SIGN REVERSAL OF THE QUANTUM HALL EFFECT AND HELICOIDAL MAGNETIC-FIELD-INDUCED SPIN-DENSITY WAVES IN ORGANIC CONDUCTORS

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Abstract. Within the framework of the quantized nesting model, we study the effect of umklapp scattering on the magnetic-field-induced spin-density-wave (SDW) phases which are experimentally observed in the quasi-one-dimensional organic conductors (TMTSF)$_2$X. We discuss the conditions under which umklapp processes may explain the sign reversals (Ribault anomaly) of the quantum Hall effect (QHE) observed in these conductors. We find that the ‘Ribault phase’ is characterized by the coexistence of two SDWs with comparable amplitudes. This gives rise to additional long wavelength collective modes besides the Goldstone modes due to spontaneous translation and rotation symmetry breaking. These modes strongly affect the optical conductivity. We also show that the Ribault phase may become helicoidal (i.e. with circularly polarized SDWs) if the strength of umklapp processes is sufficiently strong. The QHE vanishes in the helicoidal phases, but a magnetoelectric effect appears.

1. INTRODUCTION

The organic conductors of the Bechgaard salts family (TMTSF)$_2$X (where TMTSF stands for tetramethyltetraselenafulvalene) have remarkable properties in a magnetic field. In three members of this family (X=ClO$_4$, PF$_6$, ReO$_4$), a moderate magnetic field of a few Tesla destroys the metallic phase and induces a series of SDW phases separated by first-order phase transitions [1].

According to the so-called quantized nesting model (QNM) [1], the formation of the FISDWs results from the strong anisotropy of these organic materials, which can be viewed as weakly coupled chain systems (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 SDW opens a gap, but leaves closed pockets of electrons and/or holes in the vicinity of the Fermi surface. In presence of a magnetic field $H$, these pockets are quantized into Landau levels (more precisely Landau subbands). In each FISDW phase, the SDW wave vector is quantized, $Q_N = (2k_F + NG, Q_y)$ with $N$ integer, so that an integer number of Landau subbands are filled. [Here $k_F$ is the Fermi momentum along the chains, $-e$ the electron charge, $b$ the interchain spacing, and $G = eHb/\hbar$.] As a result, the Fermi level lies in a gap between two Landau subbands. In each FISDW phase, the Hall conductivity is quantized: $\sigma_{xy} = -2Ne^2/h$ per one layer of the TMTSF molecules [1]. As the magnetic field increases, the value of the integer $N$ changes, which leads to a cascade of FISDW transitions.

A striking feature of the QHE in Bechgaard salts is the coexistence of both positive and negative Hall plateaus. While most plateaus are of the same sign, referred to as positive by convention, a negative Hall effect is also observed at certain pressures (the so-called Ribault anomaly) [2].

We have recently proposed an explanation of the Ribault anomaly by taking umklapp processes into account [3,4]. The latter are allowed in the Bechgaard salts due to the half-filled electron band which results from the dimerization along the chains. Within our explanation [5], two SDWs with comparable amplitudes coexist in the negative phases, which therefore differ significantly from the positive ones. In particular, they exhibit an unusual structure of long-wavelength collective modes [4], and their polarization may become circular (helicoidal SDWs) above a critical value of the umklapp scattering strength [3].
2. SIGN REVERSAL OF THE QHE

In the absence of umklapp scattering, a magnetic field along the z axis induces a series of SDW phases characterized by a quantized wave vector \( \mathbf{Q}_N = (2k_F + NG, Q_y) \) and a quantification of the Hall effect: \( \sigma_{xy} = -2Ne^2/h \). The sign of \( N \) is entirely determined by the electron dispersion [1], which seems to preclude any sign reversal of the QHE.

Umklapp scattering transfers \( 4k_F \) and therefore couples the wave vectors \( \mathbf{Q}_N \) and \( \mathbf{Q}_N - 4k_F \). As a result two SDWs, with wave vectors \( \mathbf{Q}_N = (2k_F + NG, Q_y) \) and \( \mathbf{Q}_{-N} = (2k_F - NG, -Q_y) \) form simultaneously. In the random-phase approximation, the transition temperature is determined by the modified Stoner criterion

\[
[1 - g_2\chi_0(Q_N)][1 - g_2\chi_0(Q_{-N})] - g_3^2\chi_0(Q_N)\chi_0(Q_{-N}) = 0,
\]

where \( g_2 \) and \( g_3 \) are the scattering amplitudes of the normal and umklapp processes, respectively. \( \chi_0 \) is the spin susceptibility in the absence of electron-electron interaction.

