Destruction of Density-Wave States by a Pseudogap in High Magnetic Fields: Application to $(TMTSF)_2ClO_4$

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(Received 27 December 1994)

A model is presented for the destruction of density-wave states in quasi-one-dimensional crystals by high magnetic fields. The model is consistent with previously unexplained properties of the organic conductors $(TMTSF)_2ClO_4$ and $(BEDT\text{-}TTF)_2MHg(SCN)_4$ (M = K, Rb, Tl). As the magnetic field increases quasi-one-dimensional density-wave fluctuations increase, producing a pseudogap in the electronic density of states near the transition temperature. When the pseudogap becomes larger than the mean-field transition, temperature formation of a density-wave state is not possible.

PACS numbers: 75.30.Fv, 71.45.Lr, 74.70.Kn, 75.30.Kz

Understanding low-dimensional electronic systems continues to be one of the major challenges of condensed matter physics. Organic conductors are particularly interesting systems because they exhibit a subtle competition between metallic, superconducting, charge-density-wave (CDW), and spin-density-wave (SDW) phases. This competition is sensitive to pressure, temperature, magnetic field, and chemical substitution [1–3]. This Letter presents a theory of the destruction of density-wave phases in quasi-one-dimensional materials in high magnetic fields. The model is used to understand previously unexplained properties of the quasi-one-dimensional conductor $(TMTSF)_2ClO_4$ and the quasi-two-dimensional conductors $(BEDT\text{-}TTF)_2MHg(SCN)_4$ (M = K,Rb,Tl).

At ambient pressure and zero magnetic field $(TMTSF)_2ClO_4$ is a superconductor below 1.3 K. A spectacular cascade of transitions into field-induced spin-density-wave (FISDW) phases occurs at fields above 4 T. The quantum Hall effect is observed in these phases [1]. The phase diagram below 15 T can be explained in terms of the so-called "standard model," which is a mean-field treatment of a highly anisotropic Hubbard model [1,4]. However, there are at least five experimental observations concerning the behavior of $(TMTSF)_2ClO_4$ in high magnetic fields that cannot be explained by any existing theory. (a) At about 15 T the field-induced spin-density-wave transition temperature T_{SDW} reaches a maximum of about 5 K and then decreases at higher fields, going to zero at 27 T [5]. The high field phase is known as the "re-entrant phase." Its existence contradicts the standard model in which the transition temperature saturates at high fields. Specific heat measurements indicate that there is a well-defined phase transition from the FISDW phase to the re-entrant phase [6,7]. (b) Below (above) 2 K the specific heat of the re-entrant phase at 30 T is smaller (larger) than that of the metallic phase at zero field [6,7]. (c) Along the phase boundary at high temperatures the ratio of the specific heat jump, ΔC, to the normal state electronic specific heat, γT, is larger than the mean-field value of 1.43. For example, at 15 T this ratio is about 3.5 [6,7]. On the phase boundary near 27 T the ratio is less than the BCS value. (d) In the re-entrant phase the magnetoresistance is approximately activated in temperature [5]. (e) Thermopower measurements suggest that there is an electronic energy gap above 30 T and below 2.6 K [8].

Previously two models have been proposed to explain the existence of the re-entrant phase [9,10]. Yakovenko [9] considered how a high field could effectively confine the electronic motion to single stacks of TMTSF molecules and as a result the FISDW phase would be destroyed by one-dimensional fluctuations. However, this model does not predict the observed rapid decrease of T_{SDW} with field [6], and if commonly accepted parameter values are used the theory will only be relevant for fields of the order of 100 T [11]. The second model [10] involves changes in the band structure produced by the ordering of the ClO_4 ions at 24 K. This model predicts that there should be spikes in the transition temperature, periodic in the inverse field, at high fields. Although there is some evidence for such spikes in acoustic experiments [12], there is no evidence for such spikes in specific heat [6,7], resistivity [5], and thermopower [8] measurements. This model also does not predict the observed rapid decrease of T_{SDW} with field [6,7].

