Journal of Magnetism and Magnetic Materials 58 (1986) 297–302
North-Holland, Amsterdam

A POSSIBLE INTERPRETATION OF NON-LINEAR ARROTT PLOTS *

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Received 6 August 1985

Arrott plots are computed for a certain model of a heterogeneous system of ferromagnets. These are shown to fit very well the experimental data of Berkowitz on amorphous $Fe_{75}Si_{15}B_{10}$ particles and ribbon. They also give the general shape of the experimental data of Kaul on amorphous $Fe_{20}Ni_{60}P_{14}B_6$ alloy, even though the fine details cannot be found from the experimental data. This fit supports the view that basic non-linearity of these plots indicates heterogeneity.

The Curie temperature, T_c, of ferromagnets is often determined by plotting the experimental data of the magnetization, M, as isotherms of $M^{1/\beta}$ vs. $(H/M)^{1/\gamma}$, where H is the (internal) magnetic field. For good crystalline samples, and for properly chosen values of the exponents β and γ, the isotherms are usually straight lines near T_c, in accordance with the equation of state [1]

$$\left(\frac{H}{M}\right)^{1/\gamma} = \frac{T - T_c}{T_1} + \left(\frac{M}{M_1}\right)^{1/\beta} \tag{1}$$

Here T is the temperature, and the other symbols are adjustable parameters. As long as this equation holds, the isotherms are straight lines, the intercept of which with the $M^{1/\beta}$ axis is positive or negative when $T <$ or $T > T_c$, respectively. Interpolation between these intercepts then determines T_c more accurately than any other method.

Difficulties arise, however, when the plots are *curved* lines, and interpolation to the $M^{1/\beta}$ axis becomes impossible. Sometimes such non-linearities are caused [2] only by a poor presentation of the experimental data. Such a case is an arbitrary choice of wrong critical exponents, β and γ, or including measurements in a field which is too low to remove the vestigial domains. Other non-linearities might be due to the use of a large field, H, beyond the validity of the *first-order* relation, (1). But for the latter case, the non-linearity in crystalline materials may be taken into account to a reasonable first-approximation by generalising [3] eq. (1) to:

$$m = \left\{1 - t + q(t)\tanh\left[r(t)(h/m)^{1/\gamma}\right]\right\}^{\beta}/D(t), \tag{2}$$

with

$$t = T/T_c, \quad m = M/M_0, \quad h = H/H_0, \tag{3}$$

where

$$\begin{aligned} D(t) &= 1 - \beta t + At^{3/2} - Ct^{7/2} \quad \text{for} \quad t \leqslant 1, \\ &= 1 - \beta + A - C \quad \text{for} \quad t \geqslant 1, \end{aligned} \tag{4}$$

$$q(t) = [D(t)]^{1/\beta} + t - 1, \tag{5}$$

$$r(t) = [D(t)]^{1/\beta}/[t^k q(t)]. \tag{6}$$

In the appropriate limits, this relation contains both the Arrott and Heinrich equation [4] and eq. (1), and the saturation $m = 1$ for large H or low T. Besides β, γ and T_c, it contains 5 adjustable parameters, M_0, H_0, A, C and k.

Now that such an approximate relation exists for all temperatures and fields, it is possible to calculate theoretical Arrott plots for a heterogeneous system of ferromagnets, with a distribution of their Curie temperatures. From the many kinds of feasible distributions, a simple model [2,5] is

* Some aspects of this work were presented orally at the 1985 Intermag Conference.

adopted, which contains only one adjustable parameter, σ, the width of that distribution. It assumes a collection of clusters, each with an $M(T, H)$ curve which is p times a universal curve, given by eqs. (2)–(6). Only the gaussian distribution is somewhat modified here, and is now taken as

$$P(p) = Cp^2 \exp\left[-(p-1+2\sigma^2)^2/2\sigma^2\right], \qquad (7)$$

whose peak is at $p = 1$. The width, σ, may be taken as the 9th adjustable parameter.

