

The Effects of Broadening on the High Temperature Critical Susceptibility Exponent γ

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Abstract

A theoretical study of the effects of a distribution of ordering temperatures T_c on the high temperature critical susceptibility exponent γ is described. Analytical and numerical solutions for γ are derived for the fitting of broadened susceptibility data to the critical equation $\chi = \chi_0 \{(T - T_c)/T_c\}^{-\gamma}$. Both least squares fitting and Kouvel-Fisher analyses are considered. Using a simple model for magnetically inhomogeneous material it is shown that the inclusion of the internal demagnetizing fields greatly reduces the effect of the broadening upon the deduced critical exponent. Theory is compared with experiment for the critical susceptibility of gadolinium.

Introduction

The effects of a spread in ordering temperatures is a recurring problem in experimental studies of critical phenomena. Esipov and Mikulinski (1970) determined a simple analytical solution for the paramagnetic susceptibility χ broadened by a distribution of ordering temperatures T_c , but only for a Curie-Weiss relationship $\chi = C/(T - T_c)$. Later Hohenberg and Barmatz (1972), using numerical techniques, examined the broadening effects due to gravity on the specific heat exponent α for a liquid-gas transition. We examine here the effects of a distribution of T_c values by considering the susceptibility of a real sample to be the sum of contributions from small regions, each with a particular ordering temperature T_c and a susceptibility given by the simple critical paramagnetic equation (Stanley 1971)

$$\chi(T, T_c) = \chi_0 \varepsilon^{-\gamma_0}, \quad (1)$$

where $\varepsilon = (T - T_c)/T_c$ and γ_0 is the paramagnetic critical exponent. The aim is to determine the extent to which the susceptibility of the whole sample deviates from a simple critical equation with T_c replaced by the mean ordering temperature T_c^0 and hence to determine the effect of this upon deduced values of γ .

This study is particularly relevant to recent experimental measurements of γ for polycrystalline gadolinium which have been undertaken in this laboratory (Wantenaar *et al.* 1980), as it assists in assessing the validity of γ values determined from broadened experimental data. As well as being useful for analyses of γ , the general principles of the present study are applicable to the measurement of any critical exponent.

In the next section the effects of the broadening upon the paramagnetic susceptibility are studied both with and without allowance for the effects of internal demagnetizing fields. In later sections the effects of the broadened susceptibility functions upon the

values of T_c^0 and γ deduced by least squares fitting (LSF) and by Kouvel–Fisher (1964) analysis are determined. The calculations for Kouvel–Fisher analysis are compared with experimental results (Wantenaar *et al.* 1980) for gadolinium.

Effects of Broadening on Paramagnetic Susceptibility

Distribution of T_c Values

Neglecting the effects of internal demagnetizing fields, and assuming that each homogeneous region of the sample contributes a paramagnetic susceptibility of the form of equation (1), we have for the total susceptibility for a distribution $P(T_c)$ of Curie temperatures

$$\langle \chi(T) \rangle = \int_0^\infty \chi(T, T_c) P(T_c) dT_c. \quad (2)$$

This assumes that the data are taken over a range of temperatures T in which no region of the sample becomes ferromagnetic. The experimental technique of transient enhancement (Wantenaar *et al.* 1976) enables one to determine precisely the temperature at which thermal nucleation of ferromagnetic domains begins and hence to avoid the inclusion of data from below this temperature in the analysis.

We define $t = T - T_c^0$ and $t_c = T_c - T_c^0$ as the deviations in turn between the temperature T and Curie temperature T_c of a region and the mean Curie temperature T_c^0 . Hence we have

$$\langle \chi(T) \rangle = \chi_0 \int_{-\infty}^\infty \left(\frac{t - t_c}{t_c + T_c^0} \right)^{-\gamma_0} P(t_c) dt_c. \quad (3)$$

Assuming that the spread in Curie temperatures is small and that the measurements are made at temperatures well above the Curie temperatures (i.e. $t \gg t_c$), one may apply a binomial expansion leading to

$$\langle \chi \rangle = \chi^0 \{ 1 + \frac{1}{2} \gamma_0 (\gamma_0 + 1) (\sigma' / \varepsilon)^2 + \dots \}, \quad (4)$$

where now $\chi^0 = \chi_0 \{ (T - T_c^0) / T_c^0 \}^{-\gamma_0}$, $\varepsilon = (T - T_c^0) / T_c^0$ and $\sigma' = \sigma / T_c^0$, with the variance of $P(t_c)$ given by

$$\sigma^2 = \int_{-\infty}^\infty t_c^2 P(t_c) dt_c.$$

In deriving equation (4) it is assumed that $P(t_c)$ is an even function so that all odd terms in the expansion are zero.