This Letter presents a model for behavior near the FISDW re-entrant phase boundary that is consistent with the five observations listed above. Before describing the details a brief description is given of the basic physics. A SDW forms when the opening of an energy gap at the Fermi surface, due to the SDW, lowers the total electronic energy by more than the increase in Coulomb energy due to the SDW. The size of the energy gap is proportional to the amplitude of the SDW. However, the fluctuations in the SDW order parameter affect the electronic states. Similar effects have been seen in CDW systems [13–15]. Such effects are ignored in the standard model. Near the transition temperature the fluctuations increase and there are long range SDW correlations producing a pseudogap in the density of states. Furthermore, the pseudogap

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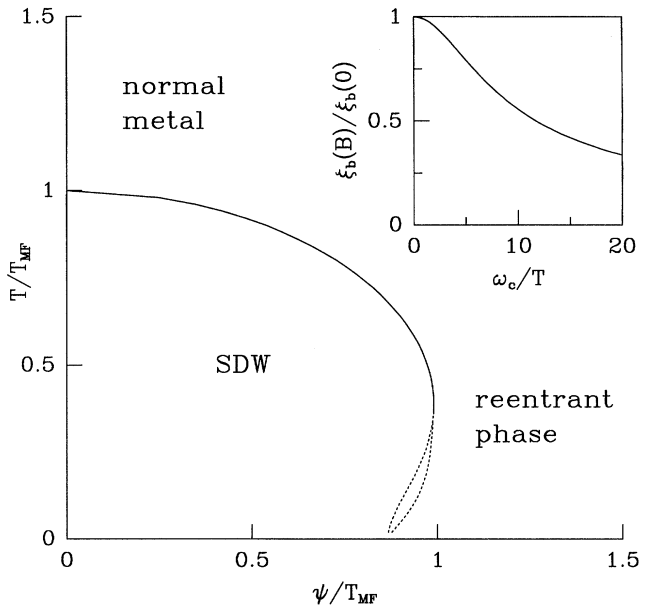

FIG. 1. Phase diagram for a spin-density-wave state in the presence of a pseudogap ψ. Both the pseudogap and the transition temperature are normalized to the mean-field transition temperature, T_{MF}. Along the dotted lines the transition is first order. There is a coexistence of phases between the dotted lines. Above the transition temperature as the field increases there is a smooth crossover from a normal metal to a nonmetal with a pseudogap. This phase diagram is consistent with that of $(\mathrm{TMTSF})_2\mathrm{ClO}_4$ at fields above 15 T and of $(\mathrm{BEDT\text{-}TTF})_2M\mathrm{Hg(SCN)}_4$ (M = K,Rb,Tl). Inset: Reduction of the SDW correlation length transverse to the chains, ξ_b, by a magnetic field parallel to the least conducting direction 16. Equation (3) then implies that ψ increases with field. ω_c is the cyclotron frequency [see Eq. (4)].

reduces the transition temperature, and for a sufficiently large pseudogap formation of a SDW is not possible (Fig. 1). The reason for this is simple. In the presence of a large pseudogap, opening an energy gap due to a SDW will not lower the total energy sufficiently to make formation of a SDW energetically favorable. The size of the pseudogap is determined by the magnitude of the SDW fluctuations which are in turn affected by the size of the SDW correlations *transverse* to the chains. Bjeliš and Maki [16] have shown that the transverse correlation length is a decreasing function of magnetic field. This provides a mechanism for the FISDW re-entrant phase transition seen in $(\mathrm{TMTSF})_2\mathrm{ClO}_4$: as the field increases the transverse correlation length decreases and the pseudogap increases above a critical value. The presence of a pseudogap is consistent with the observations (b), (d), and (e).

