Fractal model for disordered magnets

G. Gavoille

Laboratoire d'Electricité et d'Automatique, Universite de Nancy I, Boîte Postale 239, 54506 Vandoeuvre-les-Nancy Cedex, France

J. Hubsch

Laboratoire de Cristallographie et Minéralogie, Universite de Nancy I, Boîte Postale 239, 54506 Vandoeuvre-les-Nancy Cédex, France (Received 29 December 1986)

Experimental results suggest that magnetic domains in disordered magnets have a topological fractal structure. Following Mandelbrot we build up a random fractal with properties consistent with neutron diffraction experiments. We show that the structure factor of the fractal provides all the forms used in the fit of experimental data.

I. INTRGDUCTION

We are concerned with disordered magnetic systems in which the correlations are those of a pure magnetic phase over a length scale much larger than the interatomic distance. These systems are characterized, in neutron diffraction experiments, by the appearance of strong scattering around the points of the reciprocal magnetic lattice of the pure phase. Such systems have been known for a long time. One may mention the coexistence of antiferromagnetism of the first and second kind in $Mn_{0.5}Cr_{0.5}S$ (Ref. 1) or the coexistence of ferromagnetism and antiferromagnetism in mixed valency manganites.² However, there has been a recent spate of interest in such systems with the study of reentrant spin glass, random or competing anisotropy models, and random-field systems. Despite the very different nature of the disorder, all the systems have common features. In neutron scattering experiments the structure factor near the reciprocal-lattice vector G is always well fitted by the form

$$
S(Q) = AL^2 + BL \t{1}
$$

where L is the Lorentzian form

$$
L = \frac{1}{K^2 + q^2} \tag{2}
$$

with

$$
q = |Q - G| .
$$

A power-law fit $q^{-\alpha}$ (Ref. 3) or Lorentzian form at an arbitrary power $L^{x/2}$ (Ref. 4) are also well suited. We shall show that the previous forms may be deduced from topological considerations only.

The ground state of a pure magnetic phase is always degenerated. Besides the trivial degeneracy related to spin reversals, additional degeneracies may arise related to propagation vector degeneracy $(K$ domains) or to spin direction degeneracy (S domains). Only those spins that belong to the same magnetic domain will give a noticeable contribution to the neutron scattering structure factor. The latter is then the sum of partial contributions, each related to a magnetic domain. Equivalently, the neutron-diffraction pattern is the superposition of diferent patterns, each obtained by keeping only those spins that belong to a given magnetic domain. In other words, the neutron-diffraction pattern is that of a porous medium. If the degeneracy of the ground state results from a random process, we shall consider that each domain will yield the same neutron-diffraction pattern. The fit of the tail of the structure factor by a power law with an arbitrary power suggests a fractal dimension of the medium.⁵ Fractal structures, such as Sierpinski gaskets, have been previously proposed to the study of percolation.⁶ We shall, however, use a fractal proposed by Mandelbrot.⁷ Both fractals are obtained by drilling holes in matter (Fig. 1), but in the first case the holes are obtained from a regular process, while in the second case the holes are randomly distributed. As we are interested in systems where the disoder results from a random process, the second model seems more suitable. For those readers interested in the shape of that fractal, planar fractals of diferent fractal dimensions are given in Ref. 7.

FIG. 1. Planar fractals obtained by drilling holes (in black) in the matter. (a) Schematic drawing of the fractal used in that paper. (b} Sierpinski's fractal.

II. FRACTAL MODEL

We use a model proposed by Mandelbrot, 7 whose basic idea consists of drilling holes of a random size at random locations in the medium. Originally proposed for three-dimensional (3D) Euclidian space, the model may be easily extended to an arbitrary Euclidian dimension. We restrict ourself to an isotropic medium and consider spherical holes. The hypothesis of the model is the following: the number of holes that center are in a given volume V, and those whose radii lie between ρ and $p+d\rho$, obey a Poisson distribution with the mean

$$
(d - d_f) \frac{d}{S_d} \rho^{-(d+1)} V d\rho , \qquad (3)
$$

where S_d is the unit sphere area and d_f the fractal dimension. The radius ρ lie between a lower bound ϵ and an upper bound R . The following results are then easily obtained. The probability that a point belongs to the medium M or the concentration c of the occupied volume is given by

$$
c = \left[\frac{R}{\epsilon}\right]^{-(d-d_f)}.
$$
 (4)