Fig. 1 plots theoretical curves thus obtained by integrating the contribution of clusters, whose distribution is given by eq. (7). These are fitted to the actual experimental values as communicated by A.E. Berkowitz, for amorphous particles in the 20–30 μm size range, using the values of the 8 adjustable parameters as given in the figure caption. The value of the 9th parameter, M_0, is not adjusted, since it is obtained from the measure-

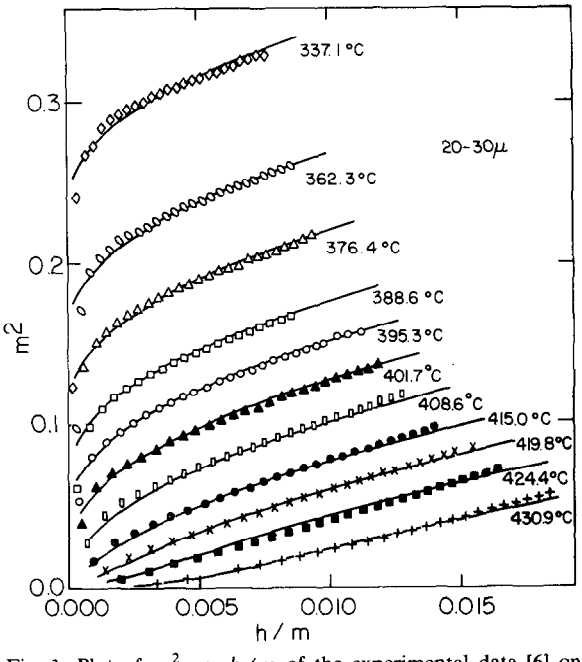

Fig. 1. Plot of m^2 vs. h/m of the experimental data [6] on amorphous $Fe_{75}Si_{15}B_{10}$ particles, with a diameter of 20–30 μm. The theoretical curves are computed by integrating over the probability distribution of eq. (7), of clusters, each with the magnetization curve of eqs. (2)–(6), with the values $T_c = 683.7$ K, $k = -0.587$, $\sigma = 0.01565$, $H_0 = 5.56 \times 10^6$ Oe, $\beta = 0.5517$, $\gamma = 2.1852$, $A = 0.1986$, $C = 0.1096$ and [7] $M_0 = 157.3$ emu.

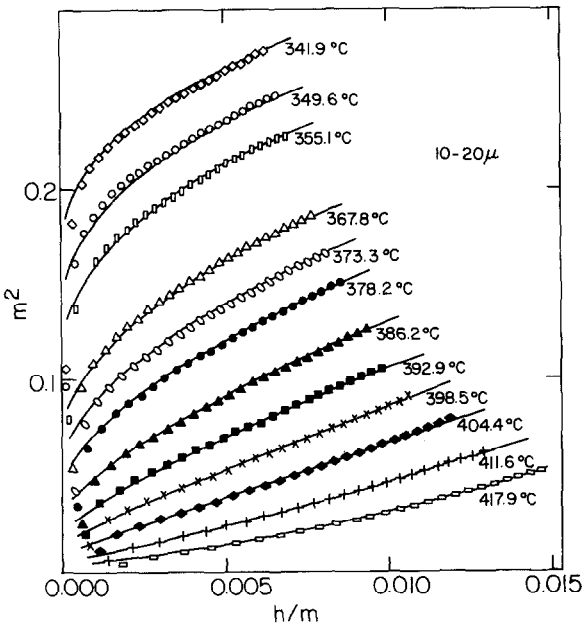

Fig. 2. Same as fig. 1, for 10–20 μm particles. The experimental data are from ref. [6], and the theoretical curves are plotted with the values $T_c = 651.0$ K, $k = -4.637$, $\sigma = 0.057$, $H_0 = 7.30 \times 10^6$ Oe, $\beta = 0.4056$, $\gamma = 2.2045$, $A = 0.2707$, $C = 0.2465$ and [7] $M_0 = 146.4$ emu.

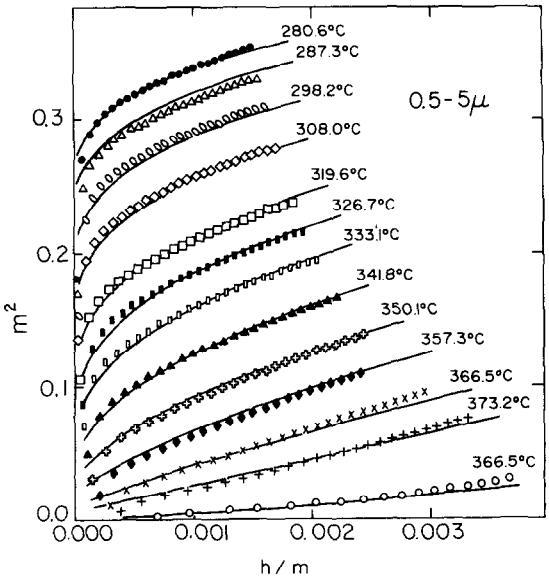

Fig. 3. Same as fig. 1, for 0.5–5 μm particles. The experimental data are from ref. [6], and the theoretical curves are plotted with the values $T_c = 611.6$ K, $k = 1.002$, $\sigma = 0.0589$, $H_0 = 2.691 \times 10^7$ Oe, $\beta = 0.2672$, $\gamma = 2.1596$, $A = 0.4628$, $C = 0.2271$ and [7] $M_0 = 132.8$ emu.

