Field-induced spin-density-wave phases in TMTSF organic conductors: Quantization versus nonquantization

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We study the magnetic-field-induced spin-density-wave (FISDW) phases in TMTSF organic conductors in the framework of the quantized nesting model. In agreement with recent suggestions, we find that the SDW wave vector **Q** deviates from its quantized value near the transition temperature T_c for all phases with quantum numbers $N>0$. Deviations from quantization are more pronounced at low pressure and higher N and may lead to a suppression of the first-order transitions $N+1\rightarrow N$ for $N\geqslant 5$. Below a critical pressure, we find that the $N=0$ phase invades the entire phase diagram in accordance with earlier experiments. We also show that at $T=0$, the quantization of **Q** and hence the Hall conductance is always exact. Our results suggest a novel phase transition/crossover at intermediate temperatures between phases with quantized and nonquantized **Q**.

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I. INTRODUCTION

Quasi-one-dimensional (Q1D) organic conductors of the $(TMTSF)_2X$ family[1] (also known as the Bechgaard salts) are highly anisotropic crystals that consist of parallel conducting chains. The electron transfer integrals along the chains (in the a direction) and transverse to the chains (in the b and c directions) are typically $t_a=250$ meV, $t_b=25$ meV, and $t_c=0.75$ meV.[2] Because of the strong anisotropy, the Fermi surface of these materials is open and consists of two disconnected sheets located near $\pm k_F$, which are the Fermi momenta along the chains. In the presence of a moderate magnetic field H along the c axis, the interplay between the nesting property of the open Fermi surface and the quantization of electron orbits due to the magnetic field leads to a cascade of magnetic-field-induced spin-density-wave (FISDW) phases.[2] These phases have long been theoretically explained in the framework of the quantized nesting model (QNM).[2–8] A central prediction of the QNM is that within each FISDW phase characterized by an integer N, the wave vector $\mathbf{Q}=(Q_x,Q_y)$ of the spin modulation is quantized: $Q_x=2k_F+NG$, where $G=ebH/\hbar c$ is the magnetic wave vector and b the interchain distance. As the field increases, the integer N varies, which leads to the FISDW cascade ($N=\ldots,4,3,2,1,0$). In each phase, the quantization of Q_x implies the quantization of the Hall effect: $\sigma_{xy}=-2Ne^2/h$ per layer of TMTSF molecules.[9,10] The ability of the QNM to explain the quantum Hall effect (QHE) observed in the Bechgaard salts[11] is one of its main successes.

Recently Lebed' called into question some fundamental aspects of the QNM.[12] He showed that due to the particle-hole asymmetry in the FISDW phases with $N\neq 0$, Q_x deviates from its quantized values. At the metal-FISDW transition, deviations from quantization are controlled by the ratio $h=\omega_c/\pi T_c$ where $\omega_c=v_F G$ (v_F is the Fermi velocity along the chains) and T_c is the transition temperature. When h reaches a critical value h_c, the first-order transitions between different FISDW phases are suppressed. Q_x then becomes a continuous function of the field. At lower temperatures, first-order transitions (i.e., discontinuous jumps of Q_x) survive although Q_x is not quantized. Lebed's results call into question our theoretical understanding of the QHE in the Bechgaard salts, since the latter relies on the quantization of the FISDW wave vector.[9,10,13]

Lebed's conclusions raise two important questions. First, the very existence of the FISDW phases, which is due to a *quantum* effect of the field, requires $h=\omega_c/\pi T_c$ to be large enough. Indeed, when $T\gg\omega_c$, the magnetic field can be treated semiclassically and we expect the FISDW cascade to disappear in favor of either the metallic phase or the SDW with $Q_x=2k_F$ (i.e., the phase $N=0$). Thus, we expect the suppression of first-order phase transitions to occur only in a small window of the parameter h. Second, the fate of the QHE can be understood only by considering explicitly the low-temperature limit. The extrapolation of results valid near T_c, as done by Lebed', is not reliable since the SDW wave vector Q_x may vary with temperature.

In this paper, we investigate the FISDW phase diagram both at $T=T_c$ and $T=0$ as a function of the strength of the electron-electron interaction. The latter is a decreasing function of pressure and can therefore be varied experimentally. We find that Q_x deviates from its quantized value near T_c for all phases $N>0$. Deviations from quantization are stronger at low pressure and higher N. When pressure is decreased, suppression of first-order phase transitions occurs for $N\geqslant 5$. At lower pressure, below a critical value P_c, we find that the $N=0$ phase invades the entire phase diagram in accordance with earlier experiments.[14] On the other hand, at $T=0$ the quantization of Q_x and hence the Hall conductance is exact for all pressures and all N, down to the critical pressure P_c below which the $N=0$ phase again invades the phase diagram. Our results suggest a novel phase transition/crossover at intermediate temperature between phases with quantized and nonquantized Q_x.