It is generally believed that the physics of the SDW phases found in the $(\mathrm{TMTSF})_2X$ salts can be described by a Hubbard model with highly anisotropic dispersion [1]. The hopping integrals t_a, t_b, and t_c associated with the three crystal axes are estimated to have values of about 250, 25, and 1 meV, respectively. The Fermi surface consists of two slightly warped planes. The sensitivity of the *imperfect nesting* of this open Fermi surface to pressure, magnetic field, and the anion X, is responsible for the rich phase diagram. A mean-field treatment of the anisotropic Hubbard model can explain the existence of the FISDW phases [1,4]. At zero field the imperfect nesting prevents formation of a SDW. A magnetic field improves the nesting because the electron motion becomes more one dimensional [4,17], resulting in SDW formation and a maximum transition temperature of about 5 K at 15 T. Since only behavior above 15 T is considered here, for simplicity, perfect nesting is assumed and a one-dimensional model of the electronic states is considered. Consequently, the effect of the orbital electron motion on the density of states and the transition temperature is neglected.

At the mean-field level the electrons move in a potential with wave vector $2k_F$, $\Delta(x)$, that is proportional to the SDW amplitude. The upper and lower components of a spinor $\Psi(x)$ describe left-moving, up-spin and right-moving, down-spin electrons, respectively, with Fermi velocity v_F. The Hamiltonian for these electrons is [1]

$$H = \int dx\, \Psi^{\dagger}\left[-iv_F\sigma_3\frac{\partial}{\partial x} + \frac{1}{2}[\Delta(x)\sigma_+ + \mathrm{H.c.}]\right]\Psi, \tag{1}$$

where σ_3 and $\sigma_{\pm} \equiv \sigma_1 + i\sigma_2$ are Pauli matrices. It is assumed that the SDW is incommensurate with the lattice and so $\Delta(x)$ is complex. The other electrons are described by a similar Hamiltonian.

In the standard mean-field treatment the order parameter $\Delta(x)$ is replaced by its expectation value. In reality $\Delta(x)$ is a dynamical field that fluctuates due to quantum and thermal effects. These fluctuations are key to the model presented here. Such spin excitations have been detected in antiferromagnetic resonance experiments [18] and have been used to explain the temperature dependence of the SDW amplitude [19]. The characteristic energy scale of these excitations is about 1 K, and they soften as the transition temperature is approached. Consequently, these excitations are treated classically here. At the Gaussian level the SDW correlation function above the transition temperature is [20]

$$\langle\Delta(x)\Delta(x')^*\rangle = \psi^2\exp[-|x - x'|/\xi_a(T)], \tag{2}$$

where $\xi_a(T)$ is the correlation length along the chains and ψ is the rms fluctuation in the order parameter. For a strictly one-dimensional system $\psi^2 \propto T\xi_a(T) \to \infty$ close to a phase transition [20]. (This divergence is related to the fact that in one dimension fluctuations prevent finite temperature phase transitions.) A more realistic model takes into account the coupling between chains and with q_0 a wave-vector cutoff gives

$$\psi^2 \propto q_0 T\frac{\xi_a(T)^2}{\xi_b(T)\xi_c(T)}, \tag{3}$$

which is finite as $\xi_i(T) \rightarrow \infty$. Here ψ will be treated as a parameter that is a measure of the SDW fluctuations. The important point is that *as the transverse correlation length ξ_b decreases ψ increases.*

Bjeliš and Maki [16] considered the effect of a large magnetic field parallel to the c direction (the least conducting direction) on the CDW and SDW correlation length in the a and b directions. They showed that, while ξ_a was not significantly affected, ξ_b is a decreasing function of magnetic field (inset of Fig. 1). This is because as the field increases the electron motion becomes more one dimensional [4,17]. If ω_c is the cyclotron frequency and T_{MF} is the mean-field transition temperature, then the size of the reduction is determined by the ratio

$$\frac{\omega_c}{T} \equiv \frac{ebv_F B}{T} \equiv \frac{B}{B_0}\frac{T_{\mathrm{MF}}}{T}. \tag{4}$$

For $(TMTSF)_2ClO_4$ ($b = 7.7$ Å, $v_F = 2 \times 10^5$ m/sec, and $T_{\mathrm{MF}} = 5$ K) $B_0 \sim 3$ T [21].