ment [7] for the same sample at low temperatures. It can be seen that the fit of the theoretical curves to the experimental data is very good, except for the points at the lowest fields. But the latter is expected [2] for fields which are too low to remove the domains, so that the measured magnetization is lower than its intrinsic value.

Similar plots are given in fig. 2 for smaller particles, in the 10–20 μm size range, and in fig. 3 for the smallest particles studied by Berkowitz, in the 0.5–5 μm range. Again, the values of M_0 are taken from experimental [7] values, and not fitted. And again, the fit between theory and experiment is very good, except for the lowest field values, for which one may assume the sample is subdivided into domains. The latter points were not included in the least-square fitting computations, since it was found that including them led to very large discrepancies in the high-field region of all the curves.

The wide variation of the 'critical' exponents, β and γ, between the different particle sizes might make one suspect the conventional idea of universality of these parameters, at least for an amorphous material, in which the immediate vicinity of one spin can be very different from that of other spins, so that there is no meaning to symmetry group classification. In a crystal, one can use the idea of superposition, but such a concept is lost in a true amorphous arrangements. Nevertheless, it is risky to jump into conclusions from the data of one system, and many more systems should be studied before concluding anything about the universality of these parameters. Besides, the determination of all 8 parameters from the few hundred data points in each of the figs. 1–3 is neither very accurate, nor necessarily unique. To demonstrate this point, the same data of fig. 3 were fitted to a completely different set of parameters, as plotted in fig. 4 (note, in particular, the factor of 10 in H_0). It is seen that the fit in fig. 4 is just as good as that in fig. 3, and actually the sum of squares of distances between points and curves is almost identical for the two cases. This makes one wonder how experimentalists manage to report any value at all, when the fit they use is not even nearly as good as in the figures reported here. A much deeper understanding of some of the parameters, or some method of eliminating some of them before fitting the others, is needed before drawing any valid conclusions.

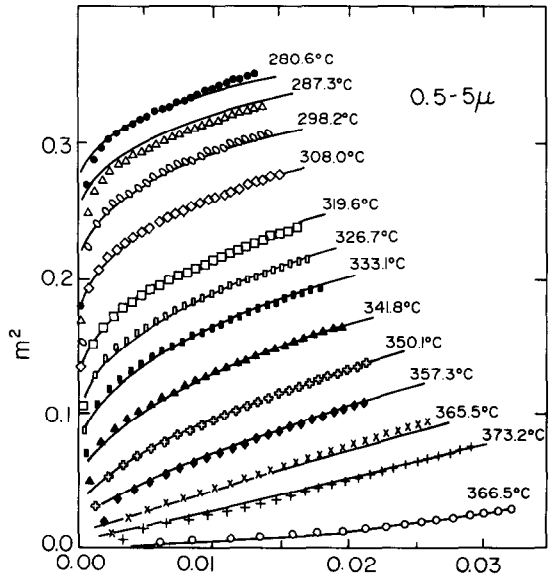

Fig. 4. Same data as fig. 3, but theoretical curves plotted with $T_c = 622.8$ K, $k = -6.806$, $\sigma = 0.0490$, $H_0 = 3.069 \times 10^6$ Oe, $\beta = 0.6086$, $\gamma = 2.4925$, $A = 0.3928$, $C = 0.4413$ and [7] $M_0 = 132.8$ emu.

Another example is a ribbon of the same amorphous composition, $Fe_{75}Si_{15}B_{10}$, plotted in fig. 5. The fit is quite good, though not as good as that of the particles in figs. 1–4. It should be noted that the distribution is quite narrow for the ribbon, indicating a large degree of short-range order, which is also found in other studies [7] of such a ribbon. And it is interesting to note that the 'critical' exponents β and γ of the ribbon are rather close to those of a crystal.