The condition probability $Pr\{P \in M \mid O \in M\}$, or the Porod function $Z(r)$ (Ref. 8) is given by

volume is given by
\n
$$
c = \left[\frac{R}{\epsilon}\right]^{-(d-d_f)}
$$
\n
$$
c = \left[\frac{R}{\epsilon}\right]^{-(d-d_f)}
$$
\nProof function $Z(r)$ (Ref. 8) is given by
\n
$$
Z_1(r) = \left[\frac{r}{2\epsilon}\right]^{-(d-d_f)}
$$
\n
$$
Z_1(r) = \frac{e^{-Kr}}{r} \text{ for } d_f = 2,
$$
\nthe minimum of
\n
$$
\int_0^\infty \left[a e^{-Kr} + b \frac{e^{-Kr}}{r} - r^{d_f - 3} e^{-Kr}\right]^{2} r^2 dr
$$
\nTherefore, $I = \frac{R}{2}$ and $I = \frac{R}{2}$

The function λ is slowly varying with r and will be replaced in the following by $\lambda(2R, d, d_f)=1$. The fractal structure of the medium is easily shown by the occupied volume V in a sphere of radius $r < 2R$, which reads as

$$
V(r) = \frac{S_d}{d_f} (2\epsilon)^{(d-d_f)} r^{d_f} . \tag{6}
$$

We shall use the Fourier transform of $Z_1(r)$ in the following. Simple analytical forms may be obtained by replacing the sharp cutoff at $2R$ by a soft exponential cutoff, such that

$$
V(2R) = S_d(2\epsilon)^{(d-d_f)} \int_0^\infty r^{-(d-d_f)} e^{-Kr_f d - 1} dr , \quad (7)
$$

where $V(2R)$ is given by Eq. (6). The soft cutoff is not expected to strongly modify the Fourier transform since the small q values are dominated by the occupied volume and the large q' arise from the small r' . The reciprocal correlation length K is given form Eq. (7) as

$$
KR = \frac{1}{2} \left[d_f \Gamma(d_f) \right]^{1/d_f} \tag{8}
$$

and $Z_1(r)$ now reads as

$$
Z_1(r) = \left(\frac{r}{2\epsilon}\right)^{-(d-d_f)} e^{-Kr} . \tag{9}
$$

The Fourier transforms of $Z_1(r)$ are listed in the following. Irrelevant factors have been deleted.

$$
d = 1 \qquad \frac{\cos(d_f \tan^{-1} q/K)}{(K^2 + q^2)^{d_f/2}} \qquad (10a)
$$

$$
d_f = 1
$$
 (10b)

$$
d = 2
$$

$$
\int_0^\infty e^{-Kr} r^{d_f - 1} J_0(qr) dr
$$
 (11a)

$$
d_f = 2
$$

\n
$$
d_f = 1
$$

\n
$$
L^{3/2}
$$

\n
$$
L^{1/2}
$$

\n(11b)
\n(11c)

$$
d_f = 1
$$

\n
$$
L^{1/2}
$$
\n(11c)
\n
$$
\frac{1}{q} \frac{\sin[(d_f - 1)\tan^{-1}q/K]}{(K^2 + a^2)^{(d_f - 1)/2}}
$$
\n(12a)

$$
d_f = 3 \qquad \qquad \frac{q}{L^2} \qquad \qquad (K^2 + q^2)^{(d_f - 1)/2} \qquad (12b)
$$

$$
d_f = 2 \qquad L \qquad (12c)
$$

One first notices that $Z_1(q)$ reduces to a power of L for all integer values of d_f . For $d=3$ one finds L^2 for $d_f = 3$ and L for $d_f = 2$. This suggests the interpolation equation (1). As

$$
Z_1(r) = e^{-Kr} \quad \text{for } d_f = 3
$$

and

$$
Z_1(r) = \frac{e^{-Kr}}{r} \quad \text{for } d_f = 2
$$

the minimum of

$$
\int_0^{\infty} \left[a e^{-Kr} + b \frac{e^{-Kr}}{r} - r^{d_f - 3} e^{-Kr} \right]_0^2 r^2 dr \quad , \tag{14}
$$

with respect to a and b , yields the following ratio between the coefficients of Eq. (1):

$$
\frac{BK^2}{A} = \frac{1}{4} \frac{3 - d_f}{d_f - 2} \tag{15}
$$

At large q values, $Z_1(q)$ is equivalent to $q^{-\alpha_f}$ for odd Euclidian space dimensions.

