

Sign Reversals of the Quantum Hall Effect and Helicoidal Magnetic-Field-Induced Spin-Density Waves in Quasi-One-Dimensional Organic Conductors

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We study the effect of umklapp scattering on the magnetic-field-induced spin-density-wave phases, which are experimentally observed in the quasi-one-dimensional organic conductors of the Bechgaard salts family. Within the framework of the quantized nesting model, we show that umklapp processes may naturally explain sign reversals of the quantum Hall effect (QHE) observed in these conductors. Moreover, umklapp scattering can change the polarization of the spin-density wave (SDW) from linear (sinusoidal SDW) to circular (helicoidal SDW). The QHE vanishes in the helicoidal phases, but a magnetoelectric effect appears. [S0031-9007(98)05849-9]

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The organic conductors of the Bechgaard salts family $(\text{TMTSF})_2X$ (where TMTSF stands for tetramethyltetraselenafulvalene) exhibit a rich phase diagram when temperature, magnetic field, or pressure are varied. In three members of this family ($X = \text{ClO}_4, \text{PF}_6, \text{ReO}_4$), a moderate magnetic field above several tesla destroys the metallic phase and induces a series of spin-density-wave (SDW) phases separated by first-order phase transitions [1]. Because of a strong quasi-one-dimensional anisotropy (the typical ratio of the electron transfer integrals in the three crystal directions is $t_a:t_b:t_c = 3000:300:10$ K), the Fermi surfaces of these materials are open. According to the so-called quantized nesting model (QNM) [1,2], the formation of the magnetic-field-induced spin-density waves (FISDW) results from an interplay between the nesting properties of the Fermi surface and the quantization of the electronic orbits in magnetic field. The wave vector of a FISDW adjusts itself to the magnetic field so that unpaired electrons completely fill an integer number of Landau levels; thus the Hall effect is quantized [3,4]. The standard QNM [2] predicts the Hall plateaus of the same sign, referred to as positive by convention, which agrees with most experiments. However, at certain pressures, a negative Hall effect is also observed [5–7]. In order to explain the sign reversals of the quantum Hall effect (QHE), Zanchi and Montambaux [8] invoke the variation of the electron dispersion law with pressure.

In this Letter, we study the effects of umklapp scattering on the FISDW phases within the framework of the QNM. Because the electron band in the $(\text{TMTSF})_2X$ materials is half-filled, the electrons are allowed to transfer the momentum $4k_F$ along the chains (k_F being the Fermi momentum) to the crystal lattice. Therefore, the interaction between electrons should include not only forward (g_2) and backward (g_1) scattering amplitudes, but also umklapp scattering amplitude (g_3) [9]. We demonstrate that, in the presence of umklapp interaction, FISDW phases with a negative QHE appear. This effect provides an alternative explanation for the sign reversals of the QHE observed in the Bechgaard salts. It differs from the

one suggested by Zanchi and Montambaux [8] in invoking the pressure dependence of g_3 rather than the electron band structure. The umklapp scattering amplitude g_3 is sensitive to pressure, because it is related to the dimerization in the crystal structure of the TMTSF chains. Moreover, we show that the polarization of the FISDW may change from linear (sinusoidal SDW) to circular (helicoidal SDW) because of umklapp interaction. The QHE vanishes in the helicoidal phases, but a magnetoelectric effect appears. These two properties are characteristic of the helicoidal phases and can be utilized to detect them experimentally. The effect of umklapp on the FISDW phases was studied before by Lebed' [10,11] using rather crude approximations, but the helicoidal phases and the sign reversals of the QHE were not discussed.

In the vicinity of the Fermi energy, the electron dispersion law in the Bechgaard salts is approximated as

$$E(k_x, k_y) = v_F(|k_x| - k_F) + t_\perp(k_y, b), \quad (1)$$

where k_x and k_y are the electron momenta along and across the one-dimensional chains of TMTSF, and $\hbar = 1$. In Eq. (1), the longitudinal electron dispersion is linearized in k_x in the vicinity of the two one-dimensional Fermi points $\pm k_F$, and $v_F = 2at_a \sin(k_F a)$ is the corresponding Fermi velocity, a being the lattice spacing along the chains. For the transverse electron dispersion, a tight-binding approximation is used:

$$t_\perp(k_y, b) = -2t_b \cos(k_y b) - 2t_{2b} \cos(2k_y b) - 2t_{3b} \cos(3k_y b) - 2t_{4b} \cos(4k_y b), \quad (2)$$

where b is the distance between the chains. The electron dispersion in the third direction along the z axis is not important for the following and is not considered here.