In order to try a different system as well, data were read from the published figure of Kaul [8], and fitted by the same procedure. It should be noted, however, that besides the inaccuracies of reading data from a small-size figure, difficulties arise from the use by Kaul [8] of the same notation for all data points. This makes it impossible to know which point belongs to which temperature, and a wrong guess can easily lead to very large errors in fitting parameters to the curves. In the first place, it is clear from figs. 1–5 that some

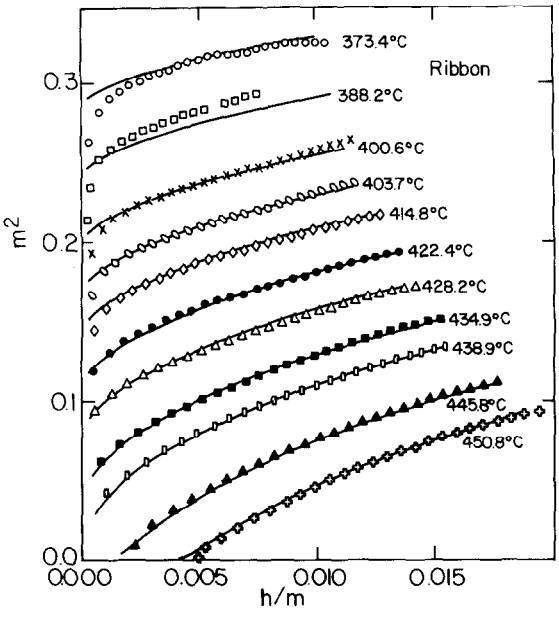

Fig. 5. Same as fig. 1, for an amorphous ribbon, with experimental data from ref. [6]. The theoretical curves are plotted for $T_c = 713.0$ K, $k = -4.303$, $\sigma = 0.00326$, $H_0 = 4.171 \times 10^6$ Oe, $\beta = 0.4080$, $\gamma = 1.675$, $A = 0.2590$, $C = 0.2023$ and [7] $M_0 = 178.4$ emu.

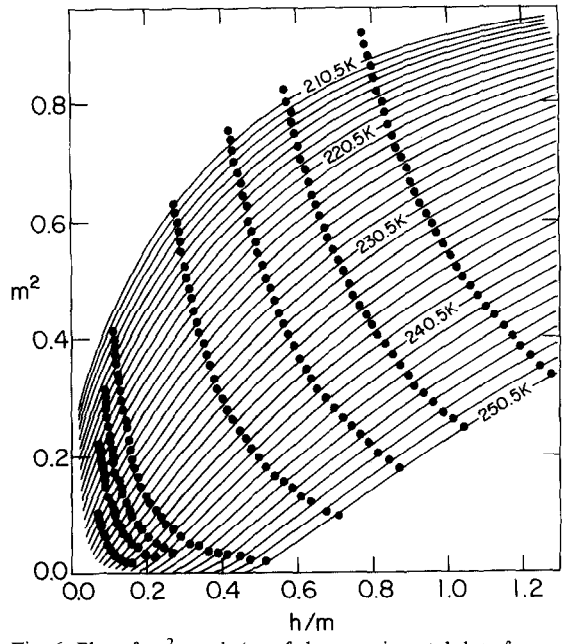

Fig. 6. Plot of m^2 vs. h/m of the experimental data from ref. [8], compared to theoretical curves computed by integrating over the probability distribution of eq. (7), of clusters, each with the magnetization curve of eqs. (2)–(6), with adjustable parameters' values given in eqs. (8)–(10).

low-field data points might belong to temperatures which are different from the line on which these points *seem* to lie. But in the case of Kaul [8], many of the points are *between* the drawn, empirical curves, and it is not clear to which of the curves they are meant to belong. Therefore, without such information, one cannot expect a good fit, and a semi-qualitative one should be good enough. Yet, for lack of other available data, it was considered worth trying this too, just in order to compare to another system. But in view of the crudeness of the data, and the fact that for this case no independent measurement is given for M_0, that has to be fitted as an extra parameter, it seemed better to reduce the number of adjustable parameters by applying the arbitrary constraint [3]

$$k = \gamma, \tag{8}$$

and by fixing A and C at the values obtained [4] for iron whiskers, namely

$$A = 0.110, \quad C = 0.129, \tag{9}$$

even though a different material is involved. The latter is chosen because it was found, in fitting figs. 1–5, that these parameters made little difference.

