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Unconventional Charge-Density Wave in the Organic Conductor α-(BEDT-TTF)$_2$KHg(SCN)$_4$

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The low temperature phase (LTP) of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ salt is known for its surprising angular dependent magnetoresistance (ADMR), which has been studied intensively in the last decade. However, the nature of the LTP has not been understood until now. Here we analyze theoretically ADMR in unconventional (or nodal) charge-density wave (UCDW). In magnetic field the quasiparticle spectrum in UCDW is quantized, which gives rise to spectacular ADMR. The present model accounts for many striking features of ADMR data in α-(BEDT-TTF)$_2$KHg(SCN)$_4$.

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The series of quasi-two-dimensional organic conductors α-(BEDT-TTF)$_2M$Hg(SCN)$_4$ [where BEDT-TTF denotes bis(ethylenedithio)tetrathiafulvalene and $M =$ K, NH$_4$, Rb, and Tl] have attracted considerable attention over the last few years due to two different ground states and rich phenomena associated with them [1].

Whereas the $M = \mathrm{NH}_4$ compound becomes superconducting below 1.5 K, other salts enter at $T_c = 8$–12 K into a specific low temperature phase (LTP) with striking angular dependent magnetoresistance (ADMR). From the magnetic phase diagram of LTP it is now believed that LTP is not spin density wave (SDW) but a kind of charge-density wave (CDW), though no detailed characterization is available [2]. We have proposed recently that unconventional (or nodal) charge-density wave (UCDW) can account for a number of features in LTP in α-(BEDT-TTF)$_2$KHg(SCN)$_4$ including the threshold electric field [3–7]. Recently UCDW and USDW have been proposed by several authors as possible electronic ground state in quasi-one-dimensional and quasi-two-dimensional crystals [8–12]. Unlike conventional density wave (DW) [13], the order parameter in UCDW $\Delta(\mathbf{k})$ depends on the quasiparticle wave vector $\mathbf{k}$. In α-(BEDT-TTF)$_2$KHg(SCN)$_4$ salts, where the conducting plane lies in the a-c plane and the quasi-one-dimensional Fermi surface is perpendicular to the a axis, we assume that $\Delta(\mathbf{k}) = \Delta\cos(ck_z)$ or $\Delta\sin(ck_z)$ [i.e., $\Delta(\mathbf{k})$ depends on $\mathbf{k}$ perpendicular to the most conducting direction], where $c = 9.778$ Å is the lattice constant along the c axis ($a = 9.914$ Å and $b = 20.420$ Å) [14]. In addition, there is a quasi-two-dimensional Fermi surface with elliptical cross section in the a-c plane. It is known also that the thermodynamics of UCDW and USDW is practically the same as the one in d-wave superconductor [11,15]. Also in spite of the clear thermodynamic signal, the first order terms in $\Delta(\mathbf{k})$ usually vanish when averaged over the Fermi surface. This implies neither clear x-ray signal for UCDW nor spin signal for USDW. Because of this fact unconventional density waves are sometimes called the phase with hidden order parameter [12].

In a magnetic field the quasiparticle spectrum is quantized as first shown by Nersesyan *et al.* [8,9]. This dramatic change in the quasiparticle spectrum is most readily seen in ADMR as it has been demonstrated recently for SDW plus USDW in (TMTSF)$_2$PF$_6$ below $T = T^*(\sim 4$ K) [16]. About a decade ago ADMR in LTP in α-(BEDT-TTF)$_2$KHg(SCN)$_4$ salts were studied intensively. In particular, ADMR for current $\mathbf{j}$ perpendicular ($\mathbf{j}\|\mathbf{b}^*$) and parallel to the a-c plane exhibits a broad peak around $\theta = 0°$(see inset of Fig. 1), where θ is the angle with which the magnetic field is tilted from the b^* axis (normal to the conducting plane). In addition, a series of dips are observed at $\theta = \theta_n$ given by [17,18]

$$\tan(\theta_n)\cos(\phi - \phi_0) = \tan(\theta_0) + nd_0, \tag{1}$$

where $\tan\theta_0 \simeq 0.5$, $d_0 \simeq 1.25$, $\phi_0 \simeq 27°$, and $n = 0, \pm1, \pm2, \ldots$. Here ϕ is the angle the projected magnetic field on the a-c plane makes with the c axis. The origin of this surprising ADMR has been discussed earlier [17–22], but only partial answers were found. These earlier models cannot describe the broad peak in the resistance around $\theta = 0°$ (see Figs. 3 and 4 below), fail to reproduce the shape of dips, and also predict a big (and quite broad) peak around $\theta = 90°$, which has never been observed in α-(BEDT-TTF)$_2$KHg(SCN)$_4$. In the following we shall show that the quasiparticle spectrum in UCDW in α-(BEDT-TTF)$_2$KHg(SCN)$_4$ salts is quantized in the presence of magnetic field. The small energy gap, which is proportional to $\sqrt{B}$ where B is the field strength, depends also on the direction of the magnetic field, and it can be seen in ADMR. As it will be shown below, we