Equation (4) indicates that $\langle \chi \rangle$ equals the unbroadened susceptibility χ^0 plus a correction term dependent upon the ratio of the reduced variance σ' to the deviation t of the sample temperature from the mean Curie temperature. As expected this expression reduces to that obtained by Esipov and Mikulinski (1970) for a Curie–Weiss law ($\gamma_0 = 1$).

Internal Demagnetizing Fields

The range of σ' values (1×10^{-5} to 3×10^{-3}) to be considered in this paper is typical for ferromagnetic samples used in critical studies and, for the case of gadolinium with $T_c \sim 290$ K, represents a width of T_c values ranging from a few mK (high purity

single crystal) to ~ 1 K (impure polycrystal). In such real samples the broadening of the Curie temperatures due to impurities must be associated with variations of composition on a macroscopic rather than an atomic scale and the spatial variation of susceptibility through the sample will result in local demagnetizing fields.

In attempting to describe the effects of internal demagnetizing fields we adopt a simplified approach to what is a complex many-body problem in micromagnetics (cf. e.g. Brown 1963); the sample is considered to be comprised of many small regions each of which can be described by an average demagnetizing factor D_r and experiencing a demagnetizing field given by $D_r(M - \langle M \rangle)/\mu_0$, where M is the field induced magnetization for the region and $\langle M \rangle$ the average magnetization of the sample, with μ_0 the permeability of free space. Hence, for each region with a given T_c , the internal field H_i for an ellipsoid can be represented as

$$H_i = H_a - D_b \langle M \rangle / \mu_0 - D_r (M - \langle M \rangle) / \mu_0, \quad (5)$$

where H_a is the applied magnetic field and D_b is the bulk demagnetizing factor for the sample. For non-ellipsoidal samples there will, of course, be no unique D_b ; an important case in AC studies of ferromagnetic critical phenomena is that of toroidal samples where D_b is zero. The magnetization for each region is given by $M = \mu_0 \chi H_i$, where χ is the relative susceptibility, so that for a toroidal sample

$$H_i = (H_a + D_r \langle M \rangle / \mu_0) / (1 + D_r \chi) \quad (6)$$

(this may be compared with the usual expression for a regular body of unique D and M of $H_i = H_a / (1 + D\chi)$). Taking $\langle M \rangle = \mu_0 \langle \chi H_i \rangle$ we obtain

$$\langle M \rangle = \mu_0 H_a \langle \chi_d \rangle / (1 - \langle D_r \chi_d \rangle), \quad (7)$$

where

$$\chi_d = \chi / (1 + D_r \chi). \quad (8)$$

For simplicity we now assume that the distribution of the demagnetizing factors may be accommodated by assuming a single effective average value of $\langle D_r \rangle = D$ (this value is probably close to that for a sphere, $D = \frac{1}{3}$). The effective susceptibility which would then be observed is

$$\chi_e = \langle M \rangle / \mu_0 H_a = \langle \chi_d \rangle / (1 - D \langle \chi_d \rangle). \quad (9)$$

It is not possible to expand χ_d as a power series for all values of D and χ but analytic solutions are possible at two limits:

(i) For $D\chi \ll 1$, χ_e approaches the result $\langle \chi \rangle$ given by equation (4) which, for each value of T , is greater than the unbroadened susceptibility χ^0 .

(ii) For $D\chi \gg 1$, χ_e approaches $\langle \chi^{-1} \rangle^{-1}$ and, as $\gamma_0 \gg 1$, this will lead to a decrease below χ^0 for all ε .

