

## Effect of magnetic fields on a spin-Peierls transition

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The effect of a magnetic field on the transition temperature of a spin-Peierls transition and the wave vector of the lattice distortion is calculated using the theory based on the Luther-Peschel-type treatment of the spin-correlation functions. Comparison with experiment offers the prospect of testing the predictions of this theory. A brief discussion of the nature of the commensurate to incommensurate transition is included.

### I. INTRODUCTION

Recently, Cross and Fisher<sup>1</sup> developed a new theory of the spin-Peierls transition using a Luther-Peschel<sup>2</sup>-type treatment of the spin Hamiltonian that should be better than the "Hartree" treatments of previous works.<sup>3-5</sup> We also showed that the mean-field nature of the transition observed<sup>6</sup> in (TTF)CuS<sub>4</sub>C<sub>4</sub>(CF<sub>3</sub>)<sub>4</sub> (TTF is tetrathiafulvalene) could be understood in terms of a phonon spectrum already (at high temperatures <200 °K) three dimensionally softened at a wave vector coincidentally on the plane of wave vectors further softened at low temperatures by the one-dimensional spin interactions. The theory developed from these two starting points should, we suggested, give a realistic description of the Heisenberg spin-Peierls transition in (TTF)CuS<sub>4</sub>C<sub>4</sub>(CF<sub>3</sub>)<sub>4</sub>. The most serious approximation is probably that of weak coupling ( $T_c \ll J$  with  $J$  the exchange interaction) only moderately well satisfied in this compound ( $T_c/J \sim 0.15$ ). The corresponding gold compound ( $T_c/J \sim 0.03$ )<sup>6</sup> is more weak coupling, but as yet less experimental information is available on it.

Although the underlying microscopic description we developed for the phase transition was significantly different from the previous works, the manifestations in most macroscopic quantities were predicted to be little changed from these works. The point is that in weak-coupling approximations the ratio of thermodynamic quantities (e.g., susceptibility  $\chi$ , specific heat) below  $T_c$  to those just above  $T_c$  will usually turn out to be functions of only  $T/T_c$  that do not look very different for different microscopic theories, except for small numerical changes (e.g., slightly different values for the initial slope of  $\Delta\chi/\chi_N$  as a function of  $T/T_c$ ). These however we were not able to accurately calculate within our theory. The relationship of  $T_c$  to  $J$  in our theory is quite different than in the Hartree theories, but depends on a microscopic coupling constant as yet unmeasured. Experi-

mental test of the new theory is not easy!

It turns out however that the effect of a magnetic field on the transition temperature  $T_c$  involves only a ratio of quantities, in which the poorly known multiplicative prefactor in our results drops out. The predictions are only sensitive to the form of certain correlation functions that we explicitly evaluated. In fact the magnetic field dependence of  $T_c$  probes rather directly the wave-vector dependence of the spin-correlation function  $\Pi_c$  — a quantity that universality arguments<sup>7</sup> suggest should be exactly given by our calculation for weak coupling. This behavior of the field is easily seen from the equivalent pseudofermion representation. There, a magnetic field, by changing the filling of the fermion bands, moves twice the Fermi wave vector  $2k_F$  (where  $\Pi_c$  is most divergent) away from the transition wave vector (where  $T_c$  effectively measures  $\Pi_c$ ). An exact probe would preferably leave the filling unchanged, and measure  $\Pi_c$  at nearby wave vectors. For small shifts it seems reasonable to neglect this difference.

Comparison with magnetic measurements therefore provides a good prospect of testing the theory. In addition, the large field behavior may give clearer proof that the phase transition is truly a spin-Peierls transition. In this paper I therefore discuss in detail the predictions of our theory for the transition temperature in a magnetic field. Bray<sup>8</sup> and Bulaevskii *et al.*<sup>9</sup> have performed these calculations for the Hartree theories, and the results presented here should be compared with these papers. In this comparison it should be borne in mind that in their approach results differing by almost 40% in the effective field (and therefore a factor of 2 in the coefficient of the quadratic field dependence) may be obtained depending on the value used for the susceptibility above  $T_c$  — the exact susceptibility (as in Ref. 1), or the larger, approximate Bulaevskii susceptibility (as in Ref. 8). In our approach we use the exact susceptibility, and the exact spin-wave velocity to define the coherence length, and no such leeway exists.