When a magnetic field H is applied along the z axis perpendicular to the (x, y) plane, it quantizes the transverse electron motion into the Wannier-Stark ladder [12]. Consequently, the static spin susceptibility $\chi_0(\mathbf{q})$, calculated at a wave vector $\mathbf{q} = (q_x, q_y)$ in the absence of interaction between electrons, diverges logarithmically at

quantized values of the longitudinal wave vector $q_x^{(n)} = 2k_F + nG$ (n integer) [1,13]:

$$\chi_0(\mathbf{q}) = \sum_n I_n^2(q_y) \chi_{1D}(q_x - nG). \quad (3)$$

Here $G = eHb/\hbar$ is the characteristic wave vector of the magnetic field (e is the electron charge), and $\chi_{1D}(q_x)$ is the susceptibility of a 1D system without interaction. The coefficients I_n depend on the transverse dispersion law of electrons:

$$I_n(q_y) = \langle e^{inu + \frac{i}{v_F G} [T_\perp(u+q_y b/2) + T_\perp(u-q_y b/2)]} \rangle, \quad (4)$$

where $T_\perp(u) = \int_0^u du' t_\perp(u')$, and $\langle \dots \rangle$ denotes averaging over u . In the absence of umklapp scattering, the transition temperature of the FISDW is determined by the Stoner criterion $1 - g_2 \chi_0(Q_x^{(N)}, Q_y) = 0$. The quantized longitudinal wave vector $Q_x^{(N)} = 2k_F + NG$ and the transverse wave vector Q_y are selected to maximize the transition temperature $T_c^{(N)}$ at a given magnetic field. Except when $N = 0$, Q_y is incommensurate: $Q_y \neq \pi/b$. The integer parameter N also determines the quantum Hall conductivity in the FISDW phase: $\sigma_{xy} = -2Ne^2/h$ per one layer of the TMTSF molecules [3,4]. As the magnetic field increases, the value of N changes, which leads to a cascade of FISDW transitions [1,2].

Umklapp scattering mixes the wave vectors $Q_x^{(N)}$ and $Q_x^{(N)} - 4k_F = -Q_x^{(-N)}$; thus two SDWs, with the wave vectors $\mathbf{Q}_N = (Q_x^{(N)}, Q_y)$ and $\mathbf{Q}_{-N} = (Q_x^{(-N)}, -Q_y)$, form simultaneously [10,11]. In the random-phase approximation, the critical temperature $T_c^{(N)}$ is determined by the modified Stoner criterion [14]:

$$[1 - g_2 \chi_0(\mathbf{Q}_N)][1 - ag_2 \chi_0(\mathbf{Q}_{-N})] - g_3^2 \chi_0(\mathbf{Q}_N) \chi_0(\mathbf{Q}_{-N}) = 0. \quad (5)$$

Of the two integers N and $-N$, we select N such that $\chi_0(\mathbf{Q}_N) > \chi_0(\mathbf{Q}_{-N})$ to label each FISDW phase.

Below $T_c^{(N)}$, the system is characterized by the order parameters $\Delta_{\beta N, \alpha}$:

$$\langle \hat{\psi}_{\alpha, \uparrow}^\dagger(\mathbf{r}) \hat{\psi}_{-\alpha, \downarrow}(\mathbf{r}) \rangle = \sum_{\beta=\pm} \Delta_{\beta N, \alpha} e^{-i\alpha \mathbf{r} \cdot \mathbf{Q}_{\beta N}}, \quad (6)$$

where $\mathbf{r} = (x, y)$ is the spatial coordinate. In Eq. (6), the index $\beta = \pm$ labels the wave vectors $\mathbf{Q}_{\pm N}$. The operators $\hat{\psi}_{\alpha, \sigma}^{(\dagger)}$ annihilate (create) electrons with spin σ and momenta close to αk_F ($\alpha = \pm$). The electron spin density has a nonzero expectation value varying in space [15]:

