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Spin-Peierls Transition in CuGeO_3: Critical, Tricritical, or Mean Field?

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Spin-Peierls Transition in $CuGeO_3$: Critical, Tricritical or Mean Field?

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The spin-Peierls phase transition in $CuGeO_3$ has been extensively studied utilizing a variety of experimental techniques. Interpretations of the phase transition behavior vary from tricritical to mean field to Ising critical to XY critical. We show that the behavior in the vicinity of the phase transition of each of the order parameter, the magnetic energy gap and the heat capacity can be quantitatively fitted with few adjustable parameters with a mean field model incorporating a tricritical to mean field critical crossover in the transition region.

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I. INTRODUCTION

The spin-Peierls transition corresponds to the dimerization of a one-dimensional S = $\frac{1}{2}$ antiferromagnetic chain coupled to a three dimensional elastic medium [1]-[6]. Until relatively recently, spin-Peierls transitions had only been observed in organic charge transfer compounds such as copper bisdithiolene (TTF-CuBDT) [5]- [8]. Experimental information obtainable in such systems has been limited both by the size of available single crystals and by the sensitivity of these materials to damage by x-rays or electrons. Nevertheless, some important information on the spin-Peierls phase transition has been obtained in a number of different organic materials. Interestingly, in most, if not all cases, the data are consistent with a simple BCS-type mean field transition [5]- [8].

Much more complete experimental work on the spin-Peierls transition has been made possible by the discovery that a structurally simple, inorganic chain compound copper germanate ($CuGeO_3$) undergoes a spin-Peierls transition at a transition temperature around 14K [9]. The crystal structure of $CuGeO_3$ is orthorhombic, space group *Pbmm*, with a unit cell of dimensions $a = 4.81$ Å, $b = 8.47$ Å and $c = 2.94$ Å at room temperature [10]. The Cu^{2+} ion carries a spin $S = \frac{1}{2}$ and forms a (CuO_2) chain with the neighboring Cu^{2+} ions along the c-axis direction. The successive Cu^{2+} S = $\frac{1}{2}$ spins are antiferromagnetically coupled through the superexchange interactions via the bridging oxygen atoms. Below the spin-Peierls transition temperature, T_{SP}, the dimerization of Cu-Cu pairs along the c-axis direction, accompanied by shifts of the bridging oxygen atoms in the ab plane, gives rise to superlattice reflections at the ($\frac{h}{2}$, k, $\frac{l}{2}$) (h, l: odd and k: integer) reciprocal-lattice positions [11]. These have been observed in electron diffraction [12], x-ray [13], and elastic neutron scattering [11] experiments. Using coarse resolution x-ray diffraction techniques, Pouget *et al.* [13] have measured the pretransitional thermal lattice fluctuations whose correlation lengths diverge anisotropically with decreasing temperature in a manner consistent with mean field theory. These same fluctuations have been studied at high resolution using synchrotron x-ray diffraction techniques by Harris *et al.* [14]. These latter authors observe within about 1K of T_{SP} large length scale fluctuations with characteristic length scales about an order of magnitude longer than those characterizing the bulk critical fluctuations.

In spite of this large amount of work, it is still not agreed whether the observed transition behavior reflects mean field or critical behavior. Extant models include: a) tricritical to 3D Ising crossover behavior [14,15]; b) mean field behavior [15]; c) 3D XY with corrections to scaling [16], and, most exotically, d) a 2D XY to 3D XY crossover as T_{SP} is approached [17]. Harris *et al.* [14] first argued that because of the one-component nature of the dimerization order parameter for a spin-Peierls phase transition, asymptotically the transition must be in the 3D Ising universality class. They argued further, that because of the coupling to the elastic strains, the precritical behavior should be tricritical-like. Similar conclusions, albeit based on different physical reasoning, were arrived at later by Werner and Gros [15]. Proponents of 3D XY behavior typically argue that the copper and oxygen displacements are independent thence yielding a two-component order parameter [18]. Implicitly, Harris *et al.* [14] assume that all of the atomic displacements accompanying the spin-Peierls transition are linearly coupled thence reducing the system to a one-component order parameter. The 3D critical behavior models seem to be supported by measurements of the order parameter [14,16,17] which for reduced temperatures $\sim 2 \times 10^{-3} < 1 - T/T_{SP} \lesssim 5 \times 10^{-2}$ exhibits power law behavior $(1 - T/T_{SP})^{\beta}$ with $\beta = 0.33 \pm 0.02$, in good agreement with both 3D Ising and XY values of $\beta = 0.325$ and 0.345 respectively [19]. The heat capacity data are equally well described by a 3D critical behavior model (Ising or XY) and by a mean field model with Gaussian fluctuations [20].