II. METAL-FISDW TRANSITION

The Hamiltonian describing the Bechgaard salts in the vicinity of the Fermi energy in the presence of a magnetic

field $\mathbf{H}=H\hat{z}$ can be written as

$$\mathcal{H}=\sum_{\alpha,\sigma}\int d^2r\,\psi^{\dagger}_{\alpha\sigma}(\mathbf{r})[v_F(-i\alpha\partial_x-k_F) +t_{\perp}(-ib\partial_y-Gx)]\psi_{\alpha\sigma}(\mathbf{r}) +\frac{g}{2}\sum_{\alpha,\sigma,\sigma'}\int d^2r\,\psi^{\dagger}_{\alpha\sigma}(\mathbf{r})\psi^{\dagger}_{\bar{\alpha}\sigma'}(\mathbf{r})\psi_{\bar{\alpha}\sigma'}(\mathbf{r})\psi_{\alpha\sigma}(\mathbf{r}). \tag{1}$$

Here the operator $\psi^{(\dagger)}_{\alpha\sigma}(\mathbf{r})$ creates (annihilates) a right ($\alpha=+$) or left ($\alpha=-$) moving electron with spin σ. We use the notation $\mathbf{r}=(x,mb)$ (m integer) and $\int d^2r=b\Sigma_m\int dx$. $v_F=\sqrt{2}t_a a$ is the Fermi velocity along the chains (with t_a the hopping amplitude and a the lattice spacing) and g the amplitude of the electron-electron interaction. We have linearized the Hamiltonian around the Fermi energy and used the gauge $\mathbf{A}=(0,Hx,0)$. $t_{\perp}(u)=-2t_b\cos(u)-2t_{2b}\cos(2u)$ describes the interchain hopping in a tight-binding approximation, t_b being the nearest-neighbor hopping. The next-nearest neighbor hopping amplitude t_{2b} destroys the perfect nesting of the Fermi surface and stabilizes the metallic phase in the absence of magnetic field. Here and in the rest of this work $\hbar=c=1$. To obtain the phase diagram near T_c, we compute the static spin susceptibility $\chi(\mathbf{q})$ within the random-phase approximation: $\chi(\mathbf{q})=\chi_0(\mathbf{q})/[1-g\chi_0(\mathbf{q})]$, where $\chi_0(\mathbf{q})$ is the bare spin susceptibility. It can be written as[8]

$$\chi_0(\mathbf{q})=\sum_{n=-\infty}^{\infty} I_n^2(q_y)\chi_{1\mathrm{D}}(q_x-nG), \tag{2}$$

$$\chi_{1\mathrm{D}}(q_x)=\frac{N(0)}{2}\left[\ln\left(\frac{2\gamma E_0}{\pi T}\right)+\Psi\left(\frac{1}{2}\right) -\mathrm{Re}\Psi\left(\frac{1}{2}+\frac{v_F}{4i\pi T}(q_x-2k_F)\right)\right], \tag{3}$$

$$I_n(q_y)=\int_0^{2\pi}\frac{du}{2\pi}e^{inu+(i/\omega_c)[T_{\perp}(u+q_yb/2)+T_{\perp}(u-q_yb/2)]}, \tag{4}$$

where $T_{\perp}(u)=\int_0^u du' t_{\perp}(u')$, $N(0)=1/\pi v_F b$ is the density of states per spin, Ψ the digamma function, E_0 an ultraviolet cutoff of the order of the bandwidth, and $\gamma\simeq 1.781$ the exponential of the Euler constant. The instability to the FISDW phase occurs when the Stoner criterion $1-g\chi_0(\mathbf{q})=0$ is satisfied.