This Letter uses the following model for the fluctuations in $\Delta(x)$. It is replaced in (1) with a *static* random potential with zero mean, $\langle \Delta(x) \rangle = 0$, and correlations given by (2). Treating $\Delta(x)$ as a *static* field is a reasonable approximation if the fluctuations can be treated classically. Similar arguments have been used to successfully model the effect of lattice fluctuations on the electronic properties of CDW compounds [13].

Sadovskiĭ [22] calculated the one-electron Green's function for the one-dimensional model (1) and (2) *exactly.* A perturbative treatment of this problem was given earlier [23]. Sadovskiĭ found that the Green's function reduced to a simple analytic form in the limit of large correlation lengths ($\xi_a \gg v_F/\psi$) [24]. Since we are interested in behavior near T_{SDW}, we will also take this limit. Then it is also possible to evaluate exactly higher-order Green's functions such as those needed to find the transition temperature. The density of states, shown in the inset of Fig. 2, is zero at the Fermi energy and suppressed on an energy scale of order ψ; i.e., there is a pseudogap. As $\xi_a\psi/v_F$ decreases the pseudogap gradually fills in [22]. In some CDW systems, at zero field, optical and susceptibility measurements near the transition temperature are consistent with a pseudogap due to fluctuations [15].

$T_{\mathrm{SDW}}/T_{\mathrm{MF}}$ is a universal function of ψ/T_{MF} (Fig. 1). (Identical results are obtained for CDW's if Zeeman splitting is neglected.) As the fluctuations increase T_{SDW} decreases. The most important point is as follows: *for $\psi > T_{\mathrm{MF}}$ formation of a SDW is not possible.*

This model is consistent with the five properties of $(TMTSF)_2ClO_4$ listed above. (a) It is postulated that the FISDW re-entrant transition is due to destruction of the FISDW phase by a pseudogap. As the field increases ψ increases due to increasing anisotropy, and when $\psi \sim T_{\mathrm{MF}}$ the re-entrant transition occurs. (b) Figure 2 shows the temperature dependence of the electronic specific heat in the presence of the pseudogap. At low (high) temperatures the specific heat is less (more) than the value in the absence of the pseudogap. For comparison the observed temperature dependence at 30 T [7] is also shown. The data are consistent with the requirement of the model that $\psi \sim T_{\mathrm{MF}} \sim 5$ K. (c) Calculation of the specific heat jump at the phase boundary requires knowledge of the temperature dependence of the pseudogap. The results for a simple model, based on Eq. (3), are shown in Fig. 2. The specific heat jump is significantly enhanced over the mean-field value of $1.43\gamma T$. This is consistent with the observed behavior at high temperatures, but inconsistent with the observed behavior at low temperatures. This disagreement could be because of the simplistic model used for the temperature dependence of ψ and because the SDW fluctuations can no longer be treated classically. (d) Because of the pseudogap the magnetoresistance will be approximately activated in temperature. (e) The thermopower data are just as consistent with a pseudogap as an absolute gap.