In this paper we present an alternative model for $CuGeO_3$, namely a Landau-Ginzburg model incorporating a tricritical to mean field crossover. As we shall show, this model describes all available data very well with few adjustable parameters. The format of this paper is as follows. In Section II we introduce the model including its genesis in studies of critical phenomena in thermotropic liquid crystals systems. Section III presents an analysis of the available data for $CuGeO_3$ using this model. Finally, in Section IV we give a summary, our conclusions and suggestions for future experiments.

II. THE MODEL

The conundrum described above for $CuGeO_3$ is reminiscent of a similar divergence of views which occurred in the interpretation of experiments on smectic A - smectic C phase transitions in thermotropic liquid crystal systems [21]- [23]. In particular, in that case, measurements of the tilt order parameter [21,23] typically reveal power law behavior $\phi \sim (1 - T/T_{AC})^\beta$ over the temperature range $5 \times 10^{-5} < (1 - T/T_{AC}) < 5 \times 10^{-3}$ with $\beta = 0.36 \pm 0.02$. However, this divergence of views was resolved by Huang and Viner [22] and Birgeneau *et al.* [23] who showed that all of the data including the heat capacity, order parameter, and tilt susceptibility, were consistent with the predictions of a simple Landau model with an anomalously large 6th order term. Clearly, it is of interest to carry out a similar analysis for the available data for the spin-Peierls transition in $CuGeO_3$.

For the Landau-Ginzburg model the free energy is given by

$$F = a\tau\phi^2 + b\phi^4 + c\phi^6.... + \frac{1}{2m_\alpha}|\nabla_\alpha \phi|^2 \qquad (1)$$

where $\tau = T/T_c - 1$.

With $\tau_0 = b^2/ac$, standard calculations yield for the order parameter, ϕ, the specific heat, C, the susceptibility, χ, and the correlation length, ξ_α,:

$$\phi = (b/3c)^{1/2}[(1 - 3\tau/\tau_0)^{1/2} - 1]^{1/2} \qquad \tau < 0 \qquad (2)$$

$$C = \begin{cases} 0 & \tau > 0 \\ (a^2T/2bT_c^2)(1 - 3\tau/\tau_0)^{-1/2} & \tau < 0 \end{cases} \qquad (3)$$

$$\chi = 1/2a\tau \qquad \tau > 0 \qquad (4)$$

$$\xi_\alpha = (2am_\alpha\tau)^{-1/2} \qquad \tau > 0 \qquad (5)$$

with similar expressions for $\tau < 0$ for χ and ξ. Eq. (2) and (3) are conveniently rewritten in the form

$$\phi = \phi_0 \left[\left(1 + 3\frac{T_{SP}-T}{T_{SP}-T_{CR}}\right)^{1/2} - 1\right]^{1/2} \qquad \tau < 0 \qquad (6)$$

$$C = \begin{cases} 0 & \tau > 0 \\ C\_T\left(1 + 3\frac{T_{SP}-T}{T_{SP}-T_{CR}}\right)^{-1/2} & \tau < 0 \end{cases} \qquad (7)$$

where T_{CR} is the crossover temperature from tricritical to mean field behavior. We note that in the above expressions the exponents are fixed and only the amplitudes and the two temperatures, T_{SP} and T_{CR}, are variable. A log-log plot of Eq. (6) reveals that for the order parameter ϕ the effective exponent β crosses over gradually from $\frac{1}{4}$ to $\frac{1}{2}$ as T varies from less than to greater than T_{CR}. In the smectic A - smectic C case the measurements span T_{CR} and accordingly intermediate exponents, $\beta \simeq 0.36$, are found even though the actual transition is mean-field-like for temperatures in the immediate vicinity of T_{AC} [23].

