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 effects 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. These two characteristic properties may be utilized to detect the magnetic-field-induced helicoidal SDW phases experimentally.

The organic conductors of the Bechgaard salts family (TMTSF)$_2$X (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=ClO$_2$, PF$_6$, 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. Because the electron band in the (TMTSF) family agrees with most experiments. However, at certain pressures, a negative Hall effect is also observed in the Bechgaard salts. 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 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 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

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

where $k_x$ and $k_y$ are the electron momenta along and across the one-dimensional chains of TMTSF, and $t_\perp = 1$. 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 $v_F = 2a t_d \sin(k_F a)$ is the corresponding Fermi velocity, $a$ being the lattice spacing along the chains. For the transverse electron dispersion, a tight-binding approximation is used:

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

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. Consequently, the static spin susceptibility $\chi_0(q)$, calculated at a wave vector $q = (q_x, q_y)$ 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) \[ 13 \]:
\[ \chi_0(q) = \sum_n I_n^2(q_y) \chi_{1D}(q_x - nG). \] (3)

Here \( G = eHb/h \) 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:
\[ I_n(q_y) = \langle e^{i n u + \int_0^q d\nu' t_\perp(u')} \cdot T_{\perp}(u + q_y b/2) + T_{\perp}(u - q_y b/2) \rangle, \] (4)
where \( T_{\perp}(u) = \int_0^u d\nu' t_\perp(u') \), and \( \langle \cdot \cdot \cdot \rangle \) denotes averaging over \( u \). In the absence of umklapp scattering, the transition temperature of the FISDW is determined by the Stoner criterion
\[ 1 - g_2 \chi_0(Q_y) = 0. \] (5)
The quantized longitudinal wave vector \( Q_y^{(N)} = 2k_F + NG \) 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 magnetic field changes, which lead to a cascade of FISDW transitions \[ 12 \].

Umklapp scattering mixes the wave vectors \( Q_x^{(N)} \) and \( Q_y^{(N)} - 4k_F = -Q_y^{(-N)} \), thus two SDWs, with the wave vectors \( Q_N = (Q_x^{(N)}, Q_y) \) and \( Q_{-N} = (Q_x^{(-N)}, -Q_y), \) form simultaneously \[ 10,11 \]. In the random-phase approximation, the critical temperature \( T_c^{(N)} \) is determined by the modified Stoner criterion \[ 4 \]:
\[ [1 - g_2 \chi_0(Q_N)] [1 - g_2 \chi_0(Q_{-N})] - g_2^2 \chi_0(Q_N) \chi_0(Q_{-N}) = 0. \] (6)

Of the two integers \( N \) and \( -N \), we select \( N \) such that \( \chi_0(Q_N) > \chi_0(Q_{-N}) \) to label each FISDW phase.

Below \( T_c^{(N)} \), the system is characterized by the order parameters \( \Delta_{\beta N}; \alpha \):
\[ \langle \psi_{\alpha, \pm}^\dagger \psi_{\alpha, \perp} \rangle = \sum_{\beta=\pm} \Delta_{\beta N, \alpha} e^{-i \alpha \sigma} Q_{\beta N}, \] (6)
where \( \mathbf{r} = (x, y) \) is the spatial coordinate. In Eq. \[ 6 \], the index \( \beta = \pm \) labels the wave vectors \( Q_{\beta N} \). The operators \( \psi_{\alpha, \sigma} \) annihilate (create) electrons with spin \( \sigma \) and momenta close to \( \alpha k_F \) (\( \sigma = \pm \)). The electron spin density has a nonzero expectation value varying in space \[ 13 \]:
\[ \langle S_\sigma(r) \rangle = \sum_{\beta=\pm} m_{\beta N}^{(x)} \cos(\mathbf{r} \cdot \mathbf{Q}_{\beta N} + \theta_{\beta N}^{(x)}), \] (7)
\[ \langle S_\sigma(r) \rangle = \sum_{\beta=\pm} m_{\beta N}^{(y)} \cos(\mathbf{r} \cdot \mathbf{Q}_{\beta N} + \theta_{\beta N}^{(y)}), \] (7)

When \( \theta_{\beta N}^{(x)} = \theta_{\beta N}^{(y)} \), Eq. \[ 6 \] 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 \( \mathbf{S}(\mathbf{r}) \) 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 \( \mathbf{S}(\mathbf{r}) \) 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:
\[ \Delta_{\beta N, \alpha} = I_{\beta N}(Q_y) (g_2 \Delta_{\beta N, \alpha} + g_3 \Delta_{-\beta N, -\alpha}), \] (8)
the Landau expansion of the free energy in the vicinity of \( T_c^{(N)} \) for the phase \( N \) has the following form \[ 10 \]:
\[ F_N(\mathbf{Q}) = \sum_{\alpha} \sum_{\beta} A_{\beta N} |\Delta_{\beta N, \alpha}|^2 + B(\Delta_{\beta N, \alpha} \Delta^*_{-\beta N, -\alpha} + c.c.) \] (9)
\[ + (K/2) \sum_{\beta} |\Delta_{\beta N, \alpha}|^4 + 2K |\Delta_{\beta N, \alpha} \Delta_{-\beta N, -\alpha}|^2 \] (9)
where the coefficients are
\[ A_{\beta N} = \frac{1}{g_2^2 - g_3^2} \chi_0(Q_{\beta N}) \] (10)
\[ B = -g_3/[N(Q_y)] I - N(Q_y)(g_2^2 - g_5^2), \] (11)
\[ K = 7\zeta(3)/16 \pi^3 v_F bT^2, \] (12)
\[ \zeta(3) \approx 1.20. \] (12)