To second order in σ'/ε these two limiting cases therefore yield

$$\chi_e = \langle \chi \rangle \approx \chi^0 \{1 + \frac{1}{2} \gamma_0 (\gamma_0 + 1) (\sigma'/\varepsilon)^2\} \quad D\chi \ll 1, \quad (10)$$

$$= \langle \chi^{-1} \rangle^{-1} \approx \chi^0 / \{1 + \frac{1}{2} \gamma_0 (\gamma_0 - 1) (\sigma'/\varepsilon)^2\} \quad D\chi \gg 1. \quad (11)$$

In experiments on critical behaviour the relative susceptibility χ could vary between about 0.1 to several hundred in the critical region while D could be between 0.01 and 1. Hence it is possible for the values of $D\chi$ to lie anywhere between the above two limiting cases. The reason why the effects of broadening are reduced when the internal demagnetizing fields are included is because they play a role similar to negative feedback, reducing the effects of those regions with the highest susceptibility (highest T_c) and vice versa via the term $-D_r(M - \langle M \rangle)/\mu_0$ in equation (5).

Determination of γ for Fixed T_c

A common approach in the analysis of critical data is to determine the critical temperature T_c independently either by experiment (for example, the peak in specific heat, Hohenberg and Barmatz 1972) or by an analytical technique (see e.g. Kouvel and Fisher 1964). This value T_c is then held constant in the LSF of the other critical parameters γ and χ_0 to the critical equation (1).

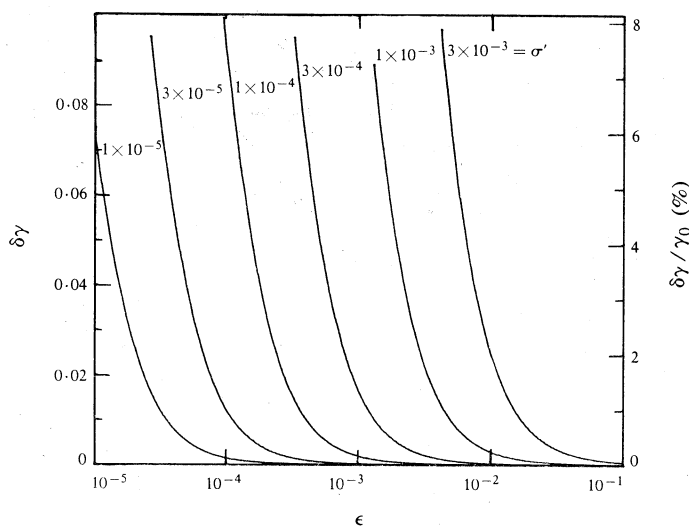


Fig. 1. Deviation $\delta\gamma$ between the deduced and unbroadened exponent (equation 13) as a function of ϵ for various fractional broadenings σ' for fitting with fixed T_c . A value $\gamma_0 = 1.23$ and the broadening expression (4) taken to second order with $D = 0$ are assumed.

Taking $D = 0$ we assume a fixed value of $T_c = T_c^0$ and also hold χ_0 constant at its unbroadened value. A fit of the broadened $\langle \chi \rangle$ of equation (3) to a simple critical equation will result in a deduced value γ^* for each narrow range of ϵ , corresponding to a mean $\langle \chi \rangle$, such that

$$\gamma^* = (\log \chi_0 - \log \langle \chi \rangle) / \log \epsilon. \quad (12)$$

The deviation of γ^* from the unbroadened value is given by

$$\delta\gamma = \gamma^* - \gamma_0 = -\{\log(1 + k\epsilon^{-2})\} / \log \epsilon, \quad (13)$$

where $k = \frac{1}{2}\gamma_0(\gamma_0 + 1)\sigma'^2$, taking equation (4) to second order in σ'/ϵ . Fig. 1 shows the variation of $\delta\gamma$ with ϵ for various σ' with $\gamma_0 = 1.23$. This γ_0 value is the theoretical prediction for a prolate anisotropic Heisenberg high spin system (Jasnow

and Wortis 1968) considered to be appropriate to gadolinium (Wantenaar *et al.* 1980). Fig. 1 shows that for a small range of ε values the deduced value γ^* will depend upon ε and that $\delta\gamma > 0$ always. For $\varepsilon \sim 5\sigma'$ a deviation $\delta\gamma \sim 1\%$ is expected. If a set of broadened data is fitted over an extended range ε_{\min} to ε_{\max} it is expected that the deviation of the fitted value γ^* from the unbroadened value would lie between $\delta\gamma(\varepsilon_{\min})$ and $\delta\gamma(\varepsilon_{\max})$.