III. ANALYSIS

We now apply this tricritical–mean field crossover model to $CuGeO_3$. The first test is T_{SP} itself or, more precisely, the ratio of the spin gap, Δ, to T_{SP}. In the mean field theory of Pytte [24], the spin-Peierls transition is BCS-like so that in the weak coupling limit $2\Delta/T_{SP} = 3.5$. In the charge transfer salts TTF - CuBDT [4,6], TTF - AuBDT [5], MEM - $(TCNQ)_2$ [7], and SBTTF - $TCNQCl_2$ [8] this ratio is found to be 3.5, 3.7, 3.1 and ≤ 3.5 respectively, in good agreement with the BCS value. Critical fluctuations, either Ising or XY in character, would act to increase this ratio. For $CuGeO_3$, $\Delta = 24.5$K and $T_{SP} \simeq 14$K implying $2\Delta/T_{SP} = 3.5$, consistent with a BCS mean field theory description [9,25]. At the minimum, this value for $2\Delta/T_{SP}$ argues against any quantitatively important effect of critical fluctuations on T_{SP} in $CuGeO_3$.

The behavior of the order parameter in $CuGeO_3$ is of particular importance since this observable appears to provide the strongest evidence for true critical rather than mean field or tricritical behavior. A number of groups have reported measurements of the temperature dependence of the order parameter in $CuGeO_3$ [14,16,17]. The measured phase transition temperature T_{SP} varies between 13.3K and 14.6K in different samples. Nevertheless, near-universal behavior is observed for the order parameter provided that it is plotted as a function of the reduced temperature T/T_{SP}. As noted above, fits of the order parameter $\phi(T/T_{SP})$ for $1 - T/T_{SP} < 0.05$ to a single power law $\phi \sim (1 - T/T_{SP})^\beta$ all yield values of $\beta = 0.33 \pm 0.02$. As discussed by Gaulin and co-workers [16], inclusion of a correction-to-scaling multiplicative factor $(1 + B|\tau|^\delta)$ in the expression for ϕ both improves the goodness of fit and, not surprisingly, extends the range of validity of the fit.

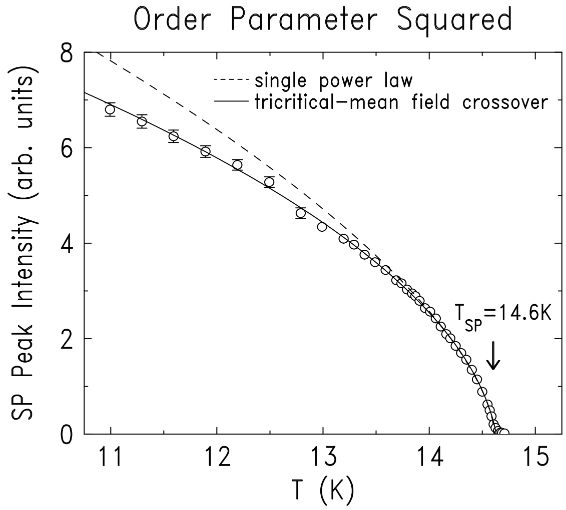

FIG. 1. (3/2, 1, 3/2) superlattice peak intensity measured with synchrotron x-ray diffraction techniques. The peak intensity is proportional to the order parameter squared, ϕ^2. The dashed line is the result of a fit of the data for $\tau = 1 - \frac{T}{T_{SP}} < 0.04$ to a single power law $\phi^2 \sim \phi_0^2(1 - \frac{T}{T_{SP}})^{2\beta}$ with $\beta = 0.314 \pm 0.01$. The solid line is the result of a fit to Eq. (6) with $\tau_{CR} = 0.006$.