As long as the quadratic part of the free energy \( F_N \) 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. \[ 10 \] 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 \[ 7 \]. Minimizing the free energy \( F_N \) \[ 8 \] with respect to \( \Delta_{N, \pm} \) at \( T < T_c^{(N)} \), we find two types of solutions depending on the value of \( g_3 \). For small \( g_3 \), when \( \sqrt{2} B \leq |A_N - A_{-N}| \), we find a solution where \( |\Delta_{N, +} | = |\Delta_{N, -} | \) and \( |\Delta_{-N, +} | = |\Delta_{-N, -} | \), which corresponds to two sinusoidal SDWs. When \( g_3 \rneds \) exceeds a certain critical value so that \( \sqrt{2} B > |A_N - A_{-N}| \), the minimum of the free energy is reached at \( \Delta_{N, +} = \Delta_{N, -} = 0 \) and \( |\Delta_{N, +} | = |\Delta_{N, -} | \neq 0 \), which corresponds to two helicoidal SDWs of opposite chiralities. Using the method of Ref. \[ 9 \], 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 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 ˜\(g_2\) = \(g_2/\pi t_F b\) and ˜\(g_3\) = \(g_3/\pi t_F b\). We vary the ratio ˜\(g_3/\tilde{g}_2\) while keeping the sum constant: ˜\(g_2 + \tilde{g}_3\) = 2/ln(2\(\gamma E_0/\pi T_c(\infty)\)). where 12 K is the transition temperature for an infinite magnetic field, 100 K is an ultraviolet cutoff of the order of \(t_\sigma\), and \(\gamma \approx 1.78\) is the exponential of the Euler constant. The calculations are performed for \(t_\sigma = 300\) K, \(t_{2b} = 20\) K, \(t_{3b} = 0\) K, \(t_{4b} = 0.75\) K, and 1000 K. The transition temperature \(T_c^{(N)}\) is obtained numerically from Eq. (4), and the polarizations of the SDWs for \(T < T_c^{(N)}\) are determined by minimizing the free energy \(F_N\) given by Eq. (5).

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 \(Q_N\) coexists with a weak SDW at the wave vector \(Q_{-N}\). In general, the value of \(Q_y\) that maximizes \(\chi_0(Q_N)\) does not maximize \(\chi_0(Q_{-N})\), so 10 \(\chi_0(Q_{-N} < \chi_0(Q_N)\). As a result, the SDW amplitude at the wave vector \(Q_{-N}\) is very small, and the polarizations of the SDWs are linear. The values of \(N\) follow the usual “positive” sequence \(N = 6, 5, 4, 3, 2, 1, 0\) 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 \(\chi_0(Q_{2N}^{(N)}, \pi/b) \approx \chi_0(Q_{2N}^{(-N)}, \pi/b)\). The latter relation implies that the two SDWs have comparable amplitudes. The two susceptibilities are strictly equal at \(t_{4b} = 0\), but when \(t_{4b} > 0\), \(\chi_0(Q_{2N}^{(N)}, \pi/b) > \chi_0(Q_{2N}^{(-N)}, \pi/b)\) (this result holds also for \(t_{3b} \neq 0\) \(\tilde{g}_3\)). This yields a negative Hall plateau, provided the SDWs are sinusoidal. Thus, for \(g_3/g_2 = 0.03\) (\(g_2 \approx 0.37\) and \(g_3 \approx 0.01\)), we find the sequence \(N = 6, 5, 4, -2, 2, 1, 0\) (see Fig. 4). 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 \(N = 8, 7, -4, 6, 5, 4, -2, 2, 1, 0\) (see Fig. 4). As discussed in Ref. (5), \(N = -2\) and \(N = -4\) correspond to the two negative QHE phases observed in experiments (6, 7). As shown in Fig. 4, the phase \(N = -2\) splits into two sub-phases: 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)$_2$PF$_6$, the pressure has to be higher than 6 kbar, since the FISDW cascade disappears below this pressure (5). 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)$_2$ReO$_4$, 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 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 \(M_z\) along the vector \(n\) which characterizes the spin polarization of a helicoidal SDW. In our case, the vector \(n\) is parallel to the magnetic field \(H\), 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 ++ electrons with spin up and the − electrons with spin down have the energy gap \(\Delta_1 = |\Delta_{N,+}|\), whereas the +k\(_F\) electrons with spin down and the −k\(_F\) electrons with spin up have the different energy gap \(\Delta_2 = |\Delta_{-N,-}|\). 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 \(M_z\):

\[
j_x = ev_F \sum_\sigma (\delta n_{+,-\sigma} - \delta n_{-,-\sigma}),
\]

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

where \(\delta n_{\alpha,\sigma}\) is the deviation of the distribution function of electrons with spin \(\sigma\) and momenta near \(\alpha k_F\) from the equilibrium one, \(g\) is the electron gyromagnetic factor, and \(\mu_B\) is the Bohr magneton. At low temperature, we can consider solely the electrons excited above the lowest gap (\(\Delta_2\) in Fig. 3). This implies that \(\delta n_{+,\uparrow} \simeq \delta n_{-,\downarrow} \simeq 0\) and \(M_z/j_x \simeq -g\mu_B/2ev_F\).

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 \(Q_N\) and \(-Q_N\)) with comparable amplitudes. We have also 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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