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THREE-DIMENSIONAL ORDERING IN HMTTF-TCNQ

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Résumé. — Nous examinons l'instabilité de Peierls dans HMTTF-TCNQ avec un modèle Ginzburg-Landau, et montrons que l'ordre à trois dimensions est dominé par le couplage diagonal entre les chaînes TCNQ du réseau alterné des chaînes HMTTF-TCNQ. Le vecteur d'onde transverse peut être incommensurable et pratiquement constant dans une région où la température est au-dessous de la température de transition commune (50 K) des deux familles de chaînes. Nous trouvons que l'ordre est dominé par les chaînes TCNQ juste au-dessous de cette température tandis que la déformation des chaînes HMTTF reste faible jusqu'à 43 K pour augmenter ensuite rapidement. Ainsi nous prédisons une seule température de transition mais deux anomalies dans les propriétés physiques. Ces résultats sont consistants avec les mesures connues de la diffusion des rayons X, des propriétés de transport et de la chaleur spécifique.

Abstract. — We examine the Ginzburg-Landau model for the Peierls instability in HMTTF-TCNQ and show that the three-dimensional ordering is dominated by the diagonal coupling between TCNQ chains in the alternating chain lattice. The transverse wave vector can be incommensurate and nearly constant in the wide temperature range below a common critical temperature (50 K) for both families of chains. We find that just below this temperature the ordering is dominated by the TCNQ chains, while the deformation of the HMTTF chains remains weak until about 43 K and then sharply increases. Thus we expect one phase transition temperature but two anomalies in physical properties. These results are consistent with the experimental observations of diffuse X-ray scattering, transport properties and specific heat.

Interesting results concerning the three-dimensional ordering in HMTTF-TCNQ were obtained in recent X-ray studies by Megtert *et al.* [1]. Below 50 K a three-dimensional ordering of chain deformations develops with wave vector component $q_a = 0.42\ a^*$, $q_b = 2\,k_F = 0.37\ b^*$, and q_c unidentified. Within the experimental error the transverse wave numbers q_a and q_c appeared to be the same over the whole temperature range of the experiment ($T > 12$ K), even though the intensity of X-ray spots in the three-dimensional order increases rapidly below 43 K.

These X-ray data show clearly that the ordering in HMTTF-TCNQ differs considerably from that in TTF-TCNQ : the ordering in the latter system [2] starts at 54 K with the commensurate value $q_a = a^*/2$. q_a decreases from this value on lowering the temperature below the second critical temperature at 49 K. Such behaviour was explained within the Ginzburg-Landau model [3-5]. It was also shown [5] that the situation in which both families of chains order together is also possible in a lattice of the TTF-TCNQ type. q_a for such ordering is in general incommensurate and varies with temperature.

The details of the three-dimensional ordering in a given chain system depend essentially on two factors : the symmetry of the chain lattice and the strength of the different (interchain) coupling constants. The chain arrangement in HMTTF-TCNQ (Fig. 1) differs considerably from that of TTF-TCNQ. While in TTF-TCNQ equivalent chains form sheets in the **c**-direction, in HMTTF-TCNQ different chains alternate in both, **a**- and **c**-directions. Due to such geometry, the coupling between the chains situated at (0, 0) and ($a/2$, $c/2$) is expected to be comparable to the coupling between neighbouring chains of the same type in the **a**- and **c**-directions.

