heisenberg ferromagnet ($\beta_H, \gamma_H, \delta_H$, and Δ_H) and those ($\beta_D, \gamma_D, \delta_D$, and Δ_D) of $d=3$, $n=3$ spin system with isotropic dipolar interactions, as elucidated below. The RG calculations, due to Bruce and Aharony[2] yield the result that $\beta_D \simeq \beta_H, \delta_D \simeq \delta_H$ while the critical exponent γ_D and the leading correction-to-scaling exponent Δ_D are roughly 0.5 and 20% *higher* than γ_H and Δ_H, respectively. The presently determined values for the quantities $\beta_D, \gamma_D, \delta_D$, and Δ_D, when compared with the most accurate theoretical estimates[28] of $\beta_H=0.365(3)$, $\gamma_H=1.386(4)$, $\delta_H=4.80(4)$, and $\Delta_H=0.55(2)$, corroborate the above relationship between the critical exponents characterizing the isotropic dipolar and isotropic Heisenberg fixed points.

Above comparison between theory and experiment asserts that amorphous ferromagnets with composition just above the percolation threshold behave as a $d=3$ isotropic dipolar ferromagnet in the asymptotic critical region and exhibit isotropic dipolar (ID) to isotropic Heisenberg crossover as the temperature is raised above the critical temperature of the ID fixed point. Moreover, the results presented in Fig. 10, when compared with the variations of γ_{eff} with ϵ predicted by the field-theoretic treatment[6] of $d=3$ isotropic Heisenberg ferromagnet with *strong* isotropic dipolar interactions for different initial values u_i of the dimensionless renormalized coupling constant (i.e., the three cases mentioned in the Introduction), demonstrate that the observed $\gamma_{eff}(\epsilon)$ corresponds to the case I (see Sec. I for the details of this case).

Several important observations, which have a direct bearing on the discussion of present results, have been made by us previously on amorphous alloys same as, or similar to, the present ones. These include: (i) Fe or Co atoms are the only moment bearing atoms; Ni atoms do not[30] carry magnetic moment, (ii) Curie temperature increases at a much steeper[20] rate with p or p' than spontaneous magnetization at $T=0$ K does, and (iii) only a small fraction[13,14,23] of the total number of magnetic moments actually participates in the ferromagnetic (FM)-paramagnetic (PM) phase transition for compositions not too far above the critical concentration and as $p \rightarrow p_c$ (or $p' \rightarrow p'_c$), this fraction reduces further at a rate much faster than a simple magnetic dilution would suggest. Since the magnetic dipole-dipole interaction energy is proportional to the magnetic moment squared and T_C reflects essentially the strength of exchange interactions, the observations (i) and (ii) indicate that with increasing Fe or Co concentration, isotropic short-range exchange interactions grow in strength more rapidly than isotropic dipolar interactions. This inference is consistent with our finding that the Hopkinson peak ($\acute{\chi}^p_{ext}$) in the measured ac susceptibility is almost completely suppressed by a dc magnetic field (H_{dc}) as low as the Earth's magnetic field ($\lesssim 0.3$ Oe) particularly for the alloy with composition just above the percolation

