Specific Heat of Random-Field Ising Systems

A recent Letter by Birgeneau *et al.* [1] reports an interesting comparison of *direct* and *indirect* specific-heat (DSH and ISH, respectively) data of the dilute antiferromagnet $Fe_{0.5}Zn_{0.5}F_2$ in applied fields (H), which is a realization of the 3D random-field Ising model (RFIM). The authors raised issues similar to the ones I raised in 1986 [2], but they overlooked some important implications in Ref. [2]. They also made an incorrect reference to the DSH data on $Fe_{0.682}Mg_{0.318}Cl_2$ and $Fe_{0.714}Mg_{0.286}Cl_2$ in Ref. [2]. I offer some clarifications here.

Ref. [1] determines the ISH by $d(M/H)/dT$, where M is the uniform magnetization. The authors suggest that it differs from DSH by a term of the form $Dt^{2\beta-1}$, where $D = D_0 + D_1H^2$, and attributed it to a prediction by Fishman and Aharony (FA) [3]. The result showed $D_0 = 0$. The authors then suggested that the same holds for other ISH techniques such as birefringence. I believe this line of reasoning is flawed because the effect predicted by FA applies only to the susceptibility $\chi = dM/dH$ in zero field, *not* M/H in finite fields. FA actually expected it to "smear away for finite fields" (see Ref. [3]). Strictly speaking, M/H is related to the Zeeman energy $E_H = -HM = -H\int_0^H \chi(T,h)\,dh$. Expanding χ for small h gives a zeroth-order term $\chi(T,0)$ which contains the effect predicted by FA, but this corresponds to the D_0-term that the authors found to be negligible. The next term in the χ-expansion, $O(H^2)$ or nonanalytic in H, is what they actually observed. As noted in Ref. [2], the T- and H-dependence of this term is unclear, but it may be approximated by $t^{2\beta}$. More importantly, Ref. [2] emphasized that other ISH techniques such as birefringence ($d\Delta n/dT$) probe the *exchange energy* E_J and, unlike χ, they *are* valid probes of the specific-heat for dilute antiferromagnets in zero field. They are questionable for finite fields because DSH measures the *total* energy $E_{\text{tot}} = E_J + E_H$. Since the DSH data in Ref. [2] (see Fig. 1) did not show the sharp peaks seen in $d\Delta n/dT$, it was conjectured that E_J and E_H contain counteracting singularities that do not appear in E_{tot}. A natural cause of this is the *cluster-flip* effect I discussed in Ref. [4] which increases E_J and decreases E_H by exactly the same amount. The resemblance between the $d(M/H)/dT$ and $d\Delta n/dT$ data in Ref. [1], which correspond to dE_H/dT and dE_J/dT, respectively, provide compelling evidence for this conjecture.

One of the main results in Ref. [1] is the absence of hysteresis between the field-cooled (FC) and zero-field-cooled (ZFC) DSH data. This contradicts Dow and Belanger's (DB) study of $Fe_{0.46}Zn_{0.54}F_2$ [5]. In this context, Birgeneau *et al.* [1] stated that no hysteresis was seen in my data. This is incorrect: a small hysteresis between heating and cooling the samples in constant fields (FH and FC, respectively) was observed in $Fe_{0.682}Mg_{0.318}Cl_2$, but not in $Fe_{0.714}Mg_{0.286}Cl_2$ [2]. This is consistent with my neutron scattering data [4] which show stronger hystereses in $Fe_{1-x}Mg_xCl_2$ than in $Fe_{1-x}Co_xCl_2$ for similar fields and concentrations. Figure 1 shows that the difference is less than 2% and only noticeable within $\pm 1\%$ of T_c. Such small effects may have been missed in Ref. [1] because of sample inhomogeneity and the small temperature cycling required in the heat-pulse method. However, the hysteresis in the ISH data is far too large to have been missed *entirely*, so the conclusion that ISH data contain an extra H-dependent term is sound.

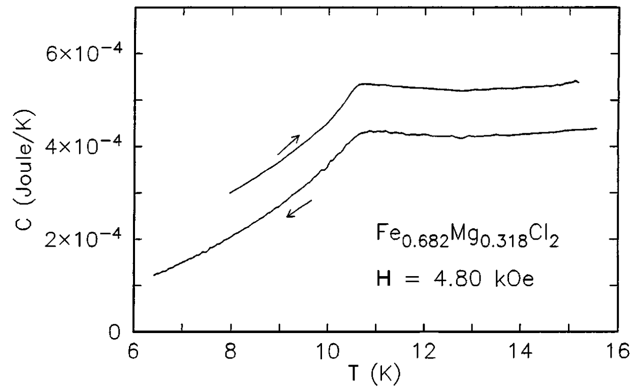

FIG. 1. The field-heating and field-cooling specific-heat data from Ref. [2] show a small hysteresis very close to the transition when the field is large enough to reduce T_c by 18%. The heating curve is shifted upward for display clarity.

An important point not mentioned by Birgeneau *et al.* is that their DSH data show a small symmetric peak at 1.5 T and it disappears at 5.5 T. DB's data on $Fe_xZn_{1-x}F_2$ [5] were limited to low fields, and the symmetric peak was interpreted as a new RFIM singularity with $\alpha \approx 0$ and a unity amplitude ratio. In the chloride systems I studied, the amplitude ratio of the peak changes continuously with increasing H, a fact which I attributed to crossover behavior [6]. The new data in Ref. [1], while they may have missed subtle hysteresis effects, *confirmed* this behavior in fluoride samples and contradict DB's claim of observing a new RFIM critical behavior.

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