Field-dependent susceptibility of a paramagnet

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The physical origin of peaks observed in the field-dependent susceptibility of a number of ferromagnets at temperatures above the critical temperature T_c is identified. As a corollary, it is shown that the field-dependent susceptibility of a paramagnet exhibits an obvious but little discussed structure at finite temperature of similar origin. This structure can be used to extract information on the zero-temperature transition that should occur in paramagnetic systems (systems which formally do not experience any interactions) in a manner similar to that used to extract information on the finite-temperature phase transition in ferromagnets. This provides strong confirmatory evidence that this type of anomaly is static rather than dynamic in origin.

Several recent publications have reported the dc field dependence of the reversible (ac) susceptibility $\chi_{ac}(H,T)$ of a number of systems undergoing a well-established second-order paramagnetic to ferromagnetic phase transition.[1-6] In a finite field H, this susceptibility exhibited a peak at a temperature T_m which occurred *above* the zero-field ordering temperature T_c (the Curie temperature). The amplitude of this peak $\chi_{ac}(H,T_m)$, decreased with increasing static field (H), while the position of this peak in temperature (T_m) increased as H increased. Two sources for this have been advanced so far. The first suggested that it originated from dynamical effects,[5] with this anomaly being linked, via relaxation effects, to a divergent behavior in the specific heat. The second indicated that this result was consistent with the predictions of *static* scaling theory applied to such a transition. In the static approach the (reduced) magnetization m is related to the conventional scaling fields t $[=(T-T_c)/T_c]$ and h $(=H/T_c)$ (neglecting numerical and spin-dependent factors) via[7]

$$m(h,t)\sim t^{\beta}F_{\pm}(h/t^{\gamma+\beta}) , \tag{1}$$

where the suffix $+$ $(-)$ refers to temperatures above (below) T_c. Equation (1) leads to the well-known asymptotic relationships

$$m(0,t)\sim t^{\beta_-},\ T<T_c , \tag{2}$$

$$m(h,0)\sim h^{1/\delta},\ \chi(h,0)\sim h^{(1/\delta)-1},\ T=T_c , \tag{3}$$

$$\chi(0,t)\sim t^{-\gamma_+},\ T>T_c , \tag{4}$$

from which the critical exponents γ_+, β_-, and δ are usually estimated experimentally. Furthermore, if the Widom equality holds[8] (in addition to the scaling hypothesis, which renders the suffices $\pm$ redundant) then these exponent values are related by

$$\gamma=\beta(\delta-1) . \tag{5}$$

In addition to these asymptotic dependences, this same static scaling law [Eq. (1)] predicts that the differential susceptibility can in general be written in the form[1,3,6]

$$\chi(h,t)=\partial m/\partial h=t^{-\gamma}F(h/t^{\gamma+\beta})=h^{(1/\delta)-1}G(h/t^{\gamma+\beta}) , \tag{6}$$

and this susceptibility arises from the same source as does that in Eqs. (3) and (4)—critical fluctuations in the magnetization. It is straightforward to show that this general form [Eq. (6)] predicts that the field-dependent susceptibility exhibits a maximum *above* T_c at a temperature t_m $[=(T_m-T_c)/T_c]$ which increases with increasing field according to

$$t_m\propto h^{1/(\gamma+\beta)} . \tag{7}$$

The susceptibility *at this peak*, $\chi(h,t_m)$, is then a function of *field alone*, decreasing with increasing field according to

$$\chi(h,t_m)\propto h^{(1/\delta)-1} . \tag{8}$$

Thus, the *peak* susceptibility exhibits the *same* asymptotic dependence on field as does the susceptibility at T_c —$\chi(h,0)$ of Eq. (3), i.e., both $\chi(h,0)$ and $\chi(h,t_m)$ are independent of temperature and vary with field with the same index δ. This latter prediction has been directly verified experimentally.[3] Furthermore, not only have these model-independent predictions of the scaling-law approach been confirmed by numerical calculations on Ising models using both high-temperature expansion techniques for various lattice structures[9] and within the *ferromagnetic* phase of the Sherrington-Kirkpartick model,[10] but Eqs. (7) and (8) have also been shown to quantitatively reproduce the observed behavior of such peaks in both metallic[1-4] and insulating ferromagnets.[6]