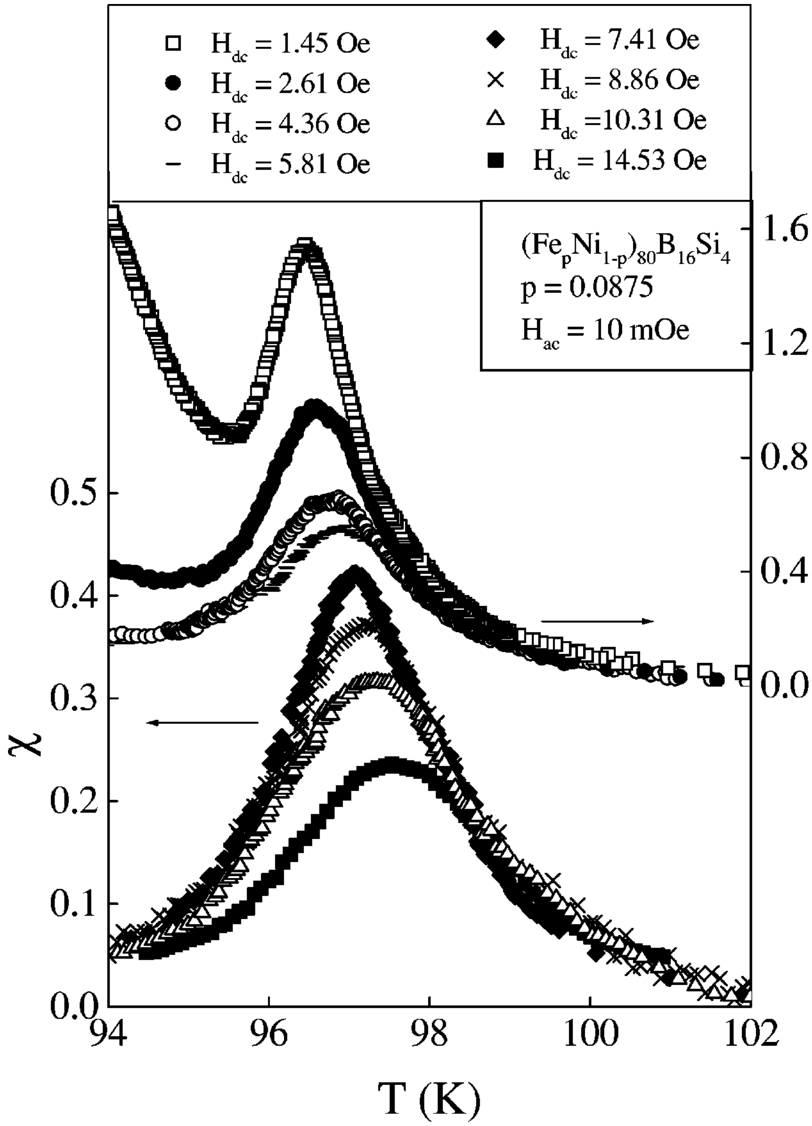

FIG. 7. Typical temperature variation of ac susceptibility for the amorphous ferromagnet that *exhibits* reentrant behavior at low temperatures observed at a few representative *fixed* static biasing field (H_{dc}) values, highlighting the suppression, and shifting to higher temperatures, of the critical peak at $T \approx T_C$ with increasing H_{dc}.

threshold and the sensitivity of $\acute{\chi}^p_{ext}$ to H_{dc} reduces as the Fe (or Co) concentration increases. Since the parameter ''g'' is the ratio of isotropic dipolar energy to isotropic exchange energy, g quickly drops in magnitude as p or p' increases. Consequently, a precipitous fall in the ID-to-IH crossover temperature $\epsilon_{co} \sim g^{1/\gamma_H}$ occurs as p (or p') progressively exceeds p_c (or p'_c) so much so that $\epsilon_{co} < 10^{-5}$ for the alloys with $p > 0.1$ or $p' > 0.2$. Hence the present experiments fail to detect this crossover in the compositions $p = 0.1125$, $p = 0.1375$, and $p' = 0.2375$ (Fig. 3) and instead the $d = 3$ isotropic Heisenberg critical behavior is observed in the temperature range $10^{-5} \lesssim \epsilon \lesssim \epsilon^*$ (Fig. 3).

As detailed elsewhere,[13–15,23,31] the infinite $d = 3$ FM network plus finite FM spin clusters model (the so-called K model[23]) provides a straightforward explanation for the observation (iii) mentioned above. Direct consequence of a rapid depletion of spins in the infinite FM network as $p \rightarrow p_c$ (or $p' \rightarrow p'_c$) is that the probability for the existence of the nearest-neighbor (NN) moment pairs is extremely small and the average intermoment spacing is much larger than the NN distance. At such distances and for temperatures $T < T_C$, *long-range* isotropic dipolar interactions provide a more effective mechanism for ordering spins than *short-range* (direct, nearest-neighbor) Heisenberg interactions. Lowering temperature below T_C results in progressive freezing[31] of finite spin clusters in random orientations and in ordering spins *better* in the infinite FM network as the spin misalignment due to thermal energy reduces. At low temperatures, a reentrant state is formed in which nearly perfect ferromagnetic long-range order coexists with cluster spin glass order. Thus, even though infinite ferromagnetic network coexists with finite spin clusters in both ferromagnetic and reentrant states, ferromagnetic state possesses *larger* entropy. The present results therefore help in resolving the long-standing reentrant spin-glass paradox[32] by explaining why the ferromagnetic state has larger entropy than the reentrant spin-glass state.

