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Unconventional spin-density wave in Bechgaard salt (TMTSF)$_2$NO$_3$

Mario Basletić[*]

Department of Physics, Faculty of Science, University of Zagreb, P. O. Box 331, HR-10002 Zagreb, Croatia

Bojana Korin-Hamzić

Institute of Physics, P. O. Box 304, HR-10001 Zagreb, Croatia

Kazumi Maki

Department of Physics and Astronomy, University of Southern California, Los Angeles California 90089-0484, USA
and Max Planck Institute for the Physics of Complex Systems, Nöthnitzer Strasse 38, D-01187, Dresden, Germany

Silvia Tomić

Institute of Physics, P. O. Box 304, HR-10001 Zagreb, Croatia

Among many Bechgaard salts, (TMTSF)$_2$NO$_3$, where TMTSF denotes tetramethyltetraselenafulvalene, exhibits very anomalous low-temperature properties. Unlike the case of the conventional spin-density wave (SDW), (TMTSF)$_2$NO$_3$ undergoes the SDW transition at $T_C \approx 9.5$ K and the low-temperature quasiparticle excitations are gapless. Also, it is known that (TMTSF)$_2$NO$_3$ does not exhibit superconductivity even under pressure, while a field-induced SDW is found in (TMTSF)$_2$NO$_3$ only for $P=8.5$ kbar and $B>20$ T. Here we show that both the angle-dependent magnetoresistance data and the nonlinear Hall resistance of (TMTSF)$_2$NO$_3$ at ambient pressure are interpreted satisfactorily in terms of an unconventional spin-density wave. Based on these facts, we propose a new phase diagram for Bechgaard salts.

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I. INTRODUCTION

(TMTSF)$_2X$ are quasi-one-dimensional molecular conductors, known as Bechgaard salts, where TMTSF denotes tetramethyltetraselenafulvalene and X is an inorganic anion with various possible symmetries: spherical (octahedral) (X =PF$_6$, AsF$_6$,...), tetrahedral (X=ClO$_4$, ReO$_4$,...), or triangular (NO$_3$). Their very complex (pressure, magnetic field, temperature) phase diagrams are well known and have a variety of electronic ground states: (conventional) spin-density wave (SDW), field-induced SDW (FISDW), superconductivity (triplet, unconventional), unconventional spin-density wave (USDW).[1–8] The observation of superconductivity in the (TMTSF)$_2X$ series requires the use of high pressure, with the exception of (TMTSF)$_2$ClO$_4$, which is superconducting under ambient pressure, and (TMTSF)$_2$NO$_3$, which never becomes a superconductor even under pressure.

NO$_3$ anions are in orientational disorder at ambient pressure and for $T>45$ K. The anion ordering (AO) transition takes place at $T_{AO}\approx 45$ K, with wave vector $\mathbf{q}=(1/2,0,0)$. Unlike AO in most other salts, here $\mathbf{q}$ has the nonzero component parallel to the most conducting direction. The SDW state develops below $T_C\approx 9.5$ K. From the resistivity data a very small activation energy was obtained, of order of 10^{-3} eV, but the curvature of the $\log_{10}R$ vs $1/T$ plot indicated that the ground state should be considered as semimetallic, rather than semiconducting.[9]

The phase diagram of Bechgaard salts under pressure is interpreted in terms of the standard model, where the approximate nesting of the quasi-one-dimensional Fermi surface (i.e., the imperfect nesting), and the repulsive Coulomb interaction between electrons are the crucial ingredients.[10–12]

The applied pressure increases the two-dimensionality of Bechgaard salts through the increase of the imperfect nesting term.[1] However, the standard model does not yet describe either triplet[4,6] superconductivity or the USDW.