The behaviour of $\delta\gamma$ shown in Fig. 1 is similar to that observed by Hohenberg and Barmatz (1972) in the specific heat exponent α for an LSF with fixed T_c to artificially generated gravity broadened data. This is expected because for $D = 0$ the behaviour of $\delta\gamma$ in equation (13) should be similar to that for any critical exponent.

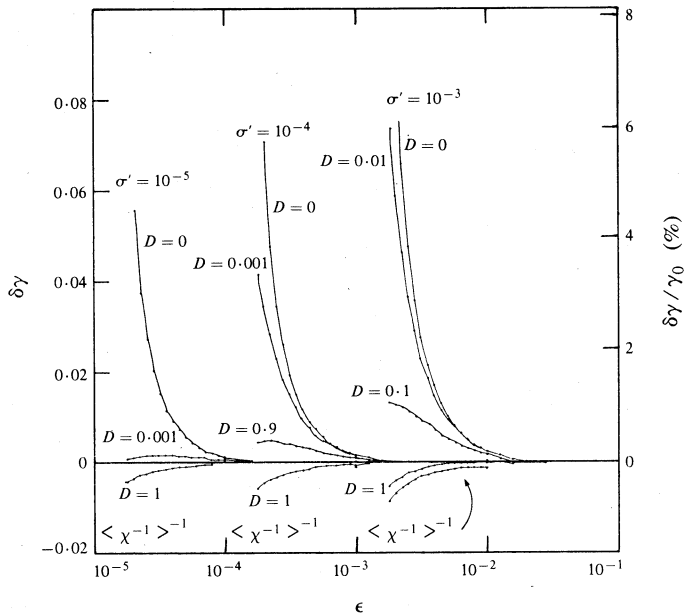


Fig. 2. Dependence of the deviation $\delta\gamma$ upon ε for $\chi_0 = 0.01$ and various values of the fractional broadening σ' and internal demagnetizing factor D , together with the result from $\langle\chi^{-1}\rangle^{-1}$ expected for $D\chi \gg 1$. For $\sigma' \lesssim 10^{-4}$ the values of $\delta\gamma$ for $D = 1$ and $D\chi \gg 1$ are indistinguishable.

Numerical Solution for $D \neq 0$

For the case $D \neq 0$ a particular functional form for the distribution of T_c values must be assumed because a numerical calculation of χ_e from equation (9) is necessary. A simple rectangular distribution of width 2τ was assumed to eliminate problems associated with components in the lower tail of a gaussian or Lorentzian distribution from becoming ferromagnetic (i.e. with negative ε). Using the rectangular distribution we rewrite equation (8) as

$$\chi_d = \frac{1}{2\sigma\sqrt{3}} \int_{-\tau}^{\tau} \frac{\chi}{1 + D\chi} dt_c, \quad (14)$$

where $\sigma = \tau/\sqrt{3} = \sigma'T_c^0$. For each value of ε , χ_e is calculated from equations (8), (9) and (14) by integration over the distribution. Replacing $\langle\chi\rangle$ by χ_e in equation (12) gives the deviation $\delta\gamma$ as a function of ε .

Fig. 2 shows the dependence of $\delta\gamma$ upon ε for distributions with various widths (i.e. various values of σ') and for various internal demagnetizing factors D . A value $\chi_0 = 0.01$ appropriate for gadolinium (Wantenaar *et al.* 1980) has been assumed. Also shown is the dependence for the limiting result $\chi_e = \langle\chi^{-1}\rangle^{-1}$ for $D\chi \gg 1$. The deviations $\delta\gamma$ indicate the increase and decrease consistent with the two extreme cases $D = 0$ and $D\chi \gg 1$ given by equations (10) and (11) respectively. The large differences in magnitude of the deviations $\delta\gamma$ between these two limits are due to the relative factors $\gamma_0 + 1$ and $\gamma_0 - 1$ in equations (10) and (11). For $D = 0$ these numerical results are similar to the second order results in Fig. 1; however at $\varepsilon = 3\sigma'$ the second order calculation is seen to underestimate $\delta\gamma$ by about 20%. This error is reduced to about 5% when the fourth order terms in equation (4) are retained, indicating that the second order expansion should not be relied upon for values of $\varepsilon \gtrsim 3\sigma'$.