## II. THEORETICAL BACKGROUND

In Ref. 1 we calculated the transition temperature, approaching from above, as the temperature at which the frequency  $\Omega$  of the soft phonon becomes zero. The frequency is calculated by a random-phase treatment

$$\Omega^2(q) = 0 = \Omega_0^2(q) + \Pi(q) \quad (1)$$

where  $\Omega_0$  is the phonon frequency in the absence of magnetic interaction [and in (TTF)CuS<sub>4</sub>C<sub>4</sub>(CF<sub>3</sub>)<sub>4</sub> is assumed to already contain significant "soft" structure] and  $\Pi$  is the relevant linear response function of the spin system to the particular perturbation given by the spin-phonon coupling. In Eq. (1)  $q$  represents the wave vector along the spin-chain direction.

The wave vector for which  $\Omega(q)$  first goes to zero as the temperature is decreased gives the lattice distortion just below the transition temperature (the transition is assumed to be second order). Normally the dominant  $q$  dependence of Eq. (1) would be expected to come from  $\Pi(q)$ . Then for the spin-Peierls transition in zero field a wave vector  $\pi/s$  (with  $s$  the lattice constant) maximizes  $-\Pi(q)$ , and leads to a lattice dimerization and a spin-singlet ground state. A magnetic field favors a distortion at some other wave vector, for which the ground state has a net magnetization and lower magnetic energy. However, two effects oppose this. First, since the commensurate wave vector is on a zone boundary, umklapp processes strongly pin the distortion wave vector. In addition, it is believed that in (TTF)CuS<sub>4</sub>C<sub>4</sub>(CF<sub>3</sub>)<sub>4</sub>  $\Omega_0^2(q)$  has a pronounced minimum at  $q = \pi/s$ , again favoring the commensurate state. To calculate this effect it is convenient to parameterize the bare phonon frequency  $\Omega_0(q)$  near this minimum as  $\Omega_0(1 + \xi_{\text{ph}}^2 Q^2)^{1/2}$ , with  $Q$  the deviation of the wave vector from  $\pi/s$ . Then  $\xi_{\text{ph}}$  acts as a phonon-induced correlation length, also important in the discussion of the mean-field nature of the observed transition (see Ref. 1). It is the competition between these opposing effects that I calculate in this paper.

The theory of Ref. 1, and also the Hartree theories,<sup>3-5</sup> were developed in terms of an equivalent pseudofermion representation (see these references for details). The spin- $\frac{1}{2}$  chain in no magnetic field corresponds to a tight-binding pseudofermion band which is half filled (a Fermi wave vector  $k_F = \pi/2s$ ). In this representation, the Heisenberg chain, believed to describe the experimental systems, differs from the exactly soluble *XY* model by the presence of large four fermion interaction terms. In Refs. 3-5 these were treated by various Hartree approximations. In particular, in the approximation used by Pytte<sup>3</sup> the Heisenberg chain in zero field behaves as an *XY* chain with a renormalized energy scale. Our calcula-

tion<sup>1</sup> treats the interactions nonperturbatively, and finds qualitatively different results for the response of the spin system to a lattice distortion.

The effect of a magnetic field on the spin-Peierls transition is readily derived in the pseudofermion representation. There, a magnetic field changes the filling

$$\delta(2k_F) = \frac{2}{\pi s} \frac{\mu H}{J} \quad (2)$$

with  $\mu$  the magnetic moment, and where the exact zero-temperature normal-state susceptibility  $\chi_N = N\mu^2/\pi^2 J$  has been used. The use of the zero-temperature value is consistent with the weak-coupling assumption  $T_c \ll J$  used throughout. The umklapp scattering is included by writing (with  $q = \pi/s + Q$ )

$$\Pi(q) = \Pi_c(2k_F + Q - \delta) + \Pi_c(2k_F - Q - \delta) \quad (3)$$

where  $\Pi_c$  is the correlation function calculated in Ref. 1 for linear bands without umklapp processes, and the second term is the umklapp term. I have written  $\delta$  for  $\delta(2k_F)$  — not to be confused with the exponent of Ref. 1. Although the calculation of  $\Pi_c$  in Ref. 1 is only strictly correct in the zero-field limit (e.g., the symmetry argument used to relate  $z$ - $z$  to  $x$ - $x$  correlation functions), I again appeal to the weak-coupling assumption to justify the use of that result for the fields of interest  $\mu H \sim T_c \ll J$ .