The UDW is a density wave whose gap function depends on the wave vector, and vanishes on certain subsets of the Fermi surface, allowing for low-energy excitations. The average of the gap function over the Fermi surface is zero, causing a lack of periodic modulation of the charge and/or spin density. As noted by Nersesyan *et al.*[13,14] the quasiparticle spectrum in an UDW is quantized in a magnetic field. This Landau quantization gives rise to the spectacular angle-dependent magnetoresistance (ADMR) and giant Nernst effect.[15,16] As we shall see later both the angle-dependent magnetoresistance[17] and the nonlinear Hall resistance[18] of (TMTSF)$_2$NO$_3$ are described nicely in terms of an USDW. We note that an earlier attempt to describe the magnetoresistance of (TMTSF)$_2$NO$_3$ in terms of a conventional SDW with a *large imperfect nesting* might not be the most appropriate model,[19] since it cannot describe the details of the resistance quantitatively. We also propose a revision of the generally accepted phase diagram, taking into account the identification of the USDW state in several Bechgaard compounds.[7,8,20]

II. IDENTIFICATION OF THE USDW

Here we summarize briefly what is known about unconventional density wave.[15,16] The unconventional density wave is a kind of density wave, where the quasiparticle energy gap vanishes along lines on the Fermi surface. In the present instance we can assume $\Delta(\mathbf{k}) \sim \cos \mathbf{b} \mathbf{k}$ or $\Delta(\mathbf{k})$

$\sim \sin \mathbf{bk}$ as in earlier analyses of the UCDW in α-(BEDT-TTF)$_2$KHg(SCN)$_4$.[21,22] Then the quasiparticle Green's function is given by

$$G^{-1}(\mathbf{k}, \omega) = \omega - \eta(\mathbf{k}) - \xi(\mathbf{k})\rho_3 - \Delta(\mathbf{k})\rho_1 \quad (1)$$

where the ρ_i's are the Pauli matrices and G operates on the Nambu spinor space.[23] The quasiparticle energy in an UDW is formed from the pole of $G(\mathbf{k}, \omega)$ as

$$\omega = \eta(\mathbf{k}) \pm \sqrt{\xi^2(\mathbf{k}) + \Delta^2(\mathbf{k})} \quad (2)$$

where $\xi(\mathbf{k})$ is the kinetic energy of electrons measured from the Fermi energy in the normal state, $\xi(\mathbf{k}) \simeq v(k_\mathbf{a} - k_F)$, $\eta(\mathbf{k})$ is the imperfect nesting term and $\Delta(\mathbf{k}) = \Delta \cos \mathbf{bk}$. Here, v denotes the Fermi velocity in the chain (**a**) direction, Δ is the order parameter for the unconventional SDW, and $\mathbf{b} = (0, b, 0)$, where b is the lattice constant.

Then in a magnetic field $\mathbf{B}$ in the $\mathbf{b}'$-$\mathbf{c}^\star$ plane with angle θ from the $\mathbf{c}^\star$ axis the quasiparticle energy changes into

$$E_n^\pm = \pm \sqrt{2neB|\cos\theta|vb\Delta} \quad (3)$$

with $n = 0, 1, 2\ldots$. Here we have neglected $\eta(\mathbf{k})$ for simplicity. Also in the following we assume $b = 7.567$ Å and $v = 3 \times 10^5$ m/s.[19] Equation (3) is a consequence of the Landau quantization of the quasiparticle spectrum in the UDW, or the *Nersesyan effect*.[13,14]

Then the conductivity tensor is constructed as

$$\sigma_{xx} = \sigma_1[1 + 2C_1\,\mathrm{sech}^2(x_1/2) + \cdots], \quad (4)$$

$$\sigma_{yy} = \sigma_2[1 + 2C_2\,\mathrm{sech}^2(x_1/2) + \cdots], \quad (5)$$

$$\sigma_{xy} = \sigma_3 n(T,B)B|\cos\theta|, \quad (6)$$

with

$$n(T,B) = n_0\{1 + 2[1 - \tanh(x_1/2)] + \cdots\}, \quad (7)$$

where $x_1 = E_1/k_B T$ and we have assumed that $x_1 \gg 1$. Also, we have assumed that σ_1, σ_2, σ_3, C_1, and C_2 are weakly dependent on T and B. Then from Eqs. (4)–(6) we can construct the resistivity tensor as