The line of susceptibility maxima $t_m\propto h^{1/\gamma+\beta}$ [along which $(\partial\chi/\partial t)_h=0$] delineates a crossover from a high-temperature region where $(\partial\chi/\partial t)_h<0$ to a lower-temperature (critical) regime within which $(\partial\chi/\partial t)_h>0$. The latter can be seen to follow directly from the scaling law [Eq. (1)] which yields (neglecting numerical and spin-dependent factors)

$$(\partial\chi/\partial t)_h \sim -t^{-(\gamma+1)}(1-h^2/t^{2\gamma+2\beta} + h^4/t^{4\gamma+4\beta}+\cdots) . \quad (9)$$

If the expression is dominated by its leading terms, the condition that $(\partial\chi/\partial t)_h$ be positive requires $h > t^{\gamma+\beta}$— as indeed would be inferred more generally from Eq. (7).

Here the physical origin of this behavior is identified and its consequences discussed.

Within this static approach, the peak structure arises as a result of a competition between level splitting induced by an internal field ($g\mu_B H_i$) and the thermal energy $\kappa_B T$, as the following argument shows. The fluctuation-dissipation theorem, in its simplest form, relates the differential susceptibility to fluctuations in the magnetization via[7]

$$\chi(H,T) \sim \frac{1}{T}(\langle S_z^2\rangle - \langle S_z\rangle^2) . \quad (10)$$

In any finite field H, if the temperature is sufficiently large, $\langle S_z\rangle \rightarrow 0$ and $\langle S_z^2\rangle \rightarrow S(S+1)/3$ and $\chi(H,T)$ will display the familiar Curie-Weiss dependence with the susceptibility decreasing with increasing temperature. This is the high-temperature regime referred to above in which $(\partial\chi/\partial t)_h < 0$; in fact, $\chi(H,T) \rightarrow 0$ as $1/T$ (for $T \gg 1$). In the opposite limit, if one approaches sufficiently close to the ordering temperature T_c from above in *nonzero* field, the magnetization is driven towards saturation, an effect which becomes more pronounced as the temperature is further reduced. Under these conditions the fluctuations in the magnetization decrease as the temperature is lowered leading, via Eq. (10), to a decrease in the differential susceptibility. Of course $\chi(H,T)$ is not zero at T_c as Eq. (3) demonstrates, but the above argument shows that it must certainly decrease with decreasing temperature as $t \rightarrow 0$ from above in a finite field.

The peak structure discussed previously thus represents the maximum that must accompany this limiting behavior as $t \rightarrow 0_+$ and $t \rightarrow \infty$. The result that such peaks move upward in temperature as the field H_i increases is also consistent with the suggestion that the peaks manifest a competition between level splitting and thermal energies.

If these arguments based on the static scaling law are correct, then as a corollary similar effects should occur in simple *paramagnetic* systems. The spins in such systems do not formally experience any interactions, and, hence, do not exhibit a cooperative phase transition at finite temperature, and while this leads to differences in the detailed behavior of the two types of systems (consider below albeit briefly) there are, nevertheless, striking similarities. Certainly such systems again display a Curie-law susceptibility in any finite field H at sufficiently high temperature so that $(\partial\chi/\partial t)_h < 0$ as $\chi(H,T) \rightarrow 0$ as $1/T$ for T very large. One difference in paramagnetic systems is that the magnetization is driven towards saturation as $T \rightarrow 0$ (rather than as $T \rightarrow T_c$ as in a ferromagnet) so that $(\partial\chi/\partial t)_h$ becomes positive as $T \rightarrow 0$ in them. Furthermore, with $\langle S_z^2\rangle$ of Eq. (10) replaced by the more general $\sum_i\sum_j\langle S_i^z S_j^z\rangle$, it is apparent that the long-range correlations that are associated with the cooperative ferromagnetic transition must substantially alter this term from the form appropriate for an uncorrelated free-spin paramagnet. This influence is greatest in the vicinity of the transition where the correlation length becomes very large, and this leads to a different susceptibility exponent in the vicinity of T_{c+} and $T=0$, respectively. However, the peak structure that must again result from this high- and low-temperature limiting behavior should convey similar information to that obtained from a ferromagnet. The validity of these arguments based on the static approach can be examined in more detail by performing numerical calculations of the differential susceptibility of paramagnetic systems in which the magnetization M is given by the Brillouin function $B_s(H,T)$, with spin per site S and internal field H_i identically equal to the uniform applied field H (so that there is no interaction between spins). It should be stressed that this simple Brillouin function model contains no dynamic, superparamagnetic, or blocking effects, so that any anomaly that arises must be static in origin.