For $\sigma \lesssim 10^{-4}$, Fig. 2 shows that $\delta\gamma$ is less than 0.01 for all realistic values of D (0.01 to 1) and of ε ($\gtrsim \sigma'/10$). In these ranges $D\chi$ is large enough for χ_e to approach $\langle\chi^{-1}\rangle^{-1}$ more closely than $\langle\chi\rangle$. It can be concluded that in many experiments the internal demagnetizing fields will largely eliminate the effects of the broadening upon the paramagnetic susceptibility and LSF would then give the correct γ over a wide range of ε values.

Determination of γ and T_c by Fitting

Because of difficulties in defining T_c independently to sufficient accuracy it is often necessary to analyse the data using LSF with both T_c and γ as floating parameters. In this section we consider the effect of allowing T_c to vary on the results obtained above for fixed T_c for $D\chi \ll 1$. We first present an analytical solution for the deviations $\delta\gamma$ and δT_c from the unbroadened values γ^0 and T_c^0 for an LSF and then compare them with a numerical LSF to artificially broadened data.

Analytical Least Squares Fit

Fitting the broadened $\langle\chi\rangle$ data to an exponent law leads to the equations

$$\frac{d}{d\xi} \int \left(\varepsilon^{-\gamma_0}(1 + k\varepsilon^{-2}) - \varepsilon^{-\gamma} \right)^2 dT = 0,$$

where $\xi = \gamma$ or T_c , and the expression (4) for $\langle\chi\rangle$ taken to second order has been used. The integration is over the fitting range from T_{\min} to T_{\max} . To first order in $\delta\gamma = \gamma - \gamma_0$ and $\delta T_c = T_c - T_c^0$ we have

$$\int \frac{d}{d\xi} \left(\varepsilon^{-\gamma_0}(\delta\gamma \ln \varepsilon - \delta T_c T_{\gamma_0}/T_c^{02} \varepsilon + k\varepsilon^{-2}) \right)^2 dT = 0.$$

Differentiation with respect to γ and T_c leads to two linear equations in $\delta\gamma$ and δT_c :

$$A \delta\gamma - B \delta T_c = -C, \quad -D \delta\gamma + E \delta T_c = F, \quad (15a, b)$$

where

$$A = \int \varepsilon^{-2\gamma_0} (\ln \varepsilon)^2 d\varepsilon, \quad B = \gamma_0 \int \varepsilon^{-(2\gamma_0+1)} (\ln \varepsilon)(1 + \varepsilon)/T_c^0 d\varepsilon,$$

$$C = k \int \varepsilon^{-(2\gamma_0+2)} \ln \varepsilon \, d\varepsilon, \quad D = B/\gamma_0,$$

$$E = \gamma_0 \int \varepsilon^{-(2\gamma_0+2)} (1+\varepsilon)^2 / T_c^{0.2} \, d\varepsilon, \quad F = \int \varepsilon^{-(2\gamma_0+3)} (1+\varepsilon) / T_c^0 \, d\varepsilon,$$

and the limits of integration are ε_1 to ε_2 corresponding to the range T_{\min} to T_{\max} respectively. With the approximation $1+\varepsilon \approx 1$ in the equations for B , E and F the integrals are easily calculated analytically. The integration was with respect to $\ln \varepsilon$ which is equivalent to LSF with data points evenly spaced in $\ln \varepsilon$.