$$R_{xx}(B, \theta) = \frac{R_0}{1 - D_1\tanh^2(x_1/2)}, \quad (8)$$

$$R_{xy}(B, T) = \frac{D_2 B}{\dfrac{n(B,T)}{n(0,T)}B^2 + D_3\dfrac{n(0,T)}{n(B,T)}\sigma_{xx}\sigma_{xy}}. \quad (9)$$

In Fig. 1 we show our fitting of the angle-dependent magnetoresistance data for (TMTSF)$_2$NO$_3$ at $T = 4.2$ K for a variety of magnetic fields.[17,24] From this fitting we obtain the USDW order parameter $\Delta = 6.3$ K and $D_1 = 0.93$. As is readily seen the fitting is excellent except for the bumpy structures. These should come from the imperfect nesting term as discussed in Refs. 8, 21, and 22. Also we note $D_1 \approx 2C_1/(1 + 2C_1)$ indicating that $C_1 = 7.1$; therefore σ_{xx} is dominated by the $n = 1$ excitations.

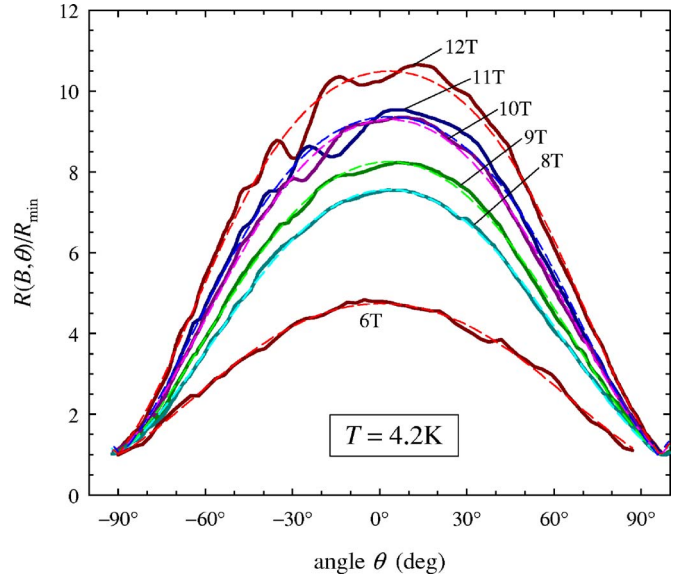In Fig. 2 we show R_{xy} fitted by Eq. (9); again we obtain

FIG. 1. (Color online) The angular dependence of the normalized resistance $R(B, \theta)/R_0$ at $T = 4.2$ K (full lines, experimental data; dashed lines, fits to the theory). Magnetic field is rotated in $\mathbf{b}'$-$\mathbf{c}^\star$ plane, and $\theta = 0°$ corresponds to $\mathbf{B} \| \mathbf{c}^\star$. Data are from Ref. 17 ($B \geq 8$ T) and Ref. 24 ($B = 6$ T).