We show in Fig. 1 our own measurements of the order parameter squared in a sample of $CuGeO_3$ with $T_{SP} = 14.6$K. These data are consistent with those measured by both ourselves and other groups in a variety of samples [14,16,17]. Fits to a single power law for $\tau < 0.04$ yield $\beta = 0.314 \pm 0.01$. However, as noted by Harris *et al.* [14] and as may be seen in Fig. 1, the data fall significantly below the power law curve for $\tau > 0.04$. We show, in addition, in Fig. 1 the results of a fit to the tricritical to mean field crossover form, Eq. (6). This fit has only three adjustable parameters, ϕ_0^2, T_{CR} and T_{SP}. This is the same number of parameters as those in the single power law fits discussed above and two less than the number of adjustable parameters in fits to a power law with corrections-to-scaling with both B and δ varied. (We note that Lumsden *et al.* [16] fix $\delta = 1/2$ whereas Lorenzo *et al.* allow δ to vary; the latter group find an optimum fit for $\delta \simeq 1$). It is evident that Eq. (6) describes the order parameter data extremely well over the complete range of temperatures. The fit yields $\tau_{CR} = 1 - T_{CR}/T_{SP} = 0.006 \pm 0.001$ implying that the crossover from tricritical to mean field behavior occurs at a quite small reduced temperature.

We now discuss the energy gap Δ. Using a simple scaling ansatz, Cross and Fisher [3] argue that $\Delta \sim \phi^{2/3}$. We show in Fig. 2 the data of Lorenzo *et al.* [17] for the magnetic energy gap for $T < T_{SP}$ in a sample of $CuGeO_3$ with $T_{SP} = 14.4$K. In part because of the apparent jump of $\Delta(T)$ at T_{SP}, Lorenzo *et al.* [17] interpret these data as indicating a 2D XY Kosterlitz-Thouless transition [26]. In fact, these data are readily explained using the model of Cross and Fisher [3] together with the tricritical-mean field crossover form for ϕ, Eq. (6). In this case we hold T_{SP} fixed at $T_{SP} = 14.4$K and set $\tau_{CR} = 0.006$ as determined above so that there is only one adjustable parameter, the overall amplitude $\Delta(0)$. The result so-obtained is shown in Fig. 2. It is evident that the tricritical-mean field model with $\Delta(T) \sim \phi^{2/3}$ describes the measured gap energy $\Delta(T)$ extremely well over a wide range of temperatures with only one adjustable parameter. Indeed, this is by far the best test to-date of the Cross-Fisher model. We should note that this model cannot explain the inferred pseudogap above T_{SP} [17]. However, the "pseudogap" is deduced using a heuristic line-shape analysis which lacks a firm theoretical basis.

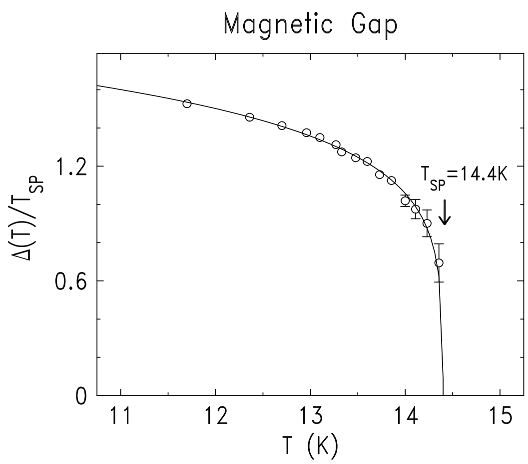

FIG. 2. Magnetic energy gap in $CuGeO_3$. These data are from Ref. 17. The solid line is the result of a fit to the form $\Delta(T) = \Delta(0)\phi^{2/3}$ where ϕ is given by Eq. (6) with τ_{CR} held fixed at 0.006.

