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# Fractal dimensionality of percolation clusters in $(Fe_pNi_{1-p})_{80}P_{20}$

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Strong, non-Lorentzian small-angle neutron scattering is observed below the Curie temperature ($T_C$) of $(Fe_pNi_{1-p})_{80}P_{20}$ for $p > p_c = 0.161$. The large-$Q$ part of this spectrum is demonstrated to be a power of a Lorentzian, the power of which gives the fractal dimensionality of the finite clusters, $D = 2.66 \pm 0.05$. The small-$Q$ region is dominated by static scattering from the infinite cluster and is shown to vanish at $T_C$ and to increase as $Q^{-2.7 \pm 0.03}$ below $T_C$, as predicted for a random-exchange model.

The magnetic properties of a dilute system, when they arise from short-range interactions among localized spins, are dominated by the geometrical arrangement of the spins.[1] Because energy-integrated neutron scattering[2] measures the instantaneous spatial Fourier transform of the spin-spin correlation function, it is an ideal means of studying such systems. Detailed studies[3,4] carried out at concentrations $p$ below the percolation point $p_c$—that concentration at which long-range order first appears—have substantiated the percolation picture of the onset of magnetic order. Comparable data reported for $p > p_c$ do not appear to fit the same simple picture. Critical scattering is not observed[5,6] along the ferromagnetic (or antiferromagnetic[3,4]) critical line $T_C(p)$, at least for modest values of momentum transfer $Q$ ($\geq 10^{-2}$ Å$^{-1}$). Below $T_C(p)$ the low-$Q$ scattered intensity diverges as $Q^{-3}$ while, at larger $Q$, it can be fitted[7] to a power of a Lorentzian (POL). Frequently[8] the sum of a Lorentzian and a Lorentzian squared, as appropriate for an Ising system in a random field, has been used and arguments given[8] that invoke random fields caused by the interactions between frozen and free-spin clusters.

In this Rapid Communication, we report small-angle neutron scattering (SANS) data over a wide range of $Q$ for percolating amorphous $(Fe_pNi_{1-p})_{80}P_{20}$ alloys. The magnetic properties of these alloys were previously shown[9] to accord well with a percolation picture.[10] We argue here that for $Q\xi_p >> 1$, where $\xi_p \approx (p-p_c)^{-\nu_p}$, the tail of the scattered intensity reflects the decay of spin correlations on the distribution of finite clusters. The POL shape of the intensity is shown to result from the fractal geometry of these clusters on length scales between the atomic scale and $\xi_p$. The width $\kappa_1$ of the POL is approximately given by $\kappa_1 = \xi_p^{-1} + \xi_1^{-1}$, where $\xi_1$ is the one-dimensional correlation length. We find the fractal dimension to be $D = 2.66 \pm 0.05$ for these alloys. The small-$Q$ behavior, on the other hand, is dominated by fluctuations in the thermal-average magnetic moment from site to site on the percolating network (the infinite cluster). This contribution is a power law in $Q$ and vanishes at $T_C(p)$, as predicted theoretically.[11]

In this note, we focus on a single concentration $p = 0.181$, which is above $p_c = 0.161$ for this system.[9] At this concentration, the infinite cluster encompasses a fraction $P_\infty(p) \approx 0.4$ of the Fe-occupied sites (7% of the transition metal sites). The complex interplay of length scales in this problem[12]—one-dimensional correlation length $\xi_1$, percolation length $\xi_p$, and thermal correlation length $\xi_T \approx |T_C(p) - T|^{-\nu_T}$—renders a complete description of the scattering cross section impossible. For $P_\infty(p)$ small, the infinite cluster will contribute little in the large-$Q$ limit, but will dominate at small $Q$, thus simplifying the analysis. Critical scattering is ignored here, although it was demonstrated in an earlier note[13] that it can be detected in the small-$Q$ limit for $T > T_C(p)$.