In this letter we suggest that the main features of

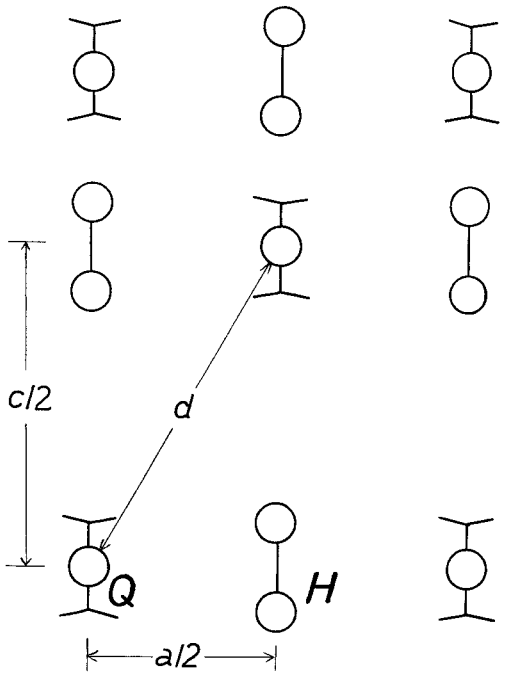

FIG. 1. — Simplified chain lattice of HMTTF-TCNQ with characteristic interchain distances.

the ordering in HMTTF-TCNQ can be explained within the Ginzburg-Landau (GL) model with this *diagonal* coupling taken into account. In other words, the main reason for the observed differences in the three-dimensional ordering in TTF-TCNQ and HMTTF-TCNQ are the different symmetries of the corresponding chain lattices.

We firstly investigate which types of ordering are possible in the chain lattice of figure 1. Afterwards we discuss in more detail the type of ordering which corresponds to the observed behaviour in HMTTF-TCNQ.

In our simplified treatment we associate one degree of freedom with each molecule. This may be the rigid displacement of the molecule or the (induced) charge density wave (CDW) averaged across the molecular dimensions. In this way the effects of the molecular tilting are neglected. This increases and simplifies the space symmetry of the model lattice. Expressed in terms of the complex deformation amplitude (or CDW) on HMTTF and TCNQ chains, $\rho_{H,Q} \exp(i\theta_{H,Q})$, the GL free energy for the ordering with the transverse wave numbers q_x, q_z then reads

$$F = a_Q \rho_Q^2 + a_H \rho_H^2 + 2[\Lambda_a \cos(q_x a) + \Lambda_c \cos(q_z c) + 2\Lambda_d \cos(q_x a/2) \cos(q_z c/2)](\rho_Q^2 + \rho_H^2) + \\ + 4[\Lambda_{a/2} \cos(q_x a/2) + \Lambda_{c/2} \cos(q_z c/2)] \cos\theta . \rho_Q \rho_H + b_Q \rho_Q^4 + b_H \rho_H^4 . \quad (1)$$

Here $\theta \equiv \theta_Q - \theta_H$, while Λ's are interchain coupling constants for the distances defined in figure 1. We take for simplicity $\Lambda_a(Q - Q) = \Lambda_a(H - H)$, etc., and also limit the following discussion to $\Lambda_a, \Lambda_c, \Lambda_d > 0$. The temperature enters F through the coefficients $a_{Q,H}$, the form of which depends upon the nature of the critical fluctuations [5].

Equation (1) implies that phase modulation of the ordering, with only one diagonal in the wave vector star ($\pm q_x, \pm 2k_F, \pm q_z$), is being activated. It can be shown in a way completely analogous to that for TTF-TCNQ, that such modulation may be unstable only when the points of the star are fourth-order commensurate [5, 6, 7] (i.e. when $q_x = a^*/4$ and/or $q_z = c^*/4$). The effects of the commensurate pinning will not be considered here.

The values of the transverse wave numbers q_a and q_c at a given temperature are the solutions of the equations $\partial F/\partial q_x = 0$, $\partial F/\partial q_z = 0$, i.e.

$$\sin(q_a a/2) \{ [2\Lambda_a \cos(q_a a/2) + \Lambda_d \cos(q_c c/2)](\rho_Q^2 + \rho_H^2) + \Lambda_{a/2} \cos\theta . \rho_Q \rho_H \} = 0 \quad (2a)$$

$$\sin(q_c c/2) \{ [\Lambda_d \cos(q_a a/2) + 2\Lambda_c \cos(q_c c/2)](\rho_Q^2 + \rho_H^2) + \Lambda_{c/2} \cos\theta . \rho_Q \rho_H \} = 0 . \quad (2b)$$