The wave vector at which  $\Pi_c$  diverges continues to be  $2k_F$ , no longer equal to  $\pi/s$  in a field. Thus the individual terms in Eq. (3) are no longer maximized at  $Q = 0$ . However, since  $\Pi_c$  is quadratic in deviations from  $2k_F$ , by expansion it is easy to verify that for small  $\delta$ , the sum of normal and umklapp terms remains largest at  $Q = 0$ : this is the pinning at the commensurate wave vector by the umklapp processes.

Equations (1)–(3) completely solve the problem, including the additional pinning due to the structure in  $\Omega_0(q)$ . They, together with expressions for  $\Pi_c$ , were derived in Ref. 1. Here I investigate their implications in detail. The calculations are reproduced in Sec. III. Those more interested in the results may immediately turn to Sec. IV.

## III. CALCULATIONS

Equation (1) may be rewritten

$$\frac{T_c}{T_c^0} = \frac{1}{2} [f(\xi(Q - \delta)) + f(\xi(Q + \delta))] - \frac{T_c}{T_c^0} \xi_{\text{ph}}^2 Q^2 \quad (4)$$

where  $T_c^0$  is the transition temperature in zero field

( $Q = \delta = 0$ ). The function  $f$  is defined by

$$f(\xi\delta q) = \frac{\Pi_c(2k_F + \delta q)}{\Pi_c(2k_F)} = \left| \frac{\Gamma(\frac{1}{4} + i\xi\delta q)}{\Gamma(\frac{3}{4} + i\xi\delta q)} / \frac{\Gamma(\frac{1}{4})}{\Gamma(\frac{3}{4})} \right|^2, \quad (5)$$

where the explicit result for  $\Pi_c$  calculated in Ref. 1 is introduced. Here  $\xi$  is a coherence length,  $\xi = c/4\pi T_c$ , and for  $c$  the exactly known long-wavelength spin-wave velocity  $c = \frac{1}{2}\pi J_s$  should be used. It is also convenient to introduce a coherence length  $\xi_0$  which is  $\xi$  evaluated at  $T_c^0$ . This is then related to the  $\xi_{in}$  of Ref. 1 by numerical factors. A convenient form for computational purposes is

$$f(x) = \prod_{n=0}^{\infty} \left[ 1 - \frac{(n + \frac{1}{2})x^2}{(n + \frac{3}{4})^2[(n + \frac{1}{4})^2 + x^2]} \right]. \quad (6)$$

The function  $f(\xi Q)$  is plotted in Fig. 1.

To continue one essentially follows Leung's<sup>10</sup> calculations for the Peierls system, except the functional dependence here is different and we have an additional pinning term given by  $\xi_{ph} \neq 0$ . Thus Eq. (4) is an implicit equation for  $T_c(Q, H)$  — note  $f$  depends on  $T_c$  through  $\xi$ . This must be maximized with respect to  $Q$ , to give  $T_c(H)$  and the optimum wave vector for the transition  $q = \pi/s + Q(H)$ . As in Leung's work, I neglect any energy tending to pin the wave vector at other commensurate wave vectors. These much smaller energies may superimpose a step structure onto the trend for the incommensurate transition temperature that I calculate.

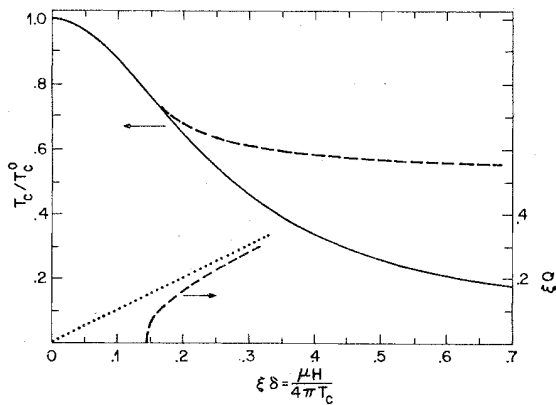


FIG. 1. Function  $f(\xi\delta)$ , which gives  $T_c/T_c^0$  implicitly for the undistorted-commensurate transition (heavy line). The dashed lines are the undistorted-incommensurate transition lines, together with the distortion wave vector, which rapidly approaches  $Q = \delta$  (dotted line).