The energy-integrated cross section for magnetic scattering is directly related to the spin-spin correlation function,

$$\left|\frac{d\sigma}{d\Omega}\right| \sim \sum_{i,j} e^{i\mathbf{Q}\cdot(\mathbf{R}_i-\mathbf{R}_j)}\langle \mathbf{S}_i\cdot\mathbf{S}_j\rangle \sim S_F(Q) + S_\infty(Q) \quad . \tag{1}$$

In (1), we have separated the contributions $S_F(Q)$ of finite clusters from $S_\infty(Q)$ due to the infinite cluster (IC), where

$$S_F(Q) = Np[1-P_\infty(p)]\sum_j e^{-i\mathbf{Q}\cdot\mathbf{R}_j}\langle\overline{\mathbf{S}_0\cdot\mathbf{S}_j}\rangle_F \quad , \tag{2}$$

and

$$S_\infty(Q) = NpP_\infty(p)\sum_j e^{-i\mathbf{Q}\cdot\mathbf{R}_j}\langle\overline{\mathbf{S}_0\cdot\mathbf{S}_j}\rangle_\infty \quad . \tag{3}$$

Here $\langle\overline{\mathbf{S}_0\cdot\mathbf{S}_j}\rangle_F$ is an average over initial sites chosen to be on a finite cluster and $\langle\overline{\mathbf{S}_0\cdot\mathbf{S}_j}\rangle_\infty$, over initial sites on the IC.

The behavior of $\langle\overline{\mathbf{S}_0\cdot\mathbf{S}_r}\rangle$ for spins a distance $r << \xi_p$ apart on the same cluster has been discussed recently by Aharony, Gefen, and Kantor.[14] The decay of correlations is determined by the one-dimensional path length $L_1(r) \sim r^\zeta$ between the spins; Coniglio[15] has suggested that $\zeta = 1/\nu_p$

 

$\approx 1/\nu_T$. The unaveraged correlation function is $\langle \mathbf{S}_0 \cdot \mathbf{S}_r \rangle \sim \exp(-r^{\zeta}/\xi_1)$, and for Heisenberg systems, $\xi_1 \sim (J/k_B T)^{\nu_T}$. For $r > \xi_1$, the decay is predicted[14] to be more rapid; there is no contribution from the finite clusters at small $Q$. Averaging over starting sites gives

$$\langle \overline{\mathbf{S}_0 \cdot \mathbf{S}_r} \rangle_F \sim [r^{2(D-d)} \exp(-r/\xi_p)] \exp(-r^{\zeta}/\xi_1) \ , \qquad (4)$$

and

$$\langle \overline{\mathbf{S}_0 \cdot \mathbf{S}_r} \rangle_\infty \sim r^{D-d} \exp(-r^{\zeta}/\xi_1) + \langle S_0^z \rangle \langle S_r^z \rangle \ . \qquad (5)$$

The factor $r^{D-d}$ comes from the decrease in density with distance for an object of fractal dimension $D$ embedded in $d$-dimensional space. The additional factors in (4) result from averaging over the distribution of cluster sizes. Close to $p_c$, where $P_\infty(p) \ll 1$, the contribution of the IC to the small-$r$ regime is much less than that of finite clusters. Note that spin correlations do not decay to zero on the IC but to a static, but nonuniform, average at temperatures below $T_C(p)$.

The Fourier transform of (4) required in (2) is not simple in general. If, however, $\zeta = 1$, it can be performed directly to give

$$S_F(Q) = [\Gamma(\mu)/Q(Q^2+\kappa_1^2)^{\mu/2}] \sin[\mu \arctan(Q/\kappa_1)] \ , \qquad (6)$$

where $\mu = 2D - d - 1$ and $\kappa_1 = 1/\xi_1 + 1/\xi_p$. For $\mu = 1$ and 2, Eq. (6) reduces to a Lorentzian and a Lorentzian squared, respectively. For intermediate values, it is indistinguishable from a power of a Lorentzian. The large-$Q$ limit of (6) should not be taken as simply[16] $Q^{2D-d}$, since the trigonometric factor is a strong function of $Q$ as $\mu \rightarrow 2$.