While the zero-field susceptibility of a paramagnet is well known to diverge at $T \rightarrow 0$ as

$$\chi(0,t) \propto t^{-\gamma} \text{ with } \gamma = 1 \text{ and } t = T \text{ (when } T_c = 0)$$

less well-known features of the magnetic response, suggested above on qualitative grounds, and which are admittedly obvious when thought about, are reproduced in Fig. 1. This figure shows that the field-dependent susceptibility $\chi(H,T)$ does indeed display the expected peak, the amplitude of which decreases but the position in temperature

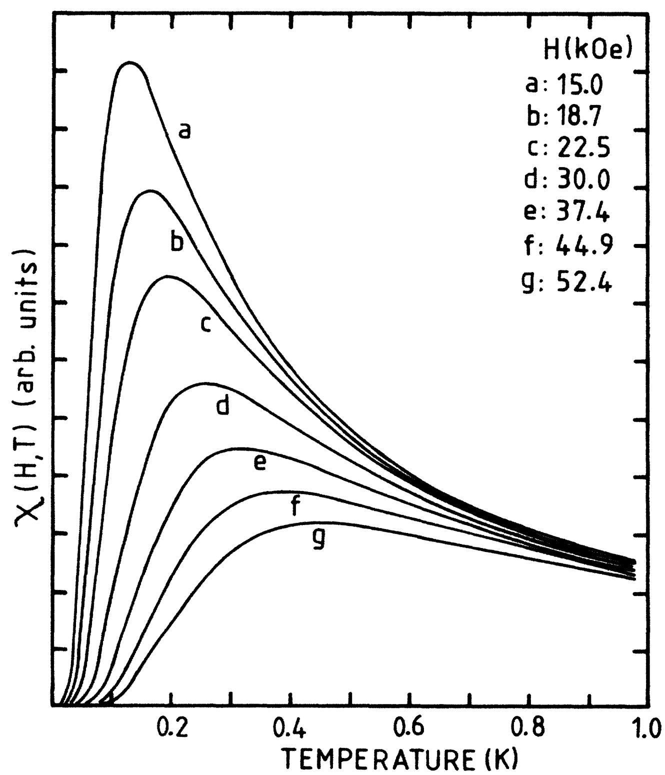

FIG. 1. The field-dependent susceptibility $\chi(H,T)$ (in arbitrary units) for a paramagnetic system with spin per site $S=\frac{1}{2}$ plotted against temperature (in K) for various applied fields (in kOe).

of which increases with field in a manner similar to that predicted and observed in ferromagnets. (It might also be argued that the correlations that build up near T_c in a ferromagnet result in a mean "molecular" field that bootstraps the response into a regime which is reached only in large external fields for a noninteracting paramagnetic system.) The response of paramagnetic systems is well known to be a universal function of H/T, from which it follows that the appropriate form of scaling function can be obtained from Eq. (1) simply by setting $\gamma=1$ and $\beta=0$ (as confirmed below), when

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

Here $h/t=H/T$, so that the argument of the scaling function, now the Brillouin function, becomes the nonlinear scaling field proposed recently by Fahnle and Souletie,[11] i.e., $(H/T)[T/(T-T_c)]\rightarrow H/T$ when $T_c=0$. Moreover, in contrast to the narrow critical regime of ferromagnetic systems, this scaling function description of the response of a free spin paramagnet is valid over the entire (h,t) plane, a result that can also be deduced from a Kadanoff-type block-spin argument.

While the zero-field susceptibility obtained from Eq. (11) directly confirms $\gamma=1$, the remaining conventional predictions of static scaling theory, the asymptotic dependences summarized by Eqs. (2) and (3), cannot be investigated for a paramagnet. A spontaneous magnetization does not appear in such a system, while along the critical isotherm, were it to be accessible experimentally (it is now at $T=0$ rather than at $T=T_c$), the magnetization is completely saturated for all $H\neq 0$ unlike a ferromagnet. In contrast, the features corresponding to the predictions of Eqs. (7) and (8) are evident at nonzero temperature, as shown in Fig. 1. While the representation of these data by a unique function of h/t is obvious (see Fig. 4), the individual power-law dependences of T_m on H and of $\chi(H,T_m)$ on H prescribed by these equations are not. Figure 2 displays a double logarithmic plot of the susceptibility peak temperature T_m from Fig. 1 against field H, for various spin values per site. The straight-line behavior evident in this figure not only confirms this power-law dependence, but also, from its slope, yields