$$\begin{aligned} \langle S_x(\mathbf{r}) \rangle &= \sum_{\beta=\pm} m_{\beta N}^{(x)} \cos(\mathbf{r} \cdot \mathbf{Q}_{\beta N} + \theta_{\beta N}^{(x)}), \\ \langle S_y(\mathbf{r}) \rangle &= \sum_{\beta=\pm} m_{\beta N}^{(y)} \cos(\mathbf{r} \cdot \mathbf{Q}_{\beta N} + \theta_{\beta N}^{(y)}). \end{aligned} \quad (7)$$

When $\theta_{\beta N}^{(x)} = \theta_{\beta N}^{(y)}$, Eq. (7) describes sinusoidal SDWs. When $m_{\beta N}^{(x)} = m_{\beta N}^{(y)}$ and $\theta_{\beta N}^{(x)} = \theta_{\beta N}^{(y)} \pm \pi/2$, it describes

helicoidal SDWs. The spin polarization vector $\langle \mathbf{S}(\mathbf{r}) \rangle$ of a generic helicoidal SDW rotates in the plane perpendicular to a vector \mathbf{n} when the coordinate \mathbf{r} varies. In our case, because of the Zeeman effect, the vector \mathbf{n} aligns itself with the magnetic field \mathbf{H} ; thus the spin polarization (7) rotates in the (x, y) plane.

The actual polarization of the SDWs is determined by minimizing the free energy of the system. In terms of the linear combinations of the order parameters,

$$\tilde{\Delta}_{\beta N, \alpha} = I_{\beta N}(Q_y) (g_2 \Delta_{\beta N, \alpha} + g_3 \Delta_{-\beta N, -\alpha}), \quad (8)$$

the Landau expansion of the free energy in the vicinity of $T_c^{(N)}$ for the phase N has the following form [16]:

$$\begin{aligned} F_N = \sum_{\alpha} \left[\sum_{\beta} A_{\beta N} |\tilde{\Delta}_{\beta N, \alpha}|^2 + B (\tilde{\Delta}_{N, \alpha} \tilde{\Delta}_{-N, -\alpha}^* + \text{c.c.}) \right. \\ \left. + (K/2) \sum_{\beta} |\tilde{\Delta}_{\beta N, \alpha}|^4 + 2K |\tilde{\Delta}_{N, \alpha} \tilde{\Delta}_{-N, \alpha}|^2 \right], \end{aligned} \quad (9)$$

where the coefficients are

$$A_{\beta N} = \frac{1}{I_{\beta N}^2(Q_y)} \left(\frac{g_2}{g_2^2 - g_3^2} - \chi_0(\mathbf{Q}_{\beta N}) \right), \quad (10)$$

$$B = -g_3 / I_N(Q_y) I_{-N}(Q_y) (g_2^2 - g_3^2), \quad (11)$$

$$K = 7\zeta(3)/16\pi^3 v_F b T^2, \quad \zeta(3) \approx 1.20. \quad (12)$$

As long as the quadratic part of the free energy F_N (9) is positively defined, the metallic state is stable. The second-order phase transition into a FISDW state takes place when the determinant of the quadratic part of Eq. (9) vanishes: $A_N A_{-N} = B^2$. With the coefficients A and B given by Eqs. (10) and (11), this condition is equivalent to the Stoner equation (5). Minimizing the free energy F_N (9) with respect to $\tilde{\Delta}_{\pm N, \pm}$ at $T < T_c^{(N)}$, we find two types of solutions depending on the value of g_3 . For small g_3 , when $\sqrt{2}|B| < |A_N - A_{-N}|$, we find a solution where $|\tilde{\Delta}_{N, +}| = |\tilde{\Delta}_{N, -}|$ and $|\tilde{\Delta}_{-N, +}| = |\tilde{\Delta}_{-N, -}|$, which corresponds to two sinusoidal SDWs. When g_3 exceeds a certain critical value so that $\sqrt{2}|B| > |A_N - A_{-N}|$, the minimum of the free energy is reached at $\tilde{\Delta}_{N, -} = \tilde{\Delta}_{-N, +} = 0$ and $|\tilde{\Delta}_{N, +}|, |\tilde{\Delta}_{-N, -}| \neq 0$, which corresponds to two helicoidal SDWs of opposite chiralities. Using the method of Ref. [4], we find that the QHE is quantized in the sinusoidal phase, $\sigma_{xy} = -2Ne^2/h$, but vanishes in the helicoidal phase. We also find that, when the magnetic field is varied, phase transitions between adjacent FISDW phases, whether sinusoidal or helicoidal, are of the first order [17]. The conclusion of Lebed' [10] that, in the presence of umklapp scattering, adjacent FISDW phases are separated by two second-order phase transitions and an intermediate phase with coexistence of four SDWs is incorrect, because he did not consider helicoidal SDWs.