At distances much greater than $\xi_p$, where the IC dominates, its density is constant, but the average spin at each site differs from the spatial average. It has been pointed out[17] that, at these length scales, the percolating system resembles a homogeneous one with a spread $\sqrt{\Delta}$ in exchange energy around the mean value. We rewrite this in the form introduced by Grinstein, Ma, and Mazenko[18]

$$\lim_{r/\xi_p \rightarrow \infty} \langle \overline{\mathbf{S}_0 \cdot \mathbf{S}_r} \rangle_\infty = C^{(s)}(r) + m^2 \ , \qquad (7)$$

where

$$C^{(s)}(r) = (\langle S_0^z \rangle - m)(\langle S_r^z \rangle - m) \ ,$$

and $m = N^{-1} \sum \langle S_r^z \rangle$. The quantity $C^{(s)}(r)$ vanishes for a uniform system. Here, however, spins on the IC have neighboring sites that lack magnetic atoms. The exchange interaction, therefore, varies from $J$ to $ZJ$, where $Z$ is the coordination number. The number of missing neighbors of the IC is called[10] the perimeter $t$, and the number of such sites per occupied site on a cluster of $s$ spins has the limiting value[19] $\lim_{s \rightarrow \infty}(t/s) = (1-p)/p$. We suggest that $\Delta \sim J^2(1/p - 1)^2$ which vanishes in the pure limit. Estimating the percolation threshold as $p_c \sim 1/Z$ gives $\Delta \sim J^2(Z-1)^2$ at the percolation threshold—a strong effect. As $T$ decreases, all spins on the IC should align regardless of the exchange coupling, causing this term to vanish at low temperatures.[11]

The behavior of $C^{(s)}(r)$ was treated by Grinstein, Ma, and Mazenko[18] in the mean-field limit. They predicted the presence of a term proportional to $M^2Q^{-4}$ in the low-$Q$ scattering cross section. Recently, Pelkovits and Aharony[11] generalized the calculation, using scaling arguments, to give a modified result,

$$C^{(s)}(Q) = \Delta t Q^{-(d-2+\eta+1/\nu)} \approx \Delta t Q^{-2.4} \ , \qquad (8)$$

where $t = 1 - T/T_C(p)$. Equation (8) is valid for $Qt^{-\nu} \gg 1$. At low temperatures, $C^{(s)}(Q)$ vanishes as $T^2$.

Restricting attention to the dominant terms, we analyze the scattered intensity using

$$I(Q) = A/(Q^2+\kappa_1^2)^{1/\alpha} + Bt/Q^y + Dt^{2\beta}\delta(Q) \ . \qquad (9)$$

The first contribution is from the short-range decay of correlations on the finite clusters, Eq. (4); the second arises from static fluctuations on the IC, Eq. (8); and the last is the true Bragg scattering which cannot be observed in a SANS experiment. Equation (9) is tractible, but ignores several important contributions: critical scattering from the IC in the vicinity of $T_C(p)$, the decay of short-range correlations on the IC, and the short-distance fluctuations of the average moment on the IC. The latter two contributions would reflect the density of IC as in Eq. (5) and would resemble the first term in (9), but with a smaller value of $\alpha$. We will argue below that these terms are not important in the immediate vicinity of the percolation concentration.

Neutron scattering data were obtained on the D11 diffractometer at the Institut Laue-Langevin. For each concentration, 5 g of the amorphous ribbon was wound on a thin-walled, 2.5-cm-diameter aluminum cylinder approximately 5-cm long. Only the central 1 cm of the sample, defined by the collimation of the beam and a Cd aperture, was in the neutron flux. Data were taken with the area detector at the 5 and 20-m positions with an incident wave vector of 1.0 $\text{Å}^{-1}$ ($\Delta\lambda/\lambda \approx 0.09$); this permits measurements in the range $0.0029\ \text{Å}^{-1} \leq Q \leq 0.053\ \text{Å}^{-1}$, with overlap near $Q = 0.01\ \text{Å}^{-1}$. The resolution is determined by the solid angle subtended by each area-detector cell, and is $5 \times 10^{-4}\ \text{Å}^{-1}$. A beam stop limited the small-$Q$ end of the spectrum to the range noted above. A radial average was performed over cells at a constant $Q$, after calibration of the detector efficiency by the incoherent scattering of a Plexiglas sheet placed in the sample position. The magnetic scattering was separated from nuclear and background scattering by subtracting data taken at high temperatures ($\gtrsim 2T_C$) from the low-temperature runs. Because the magnetic scattering persists well above $T_C(p)$ for large $Q$, this process causes us to underestimate the magnetic scattering at high temperatures. We have not included data here unless the magnetic contribution is at least twice the background.