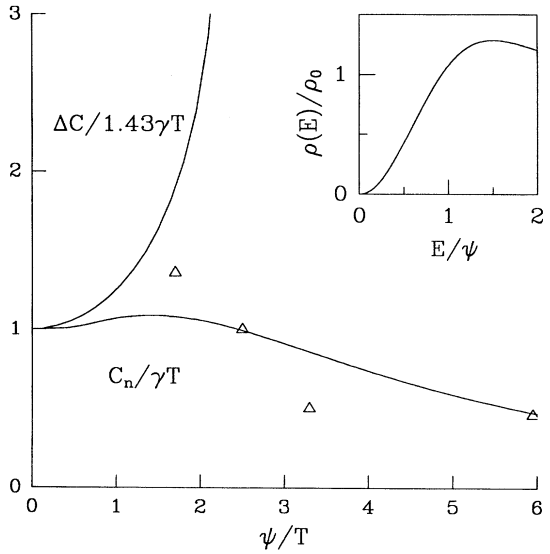

FIG. 2. Dependence of the normal state electronic specific heat C_n and the specific heat jump at the phase boundary ΔC on the pseudogap. At low (high) temperatures C_n is smaller (larger) than its value, γT, in the absence of the pseudogap. ΔC is enhanced compared to the mean-field value of $1.43\gamma T$. As the pseudogap ψ increases ΔC increases until $\psi \sim 2.6T$ where the transition becomes first order. The triangles are C_n data for $(TMTSF)_2ClO_4$ at 30 T [7], with $\psi = T_{\mathrm{MF}} = 5$ K. Inset: Pseudogap in the density of states near the transition temperature. The energy is relative to the Fermi energy. The density of states is normalized to the free-electron value ρ_0.

Several key experiments could test this model. Far-infrared or NMR measurements could reveal the pseudogap near the phase boundary. As the field increases above 27 T, ξ_a decreases, and the pseudogap will fill in [22]. The anion gap model [10] predicts that the re-entrant phase only exists due to the anion ordering in $(TMTSF)_2ClO_4$. In contrast the model presented here predicts the destruction

of SDW phases in *any* material at sufficiently high fields. The difference between the two models could be tested by searching for the re-entrant phase above 30 T in several materials. (i) $(TMTSF)_2ClO_4$: at pressures above 5 kbar the anion ordering is destroyed [3]. (ii) $(TMTSF)_2PF_6$ has no anion ordering. At ambient pressure $T_{SDW} \sim 12$ K up to 30 T. Some of these experiments are planned at the Australian National Pulsed Magnet Laboratory, which provides access to fields up to 60 T at temperatures down to 60 mK.

This theory is also applicable to the quasi-two-dimensional materials $(BEDT\text{-}TTF)_2MHg(SCN)_4$ (M = K, Rb, Tl). A coexisting quasi-one-dimensional Fermi surface is believed to be responsible for the formation of a density-wave state in these materials at low temperatures. There is some controversy as to whether this is a CDW or SDW state [2]. This state is destroyed above the "kink field," H_k [2,25]. (For M = Rb, H_k = 32 T.) The observed phase diagram is consistent with Fig. 1, including the observation of first-order hysteretic behavior near H_k at low temperatures [2,25].

In conclusion, a theory has been presented to show how fluctuations enhanced by a high magnetic field can destroy density-wave states in quasi-one-dimensional materials. The model is consistent with previously unexplained properties of $(TMTSF)_2ClO_4$ and $(BEDT\text{-}TTF)_2MHg(SCN)_4$ (M = K,Rb,Tl). For calculational and conceptual simplicity, the effects of anion ordering and imperfect nesting of the Fermi surface are neglected. A complete description of these materials must include the effects of anion ordering, imperfect nesting, and fluctuations. Finally, I hope this work will stimulate others to use sophisticated many-body techniques to test whether the model presented here gives a good description of the effects of spin-density-wave fluctuations on the electronic properties of quasi-one-dimensional systems.

This work was stimulated by the visit of J. S. Brooks, supported by NSF Grant No. DMR 92-14889, to the Australian National Pulsed Magnet Laboratory. I have benefited greatly from discussions with J. S. Brooks. I thank him for pointing out the relevance of this model to $(BEDT\text{-}TTF)_2MHg(SCN)_4$. Work at UNSW was supported by the Australian Research Council. Early stages of this work were performed at The Ohio State University and supported by the U.S. Department of Energy, Basic Energy Sciences, Division of Materials Science. I thank J. W. Wilkins, R. A. Lewis, and D. J. Evans for helpful discussions.