Fig. 6 plots theoretical curves similarly obtained by integrating the contribution of clusters, whose distribution is given by eq. (7), using the particular values:

$$T_c = 225.6 \text{ K}, \quad M_0 = 46.0 \text{ emu}, \quad H_0 = 13.6 \text{ kOe},$$
$$\beta = 0.556, \quad \gamma = 1.46, \quad \sigma = 0.0193, \tag{10}$$

for the parameters, and these are compared to the experimental data of Kaul [8] on amorphous $Fe_{20}Ni_{60}P_{14}B_6$ alloy. The fit is not particularly good, especially for the highest fields, at the lowest temperatures, which is rather similar to what one obtains in figs. 1–5 when the *lowest* field points are included. But the general shape of the theoretical and experimental curves is quite similar, which is all one can expect for the uncertainties involved in the temperatures. This similarity is even more obvious in fig. 7, where the same theoretical curves and the same experimental data points are plotted in the more appropriate fashion [2,9] of $m^{1/\beta}$ vs.

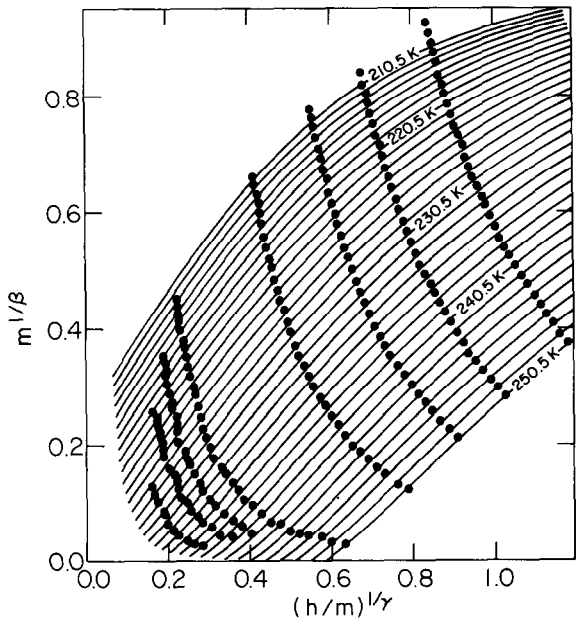

Fig. 7. Same data of fig. 6, plotted with the appropriate values of β and γ.

$(h/m)^{1/\gamma}$. At any rate, the fit is not much worse than that of the *empirical* curves which Kaul draws between the points.

In considering the discrepancy between theory and this particular experiment, it should also be noted that a highly specialized distribution is chosen. In particular, there is no physical reason that a 'cluster' with a high Curie point would also have a high magnetization at low temperatures, and vice versa. Also, the basic relation of eq. (2) had only a fair success in describing crystalline materials [3], and the t^k dependence in eq. (6) is a mere geuss. The latter cannot even be checked for crystals, for which data are available only near the Curie point, while very different temperatures contribute to the integration in the model for amorphous alloys, and for all one knows at this stage, the good fit of Berkowitz's data *might* be just a coincidence.

In view of these limitations, and the high inaccuracy and uncertainty involved in reading the data from a published figure, it was decided to save some of the impossibly-large computation time involved in a large number of integrations over the function m, which has to be evaluated by tedious successive iterations of eq. (2). Therefore, the least-square fitting was done, in this case, to a low accuracy, by integrating over few points only. But the curves in all the figures were computed with adequate number of integration points, so that the comparison in figs. 6 and 7 is correct for eq. (10). It is just not certain if the parameters of eq. (10) actually give the best fit to the data.

To demonstrate how unreliable the values of eq. (10) are, fig. 8 compares the *same* Kaul data to theoretical curves plotted with the values:

$$T_c = 218.8\ \text{K}, \quad M_0 = 50.2\ \text{emu}, \quad H_0 = 17.2\ \text{kOe},$$
$$\beta = 0.416, \quad \gamma = 1.61, \quad \sigma = 0.0691. \qquad (11)$$

Although these are considerably different than those of eq. (10), the fit is not much worse than in fig. 6. To the eye it might even look better that no point is far from a curve. It is not clear how Kaul [8] could deduce, from the same data, parameter values to a high accuracy, without even assuming a distribution.