Low-field magnetization data for our $p = 0.161$ sample are shown[20] in the inset to Fig. 1. The decrease in magnetization at low temperatures reflects a tendency toward spin-glass-like behavior in the low-field–low-temperature region. The main part of Fig. 1 shows the low-$Q$ SANS data. There is no evidence for critical scattering near $T_C = 92$ K but very strong scattering is evident down to 2 K, the non-Lorentzian nature of which is evident in the inset of Fig. 2. The upward curvature of such plots is commonly found[4-8] for diluted magnetic systems. The behavior is much improved by taking an inverse power of the intensity, as shown in Fig. 2 for several temperatures. The choice $\alpha = 0.8 \pm 0.03$ straightens the large-$Q$ portion of all the runs and gives positive intercepts; that is, these are POL's. The straight lines in Fig. 2 are fits to the large-$Q$ data which we assert represents the first term in Eq. (9). The effective one-dimensional inverse correlation length $\kappa_1$ extracted from the fit is shown as a

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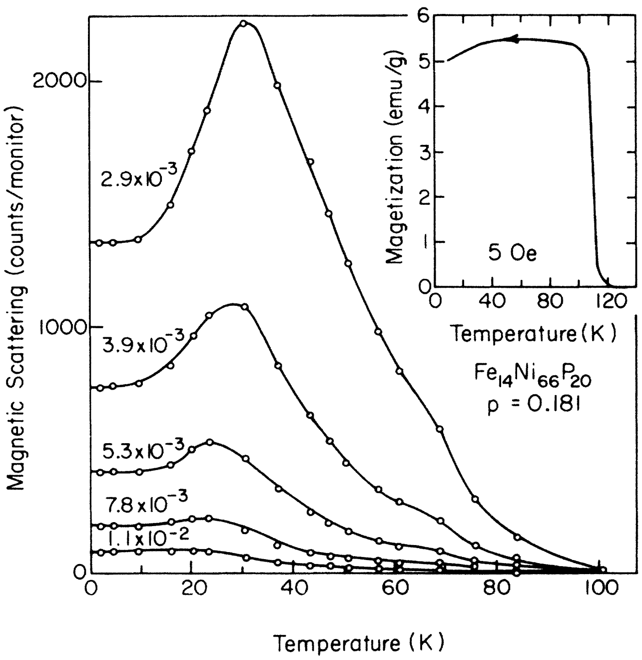


FIG. 1. Small-angle neutron scattering intensity at various values of the momentum transfer $Q$ (in Å$^{-1}$) vs temperature. The inset shows low-field magnetization data (field cooling). Note the absence of critical scattering near the ferromagnetic transition ($T_C = 92$ K) and the presence of a shoulder that moves toward $T_C$ as $Q$ decreases.

function of temperature in Fig. 3 (open circles), along with results of a similar analysis for two concentrations below and one above $p_c$. The straight line has unity slope, suggesting that $\xi_1 \approx T^{-\nu_p}$ with $\nu_p \approx 1.0$. This justifies our choice $\zeta = 1$. The data for $p < p_c$ are especially important, demonstrating that the large-$Q$ scattering is entirely dominated by finite clusters. The constant value of $\kappa_1$ at low temperatures measures[14] the inverse of the percolation length $\xi_p$.