VI. SUMMARY AND CONCLUSION

We report the results of extensive ac susceptibility measurements performed on well-characterized amorphous $(Fe_pNi_{1-p})_{80}B_{16}Si_4$ ($p = 0.0540, 0.0875, 0.1125, 0.1375$) and $(Co_{p'}Ni_{1-p'})_{80}B_{16}Si_4$ ($p' = 0.1125, 0.1375, 0.2375$) alloys at the ac driving field of rms amplitude $H_{ac} = 10$ mOe and frequency $\nu = 87$ Hz in the presence and absence of superposed

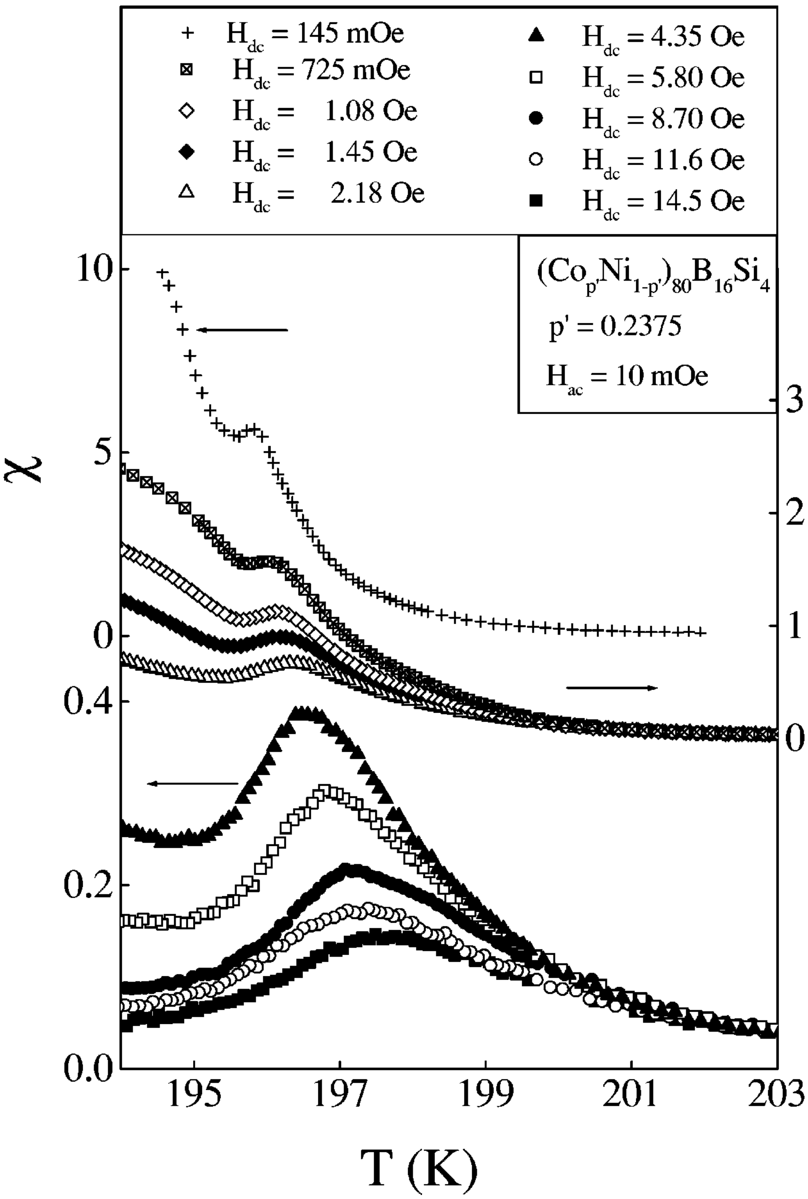

FIG. 8. Typical temperature variation of ac susceptibility for the amorphous ferromagnet that *does not* exhibit reentrant behavior at low temperatures observed at a few representative *fixed* static biasing field (H_{dc}) values, highlighting the suppression, and shifting to higher temperatures, of the critical peak at $T \approx T_C$ with increasing H_{dc}.