The porous medium built up by the random process described above has fractal correlations up to a cutoff 2R. The Fourier transforms of the fractal correlations yield all the analytical forms used in the fit of the data of neutron-diffraction experiments. We shall now consider some experimental results in order to check the reliability of the model.

III. RANDOM-FIELD SYSTEMS

Random-field systems are Ising antiferr which behave in an uniform field as an Ising ferromagn in a random field. ' 10 We shall consider the latter, which is split into domains. The porous medium is considered to be formed by the up (down} spins and, consequently, Eq. (4) becomes

$$
c = \left(\frac{R}{\epsilon}\right)^{-(d-d_f)} = \frac{1}{2} \tag{16}
$$

R and ϵ are, respectively, the largest and the smallest radius of curvature of the boundary of the medium, i.e.,

of the domain walls. ϵ may be taken as the domain-wall or the domain walls. ϵ may be taken as the domain-wall
width, and it has been shown that R scales as $H^{-\nu}$ in width, and it has been shown that *K* scales as *H* in field-cooled experiments.^{11,12} We then expect d_f to be very near d [from Eq. (16)] in very low fields. As the spin correlations have a cutoff at $r=2R$, the structure factor is the Fourier transform of $Z_1(r)$. L^2 and $L^{3/2}$ forms have been, respectively, obtained in the 3D $Fe_{x}Zn_{1-x}F_{2}$ (Ref. 13) and the 2D $Rb_{2}Co_{0.7}Mg_{0.3}F_{4}$ (Ref. 4) compounds, in agreement with Eqs. (12b) and (11b). An interesting feature has been observed in $K_2Ni_xZn_{1-x}F_4$ (Ref. 14), where the interplanar interaction is much smaller than the in-plane interaction. The 30 order breaks up in very low fields, while higher fields are needed to break up the 2D order. For scans along the c axis the system behaves as $1D$ system in low fields and the correlations have an exponential decay, implying $d_f = 1$. In higher fields, the tail of $S(q)$ has a q^{-3} dependence for scans in the plane. This is consistent with a 2D system with $d_f = 2$ [Eq. (11b)]. In very high fields, d_f should be different from d if Eq. (16) is satisfied. It has been observed in the 3D Co_{0.3}Zn_{0.7}F₂ compound⁴ that $S(Q)$ changes from a dominant L^2 form in low fields to a L form as the field increases. At 8,4 kG a good fit is obtained with $L^{1,2}$, suggesting $d_f = 2.4$ from Eq. (12a). The experimental data considered here seem to be consistent with the model; it would be, however, interesting to check if Eq. (16) holds; namely, if

$$
v(d-d_f)\ln H = -\ln 2 + \lambda (d-d_f) , \qquad (17)
$$

where λ is a constant.

IV. FRUSTRATED SYSTEMS

We restrict ourself to frustrated ferromagnets, which undergo a ferromagnetic to spin-glass transition as the concentration in magnetic species decreases. We shall first consider a single ferromagnetic domain with spinglass inclusions. This corresponds to the semi-spin-glass phase considered by Villain.¹⁵ The fractal to be considered consists of the ferromagnetic phase, for which Porod function may be written as

$$
Z(r) = \begin{cases} c & \text{for all } r \\ \left[Z_1(r) - c \right] & \text{for } r < 2R \end{cases} . \tag{18}
$$

The first term in Eq. (18} yields Bragg scattering, while the second one, which gives the diffuse scattering, may be reduced to $Z_1(r)$ for $q \gg 1/R$. For 3D systems we then expect $S(Q)$ to be given by Eq. (12a) or (1)

and (15). The small-angle structure factor of $(Fe_{0.75}Mn_{0.25})_{75}P_{16}B_6A1_3$ has been fitted by the power law $q^{-2,54}$ and by the form (1). The data have been recorded for $q \gg K$ and the best fit is obtained with the power-law form.³ The experimental parameters of the form (1) , together with Eq. (15), yield $d_f = 2.66$; this is consistent with the value given by the power law fit. Qn the other hand, we have obtained a good fit of the data for $q > 3K$ with Eq. (12a). The fractal dimension is 2.59 and the reciprocal correlation length 0.026 \AA^{-1} , while Eq. (1) gives 0.015 A^{-1} . The agreement is satisfactory if we consider that the latter fit is better at low q than at large q values.