Since the 1D susceptibility has a logarithmic divergence for $q_x=2k_F$, Eq. (2) suggests that the SDW instability will occur with a quantized wave vector $\mathbf{Q}=(2k_F+NG,Q_y)$ (N integer). However, the parameter $N=(Q_x-2k_F)/G$ obtained from the Stoner criterion is not in general an integer. This can easily be shown analytically. Writing $Q_x=2k_F+(N+\epsilon)G$ with N integer and $\epsilon\ll 1$, the maximum of the susceptibility χ_0 (which will give the highest transition temperature) is found to be determined by

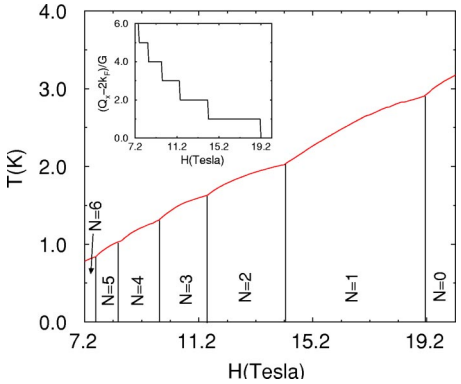

FIG. 1. Phase diagram for $\tilde{g}=0.38$. The inset shows the parameter $N=(Q_x-2k_F)/G$ at the metal-FISDW transition ($T=T_c$) as a function of the magnetic field. The vertical lines are guides to the eyes and indicate first-order transitions. [They do not correspond to the actual transition lines, which are not strictly vertical.]

$$\epsilon(T,Q_y)=\frac{8\pi^2T^2}{7\zeta(3)\omega_c^2}\sum_{n\neq 0}\frac{I_{N+n}^2}{nI_N^2}, \tag{5}$$

where ζ is the Riemann zeta function and $I_n\equiv I_n(Q_y)$. ϵ vanishes only in the phase $N=0$ (for which $Q_y=\pi/b$) due to the particle-hole symmetry which implies $I_n(\pi/b)=I_{-n}(\pi/b)$.

The phase diagram obtained by numerical solution of the Stoner criterion is shown in Figs. 1 and 2 for different values of the dimensionless interaction constant $\tilde{g}=gN(0)$. Since $\tilde{g}\propto 1/t_a$, increasing $\tilde{g}$ can be experimentally achieved by decreasing pressure.[15] For small $\tilde{g}$, the quantization of Q_x is essentially exact (see Fig. 1 obtained for $\tilde{g}=0.38$). Figure 2, which is obtained for $\tilde{g}=0.43$, shows that strong deviations from quantization appear for a sufficiently strong interaction. These deviations are more pronounced for high values of N. For $\tilde{g}=0.43$, the first-order transitions $N\simeq 6\rightarrow N\simeq 5, N\simeq 7\rightarrow N\simeq 6,\ldots$ are suppressed. The parameter $N=(Q_x-2k_F)/G$ varies continuously in the corresponding field

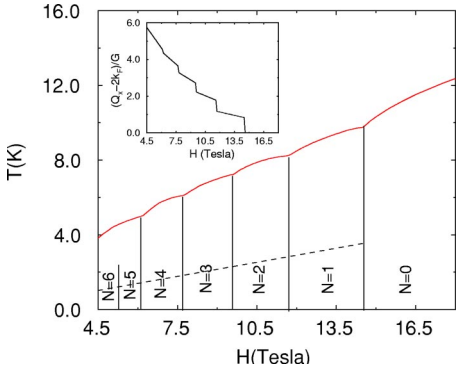

FIG. 2. Same as Fig. 1 for $\tilde{g}=0.43$. The first-order transition line between the phases $N=6$ and $N=5$ terminates by a second-order critical point above which N varies continuously with the field. The dashed line schematically indicates a phase transition/crossover at intermediate temperature between phases with quantized and nonquantized Q_x (see text).

range. Since Q_x is exactly quantized at $T=0$ (see below), the low-temperature first-order transition line between the phases $N=6$ and $N=5$ terminates by a second-order critical point above which the first-order transition is suppressed (Fig. 2). We find that first-order transitions $N+1\rightarrow N$ with $N<5$ are never suppressed. Indeed, if one increases $\tilde{g}$ beyond the critical value $\tilde{g}_c=0.433$, the phase $N=0$ invades the entire phase diagram. This latter result agrees with the experimental results showing that the same SDW phase is stable for any value of the field below a critical pressure $P_c\sim 6$ kbar.[14]