The inset to Fig. 3 shows the results of fitting Eq. (6) to a POL, which permits us to perform the analysis in Fig. 2. The value $1/\alpha = 1.25 \pm 0.05$ corresponds to $\mu = 1.33 \pm 0.05$ and leads to $D = 1/2(d + \mu + 1) = 2.66 \pm 0.05$. This is consistent with the result $D = d - 2\beta/\nu_p$ when we use our value $\beta = 0.4$ from the magnetization data[9] and $\nu_p \approx \nu_T \approx 1$ from Fig. 3. Our value is significantly different from the $d = 3$ series value $D = 2.5$ which leads to $\mu = 1$, and a purely Lorentzian large-$Q$ tail.

The small-$Q$ behavior is, according to (9), dominated by the IC contribution. We have analyzed this contribution by subtracting the POL's determined in Fig. 2 from the data. The remaining intensity has been divided by $T_C - T$, with $T_C = 92$ K and plotted versus $Q$ in Fig. 4. The solid line corresponds to $y = 2.4$ as predicted in Eq. (8), and a fit to the data gives $y = 2.7 \pm 0.3$. Equation (8) holds only for $Qt^{-\nu} >> 1$; in the opposite limit, $C^{(s)}(Q)$ is predicted to decrease as $t^{-\nu-\alpha}$. In Fig. 1 a shoulder can be discerned near 70 K which moves to higher temperatures with decreasing $Q$, but not the predicted maximum. The maximum which is observed at low temperatures is relatively $Q$ in-

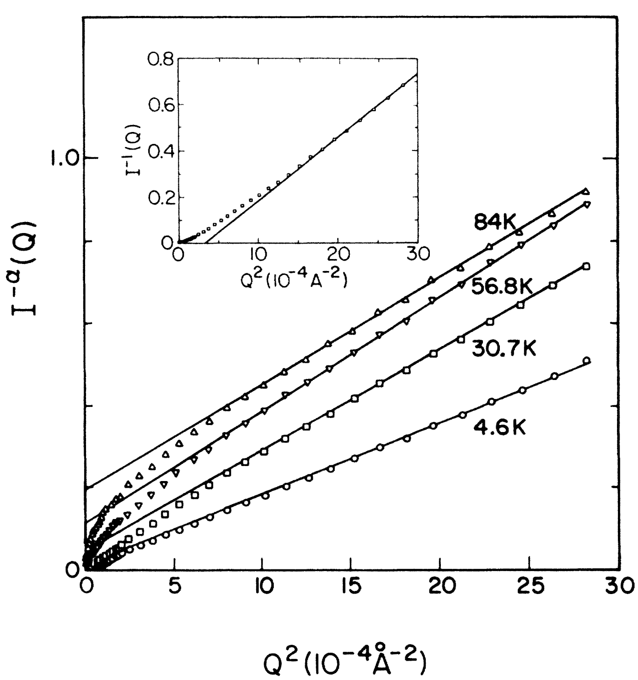


FIG. 2. The scattered intensity raised to the negative power $\alpha = 0.8$ vs $Q^2$ at various temperatures. The inset shows the strongly non-Lorentzian shape at 30.7 K. The straight lines are fits to the data for $Q \gtrsim 0.025$ Å$^{-1}$. They are Lorentzians to the power 1.25, and have temperature dependent widths $\kappa_1$.