Instead of describing all the criteria under which a particular solution of these equations is stable, we shall only briefly point out the main characteristics of each type of ordering. Three solutions are to be distinguished :

(i) $$q_a/2 = n_a a^*/2 , \quad q_c/2 = n_c c^*/2 , \quad n_a, n_c = 0 \text{ or } 1 . \quad (3)$$

After substitution of the solution (3) and the minimization over θ, the free energy (1) reduces to

$$F = a_Q \rho_Q^2 + a_H \rho_H^2 + 2(\Lambda_a + \Lambda_c \pm 2\Lambda_d)(\rho_Q^2 + \rho_H^2) - 4 | \Lambda_{a/2} \pm \Lambda_{c/2} | \rho_Q \rho_H + b_Q \rho_Q^4 + b_H \rho_H^4 . \quad (4)$$

Here the upper and lower sign correspond to $n_a + n_c = 0$ or 2 and $n_a + n_c = 1$ respectively. Equation (4) represents the standard GL problem of two order parameters coupled through the bilinear $\rho_Q \rho_H$ term. The critical temperature for the ordering (3) is given as the highest solution of the equation

$$\tilde{a}_Q . \tilde{a}_H = \Lambda_{QH}^2/4 , \quad (5)$$

where $\tilde{a}_Q$, $\tilde{a}_H$ and Λ_{QH} are respectively the coefficients in front of ρ_Q^2, ρ_H^2 and $\rho_Q \rho_H$ in equation (4). Below this

temperature both deformation amplitudes, ρ_Q and ρ_H, are finite : the transverse periodicity (3) can occur only when both chain families are ordered.

(ii) $$q_c/2 = n_c\, c^*/2\,, \qquad n_c = 0 \quad \text{or} \quad 1\,, \tag{6}$$

and q_a given by the zero of the curly bracket in equation (2*a*). After minimization over θ, equations (1) and (2*a*) read

$$F = (a_Q - 2\,\Lambda_a + 2\,\Lambda_c - \Lambda_d^2/\Lambda_a)\,\rho_Q^2 + (a_H - 2\,\Lambda_a + 2\,\Lambda_c - \Lambda_d^2/\Lambda_a)\,\rho_H^2 - \\ - \frac{\Lambda_{a/2}^2}{\Lambda_a}\cdot\frac{\rho_Q^2\,\rho_H^2}{\rho_Q^2 + \rho_H^2} - 4\left|\Lambda_{c/2} - \frac{\Lambda_{a/2}\,\Lambda_d}{2\,\Lambda_a}\right|\rho_Q\,\rho_H + b_Q\,\rho_Q^4 + b_H\,\rho_H^4\,, \tag{7}$$

$$\cos(q_a\,a/2) = (-1)^{n_c+1}\left[\frac{\Lambda_d}{2\,\Lambda_a} - \frac{\Lambda_{a/2}}{2\,\Lambda_a}\,\frac{\rho_Q\,\rho_H}{\rho_Q^2 + \rho_H^2}\,\mathrm{sign}\left(2\,\Lambda_{c/2} - \frac{\Lambda_{a/2}\,\Lambda_d}{\Lambda_a}\right)\right]. \tag{8}$$

The solutions (6, 8) with $n_c = 0$ and $n_c = 1$ are degenerate and structurally equivalent.

Equations (7, 8) can be further simplified by introducing the approximation

$$\frac{\Lambda_{a/2}}{\Lambda_a}\cdot\frac{\rho_Q\,\rho_H}{\rho_Q^2 + \rho_H^2} \approx \frac{\Lambda_{a/2}}{\Lambda_a}\cdot\frac{\rho_H}{\rho_Q} < 1\,, \tag{9}$$

which is valid whenever $\rho_H \ll \rho_Q$. This approximation necessarily goes together with $|\Lambda_{a/2}|/\Lambda_a > 1$, which is to be reasonably expected in the simplest picture of interchain coupling, used in setting up equation (1). Moreover, the condition $\rho_H/\rho_Q \ll 1$ is consistent with the result for ρ_H and ρ_Q which follows from equation (7) and will appear to be the most relevant one for HMTTF-TCNQ.