The distribution width, $\sigma = 0.0193$, needed for plotting figs. 1 and 2, is quite small, and not easy to detect directly. This can be seen from the average magnetization for this model, which is plotted

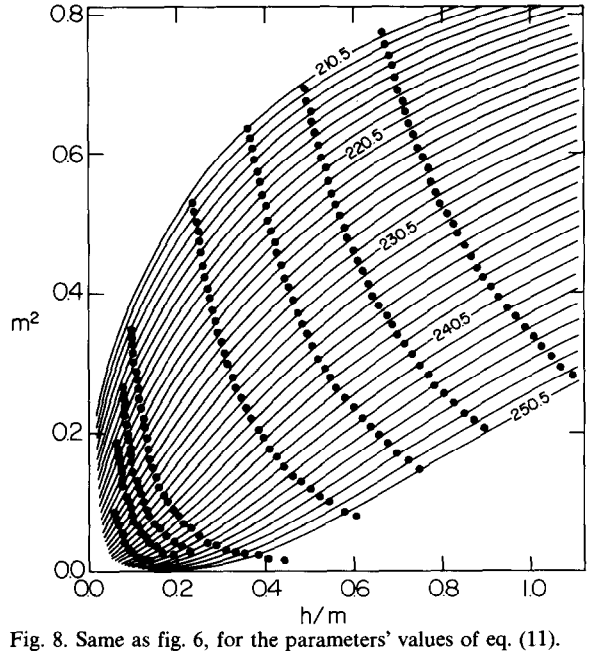

Fig. 8. Same as fig. 6, for the parameters' values of eq. (11).

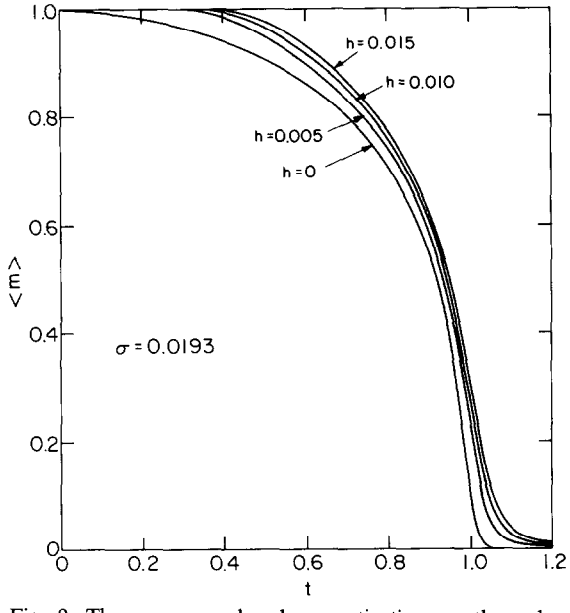

Fig. 9. The average reduced magnetization vs. the reduced temperature, for the distribution of clusters, whose Arrott plots are shown in fig. 6. Values of the reduced field, h, refer to the *internal* field.

in fig. 9 for this σ. The curve for $h = 0$ is a theoretical plot of what should be observed, if sufficiently accurate data could be taken without applying a field (like in the Mössbauer effect), or if data could be *properly* extrapolated down to zero field. The latter is not trivial because experimental data at low fields depend strongly on demagnetization, which increases [5] the 'smearing' near T_c.

It may, thus, be concluded that the intrinsic curvatures of Arrott plots in amorphous ferromagnets, are consistent with those of a heterogeneous system of ferromagnets. Instead of one, well-defined, Curie temperature, such a system has only a *distribution* of such points. But, obviously, a good analysis of amorphous ferromagnets is not possible before much more details are known on the behaviour of crystalline ferromagnets. And the real problem is to find a sufficiently accurate formular for $m(t, h)$ in crystals, preferably one that does not call for numerical iterations.

References

[1] A. Arrott and J.E. Noakes, Phys. Rev. Lett. 19 (1967) 786.
[2] A. Aharoni, J. Appl. Phys. 56 (1984) 3479.
[3] A. Aharoni, J. Appl. Phys. 57 (1985) 648.
[4] A.S. Arrott and B. Heinrich, J. Appl. Phys. 52 (1981) 2113.
[5] A. Aharoni, J. Appl. Phys. 53 (1982) 7719
[6] A.E. Berkowitz, private communication.
[7] A.E. Berkowitz, J.L. Walter and K.F. Wall, Phys. Rev. Lett. 46 (1981) 1484.
[8] S.N. Kaul, Phys. Rev. B23 (1981) 1205.
[9] S.N. Kaul, IEEE Trans. Magn. MAG-20 (1984) 1290.