dc fields over a wide temperature range embracing the critical region near the ferromagnetic (FM)-paramagnetic (PM) phase transition. In the present experiments, unprecedented (relative) accuracy in the measurement of susceptibility has been achieved so that the field dependence of the *critical peak* in susceptibility could be studied even at fields as low as 0.1 Oe and the critical point T_C has been approached closer by *three decades* in reduced temperature $\epsilon=(T-T_C)/T_C$ than in previous investigations[33] on similar amorphous alloy systems. Closer approach to T_C, ability to detect the critical peak in susceptibility even at very low fields and higher accuracy in zero-field ac susceptibility measurements combined with an elaborate data analysis have made it possible to arrive at the true values of the *asymptotic* critical exponents β, γ, and δ for spontaneous magnetization, susceptibility, and critical $M-H$ isotherm, respectively.

Consistent with the theoretically predicted[2–6,19] behavior of a $d=3$ ferromagnet with both isotropic Heisenberg (IH) short-range exchange and isotropic dipolar (ID) long-range interactions, *effective* critical exponent for zero-field susceptibility $\gamma_{eff}(\epsilon)$ obeys Eq. (6) in the asymptotic critical region (Fig. 4), goes through a deep *minimum* at ϵ_{min} in the crossover region and approaches the $d=3$ isotropic Heisenberg (IH) value at temperatures ϵ^{**} well above $T_C(\epsilon \gg \epsilon_{min})$ for the amorphous $(Fe_pNi_{1-p})_{80}B_{16}Si_4$ and $(Co_{p'}Ni_{1-p'})_{80}B_{16}Si_4$ alloys with composition in the range $p_c \lesssim p \lesssim 0.1$ or $p'_c \lesssim p' \lesssim 0.2$ (Figs. 3 and 10) that exhibit *reentrant behavior* at low temperatures ($T \ll T_C$). Scaling equation of state analysis, based on Eqs. (13) and (14), of the ''in-field'' susceptibility $\chi(h,t)$ data on these systems yields the values for the *asymptotic* critical exponents β_D and δ_D for spontaneous magnetization and critical $m-h$ isotherm that are consistent with the value of *asymptotic* critical exponent for susceptibility γ_D deduced from the zero-field susceptibility data in that the presently determined values of the exponents β_D, γ_D, and δ_D satisfy the Widom scaling relation $\beta+\gamma=\beta\delta$ accurately. Now that the exponents β_D, γ_D, and δ_D characterize the *isotropic dipolar* (ID) fixed point, above observations assert that the asymptotic critical behavior of amorphous reentrant ferromagnets with $p \lesssim 0.1$ or $p' \lesssim 0.2$ is that of a $d=3$ isotropic dipolar ferromagnet. The