#### IV. RESULTS

Let us first suppose the transition occurs at the commensurate wave vector ( $Q = 0$ ). This is always true for small fields, and remains true in larger fields if  $\xi_{ph}$  is large compared with  $\xi_0$ , the intrinsic coherence length. Then Eq. (4) reduces to

$$\frac{T_c}{T_c^0} = f\left(\frac{\mu H}{4\pi T_c^0} \frac{T_c^0}{T_c}\right), \quad (7)$$

where the function  $f$  is plotted in Fig. 1. From these results the direct dependence of the commensurate  $T_c/T_c^0$  on  $H$  is easily calculated numerically (Fig. 2).

For small  $H$  the depression of  $T_c$  is quadratic in the field

$$\frac{\Delta T_c}{T_c^0} = -14.4 \left(\frac{\mu H}{4\pi T_c^0}\right)^2. \quad (8)$$

Using the values for (TTF)CuS<sub>4</sub>C<sub>4</sub>(CF<sub>3</sub>)<sub>4</sub>,  $T_c = 12$  K and  $\mu = 1.9 \times 10^{-20}$  ergs/G, leads to the prediction  $\Delta T_c/H^2 = -1.3 \times 10^{-4}$  K/kG<sup>2</sup>. It should be emphasized that the depression of  $T_c$  quadratic in the applied field immediately follows from the original observation<sup>6</sup> of the decrease in magnetic susceptibility below  $T_c$ , together with the measured specific-heat change,<sup>11</sup> and does not give new information or additional evidence for the phase transition being truly a spin-Peierls transition. Nevertheless, this may provide a more accurate measurement of these parameters by which the theory may be tested.

For large fields the commensurate  $T_c$  shows an unusual reentrant behavior, that can ultimately be

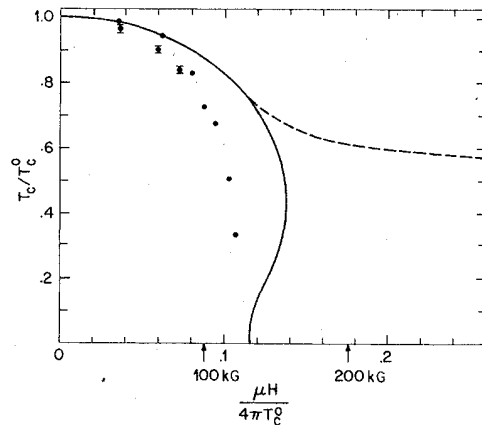


FIG. 2. Explicit field dependence of undistorted-commensurate (heavy line) and undistorted-incommensurate (dashed line) transition temperatures. The direct commensurate-incommensurate line is not shown. Points are from experiments (Ref. 12) on (TTF)CuS<sub>4</sub>C<sub>4</sub>(CF<sub>3</sub>)<sub>4</sub>: full circle with error bar, neutron scattering; ● magnetization (no errors given). The fields quoted are for this compound.

traced to the increase of the intrinsic correlation length as  $T^{-1}$ . There is a maximum field in which the commensurate spin-Peierls transition may occur, given by  $\mu H/4\pi T_c^0 = 0.138$ , corresponding to a field of about 157 kG in (TTF)CuS<sub>4</sub>C<sub>4</sub>(CF<sub>3</sub>)<sub>4</sub>.

If the assumption  $Q = 0$  is relaxed Eq. (4) for  $T_c/T_c^0$  must first be maximized to find the optimum  $Q$ . As  $H$  is increased from zero  $Q$  remains zero (i.e., the transition is to the commensurate state) until a critical field given by (cf. Leung)

$$f''\left(\frac{\mu H}{4\pi T_c}\right) = 2\left(\frac{T_c}{T_c^0}\right)^3 \left(\frac{\xi_{\text{ph}}}{\xi_0}\right)^2 \quad (9)$$