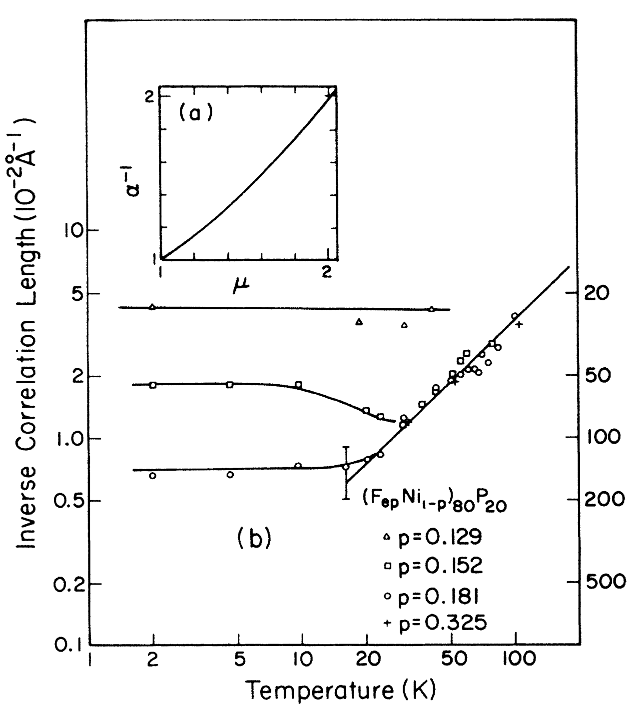


FIG. 3. Inverse correlation length $\kappa_1$, obtained from power-of-Lorentzian fits as in Fig. 2, as a function of temperature. The open circles are for the present sample; other symbols, for comparable analysis on samples above and below the percolation concentration $p_c = 0.161$. The straight line corresponds to $\kappa_1 \sim T$.

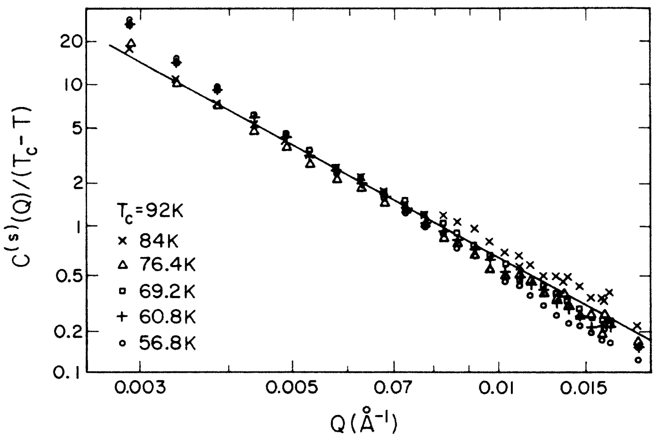


FIG. 4. Low-$Q$ scattering, after subtraction of the power of a Lorentzian, divided by the temperature difference $T_C-T$. The straight line is the predicted slope (2.4) for a dilute Heisenberg model. A fit gives a slope of 2.7 ±0.3.

dependent and seems to reflect the disappearance of this contribution at low temperatures.

We have argued that the non-Lorentzian shape of the scattered intensity reflects the fractal nature of the finite clusters both above and below the percolation concentration $p_c$. Quite recently, a similar interpretation[21] of the SANS from silica aggregates (a single fractal object) led to $D=2.6 \pm 0.1$ for dry aggregates, fortuitously close to our value. While consistent with values of $\beta$ and $\nu_T$, $D$ is smaller than calculated for site percolation, meaning that the structure is less open than predicted. Further-neighbor interactions or dipolar contributions may induce correlations between neighboring clusters, thereby increasing the effective density. It is interesting to note that a wide variety of dilute magnetic materials,[4,7] $(Eu_pSr_{1-p})S$, $(Fe_pMn_{1-p})_{75}P_{16}B_6Al_3$, and the present amorphous alloys all show low-temperature $Q^{-2.6}$ tails in the scattered intensity, suggesting that the value $D=2.66$ is universal.

The effective correlation length at low temperatures levels off at approximately 200 Å, although the error bars are large. We take this to be a measure of $\xi_p$. The number of spins in the largest cluster will be $\sim p(\xi_p/a)^D \sim 1.3\times10^4$. In our previous analysis, we were unable to determine the cutoff of the cluster size distribution, which introduced an error in our determination of $P_\infty(p)$. With this estimate, we can state that the error in the analysis presented there is on the order of 10%, well within the error limits stated.

The present analysis demonstrates that the percolation picture, which has been shown to work well for $p<p_c$, is adequate in the percolating regime as well. The competition between finite-cluster effects, which dominate at small distances, and infinite-cluster ordering, which sets in on large length scales, has made adequate analysis difficult. It is satisfying that *ad hoc* random-field arguments[8] are neither necessary, nor appropriate, to understand the behavior of dilute magnets near the percolation limit.

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