The specific heat in $CuGeO_3$ has proven to be the most difficult thermodynamic quantity to interpret unambiguously [20]. This is, in part, because of the extreme sensitivity of the specific heat near T_{SP} to sample inhomogeneities and, in part, because of the inevitable large number of adjustable parameters required to describe the critical specific heat in any physically relevant model. Fig. 3 shows high resolution magnetic specific heat (C_M) data for a sample of $CuGeO_3$ with $T_{SP} = 14.24$K from Lasjaunias and coworkers [20]. Hegman *et al.* [20] have carried out an extensive analysis of these data using both a mean field "BCS plus Gaussian fluctuation" model and a critical behavior model. They find that both models describe C_M quite well in the immediate vicinity of T_{SP}, albeit at the cost of a rather large number of adjustable parameters. The critical behavior model fits give a value for the specific heat exponent, α, near 0. On the other hand, the Gaussian fluctuation analysis implies that the true critical behavior is confined to the region $|\tau| < 0.0006$.

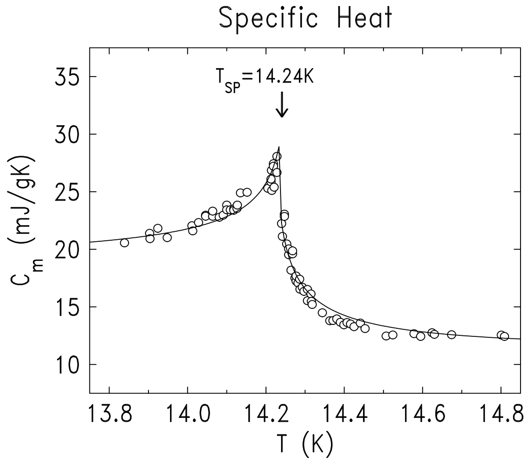

FIG. 3. Magnetic specific heat in $CuGeO_3$. These data are from Ref. 20. The solid line is the result of a fit to Eq. (8) with τ_{CR} held fixed at 0.006.

Given the uncertainties connected with the fits described above, the best one can hope for is to determine whether or not the tricritical-mean field crossover model is consistent with the experimental results for C_M shown in Fig. 3. First, it is evident that Eq. (7) will be inadequate since one must, at the minimum, include Gaussian fluctuations above T_{SP}. We therefore include the fluctuations above T_{SP} in the simplest way possible by replacing Eq. (7) by

$$C_M = \begin{cases} C_M^+ T \left(1 + 3\frac{T-T_{SP}}{T_{SP}-T_{CR}}\right)^{-1/2} + \gamma T & \tau > 0 \\ C_M^- T \left(1 + 3\frac{T_{SP}-T}{T_{SP}-T_{CR}}\right)^{-1/2} + B_- & \tau < 0 \end{cases} \tag{8}$$

where γT is the regular linear term for a 1D Heisenberg antiferromagnet and B_- is the background term below T_{SP}. The background B_- should, in general, be temperature dependent; however, given the narrow range of temperatures we consider, a constant background is adequate. Eq. (8) is closely similar to the BCS plus Gaussian fluctuation model considered by Hegman *et al.* [20] since the Gaussian fluctuations give rise to a $|\tau|^{-1/2}$ contribution to C_M both above and below T_{SP}. The solid lines in Fig. 3 correspond to fits to Eq. (8) with τ_{CR} fixed at 0.006 and C_M^+, C_M^-,γ, B_- and T_{SP} varied. Clearly Eq. (8) describes C_M quite well; indeed the fit appears to be better than those for either of the models tested by Hegman *et al.* [20]. The fit shown in Fig. 3 gives $C_M^+/C_M^- = 1.1 \pm 0.13$; this ratio is expected to be non-universal so it cannot be simply interpreted. We conclude, therefore, that the tricritical-mean field crossover model describes C_M well although not uniquely so.