First considering the case where the additional pinning is small,  $\xi_{\text{ph}} \rightarrow 0$ , the critical field is defined by

$$f''(x) = 0,$$

which, as can be seen from Fig. 1 is at  $x = 0.145$ , corresponding to  $T_c/T_c^0 = 0.77$  and  $\mu H/4\pi T_c^0 = 0.11$  [a field of 125 kG in (TTF)CuS<sub>4</sub>C<sub>4</sub>(CF<sub>3</sub>)<sub>4</sub>]. As the field is increased beyond this value the wave vector of the transition moves rapidly to the value of  $2k_F$  for the field as shown in Fig. 1, where also the transition temperature to this incommensurate state is shown (and see Fig. 2). This description is qualitatively the same as the analysis of Bray and Bulaevskii for the Hartree approximation: the numerical values are changed however due to the different functional form of  $\Pi$ . The predictions are however quite different for large fields (still assuming  $\xi_{\text{ph}} = 0$ ). As  $2k_F$  and the transition wave vector move away from  $\pi/s$  with increasing field, the umklapp scattering term in Eq. (3) involves  $\Pi_c$  well away from  $2k_F$ , and becomes much smaller than the normal term. Thus, the effective coupling constant is decreased by a factor of 2. (This, of course, is the reason for the strong pinning at the commensurate wave vector for the half-filled band.) In the calculations of Bray<sup>8</sup> and Bulaevskii<sup>9</sup> where a BCS-like  $\exp(-1/\lambda)$  dependence on the spin-phonon coupling constant  $\lambda$  is assumed,  $T_c/T_c^0$  decreases rapidly to the order of  $T_c^0/J$ . On the other hand our theory gives a linear dependence of  $T_c$  on  $\lambda$ , and  $T_c/T_c^0$  will only decrease to  $\frac{1}{2}$ . Clearly observation of this behavior would dramatically confirm this unusual dependence on the coupling constant.

Unfortunately, the additional pinning effect of the already soft phonon will tend to mask this behavior. Equation (4) provides the formalism for including this pinning, once  $\xi_{\text{ph}}$  is known. The detailed results for  $T_c/T_c^0$  depend sensitively on  $\xi_{\text{ph}}$ , but the ratio will no longer saturate at  $\frac{1}{2}$ , although a slower decrease for an incommensurate  $T_c$  than predicted by the Hartree theories may be expected. At first sight we might expect saturation to occur instead at  $T_c/T_c^0$  about  $\Omega_0^2(\pi/s)/\Omega_0^2$ , where  $\Omega_0$  is the phonon frequen-

cy away from  $\pi/s$ . However as the wave vector leaves the neighborhood of the already softened phonon, fluctuation effects will probably further reduce  $T_c$ .

In Ref. 1 we estimated for (TTF)CuS<sub>4</sub>C<sub>4</sub>(CF<sub>3</sub>)<sub>4</sub> a value of  $\xi_{\text{ph}} \sim 1.5\xi_0$  from the diffuse x-ray scattering measurements.<sup>11</sup> This is probably a lowest reasonable estimate of  $\xi_{\text{ph}}$ . For this value we estimated the critical field and temperature for transition to the incommensurate state to be changed only by a small amount. Larger values of  $\xi_{\text{ph}}$  will change the picture significantly. For example, increasing  $\xi_{\text{ph}}$  to only about  $6\xi_0$  raises the critical field to the maximum field in which the commensurate transition occurs.

## V. COMPARISON WITH EXPERIMENT TO DATE

The effect of magnetic fields up to about 80 kG on the transition temperature of (TTF)CuS<sub>4</sub>C<sub>4</sub>(CF<sub>3</sub>)<sub>4</sub>, together with neutron scattering measurement of the transition wave vector, has been studied by Bray *et al.*<sup>13</sup> As expected no transition away from the commensurate wave vector was observed. The depression of  $T_c$  observed is significantly faster than predicted by either our theory (see Fig. 2), or the Hartree theory. It is possible that this is a consequence of the weak-coupling approximation  $T_c \ll J$ . For example, the zero-temperature susceptibility used to derive Eq. (1) is about 16% too small for  $T = 12$  K. Using  $\chi_N(12$  K) would increase the effective field by this amount, and bring theory into better agreement with experiment. The required correction is more like 30% in the effective field, however. As pointed out in Ref. 1, the measured coefficient of the quadratic dependence of  $T_c$  on  $H$  is also larger than would be estimated from specific-heat and susceptibility measurements.

In addition these authors<sup>13</sup> have attempted to identify  $T_c$  in higher fields from steps or knees in magnetization curves. Although less confidence can be placed in these estimates, they apparently continue the trend of the neutron scattering measurements: again  $T_c$  is seen to be depressed more than expected. No evidence for a change in dependence of  $T_c$  on field corresponding to the transition to an incommensurate state is seen up to 150 kG. Note however that since the incommensurate phase should have a magnetization very close to that of the undistorted phase in the same field, these authors may be observing the commensurate-incommensurate transition rather than the commensurate-undistorted transition (see Sec. VI).