Having gained an analytical insight into the problem, we study the phase diagram in the presence of umklapp scattering numerically. It is convenient to characterize the interaction by dimensionless coupling constants

$\tilde{g}_2 = g_2/\pi v_F b$ and $\tilde{g}_3 = g_3/\pi v_F b$. We vary the ratio \tilde{g}_3/\tilde{g}_2 while keeping the sum constant: $\tilde{g}_2 + \tilde{g}_3 = 2/\ln(2\gamma E_0/\pi T_c^{(\infty)})$, where $T_c^{(\infty)} = 12$ K is the transition temperature for an infinite magnetic field, E_0 is an ultraviolet cutoff of the order of t_a , and $\gamma \approx 1.781$ is the exponential of the Euler constant. The calculations are performed for $t_b = 300$ K, $t_{2b} = 20$ K, $t_{3b} = 0$ K, $t_{4b} = 0.75$ K, and $E_0 = 2000$ K. The transition temperature $T_c^{(N)}$ is obtained numerically from Eq. (5), and the polarizations of the SDWs for $T < T_c^{(N)}$ are determined by minimizing the free energy F_N given by Eq. (9).

A very small g_3 does not change the phase diagram qualitatively compared to the case $g_3 = 0$. Now the main SDW at the wave vector \mathbf{Q}_N coexists with a weak SDW at the wave vector \mathbf{Q}_{-N} . In general, the value of Q_y that maximizes $\chi_0(\mathbf{Q}_N)$ does not maximize $\chi_0(\mathbf{Q}_{-N})$, so $\chi_0(\mathbf{Q}_{-N}) \ll \chi_0(\mathbf{Q}_N)$. As a result, the SDW amplitude at the wave vector \mathbf{Q}_{-N} is very small, and the polarizations of the SDWs are linear. The values of N follow the usual ‘‘positive’’ sequence $N = \dots, 6, 5, 4, 3, 2, 1, 0$ with increasing magnetic field.

A larger value of g_3 increases the coupling between the two SDWs. This leads to a strong decrease of the critical temperature or even the disappearance of the SDWs. However, for even N , there exists a critical value of g_3 above which the system prefers to choose the transversely commensurate wave vector $Q_y = \pi/b$ for both SDWs. The reason is that, for even N (as opposed to odd N), $Q_y = \pi/b$ corresponds to a local maximum of the susceptibilities and $\chi_0(Q_x^{(-N)}, \pi/b) \approx \chi_0(Q_x^{(-N)}, \pi/b)$. The latter relation implies that the two SDWs have comparable amplitudes. The two susceptibilities are strictly equal at $t_{4b} = 0$, but when $t_{4b} > 0$, $\chi_0(Q_x^{(-N)}, \pi/b) > \chi_0(Q_x^{(N)}, \pi/b)$ (this result holds also for $t_{3b} \neq 0$) [8]. This yields a negative Hall plateau, provided the SDWs are sinusoidal. Thus, for $g_3/g_2 = 0.03$ ($\tilde{g}_2 \approx 0.37$ and $\tilde{g}_3 \approx 0.01$), we find the sequence $N = \dots, 6, 5, 4, -2, 2, 1, 0$ (see Fig. 1). The phase $N = 3$ is suppressed, and the negative commensurate phase with $N = -2$ and $Q_y = \pi/b$ appears in the cascade. All the phases are sinusoidal, so the Hall effect is quantized ($\sigma_{xy} = -2Ne^2/h$).