III. ZERO-TEMPERATURE PHASE DIAGRAM

To obtain the phase diagram at $T=0$, one should calculate the condensation energy of the system and look for its minimum as a function of **Q** (at fixed electron density). According to the QNM, each FISDW phase is characterized by a series of gaps $\Delta_n=gI_n^2\Delta$ where $\Delta=\langle\psi_{\downarrow -}^{\dagger}(\mathbf{r})\psi_{\uparrow +}(\mathbf{r})\rangle e^{-i\mathbf{Q}\cdot\mathbf{r}}$ is the SDW order parameter. The gap with the largest amplitude, Δ_N, opens up at the Fermi energy. Here we allow for a nonquantized wave vector $Q_x=2k_F+NG+z/v_F$ (N integer) and assume that $|z|\ll\Delta_N$. If Q_x is not quantized ($z\neq 0$), the particle number conservation implies a shift $\delta\mu=z+\mathrm{sgn}(z)(z^2+\Delta_N^2)^{1/2}$ of the chemical potential. As a result, the chemical potential does not lie in a gap (since $|\delta\mu|>\Delta_N$), and the Hall conductance is not quantized. In order to determine the value of z, we use the method of Ref. 8. We take into account the main gap Δ_N exactly, and consider the gaps $\Delta_{n\neq N}$ which open away from the Fermi level within perturbation theory. Skipping technical details, we obtain the condensation energy

$$\Delta E_N=-\frac{N(0)}{2}\Delta_N^2+N(0)[|z|(z^2+\Delta_N^2)^{1/2}-z^2], \qquad (6)$$

$$\frac{2}{\tilde{g}I_N^2}=\ln\frac{2E_0}{\Delta_N}+\sum_{n\neq 0}\frac{I_{N+n}^2}{I_N^2}\ln\frac{2E_0}{|n\omega_c|}-\mathrm{arcsinh}\frac{|z|}{\Delta_N}-\frac{z}{\omega_c}\sum_{n\neq 0}\frac{I_{N+n}^2}{nI_N^2}. \qquad (7)$$

Equation (6) shows that for a given value of Δ_N the energy is minimum for $z=0$. Therefore, in order to stabilize a phase with $z\neq 0$, a necessary condition is $\Delta_N(|z|>0)>\Delta_N(z=0)$, i.e. [see Eq. (7)],

$$\mathrm{arcsinh}\frac{|z|}{\Delta_N}+\sum_{n\neq 0}\frac{z}{\omega_c}\frac{I_{N+n}^2}{nI_N^2}<0. \qquad (8)$$

From Eq. (8), we conclude that a sufficient condition for Q_x to be quantized is

$$\alpha_N(H)=\left|\frac{\Delta_N}{\omega_c}\sum_{n\neq 0}\frac{I_{N+n}^2}{nI_N^2}\right|<1, \qquad (9)$$

where we have used $|z|\ll\Delta_N$. Given that the I_n coefficients satisfy the sum rule $\Sigma_n I_n^2=1$ and $\Delta_N\lesssim\omega_c$,[8] we expect the inequality (9) to be satisfied. Our numerical results for $\tilde{g}=0.43$ confirm this expectation (Fig. 3). We find that $\alpha_N(H)$ increases with N, but is always much less than unity. For $\tilde{g}<0.43$, α_N further decreases. We therefore conclude that, while it is never quantized near T_c for $N\neq 0$ [see Eq. (5)], Q_x is strictly quantized at $T=0$ for all values of N.

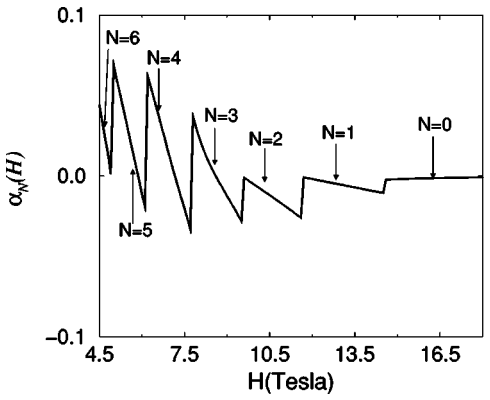

FIG. 3. $\alpha_N(H)$ [Eq. (9)] vs field for $\tilde{g}=0.43$. The condition $\alpha_N<1$ implies that quantization of Q_x and hence the Hall conductance is exact at $T=0$.

The zero-temperature phase diagram, obtained by solving Eqs. (6) and (7) is shown in Fig. 4 as a function of field and electron-electron interaction strength. For $T=0$, we find that the $N=0$ phase again invades the phase diagram at $\tilde{g}_c=0.433\pm 0.001$. At low temperature, corrections to the $T=0$ condensation energy will be exponentially small ($\propto e^{-\Delta_N/T}$). Thus, the quantization of Q_x will persist in a finite temperature range. This implies that for $\tilde{g}\lesssim\tilde{g}_c$, at some intermediate temperature $T^*(g,H)$ between $T=0$ and $T=T_c$, there must be a phase transition or a crossover between phases with quantized and nonquantized Q_x. This phase transition/crossover is schematically indicated by a dotted line in Fig. 2. The details of this transition/crossover is beyond the scope of our present study.