reasonable fitting with $D_3 \simeq 80\ \Omega$ T. Figure 3 shows the temperature dependence of the parameter D_2, along with the temperature dependence of the resistance R_{xx}. There appears to be a slight change of the parameter D_2 across $T^\star \approx T_C/3 = 3$ K: for $T \gtrsim 3$ K it follows the temperature dependence of the resistance R_{xx}, while for lower temperature it is nearly constant. It signals the possible occurrence of yet another phase transition at 3 K—as in (TMTSF)$_2$PF$_6$—in agreement with several other suggestions.[20,25,26]
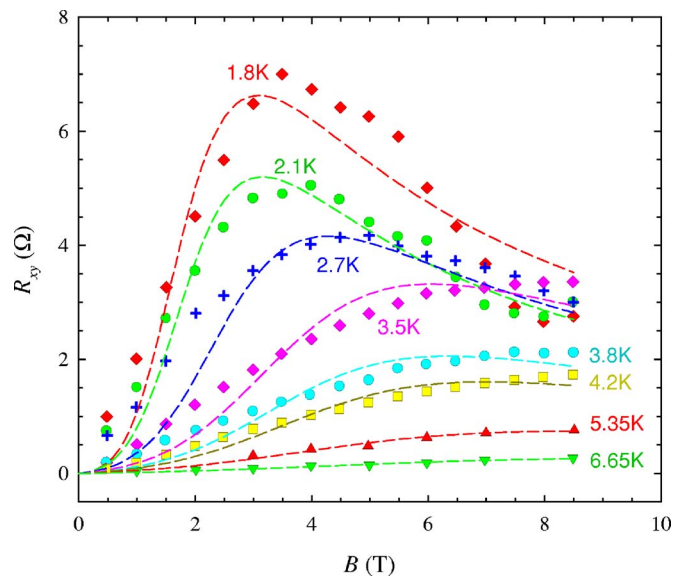

FIG. 2. (Color online) The magnetic field B dependence of Hall resistance R_{xy} at several temperatures (points, experimental data; dashed lines, fit to the theory). Data are from Ref. 18.
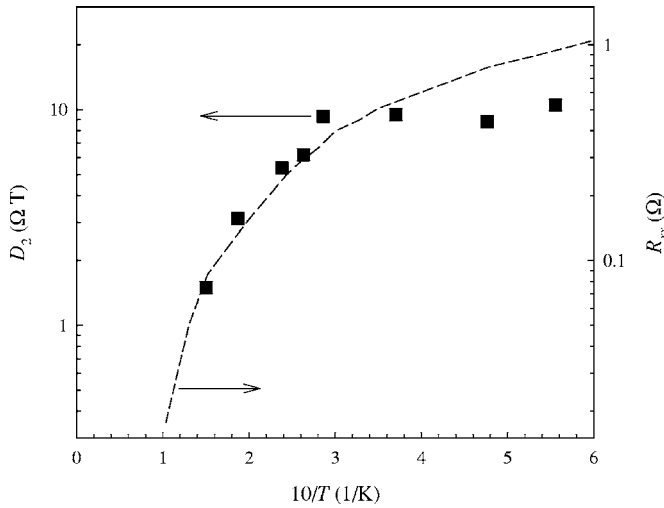
FIG. 3. The temperature dependence of the fitting parameters D_2 (■)—see text. Dashed line shows temperature dependence of the resistance R_{xx} of $(TMTSF)_2NO_3$ for $B=0$.

FIG. 4. The schematic pressure-temperature phase diagram for Bechgaard salts. Arrows denote position of $(TMTSF)_2PF_6$, $(TMTSF)_2ClO_4$, and $(TMTSF)_2NO_3$ in the phase diagram at ambient pressure.

III. THE NEW PHASE DIAGRAM OF BECHGAARD SALTS

Recently, one of us proposed a phase diagram for Bechgaard salts with an octahedral (centrosymmetric) anion like PF_6 which exhibit metallic behavior down to the SDW transition at $T_C \approx 12$ K (see Ref. [16]). The salts with noncentrosymmetric anions undergo the AO transition and become insulating at ambient pressure, except for $X=ClO_4$ and NO_3. Here, we propose an extension or revision of the phase diagram (see Fig. [4]). As indicated in Fig. [4], $(TMTSF)_2PF_6$ at ambient pressure undergoes yet another transition around $T^\star \approx T_C/3 \approx 4$ K. The drastic change in the quasiparticle spectrum through $T^\star$ has been interpreted as the appearance of a SDW+USDW.[20] Further, from the angle-dependent magnetoresistance of $(TMTSF)_2PF_6$ and $(TMTSF)_2ReO_4$ for $P > 8$ kbar the existence of an USDW in the high-pressure range is inferred.[7,8] Then, it is customary to put $(TMTSF)_2ClO_4$ at ambient pressure around $P=8$ kbar in Fig. [4], where the transition the from metallic to the superconducting state takes place. In this way we may understand the superconductivity at ambient pressure. Similarly, we may put $(TMTSF)_2NO_3$ at ambient pressure around $P \simeq 8.5$ kbar, since the transition from the metallic to the density wave state takes place at $T_C \approx 9.5$ K. The further behavior of T_C vs pressure is based on experiments, which have shown that T_C is gradually suppressed under increasing pressure.[27] Then the absence of superconductivity, and appearance of a FISDW only at high pressure and high magnetic fields ($P \gtrsim 8$ kbar, $B > 20$ T),[28] are very surprising.