The strength of umklapp scattering is very sensitive to pressure, because hydrostatic pressure reduces the dimerization in the crystal structure of the TMTSF chains, which diminishes g_3 . Therefore, we conclude that the sign reversals of the QHE can be induced by varying pressure. In our simplified model, this effect requires $t_{4b} > 0$. Our results provide a new explanation of the negative Hall plateaus in the Bechgaard salts.

If g_3/g_2 is increased to 0.06, a second negative phase ($N = -4$) appears, and the cascade becomes $N = \dots, 8, 7, -4, 6, 5, 4, -2, 2, 1, 0$ (see Fig. 2). As discussed in Ref. [8], $N = -2$ and $N = -4$ correspond to the two negative QHE phases observed in experiments [6,7]. As shown in Fig. 2, the phase $N = -2$ splits into two subphases: helicoidal (the dark shaded area) and sinusoidal (the light shaded area).

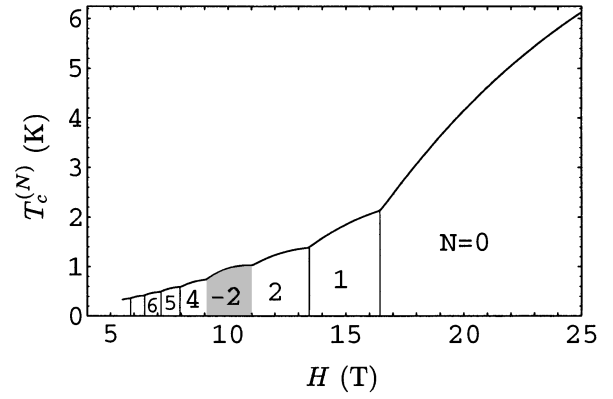


FIG. 1. Phase diagram for $g_3/g_2 = 0.03$ ($\tilde{g}_2 \approx 0.37$ and $\tilde{g}_3 \approx 0.01$). The phase $N = 3$ is suppressed, and the negative commensurate phase with $N = -2$ and $Q_y = \pi/b$ appears in the cascade (the shaded area). All the phases are sinusoidal, and the Hall effect is quantized: $\sigma_{xy} = -2Ne^2/h$. The vertical lines are only guides for the eyes and do not necessarily correspond to the actual first-order transition lines.

into two subphases: helicoidal and sinusoidal. *Not only does umklapp scattering stabilize negative phases, but, as g_3 increases, these negative phases are likely to become helicoidal.* Therefore, in order to observe the helicoidal phase experimentally, it would be desirable to stabilize the negative phase $N = -2$ at the lowest possible pressure (which corresponds to the strongest g_3). In (TMTSF)₂PF₆, the pressure has to be higher than 6 kbar, since the FISDW cascade disappears below this pressure [1]. In the experiment [7], where the phase $N = -2$ has been observed at 8.3 kbar, the pressure could be reduced by only about 2 kbar. Nevertheless, such a pressure reduction could induce a significant increase of g_3 . (TMTSF)₂ReO₄, where the sign reversals of the QHE have been observed under pressure [18], could also be a good candidate for detecting helicoidal phases.

The helicoidal FISDW phases exhibit a kinetic magnetoelectric effect and vanishing QHE. The magnetoelectric

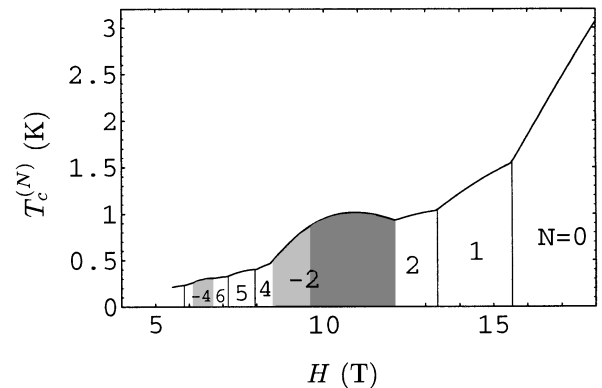


FIG. 2. Phase diagram for $g_3/g_2 = 0.06$ ($\tilde{g}_2 \approx 0.36$ and $\tilde{g}_3 \approx 0.02$). Two negative phases, $N = -2$ and $N = -4$, are observed (the shaded areas). The phase $N = -2$ splits into two subphases: helicoidal (the dark shaded area) and sinusoidal (the light shaded area).