$$\gamma+\beta=1 \ .$$

With $\gamma=1$, then β must be zero, as suggested previously; the latter cannot be tested using conventional methods [Eq. (2)], it can only be obtained from the variation of these *finite* temperature peak positions with field. Figure 3, in which the susceptibility at the peak $\chi(H,T_m)$ is plotted on a double logarithmic scale against the applied field H, verifies the prediction of Eq. (8), and from its slope one obtains

$$\delta=\infty \ .$$

The expectation that the magnetization of a paramagnet (with no formal interactions) is saturated along the critical isotherm is thus confirmed [with $\delta=\infty$ Eq. (3) indicates that the magnetization m is field independent for all $H\neq 0$] although this critical isotherm is not accessible experimentally. In contrast, the susceptibility *peaks* occur above $T=0$ at a nonzero temperature in finite field.

The values deduced from the various exponents

$$\gamma=1, \ \beta=0, \ \delta=\infty \ ,$$

while enabling the Widom equality to be balanced, obvi-

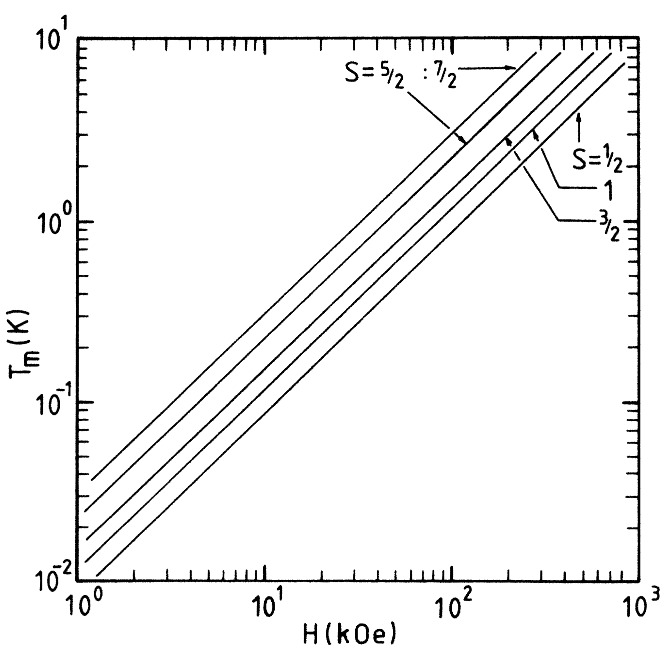

FIG. 2. The temperature of the susceptibility peak T_m (in K) plotted against the applied field H (in kOe) on a double logarithmic scale for various spin per site values. The slope of each line is +1, so that $\gamma+\beta=1$.

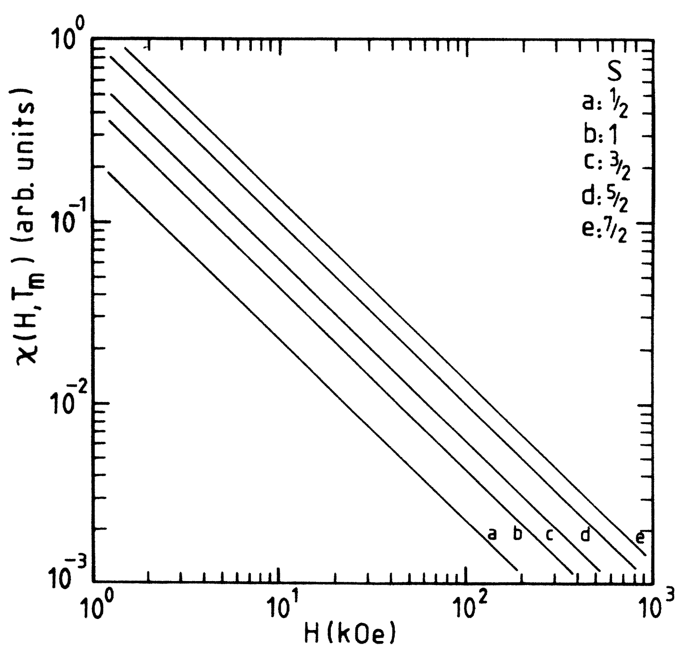

FIG. 3. The peak susceptibility $\chi(H,T_m)$ (in arbitrary units) plotted on a double logarithmic scale against the applied field H (in kOe) for various spin-per-site values. The slope of each line $(1/\delta)-1$ is -1, which means that $\delta=\infty$.

ously indicate that the paramagnet and ferromagnet are not in the same universality class.