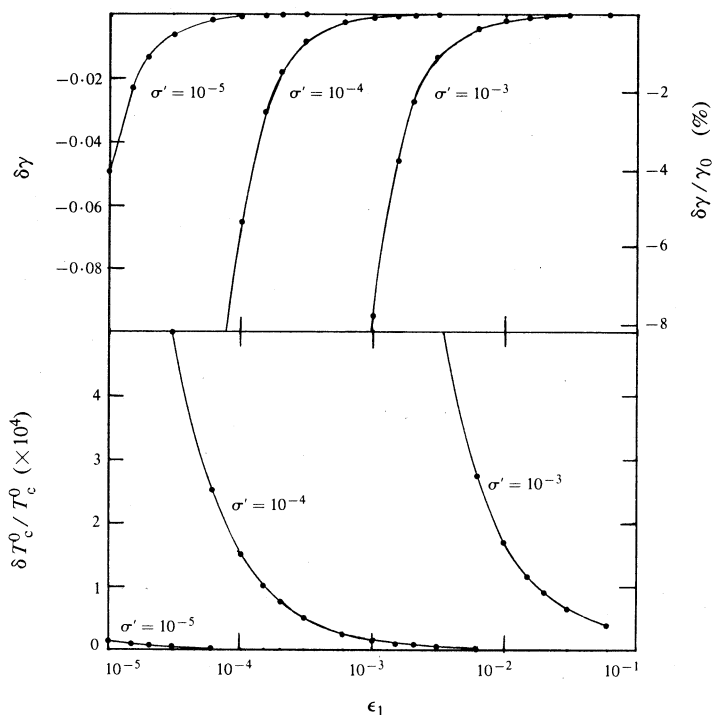


Fig. 3. The deviations $\delta\gamma$ and $\delta T_c^0/T_c^0$ predicted for a two-parameter LSF as a function of the lower range limit ε_1 , keeping the upper limit fixed at $\varepsilon_2 = 0.1$ for various σ' .

The calculated dependences of $\delta\gamma$ and δT_c upon ε_1 for $\varepsilon_2 = 0.1$ and various σ' are shown in Fig. 3. Comparing these results with those of Fig. 1 for the deviation $\delta\gamma$ with fixed T_c shows that, for $D\chi \ll 1$, the deviation $\delta\gamma$ is almost halved in magnitude and has its sign changed when T_c also is allowed to vary in the fitting; the deduced value of T_c will be larger than the mean value T_c^0 .

Numerical Least Squares Calculations

To test the validity of the various approximations made above, and to determine also the effects of allowing χ_0 to vary in the fitting, the broadening equation (4) was taken to second order and used to generate broadened data at points equally spaced in $\ln \varepsilon$. An LSF computer program (Wantenaar 1978) was then used and involved searching

the chi-squared hypersurface for its minimum. Fig. 4 shows the results obtained for both fixed χ_0 and when χ_0 is also permitted to vary; it indicates that, if χ_0 is determined as an adjustable fitting parameter, there can be quite significant effects upon the deduced values of γ and T_c .

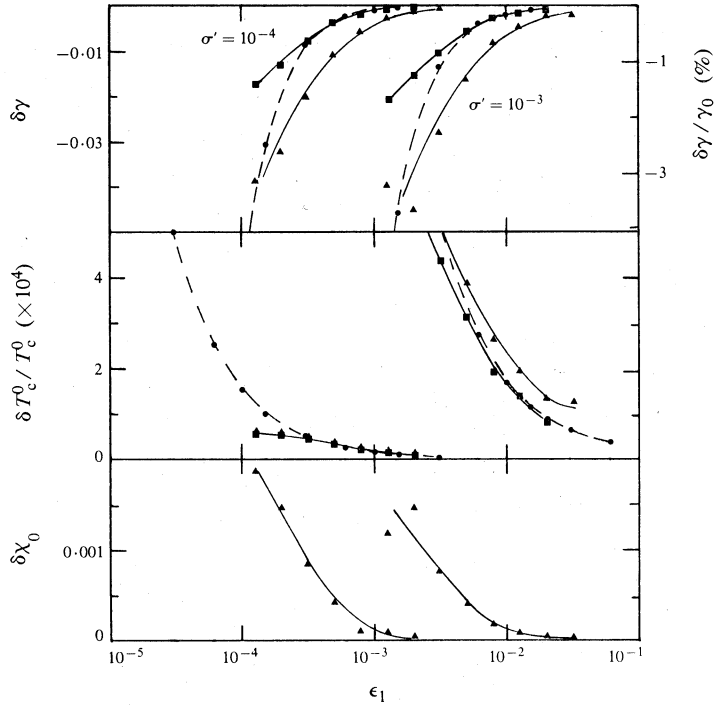


Fig. 4. The deviations $\delta\gamma$, $\delta T_c^0/T_c^0$ and $\delta\chi_0$ as a function of the lower range limit ϵ_1 for $D = 0$ and $\epsilon_2 = 0.1$. Squares are the LSF with χ_0 held constant, triangles the LSF with χ_0 allowed to vary, and circles are for the analytical treatment using a second order expression for the broadened susceptibility.