Through equation (9) the free energy (7) reduces again to the *bilinear* form (4), but the harmonic coefficients are now given by

$$\tilde{a}_Q = a_Q - 2\,\Lambda_a + 2\,\Lambda_c - \Lambda_d^2/\Lambda_a\,, \tag{10a}$$

$$\tilde{a}_H = a_H - 2\,\Lambda_a + 2\,\Lambda_c - \Lambda_d^2/\Lambda_a - \Lambda_{a/2}^2/\Lambda_a\,, \tag{10b}$$

$$\Lambda_{QH} = 4\,|\,\Lambda_{c/2} - \Lambda_{a/2}\,\Lambda_d/2\,\Lambda_a\,|\,. \tag{10c}$$

As in the case (i), both chain families again order together, but now with an incommensurate value of q_a. The details of the temperature behaviour of ρ_Q and ρ_H below the critical temperature T_c (Eq. (5)) will be discussed later.

The solution (8) is stable provided that the absolute value of the bracket on the r.h.s. is smaller than unity. However if both terms in this bracket are larger than unity but their sum smaller than unity, the stability condition $\partial^2 F/\partial q_x^2 > 0$ is not satisfied. Having thus both terms in the bracket smaller than unity, we distinguish two different limits, depending on which of these terms dominates.

If the first temperature independent term prevails, q_a is mainly determined by the coupling constants among the chains of the same type, and is thus weakly temperature dependent. This limit includes the special case of partly ($\Lambda_{a/2} = 0$) or completely ($\Lambda_{a/2} = \Lambda_{c/2} = 0$) decoupled H and Q chains, for which q_a is entirely temperature independent. In this latter case

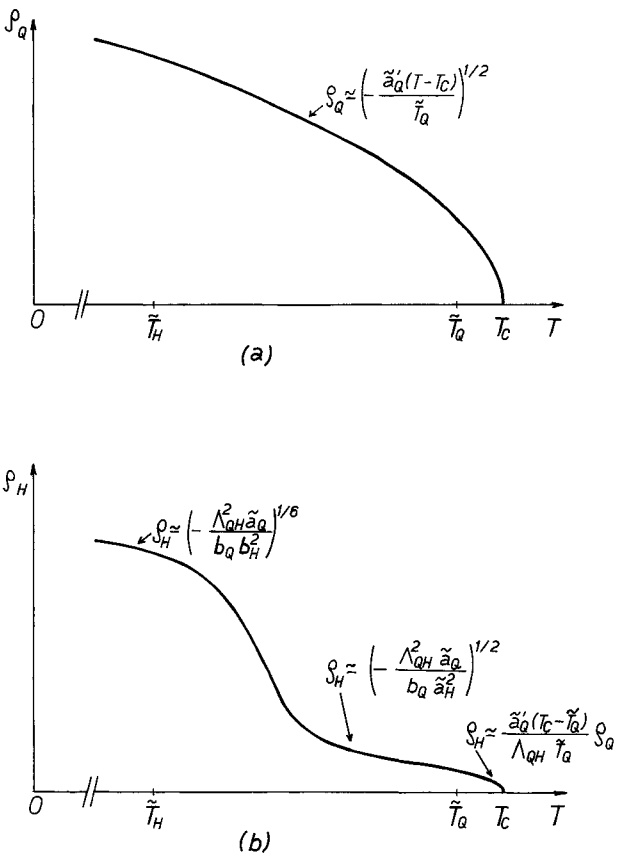

FIG. 2. — Qualitative behaviour of ρ_Q (a) and ρ_H (b) in the limit defined by equations (7), (10) and (17). ρ_Q and ρ_H in arbitrary units ; $\rho_H \ll \rho_Q$ in the whole temperature range.