The prospect of performing such experiments on the similar gold compound (TTF)AuS<sub>4</sub>C<sub>4</sub>(CF<sub>3</sub>)<sub>4</sub> offers hope of more stringently testing the theories, largely due to the lower  $T_c$  (about 2 K) for a similar exchange constant. Then, the weak-coupling approximation is much better satisfied, and comparison with theory can be made with more confidence. Also, the

fields needed  $\mu H \sim T_c$  are reduced by a factor of 6 from those in  $(\text{TTF})\text{CuS}_4\text{C}_4(\text{CF}_3)_4$  and are more accessible experimentally. In addition the lower  $T_c/J$  suggests a less dramatically softened bare phonon and so a smaller  $\xi_{\text{ph}}$ , a result also suggested by the apparent greater rounding of the transition. In any case  $\xi_0 \propto T^{-1}$  is larger and so  $\xi_{\text{ph}}/\xi_0$  is smaller in the gold compound, making any additional pinning less important [the factor on the right of Eq. (8) is 36 times smaller, even if  $\xi_{\text{ph}}$  is unchanged]. The exciting possibility of observing the transition away from the commensurate phase looks very promising. This would not only be the first observation of such a driven commensurate-incommensurate transition, but also, as we have seen, provides a clear discrimination between rival theories.

## VI. COMMENSURATE-INCOMMENSURATE TRANSITION

This paper has dealt so far with the transition from the undistorted phase as the temperature is reduced in a fixed magnetic field. An alternative experimental approach would be to study the commensurate to incommensurate transition as the field is increased at fixed temperature below  $T_c$ . The question then naturally arises whether the transition is first order or continuous. This has not been discussed before in detail for the spin-Peierls system, although the formalism is readily available from work on other systems. The answer is probably not sensitive to details of the microscopic theory — the main interest of this paper — but the analysis is relevant to the experimental signature of the incommensurate state to be expected, and seems worth including here.

At first sight a commensurate to incommensurate transition might be thought necessarily first order, and indeed this is true if the incommensurate state is a single Fourier component at the wave vector minimizing the distortion energy. However an alternative scheme was realized long ago,<sup>14</sup> in which the transition occurs continuously as a continuously increasing number of domain walls separating commensurate regions. The scattering by these two states is, of course, quite different. The latter transition has been calculated in detail for the usual Ginzburg-Landau description of a charge-density wave transition in terms of an array of sine-Gordon solitons.<sup>15,16</sup> The smoothness of the transition as the chemical potential is raised is then easily understood: although formation of a domain boundary eventually becomes energetically favorable, the mutual repulsion of like sine-Gordon solitons makes the growth of their number a continuous function of the driving field.

For the spin-Peierls system (and the similar case of the one-dimensional charge-density wave in a half-filled band) the description of the distorted state, and

the resulting phenomenological Ginzburg-Landau theory, show two new features. First, in zero field the order parameter has only a  $\pm$  degeneracy, and may be taken to be real. Second, even in an applied magnetic field the distortion energy is an even function about the commensurate zone-boundary wave vector. In fact the Ginzburg-Landau free energy is

$$F = \alpha \int dx \psi(x) \left[ - \left( 1 - \frac{T}{T_c} \right) + \frac{1}{2} b \psi^2(x) + G(-i\nabla_x) \right] \psi(x), \quad (10)$$

where  $-G(Q)$  is the right-hand side of Eq. (4) less its  $Q=0$  limit, explicitly an even function of  $Q$ , and  $\alpha$  and  $b$  may be taken as the parameters of Ref. 1. The transition temperature  $T_c$  is that to the commensurate state and  $\psi(x)$  is a real order parameter, such that the lattice displacement at  $x = ns$  is proportional to  $(-1)^n \psi(x)$ . As we have seen, for  $\xi\delta > 0.145$  the distortion energy  $G(Q)$  is minimized for nonzero  $Q = Q_0$ , say.