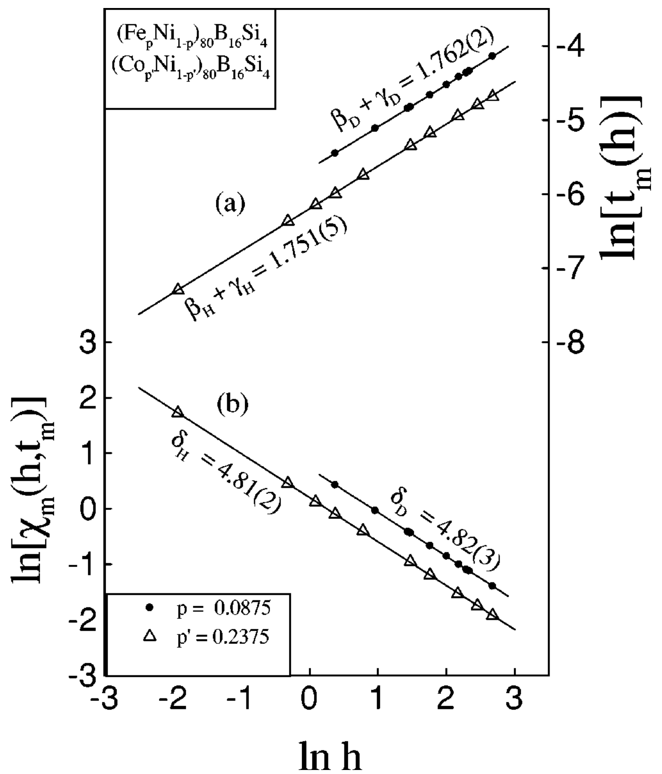

FIG. 9. (a) The reduced peak temperature t_m versus the internal field h on a double logarithmic plot. The inverse slopes of the straight line fits, based on Eq. (13) of the text, yield the gap exponent $\beta+\gamma$. (b) The peak value at t_m of the critical peak in the in-field susceptibility χ_m versus the internal field h on a double logarthmic plot. The slopes of the straight line fits, based on Eq. (14) of the text, determine the values of the critical exponent δ of the critical isotherm.

unique property of such systems that isotropic long-range dipolar and isotropic short-range Heisenberg interactions of comparable magnitude coexist in them thus permits the first unambiguous observation of ID fixed point and ID→IH crossover. As the Fe or Co concentration in these alloys exceeds $p\simeq 0.1$ or $p'\simeq 0.2$, isotropic Heisenberg exchange interactions grow in strength at a much faster rate than isotropic dipolar interactions do, with the result that the reentrant behavior is completely suppressed and the ID→IH crossover temperature falls below $\epsilon\simeq 10^{-5}$, the minimum temperature deviation from the Curie point discerned in the present experiments. Hence neither the ID fixed point nor the ID→IH crossover but only the $d=3$ isotropic Heisenberg critical behavior is observed in the temperature range $10^{-5}\lesssim\epsilon\lesssim\epsilon^*\simeq 10^{-2}$.

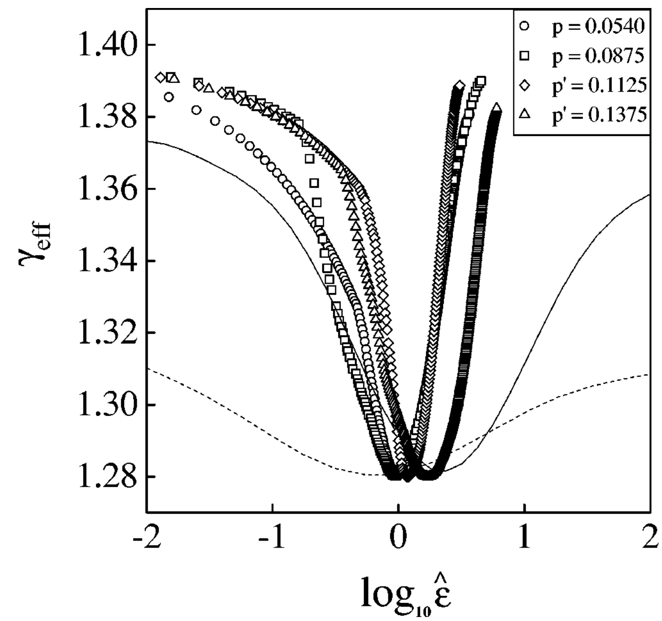

FIG. 10. Comparison of the experimentally observed variation of γ_{eff} with $\log_{10}\hat{\epsilon}$ for the amorphous ferromagnetic alloys that exhibit reentrant behavior at low temperatures with that predicted by the renormalization-group theories: continuous curve (Ref. 4) and dashed curve (Refs. 6 and 29).

The main findings of the present investigation together with those previously reported by us on the glassy alloys same as, or similar to, the ones under consideration, when viewed within the framework of the infinite three-dimensional ferromagnetic (FM) network plus finite FM spin clusters model (the so-called K model) originally proposed by Kaul,[23] strongly suggest that long-range isotropic dipolar interactions are largely responsible for establishing long-range ferromagnetic order in such dilute magnetic systems. With this input, the K model convincingly explains as to why the ferromagnetic state has larger entropy than the reentrant spin-glass state and thereby offers a possible way to resolve the long-standing reentrant spin-glass paradox.[32]

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