effect may exist if time-reversal and space-inversion symmetries are broken [19]. Gor'kov and Sokol found the kinetic magnetoelectric effect for a single helicoidal SDW [20]. The effect also exists in the presence of two helicoidal SDWs of opposite chiralities, provided their amplitudes are not equal. An electric current along the chains, j_x , induces a uniform magnetization \mathbf{M} along the vector \mathbf{n} that characterizes the spin polarization of a helicoidal SDW. In our case, the vector \mathbf{n} is parallel to the magnetic field \mathbf{H} , which is oriented along the z axis; thus $M_z \propto j_x$. (Here M_z is the additional spin magnetization induced by j_x in excess of the magnetization induced by the magnetic field without j_x .) The effect can be understood by considering the spectrum of electronic excitations in the helicoidal FISDW phase shown in Fig. 3. The $+k_F$ electrons with spin up and the $-k_F$ electrons with spin down have the energy gap $\Delta_1 = |\tilde{\Delta}_{N,+}|$, whereas the $+k_F$ electrons with spin down and the $-k_F$ electrons with spin up have the different energy gap $\Delta_2 = |\tilde{\Delta}_{N,-}|$. To produce a current j_x along the chains, electrons need to be transferred from $-k_F$ to $+k_F$. For $\Delta_1 \neq \Delta_2$ ($\Delta_1 \neq \Delta_2$ if $t_{4b} \neq 0$), this redistribution of electrons results in a uniform magnetization M_z :

$$j_x = ev_F \sum_{\sigma} (\delta n_{+, \sigma} - \delta n_{-, \sigma}), \quad (13)$$

$$M_z = \frac{g\mu_B}{2} \sum_{\alpha=\pm} (\delta n_{\alpha, \uparrow} - \delta n_{\alpha, \downarrow}),$$

where $\delta n_{\alpha, \sigma}$ is the deviation of the distribution function of electrons with spin σ and momenta near αk_F from the equilibrium one, g is the electron gyromagnetic factor, and μ_B is the Bohr magneton. At low temperature, we can consider solely the electrons excited above the lowest gap (Δ_2 in Fig. 2). This implies that $\delta n_{+, \uparrow} \approx \delta n_{-, \downarrow} \approx 0$ and $M_z/j_x \approx -g\mu_B/2ev_F$.

In conclusion, we have shown that the negative phases (i.e., with a sign reversal of the QHE) observed in the Bechgaard salts can be explained by considering umklapp processes. These phases are characterized by the coexistence of two linearly polarized SDWs (with the wave vectors \mathbf{Q}_N and \mathbf{Q}_{-N}) with comparable amplitudes. We have also

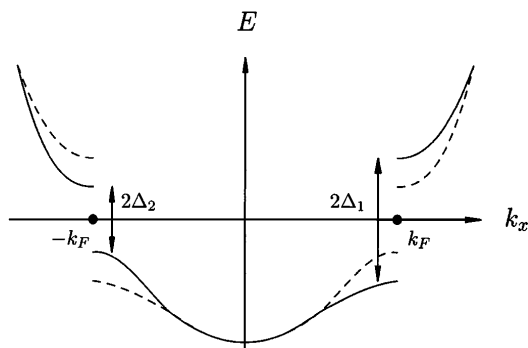


FIG. 3. Spectrum of electronic excitations in the helicoidal FISDW phase. The solid (dashed) line corresponds to up (down) spins.

shown that these negative phases are likely to become helicoidal under low pressure. The helicoidal phases have no QHE and exhibit a magnetoelectric effect. The latter effect can be utilized to detect the helicoidal phases experimentally by looking for a spin magnetization proportional to the current along the chains.

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