IV. COMPARISON WITH PREVIOUS THEORETICAL RESULTS

The overall phase diagram that we obtain is therefore qualitatively different from those obtained in the previous

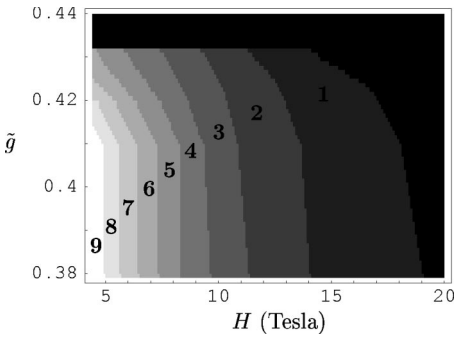

FIG. 4. Zero-temperature phase diagram showing the quantum number N vs field and interaction strength $\tilde{g}$. For $\tilde{g}>\tilde{g}_c=0.433$, the phase $N=0$ (black area) becomes stable for all values of H.

studies.[3–8,12] Near the metal-FISDW transition, we find that Q_x deviates from its quantized values, in accordance with Ref. 12. However, in contrast to Ref. 12, our study indicates that this deviation is large enough to suppress the first-order transitions only in a very limited region of the phase diagram corresponding to high values of N and $\tilde{g}$ close to $\tilde{g}_c$ (Fig. 2). In Ref. 12, it is assumed that $Q_y=\pi/b$. When $N\neq 0$, this assumption is not correct and one has to look for the value of Q_y which maximizes the transition temperature. Even with the assumption $Q_y=\pi/b$, we are unable to reproduce Lebed's results. Instead of the FISDW cascade, we find that only the phase $N=0$ is stable at low temperature albeit with a very low T_c. Furthermore, at low temperature ($T\ll T_c$), we find that the quantization of Q_x is exact (implying the quantization of the $T=0$ Hall conductance), which contradicts the prediction of Ref. 12 based on an extrapolation of results obtained near T_c. This suggests a novel phase transition/crossover at intermediate temperatures between phases with quantized and non-quantized Q_x. Also, below a critical pressure, we find that the $N=0$ phase invades the entire phase diagram.

V. COMPARISON WITH EXPERIMENTS

The overall phase diagram that we obtain agrees with the experimental observations in the compound $(TMTSF)_2PF_6$.[14] Above a critical pressure P_c (which corresponds to $\tilde{g}<\tilde{g}_c$ in our theoretical analysis), we describe the cascade of FISDW phases. When $P<P_c$ we find that the phase $N=0$ invades the entire phase diagram. Thus our study shows that the SDW phase below P_c is nothing else but the phase $N=0$ of the FISDW cascade (Fig. 4). This is also the conclusion obtained in Ref. 14. To our knowledge, the sudden disappearance of the FISDW cascade below the critical pressure P_c has not been explained before.

Recent magnetoresistance measurements by Kornilov *et al.*[16] found that hysteretic behavior occurs at low temperature at the transitions between successive FISDW phases. The hysteresis weakens at higher temperature and disappears above a characteristic temperature T_0 ($T_0<T_c$) for all $N>0$. This behavior was ascribed to the suppression of the first-order transitions in the temperature range $T_0\lesssim T\lesssim T_c$ in agreement with Lebed's predictions.[12] However, this interpretation is inconsistent with our result that the first-order phase transitions can be suppressed only for $N\geqslant 5$. We cannot exclude, even if it seems quite unlikely, that in a more realistic model (for instance, taking account of the triclinic structure of the Bechgaard salts) the suppression of the first-order phase transitions would also occur for $N<5$. In our opinion, the conclusion that the absence of hysteresis observed in experiments originates from the suppression of the first-order transitions should be taken cautiously. Such an absence of hysteresis could also be due to the weak first-order character of the transitions near T_c as was originally thought.[11] Our results suggest to perform experimental studies close to P_c, since the suppression of the first-order transitions should primarily be observed in the close vicinity of the critical pressure P_c (i.e., $P\gtrsim P_c$) below which the FISDW cascade disappears.

ACKNOWLEDGMENTS

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