In summary, both the above analytical and numerical LSF calculations show that fitting $\langle\chi\rangle$ (calculated to second order in σ'/ϵ from equation 4) to the simple critical equation (1) gives a decrease in the fitted γ and an increase in T_c as the minimum of the fitting range approaches T_c^0 . If χ_0 is held constant we have $\delta\gamma \lesssim 1\%$ for $\epsilon > 3\sigma'$, whereas if χ_0 is allowed to vary $\delta\gamma \lesssim 1\%$ for $\epsilon > 6\sigma'$.

Kouvel–Fisher Analysis

A common approach in the analysis of critical susceptibility data is to eliminate χ_0 by determining the temperature derivative and fitting to the straight line

$$\chi/(d\chi/dT) = (T - T_c)/\gamma. \tag{16}$$

This approach was first used by Kouvel and Fisher (1964) with $d\chi/dT$ values being obtained by differentiation of the susceptibility data. Wantenaar *et al.* (1980) recently extended this to include the determination of $d\chi/dT$ experimentally using the temperature modulation technique.

Kouvel-Fisher Analysis for Artificially Broadened Data

For fixed values of σ' and D , sets of values of χ_e were determined for a rectangular distribution of T_c values with a half-width of $\sigma\sqrt{3}$ (equations 9 and 14). The derivative $d\chi_e/dT$ was calculated using the method of central differences. These data were then fitted to equation (16) using the LINFIT program (Bevington 1969) modified by Wantenaar *et al.* (unpublished) for analysing experimental susceptibility data. As in the previous section, data are evenly spaced in $\ln \varepsilon$ with a range maximum $\varepsilon_2 = 0.1$. Fig. 5 shows the parameters $\delta\gamma$ and δT_c^0 as functions of ε_1 , the lower range limit, for $\sigma' = 10^{-4}$ and 10^{-3} , and $\chi_0 = 0.01$, with several values of D . As expected the behaviour of $\delta\gamma$ and δT_c^0 for $D = 0$ is very similar to that for the LSF of the previous section except that for $\varepsilon_1 \gtrsim 5\sigma'$ differences begin to appear because of the approximation to second order in σ'/ε used previously.

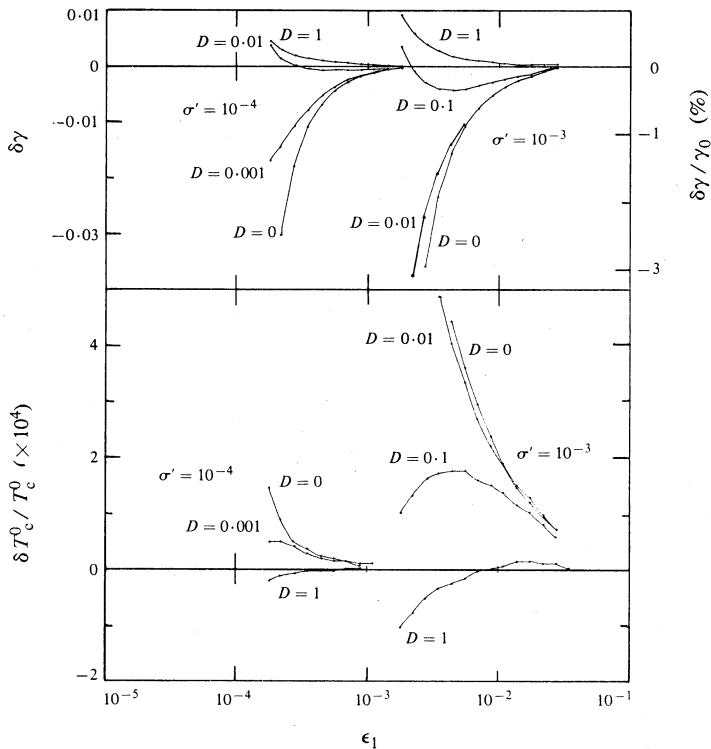


Fig. 5. Plot of the calculated deviations $\delta\gamma$ and $\delta T_c^0/T_c^0$ versus ε_1 for Kouvel-Fisher analysis with $\varepsilon_2 = 0.1$, $\chi_0 = 0.01$ and for $\sigma' = 10^{-4}$, 10^{-3} and several values of D .