$\Lambda_d/2\,\Lambda_a < 1$ and $\Lambda_d^2/4\,\Lambda_a\,\Lambda_c > 1$ are the conditions under which the Q sublattice alone orders incommensurately [1].

When the second term in equation (8) is dominant, q_a is directly governed by the temperature dependent quantities ρ_Q and ρ_H. Although the temperature variations of ρ_Q and ρ_H may cancel to a great extent in the ratio ρ_H/ρ_Q, any strong change in the behaviour of only one deformation amplitude in a certain temperature range will necessarily cause significant variation of the second term in equation (8) and

consequently of q_a. We note that the dependence of q_a on ρ_Q and ρ_H, as given by equation (8) without the first term, was already met in the case of the TTF-TCNQ lattice [5].

(iii) The brackets in both equations (2*a*) and (2*b*) vanish. The free energy (1) is then given by

$$F = (a_Q - 2\Lambda_a - 2\Lambda_c)\rho_Q^2 + (a_H - 2\Lambda_a - 2\Lambda_c)\rho_H^2 + \\ + \frac{8(\Lambda_{a/2}^2\Lambda_c + \Lambda_{c/2}^2\Lambda_a - \Lambda_{a/2}\Lambda_{c/2}\Lambda_d)}{\Lambda_d^2 - 4\Lambda_a\Lambda_c}\frac{\rho_Q^2\rho_H^2\cos^2\theta}{\rho_Q^2 + \rho_H^2} + b_Q\rho_Q^4 + b_H\rho_H^4 . \quad (11)$$

In this equation ρ_Q and ρ_H are coupled only through the third term on the r.h.s. ; there is no bilinear term like in equations (4) and (7). If the coefficient in front of this term is negative, the relative phase is given by

$$\cos^2\theta = 1 . \quad (12)$$

The free energy then reduces to the form which was already discussed in detail for the case of the TTF-TCNQ lattice [5, 8]. Here we only briefly recall the main results.

The ordering can occur in two ways, depending on parameters in equation (1). In the first case the Q chains order at the temperature T_Q defined by

$$a_Q - 2\Lambda_a - 2\Lambda_c = 0 , \quad (13)$$

while H chains order at some lower temperature T'_H, defined by

$$a_H - 2\Lambda_a + 2\Lambda_c - \Lambda_d^2/\Lambda_a - \Lambda_{a/2}^2/\Lambda_a = 0 . \quad (14)$$

For $T'_H < T < T_Q$, $\rho_Q \neq 0$, $\rho_H = 0$, and the wave numbers q_a and q_c stay at the boundary of the Brillouin zone ($q_a = a^*/2$, $q_c = c^*/2$). For $T < T'_H$, q_a and q_c start to move into the Brillouin zone. This behaviour is analogous to that observed in TTF-TCNQ. Here it can occur if the *diagonal* coupling Λ_d is sufficiently weak, as is seen from the note (1) and the condition for equation (12).

If $T'_H > T_Q$, Q and H chains order together at some intermediate temperature and with the finite values of $\cos(q_a a/2)$ and $\cos(q_c c/2)$ proportional to ρ_H/ρ_Q [5], i.e. there is no constant term as in equation (8).

Finally, for

$$\frac{(\Lambda_{a/2}^2\Lambda_c + \Lambda_{c/2}^2\Lambda_a - \Lambda_{a/2}\Lambda_{c/2}\Lambda_d)}{(\Lambda_d^2 - 4\Lambda_a\Lambda_c)} > 0 \quad (15)$$

$\cos^2\theta = 0$, and ρ_Q and ρ_H in equation (11) are completely decoupled. They start to develop at temperatures defined by equations (13) and

$$a_H - 2\Lambda_c - 2\Lambda_a = 0 \quad (16)$$

respectively. Below this latter temperature the wave numbers remain at the boundary of the Brillouin zone, although the deformation amplitudes of both chain families are finite. The relative phases between the deformation amplitude on the H chain and two deformation amplitudes at neighbouring Q chains in **a**- and **c**-directions are 0 and π. The corresponding interchain terms in the free energy cancel each other by interference and the order parameters on Q and H chains appear to be effectively independent. Note from equation (15) that such type of ordering is possible due to the finite value of Λ_d.