Finally, we discuss the correlation length and the staggered susceptibility. Pouget *et al.* [13] have found that the correlation length over a wide temperature range follows the behavior $\xi \sim (T/T_{SP} - 1)^{-1/2}$, consistent with mean field theory; however, the number of data points in their experiment near T_{SP} is sufficiently small that their results do not meaningfully differentiate between various theoretical models. Harris *et al.* [14] have reported a high resolution synchrotron x-ray study of the critical fluctuations above T_{SP} in $CuGeO_3$. They find pretransitional lattice fluctuations within 1K above T_{SP} whose length scale is about an order of magnitude longer than those characterizing the bulk thermal fluctuations. The lineshape of the large length scale fluctuations is consistent with a Lorentzian-squared form. The measured critical exponents are $\nu = 0.56 \pm 0.09$ and $\bar{\gamma} = 2.0 \pm 0.3$ where $\bar{\gamma}$ is the exponent characterizing the divergence of the disconnected staggered susceptibility [27]. The mean field predictions for these exponents are $\nu = 1/2$ and $\bar{\gamma} = 2\gamma = 2$ whereas for 3D Ising (XY) critical behavior one expects $\nu = 0.63$ (0.67) and $\bar{\gamma} = 2.5(2.64)$. Thus the Harris *et al.* [14] data favor the tricritical-mean field model but 3D Ising or XY critical models are not excluded. Precise measurements of the bulk staggered susceptibility using neutrons should yield accurate values for ν and $\bar{\gamma}$ and this, in turn, would definitively choose between the models.

IV. DISCUSSION

In summary, each of the order parameter, magnetic energy gap, specific heat, correlation length and disconnected staggered susceptibility are well-described by a simple Landau-Ginzburg model exhibiting a tricritical-mean field crossover near T_{SP}. Further, the ratio of the energy gap to T_{SP} is consistent with the value for a BCS mean-field transition. We conclude, therefore, that $CuGeO_3$, in common with the organic change transfer salts, exhibits a mean field spin-Peierls transition for reduced temperatures $|\tau| > 0.001$.

The principal remaining issue is the microscopic origin of the tricritical behavior. Harris *et al.* [14] argue that this is caused by a diminution in the effective fourth order term in Eq. (1), $b\phi^4$, because of coupling to the macroscopic strain. It also seems possible that competing nearest and next-nearest neighbor exchange interactions along the chain could generate the tricritical instability [15,28]. Specifically, Castilla *et al.* [28] argue that the ratio of the next nearest neighbor to nearest neighbor exchange interaction along the chain is close to the critical value for spontaneous formation of a magnetic gap independent of coupling to the lattice. Heuristically, it seems that this could generate tricritical behavior in the phase diagram. Another possible source of tricritical behavior is competition between the Néel state and the spin-Peierls state, that is, competition between the coupling of the $S = 1/2$ chain to the lattice and the interchain exchange coupling. Clearly, a multidimensional theoretical analysis of the spin-Peierls phase diagram including magnetostriction, competing intrachain exchange interactions together with the interchain magnetic and elastic coupling is required.

Of course, the mean field behavior itself in all of these spin-Peierls systems is not yet well understood. In TTF-CuBDT there is evidence for a soft phonon at very high temperatures [6] and Cross and Fisher [3] speculate that the precursive soft mode accounts for the large length scale underlying the mean field behavior. In $CuGeO_3$, no soft phonon at all has yet been seen. Thus, the microscopic origin of the large length scale in $CuGeO_3$ remains to be elucidated.

Finally, it would be very interesting to see if the putative nearby tricritical point could be accessed by changing some variable such as pressure, uniaxial stress or doping. Masuda *et al.* [29] have shown that replacement of Cu by Mg both depresses T_{SP} and appears to drive the spin-Peierls transition first order. The concomitant tricritical point could well account for the observed tricritical-mean field crossover in pure $CuGeO_3$. We note, however, that the actual physics of magnetic dilution in $CuGeO_3$ is quite complex since dilution introduces frustration of the interchain elastic interaction [30]. Replacement of Cu^{2+} by Cd^{2+} (Ref. [31]) or Ge^{4+} by Ga^{4+} (ref. [32]) both lead to mean field behavior over quite wide temperature ranges; that is, doping with these ions moves $CuGeO_3$ away from the tricritical point into the pure mean field regime. Again, further research, both experimental and theoretical, is required to elucidate these effects more completely.

V. ACKNOWLEDGEMENTS

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