We think that the lack of inversion symmetry in NO_3 is at the heart of the absence of superconductivity and a FISDW (for low pressure, $P < 8.5$ kbar) in $(TMTSF)_2NO_3$. For example Anderson[29] speculated that a triplet superconductor cannot exist in a crystal without inversion symmetry. Also the nature of superconductivity in $CePt_3Si$, a crystal without inversion symmetry, is hotly discussed in the current literature.[30,31] The inversion symmetry breaking is usually characterized by E_{ch}, the chiral symmetry breaking or
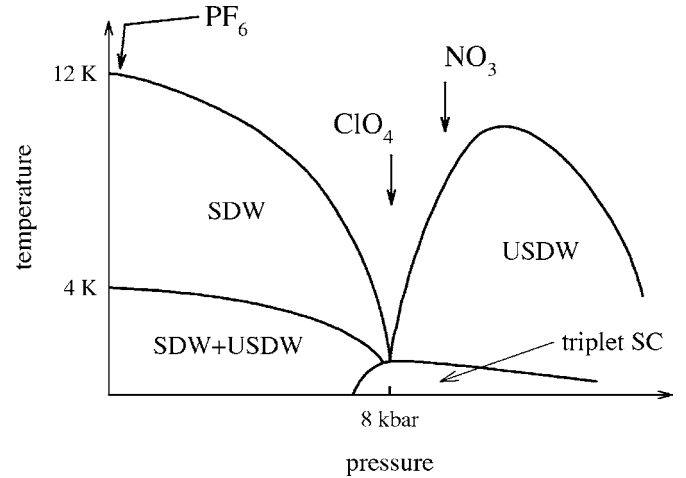
Rashba term.[31–33] Both the absence of triplet superconductivity (for $T_{SC} < 1$ K) and the appearance of a FISDW for $B > 20$ T suggest $E_{ch} \sim 7–8$ K. Also, this E_{ch} appears to be consistent with $T_{AO} \approx 45$ K ($T_{AO} \gg E_{ch}$).[34] We believe that further study of the electronic properties of $(TMTSF)_2NO_3$ is of great interest.

IV. CONCLUDING REMARKS

We have shown that the anomalous low-temperature behavior of ADMR and Hall resistance of the Bechgaard salt $(TMTSF)_2NO_3$ could be interpreted in terms of an unconventional spin-density wave, indicating that the possible ground state below $T_C \approx 9.5$ K is an USDW. This is consistent with the new phase diagram of Bechgaard salts proposed recently,[8,16] which we—in addition—revised and extended in this paper. Therefore, it will be of great interest to study how the USDW order parameter Δ changes as the pressure is applied. This will provide a first step to explore the wider phase diagram. Another question is if there are other candidates among the Bechgaard salts which exhibit an USDW under ambient pressure.

We have also proposed a possible explanation for the absence of superconductivity. Both the absence of a superconductivity and partial suppression of a FISDW (i.e., the absence of a FISDW for $P < 8$ kbar, $B < 20$ T) are due to the inversion symmetry breaking associated with the NO_3 anion ordering. The details of this will be presented elsewhere.[34]

*Electronic address: basletic@phy.hr

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