We conclude the discussion of equations (2*a*, *b*) by emphasizing that the chains order separately, i.e. there is a temperature range in which $\rho_Q \neq 0$, $\rho_H = 0$, only if Q chains order in such a way that the resulting mean field vanishes at the positions of the H chains. This is realized only within the solution (iii) when $q_a = a^*/2$ and $q_c = c^*/2$. For all other wave numbers both chain families are ordered simultaneously.

Let us now return to the actual ordering in HMTTF-TCNQ. We have seen that the formation of three-dimensionally ordered deformations with the incommensurate value of q_a is realized in the case (ii) as well as in the case (iii) provided that equation (12) is valid and that $T'_H > T_Q$. However detailed examination of equation (11) shows that the observed temperature dependence of X-ray spot intensities [1] cannot be understood within the latter case. In this case it appears that for all possible physical values of parameters appearing in equation (11) the rates of change of both deformation amplitudes ρ_Q and ρ_H are smooth in the whole temperature range below T_c. This does not agree with the experimental data which show two anomalies. Furthermore, the variation of q_a and q_c is usually such that it could be seen in X-ray experiments.

On the contrary equation (7) of case (ii) may lead to a temperature behaviour of ρ_Q and ρ_H consistent with the observations. Such behaviour occurs provided the interchain coupling constant Λ_{QH} is smaller than the parameters $\tilde{a}'_Q$, $\tilde{a}'_H$ defined by

$$\tilde{a}_{Q,H} \simeq \tilde{a}'_{Q,H}(T/\tilde{T}_{Q,H} - 1) ,$$

(1) The other two types of ordering for the sole Q sublattice are : $q_a/2 = 0$, $q_c/2 = \pi$ or *vice versa*, for $\Lambda_d > 2\Lambda_a$, $\Lambda_d > 2\Lambda_c$ and $q_a = a^*/2$, $q_c = c^*/2$ for $\Lambda_d^2/4\Lambda_a\Lambda_c < 1$.

and if the temperatures $\tilde{T}_Q$ and $\tilde{T}_H$ are not too close to one another. Then the condition

$$\frac{4\,\tilde{T}_Q\,\tilde{T}_H}{(\tilde{T}_Q - \tilde{T}_H)^2} \cdot \frac{\Lambda_{QH}^2}{\tilde{a}'_Q\,\tilde{a}'_H} \ll 1 \qquad (17)$$

is satisfied and the critical temperature T_c lies slightly above the higher of the temperatures $\tilde{T}_Q$, $\tilde{T}_H$ (say $\tilde{T}_Q$), so that $T_c - \tilde{T}_Q \ll \tilde{T}_Q - \tilde{T}_H$. The deformation amplitude ρ_Q then has roughly the simple smooth GL behaviour in the whole temperature range below T_c. In contrast, the other deformation amplitude ρ_H increases abruptly just below T_c. This happens somewhat above $\tilde{T}_H$, i.e. in the temperature range of the width

$$(T - \tilde{T}_H)/\tilde{T}_H \lesssim \frac{\Lambda_{QH}^{2/3}\,\tilde{a}_Q'^{1/3}}{\tilde{a}'_H}(1 - \tilde{T}_H/\tilde{T}_Q)^{1/3}\,.$$

Condition (17) is essential for such behaviour. When it does not hold, i.e. when $T_c - \tilde{T}_Q \gg \tilde{T}_Q - \tilde{T}_H$, ρ_H varies smoothly with temperature.