A very small \( g_3 \) does not qualitatively change the phase diagram compared to the case \( g_3 = 0 \). Now the main SDW at wave vector \( \mathbf{Q}_N \) coexists with a weak SDW at wave vector \( \mathbf{Q}_{-N} \). The values of \( N \) follow the usual ‘positive’ sequence \( N = \ldots, 4, 3, 2, 1, 0 \) 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 transition 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. [This follows from the structure of the bare spin susceptibility \( \chi_0(Q) \) at \( Q_y = \pi/b \).] The two SDWs have now comparable amplitudes. For certain dispersion laws [6], the main SDW corresponds to the wave vector \( \mathbf{Q}_{-|N|} \), which yields a negative Hall plateau. Thus, for \( g_3/g_2 \simeq 0.03 \), we find the sequence \( N = \ldots, 5, 4, -2, 2, 1, 0 \) (Fig. 1a). In Bechgaard salts, the strength of umklapp scattering is very sensitive to pressure. Therefore, we conclude that sign reversals of the QHE can be induced by varying pressure as observed experimentally [2].

\[\text{Figure 1: (a) Phase diagram for } g_3/g_2 = 0.03. \text{ The phase } N = 3 \text{ is suppressed, and the negative commensurate phase with } N = -2 \text{ and } Q_y = \pi/b \text{ appears in the cascade (the shaded area). All the phases are sinusoidal, and the Hall effect is quantized: } \sigma_{xy} = -2Ne^2/h. \text{ The vertical lines are only guides for the eyes and do not necessarily correspond to the actual first-order transition lines. (b) Phase diagram for } g_3/g_2 = 0.06. \text{ Two negative phases, } N = -2 \text{ and } N = -4, \text{ are observed (the shaded areas). The phase } N = -2 \text{ splits into two subphases: helicoidal (the dark shaded area) and sinusoidal (the light shaded area).}\]

3. HELICOIDAL PHASES

We have also shown that umklapp scattering can change the polarization of the SDWs from linear (sinusoidal SDWs) to circular (helicoidal SDWs) [3]. The QHE vanishes in the helicoidal phases, but
a magnetoelectric effect appears. For \( g_3/g_2 \simeq 0.06 \), there are two negative phases: \( N = -2 \) and \( N = -4 \) (Fig. 1b). The phase \( N = -2 \) splits into two subphases: helicoidal and sinusoidal. In order to observe helicoidal phases experimentally, it would be desirable to stabilize the negative phase \( N = -2 \) at the lowest possible pressure (which corresponds to the strongest \( g_3 \)).

4. COLLECTIVE MODES

The Ribault phase exhibits an unusual structure of long-wavelength collective modes. The coexistence of two SDWs in this phase gives rise to additional modes besides the Goldstone modes resulting from spontaneous rotation and translation symmetry breaking [4]. Below we discuss only the sliding modes, restricting ourselves to longitudinal (i.e. parallel to the chains) fluctuations.

There is a Goldstone mode with a linear dispersion law
\[
\omega = v_F q_x,
\]
which corresponds to out-of-phase oscillations of the two SDWs: \( \theta_N(q_x, \omega) = -\theta_{-N}(q_x, \omega) \), where the phases \( \theta_N \) and \( \theta_{-N} \) determine the positions of the two SDWs with respect to the crystal lattice. The fact that the two SDWs can be displaced in opposite directions without changing the energy of the system is related to the pinning that would occur for a single commensurate SDW.

Besides the Goldstone mode, there is a gapped mode
\[
\omega^2 = \omega_0^2 + v_F^2 q_x^2,
\]
which corresponds to in-phase oscillations of the two SDWs. \( \omega_0 \) depends on the strength of umklapp scattering and is generally larger than the mean-field gap. We therefore expect this mode to be strongly damped due to the coupling with the quasi-particle excitations.

The presence of two SDWs can be detected by measuring the optical conductivity. In the limit \( q = 0 \), the dissipative part of the conductivity is given by
\[
\text{Re}[\sigma(\omega)] = \frac{\omega_p^2}{4} \left( \delta(\omega) - \left( \frac{1 - \tilde{\gamma}^2}{3 + 5\tilde{\gamma}^2} + \delta(\omega \pm \omega_0) \right) \right),
\]
where \( \tilde{\gamma} \) is the ratio of the SDW amplitudes, and \( \omega_p \) the plasma frequency. Eq. (2) satisfies the conductivity sum rule \( \int_{-\infty}^{\infty} \text{Re}[\sigma(\omega)] = \omega_p^2/4 \). Quasi-particle excitations above the mean-field gap do not contribute to the optical conductivity, a result well known in clean SDW systems. Because both modes contribute to the conductivity, the low-energy (Goldstone) mode carries only a fraction of the total spectral weight. We obtain Dirac peaks at \( \pm \omega_0 \) because we have neglected the coupling of the gapped mode with quasi-particle excitations. Also, in a real system (with impurities), the Goldstone mode would broaden and appear at a finite frequency (below the quasi-particle excitation gap) due to pinning by impurities.

References

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