In reentrant spin glass in decreasing temperatures the correlation length increases up to a maximum at T_G and decreases as the temperature further decreases. The latter behavior is usually interpreted as the breakdown of the long-range ferromagnetic order. The power-law fit of the structure factor of $(Fe_{0.70}Mn_{0.30})_{75}P_{16}B_6Al_3$ (Ref. 3) shows an increase of the exponent up to 2.4 at T_G and then a decrease up to $\alpha \approx 2.1$. With Eqs. (8) and (4) we may conclude that the volume of the fractal strongly decreases below T_G , in agreement with the picture of the breakdown of the ferromagnetic phase into ture or the breakdown of the refromagnetic phase into
domains. As for the random-field systems, $R \sim K^{-1}$ is the largest radius of curvature of the domain wall. The domain walls are pinned and K scales as the smallest pinning force.¹⁶

V. CONCLUSION

We hope that new experimental data considered along the lines developed in this paper will appear and confirm the reliability of the model. The main result is that magnetic domains may be fractal. From a topological point of view, a fractal dimension near ² in a 30 system corresponds to a gasket in which the largest radius of curvature of the boundary is given by K^{-1} . This picture strongly differs from that of spherical domains of radius K^{-1} . We have considered a fractal whose occupied part of the space has an uniform density. Near a phase transition the thermodynamic fluctuations become relevant, the density is no longer uniform, and the model breaks down in a nontrivial way. However, at low temperatures, thermal variations of K and d_f may be related to a temperature-dependent topology of the system. Magnetic systems of interest are diluted and care must be taken of percolation, since the infinite cluster near p_c is fractal itself.

ACKNOWLEDGMENT

The Laboratoire de Cristallographic et Minéralogie is Unite associe au Centre National de la Recherche Scientifique, UA No. 809.

¹P. Burlet, Ph.D. thesis, Université de Grenoble, 1968 (unpublished).

Phys. Rev. B 28, 5160 (1983}.

 $2E.$ O. Wollan and W. C. Koehler, Phys. Rev. 100, 545 (1955).

³G. Aeppli, S. M. Shapiro, R. J. Birgeneau, and H. S. Chen,

⁴H. Yoshizama, R. A. Cowley, G. Shirane, R. J. Birgeneau, H. J. Guggenheim, and H. Ikeda, Phys. Rev. Lett. 48, 438 $(1982).$

- 5T. Freltoft, J. K. Kjems, and S. K. Sinha, Phys. Rev. 8 33, 269 (1986).
- 6A. Aharony, Y. Gefen, and Y. Kantor, J. Stat. Phys. 36, 795 (1984).
- ⁷B. Mandelbrot, C. R. Acad. Sci. 288A, 81 (1979). B. Mandelbrot, Fractals —Form, Chance and Dimension (Freeman, San Francisco, 1977), p. 178.
- ⁸A. Guinier, G. Fournet, C. B. Walker, and K. L. Yudowitch, in Small-Angle Scattering of X-Rays (Wiley, New York, 1955), p. 78.
- ⁹S. Fishman and A. Aharony, J. Phys. C 12, L729 (1979).
- ¹⁰Y. Imry and S.-k. Ma, Phys. Rev. Lett. 35, 1399 (1975).
- ¹¹J. Villain, Phys. Rev. Lett. 52, 1543 (1984).
- ¹²R. Bruinsma and G. Aeppli, Phys. Rev. Lett. 52, 1547 (1984).
- ¹³R. A. Cowley, H. Yoshizawa, G. Shirane, and R. J. Birgeneau, Z. Phys. 8 58, 15 (1984).
- ¹⁴B. J. Dikken, A. F. M. Arts, H. W. de Wijn, and J. K. Kjems, Phys. Rev. 8 30, 2970 {1984).
- I5J. Villain, Z. Phys. 8 33, 31 (1979).
- ¹⁶I. M. Lifshitz, Zh. Eksp. Teor. Fiz. 42, 1354 (1962); [Sov. Phys.—JETP 15, ⁹³⁹ (1962)].