Since the sulphur atoms in HMTTF molecules are the dominant scatterers for X-rays [1], the increase of ρ_H has a strong impact on spot intensities, in agreement with the experimental results of Megtert *et al.* [1]. We thus identify T_c to be equal to 50 K, while the steep increase of ρ_H starts at about 43 K. Provided that the gap on HMTTF chains is less than $k_B T$, this is consistent with the conductivity [9, 10] and specific heat [11] measurements in which 50 K and 43 K anomalies were observed. Furthermore the ESR data [12, 13] indicate that these anomalies correspond to the change from the metallic to the insulator state on TCNQ and HMTTF chains respectively.

Finally, we interpret the observed temperature independence of q_a and q_c in HMTTF-TCNQ [1]. The constancy of q_c is consistent with our result ([2]) $q_c = c^*$ (Eq. (6)) in the solution (ii). Regarding the other transverse direction, it was already pointed out that q_a can be indeed very weakly temperature dependent, provided that the first, temperature independent term in equation (8) is much larger than the second term containing the ratio ρ_H/ρ_Q. The ratio ρ_H/ρ_Q varies strongly in the region of the second anomaly in ρ_H, and reaches the value $[b_Q\,\tilde{a}'_H/b_H\,\tilde{a}'_Q]^{1/2}$ at low temperatures. We expect that the resulting temperature variation of q_a which is largest at about 43 K is still smaller than the error bars of the X-ray data. The temperature independent and incommensurate value of q_a should be thus predominantly determined by the coupling between TCNQ chains in the diagonal and the **a**-direction of figure 1. The value $q_a = 0.42$ corresponds to the reasonable ratio $\Lambda_d/\Lambda_a \simeq 0.3$.

It is gratifying that basically the same theory accounts for the observation in crystals as different as HMTTF-TCNQ and TTF-TCNQ. The present understanding of the structural properties of chain conductors *via* the Ginzburg-Landau approach [3-8] does not account for the fact that the transverse order observed at 100 K in $K_2Pt(CN)_4Br_{0.3}\cdot 3\,H_2O$ [15], at 54 K and 49 K in TTF-TCNQ [14] and at 50 K in HMTTF-TCNQ [1] is incomplete. The reason for this absence of long range order is not yet resolved, and might well be different for the different materials. Although our free energy minimization procedure covers only the regime below the temperature of three-dimensional ordering in both limits of strong and weak interchain coupling [5], we do not expect that any of the above temperatures can be identified as a crossover temperature [1]. This is because for one set of chains the crossover by itself does not lead to thermodynamic anomalies [16, 17]. Pronounced anomalies above the long range phase ordering temperature are related to the amplitude ordering which sets in near to the single chain mean field transition temperature [18]. However for the crossover picture to be meaningful, the mean field temperature should be much more than a few degrees above the phase ordering temperature. In other words we believe that such closely spaced anomalies as observed in HMTTF-TCNQ cannot be ascribed to the crossover from 1-dim to 3-dim regimes.

The striking difference in the behaviour of the anomalies in HMTTF-TCNQ and TTF-TCNQ is thus attributed here to the difference in the lattice geometries rather than to the differences in the underlying physics. The alternation of chains in both transverse directions led to the *bilinear* form of the Ginzburg-Landau expansion in terms of ρ_Q and ρ_H (Eqs. (8-10)) and subsequently to the possibility of behaviour of ρ_Q and ρ_H resembling two phase transitions but without large variations in q_a. Of course there is only one critical temperature, but the order parameter on one set of chains increases sharply near 43 K giving rise to a second anomaly in the transport properties [9, 10] and specific heat [11]. Furthermore, due to the diagonal coupling Λ_d which is smaller than Λ_c, q_a may have a nearly constant incommensurate value in a wide range of temperatures. Such behaviour could not be realized in e.g. the TTF-TCNQ like lattice, for which the above discussion as well as the earlier work [5] show that the incommensurate ordering always goes together with substantial temperature dependence in the wave number q_a.

We have benefited from discussions with R. Comès, D. Jérome and J. P. Pouget.

([2]) This result has indeed been confirmed by Pouget *et al.* [14] after the present text was submitted.

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