In conventional scaling theory the form of the scaling function, F or G previously, cannot be specified for arbitrary values of its argument. However, its form can be determined experimentally from field-dependent susceptibility data. From Eq. (6)

$$\frac{\chi(h,t)}{\chi(h,t_m)} = [G(h/t^{\gamma+\beta})/G(h/t_m^{\gamma+\beta})] \rightarrow G(h/t^{\gamma+\beta}) \ , \quad (12)$$

since Eq. (7) indicates that the denominator is a constant. Thus, a plot of the susceptibility (measured in a number of fixed fields as a function of temperature and normalized to its peak value) against $h/t^{\gamma+\beta}$ should produce a universal curve, the form of which specifies that of the scaling function G for arbitrary values of its argument (the latter is simply the ratio H/T in a paramagnet, but is more complicated in a ferromagnet).

The predictions of Eq. (12) are very simple to demonstrate for a paramagnet, for which the scaling function takes an obvious and simplified form, the derivative of the Brillouin function, as shown in Fig. 4. The reason for including this figure is not to demonstrate the expected result that these susceptibility data (for a given spin value) can be represented by a unique function of H/T, but rather to show that the sharpness of the peak structure in these data increases with increasing spin per site value (S). A complimentary effect appears in the Schottky peak widths measured in the specific heat of paramagnets (in which the level splitting is induced by either an applied field or through crystal-field effects), an effect which is more widely documented. This result may be inferred directly from the spin dependence of the Brillouin function (a point that does not seem to have been made generally for the susceptibility); the corresponding behavior has been observed only recently in the magnetic response of ferromagnets,[12] although it origin was not identified.

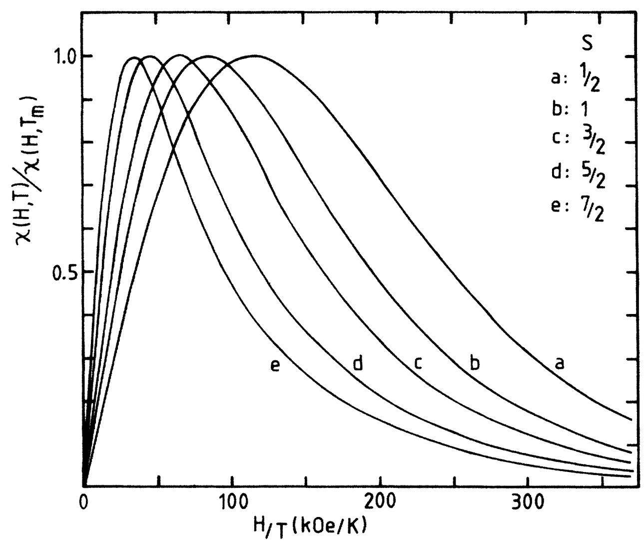

FIG. 4. The complete field and temperature-dependent susceptibility (normalized to its peak value in each case) plotted against the ratio H/T (in kOe/K), for various spin-per-site values. The $S=\frac{1}{2}$ curve represents a superposition of *all* the data from Fig. 1, for example, demonstrating the universal behavior expected for a paramagnet with a given spin value. This sharpening of the peak structure as S increases is clearly evident.

In summary, the physical origin of a peak structure observed in the field-dependent susceptibility of ferromagnets at temperatures above the critical temperature T_c is identified. As a consequence we have shown that paramagnets should exhibit similar structures in their field-dependent susceptibility which could be used to extract information on the zero-temperature phase transition that occurs in spin systems which are formally noninteracting. This information would normally not be experimentally accessible. While these features can be obtained directly from existing standard treatments of paramagnetic systems, they do not generally appear to be well known. These results provide strong confirmatory evidence that such anomalies in ferromagnets are static, not dynamic, in origin. They also indicate that the field-dependent susceptibility can be used to establish the general form of the scaling function.

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