Bruce *et al.*<sup>17</sup> have analyzed a free-energy functional equivalent to Eq. (10) with  $G$  expanded up to order  $Q^4$ , so that

$$G(Q) = \gamma^4 Q^2 (c + \frac{1}{3} Q^2), \quad (11)$$

where  $\gamma(\delta)$  and  $c(\delta)$  are expansion parameters with  $\gamma$  nonzero and  $c$  going linearly through zero for  $\xi\delta = 0.145$ , and  $Q_0^2 = -\frac{3}{2}c$  for  $c$  negative above this critical  $\delta$ . Their numerical analysis then shows that although a domain boundary stationary solution to Eq. (10) exists, it becomes energetically favorable only for  $\gamma^2 Q_0^2 > 0.987(1 - T/T_c)^{1/2}$ . On the other hand a single Fourier component with wave vector  $Q_0$  ansatz for the incommensurate state becomes of lower energy than the commensurate state for  $\gamma^2 Q_0^2 > 0.821(1 - T/T_c)^{1/2}$ , corresponding to a smaller  $\delta$  and magnetic field. Their more complete analysis predicts that as the field is increased, there is indeed a first-order transition to an incommensurate state rather well described by a single Fourier component at a wave vector close to  $Q_0$ . It is tempting to suppose that the difference from the sine-Gordon case is that here the domain boundaries must attract, and coalesce to give the plane-wave distortion and a first-order transition at a lower driving field.

A similar conclusion arises comparing two calculations of the zero-temperature behavior of half-filled Peierls systems. These therefore apply directly to the XY spin-Peierls system, and perhaps qualitatively to the Heisenberg spin-Peierls system. Both calculations take particular values for the zero-temperature gap and bandwidth. However, in the weak-coupling limit the bandwidth should not be involved in calculating the nature of the commensurate-incommensurate transition (merely setting a length scale, and a cutoff)

and energy differences calculated should scale with the zero-temperature gap. I use their calculations to estimate this weak-coupling limit.

Kotani<sup>18</sup> has numerically calculated the energy of the incommensurate state assuming the wave vector of the distortion to be  $2k_F \neq \pi/s$  (together with harmonics). This must be compared with the energy of the commensurate state for the same chemical potential (magnetic field), and not for the same *number* which is Kotani's interest. (This point was, I believe, overlooked by Refs. 8 and 9, changing completely the conclusions.) On the other hand, Su *et al.*<sup>19</sup> have numerically calculated the energy of formation of a single-domain boundary for a particular half-filled Peierls system.

A comparison of these results shows that the ansatz of a distortion at the wave vector corresponding to the chemical potential (Kotani) leads to a lower energy than the ansatz of a single-domain wall (Su *et al.*) — the transition is therefore suggested to be first order.

## VII. CONCLUSIONS

I have considered in detail the effect of a magnetic field on a mean-field spin-Peierls transition, using the spin-correlation functions calculated in Ref. 1 by a Luther-Peschel-type treatment. This is a particularly interesting question, since it probes rather directly the wave-vector dependence of the correlation functions. As in previous calculations using the Hartree approximation for the spin-correlation functions, I find the transition temperature to be depressed by the field, and above a critical field, the transition is no longer to the dimerized state. I have calculated the transition temperature to the incommensurate

state above this field. Additional pinning energies may add a step structure at other commensurate wave vectors.

Although the numerical details differ a little between the theories below and just above the critical field, qualitatively the results are similar. Well above the critical field however the predictions are quite different, and measurements in this region may provide a good test of the new theory. For large fields the effective coupling constant is reduced by a factor of 2; the dependence of  $T_c$  on the coupling constant is thereby tested. An extra pinning effect of an already soft phonon, as expected in  $(\text{TTF})\text{CuS}_4\text{C}_4(\text{CF}_3)_4$ , may however complicate the prediction.

Measurements of  $T_c$  in a magnetic field for  $(\text{TTF})\text{CuS}_4\text{C}_4(\text{CF}_3)_4$  do not agree well with either theory. It is to be hoped that, if confirmed by further experiment, this disagreement may be explained as a consequence of the breakdown of the weak-coupling approximation,  $T_c \ll J$ . Then experiments on the similar gold compound,  $T_c/J = 0.03$ , are to be eagerly awaited. Another possibility is that the lack of agreement may rather arise from the "random-phase"-type approximation common to all present theories. If this approximation is not adequate, then an accurate description of the spin-Peierls transition in these compounds will be much harder to calculate.

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