The most interesting behaviour occurs for $D \neq 0$. As in the LSF calculations the effect of including internal demagnetizing fields is to reduce the deviations $\delta\gamma$ and δT_c^0 and, for large enough $D\chi$, these deviations change sign. When $\sigma' = 10^{-4}$, for all realistic values of D the variation in $\delta\gamma$ for $\varepsilon_1 > 2 \times 10^{-4}$ is $\lesssim 0.005$. Thus, even for such an 'intermediate purity' sample, the effects of the broadening have been greatly reduced by the demagnetizing fields, as occurred for $\delta\gamma$ with fixed T_c (Fig. 2). For the 'impure sample' ($\sigma' = 10^{-3}$) the effects of the demagnetizing fields are similar though somewhat reduced.

Comparison with Experiment

The Kouvel–Fisher technique has recently been used in the analysis of the critical susceptibility for 96 at. % pure polycrystalline gadolinium with $T_c \sim 291$ K (Wantenaar *et al.* 1980). Transient enhancement measurements indicated a distribution in T_c values with $\sigma \sim 0.5$ K (i.e. $\sigma' \sim 1.7 \times 10^{-3}$). Fig. 6 shows the values of γ and T_c deduced from a Kouvel–Fisher analysis as a function of ϵ_1 keeping ϵ_2 fixed at 4×10^{-2} . For ϵ_1 in the range $(1-2) \times 10^{-2}$, γ is constant to within $\pm 1\%$ and T_c is constant to within $\pm 0.01\%$. The larger deviations for $\epsilon_1 \gtrsim 2 \times 10^{-2}$ are simply due to inaccuracies arising as the range $\epsilon_2 - \epsilon_1$ becomes too small. Comparison with Fig. 5 shows that the relative constancy of γ and T_c for ϵ_1 in the range $(1-2) \times 10^{-2}$ and the polarity of the small variations in this range cannot be explained in terms of the broadening alone (i.e. with $D = 0$); however the experimental results are quite consistent with the theoretical curves if internal demagnetizing fields with $D \gtrsim 0.2$ are included.

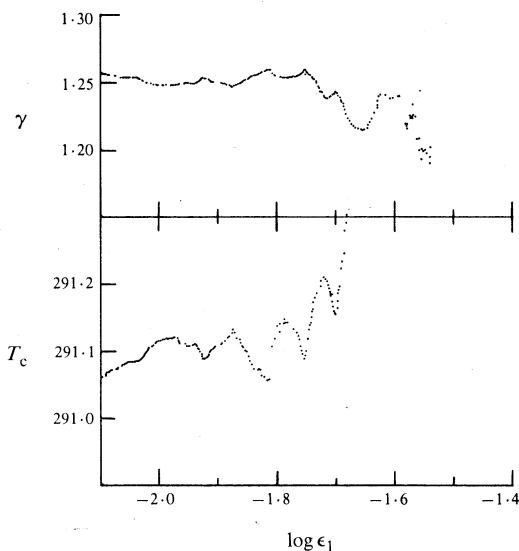


Fig. 6. Dependence of the deduced values of γ and T_c (in K) upon ϵ_1 for Kouvel–Fisher analysis of experimental data for polycrystalline gadolinium. The upper range limit ϵ_2 was held constant at 4×10^{-2} .

The theoretical analysis shows that, under these conditions, a γ value which is independent of ϵ_1 should be within ~ 0.005 of the true unbroadened value γ_0 . Therefore these experiments (Wantenaar *et al.* 1980), for which studies of several samples yielded the value $\gamma_0 = 1.24 \pm 0.03$, should yield the unbroadened susceptibility exponent for gadolinium.

Conclusions

The fitting of a simple critical equation (1) to susceptibility data broadened by a distribution of critical temperatures has been studied with and without allowance for the effects of internal demagnetizing fields. In each case studied the inclusion of internal demagnetizing fields reduces the deviations of the fitted parameters from their unbroadened values. The observed weak dependences of the fitted values of

γ and T_c in a Kouvel-Fisher analysis of experimental data for polycrystalline gadolinium agree well with the theoretical results provided internal demagnetizing fields are included. In such cases it is possible to determine accurately the unbroadened critical exponents in the presence of significant distributions of critical temperatures.

Acknowledgments

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