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[1] For a review, see T. Ishiguro and K. Yamaji, *Organic Superconductors* (Springer-Verlag, Berlin, 1990).
[2] J. S. Brooks, Mater. Res. Soc. Bull. **18**, 29 (1993).
[3] W. Kang, S. T. Hannahs, and P. M. Chaikin, Phys. Rev. Lett. **70**, 3091 (1993).
[4] L. P. Gor'kov and A. G. Lebed', J. Phys. (Paris), Lett. **45**, L433 (1984).
[5] M. J. Naughton *et al.*, Phys. Rev. Lett. **61**, 621 (1988).
[6] N. A. Fortune *et al.*, Phys. Rev. Lett. **64**, 2054 (1990).
[7] J. S. Brooks *et al.*, Mater. Res. Soc. Symp. Proc. **173**, 217 (1990).
[8] R. C. Yu *et al.*, Phys. Rev. Lett. **65**, 2458 (1990).
[9] Y. M. Yakovenko, Zh. Eksp. Teor. Fiz. **93**, 627 (1987) [Sov. Phys. JETP **66**, 355 (1987)]; Phys. Rev. Lett. **61**, 2276 (1988); L. Hubert and C. Bourbonnais, Synth. Met. **55–57**, 4231 (1993).
[10] A. G. Lebed' and P. Bak, Phys. Rev. B **40**, 11 433 (1989); T. Osada, S. Kagoshima, and N. Miura, Phys. Rev. Lett. **69**, 1117 (1992); L. P. Gor'kov and A. G. Lebed', Phys. Rev. B **51**, 3285 (1995).
[11] R. V. Chamberlin *et al.*, Phys. Rev. Lett. **61**, 2277 (1988).
[12] X. D. Shi, W. Kang, and P. M. Chaikin, Phys. Rev. B **50**, 1984 (1994).
[13] R. H. McKenzie and J. W. Wilkins, Phys. Rev. Lett. **69**, 1085 (1992); K. Kim, R. H. McKenzie, and J. W. Wilkins, *ibid.* **71**, 4015 (1993).
[14] F. H. Long, S. P. Love, B. I. Swanson, and R. H. McKenzie, Phys. Rev. Lett. **71**, 762 (1993); L. Degiorgi *et al.*, Phys. Rev. B **49**, 14 754 (1994).
[15] B. P. Gorshunov *et al.*, Phys. Rev. Lett. **73**, 308 (1994), and references therein.
[16] A. Bjeliš and K. Maki, Phys. Rev. B **44**, 6799 (1991).
[17] P. M. Chaikin and R. L. Greene, Phys. Today **39**, No. 5, 24 (1986), Fig. 7.
[18] J. B. Torrance, H. J. Pedersen, and K. Bechgaard, Phys. Rev. Lett. **49**, 881 (1982); W. M. Walsh *et al.*, *ibid.* **49**, 885 (1982).
[19] L. P. Le *et al.*, Phys. Rev. B **48**, 7284 (1993).
[20] H. J. Schulz, in *Low-Dimensional Conductors and Superconductors,* edited by D. Jérome and L. G. Caron (Plenum, New York, 1986), p. 95.
[21] Recent studies on $(TMTSF)_2AsF_6$ at fields up to 12 T [G. Kriza *et al.*, Europhys. Lett. **16**, 585 (1991)] and on $(TMTSF)_2PF_6$ at fields up to 7 T [J. Coroneus, B. Alavi, and S. E. Brown, Phys. Rev. Lett. **70**, 2332 (1993)] note that they do not see evidence of the predicted effects. However, since in these materials $T_{MF} \sim 12$ K, only a 10% change in ξ_b is expected at these fields.
[22] M. V. Sadovskiĭ, Zh. Eksp. Teor. Fiz. **77**, 2070 (1979) [Sov. Phys. JETP **50**, 989 (1979)].
[23] M. J. Rice and S. Strässler, Solid State Commun. **13**, 1389 (1973); P. A. Lee, T. M. Rice, and P. W. Anderson, Phys. Rev. Lett. **31**, 462 (1973).
[24] M. V. Sadovskiĭ, Zh. Eksp. Teor. Fiz. **66**, 1720 (1974) [Sov. Phys. JETP **39**, 845 (1974)].
[25] J. S. Brooks *et al.*, Phys. Rev. Lett. **69**, 156 (1992).