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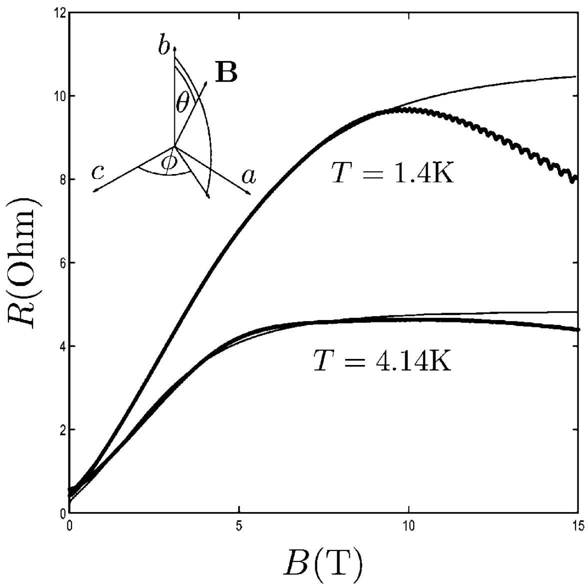

FIG. 1. The magnetoresistance is plotted for $T = 1.4$ and 4.14 K as a function of magnetic field. The thick solid line is the experimental data, and the thin one denotes our fit based on Eq. (7). The inset shows the geometrical configuration of the magnetic field with respect to the conducting plane.

can describe salient aspects of ADMR seen in LTP of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ very consistently. Therefore we may conclude that ADMR in α-(BEDT-TTF)$_2$KHg(SCN)$_4$ provides a strong argument in favor of the UCDW nature of the LTP. We stress that the Landau quantization as proposed by Nersesyan *et al.* [8,9] should be readily accessible in other UCDW and USDW systems. In this respect experimental analysis of ADMR in the pseudogap phase in high T_c cuprate superconductors [23] and the glassy phase in κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br salt [24] will be of great interest. In LTP we assume that UCDW appears on the quasi-one-dimensional Fermi surface with quasiparticle energy given by

$$E(\mathbf{k}) = \sqrt{\xi^2 + \Delta^2(\mathbf{k})} - \varepsilon_0 \cos(2\mathbf{b}' \cdot \mathbf{k}), \qquad (2)$$

where $\xi \approx v_a(k_a - k_F)$, v_a is the Fermi velocity, $\Delta(\mathbf{k}) = \Delta \cos(ck_z)$, $\mathbf{b}'$ is the vector lying outside of the a-c plane, and ε_0 is the parameter describing the imperfect nesting [25–28]. In fitting the experimental data we discovered the following: (1) Eq. (2) gives only one single dip in ADMR, and (2) therefore the imperfect nesting term has to be generalized as

$$\varepsilon_0 \cos(2\mathbf{b}' \cdot \mathbf{k}) \longrightarrow \sum_n \varepsilon_n \cos(2\mathbf{b}'_n \cdot \mathbf{k}), \qquad (3)$$

where $\mathbf{b}'_n = b'[\hat{\mathbf{r}}_b + \tan(\theta_n)(\hat{\mathbf{r}}_a \cos\phi_0 + \hat{\mathbf{r}}_c \sin\phi_0)]$, $\varepsilon_n = \varepsilon_0 2^{-|n|}$, $\tan(\theta_n) = \tan(\theta_0) + nd_0$. From this, the origin of the generalized imperfect nesting terms can be regarded as an effective tight binding approximation, where hopping takes place between sites in the $\hat{\mathbf{r}}_b$ direction and along nearest neighbor chains oriented in the $\hat{\mathbf{r}}_a \cos\phi_0 + \hat{\mathbf{r}}_c \sin\phi_0$ direction. The orientation of the unit vectors can be seen in the inset of Fig. 1. As seen from Eq. (2), the quasiparticle spectrum is gapless and LTP is metallic in sharp contrast to conventional CDW. In a magnetic field the first term of the quasiparticle spectrum changes to

$$E_n = \pm\sqrt{2nv_a \Delta ce|B \cos\theta|}, \qquad (4)$$

where $n = 0, 1, 2, \ldots$. This is readily obtained following Refs. [8.9]. The contribution from the imperfect nesting term is considered as a perturbation, and the lowest order corrections to the energy spectrum are given by

$$E_0^1 = E_1^1 = -\sum_m \varepsilon_m \exp(-y_m), \qquad (5)$$

$$E_1^2 = -\sum_m \varepsilon_m (1 - 2y_m) \exp(-y_m), \qquad (6)$$

where $y_m = v_a b'^2 e|B \cos(\theta)|[\tan(\theta)\cos(\phi - \phi_o) - \tan(\theta_m)]^2/\Delta c$. The $n = 1$ level was twofold degenerate, but the imperfect nesting term splits the degeneracy by E_1^1 and E_1^2. Also the imperfect nesting term breaks the particle-hole symmetry. When $\beta E_1 \gg 1$ [$\beta = (k_B T)^{-1}$], the quasiparticle transport in the quasi-one-dimensional Fermi surface is dominated by the quasiparticles at $n = 0$ and $n = 1$ Landau levels. Considering that there are two conducting channels and only the quasi-one-dimensional one is affected by the appearance of UCDW, the ADMR is written as

$$R(B, \theta, \phi)^{-1} = 2\sigma_1 \left(\frac{\exp(-\beta E_1) + \cosh(\beta E_1^1)}{\cosh(\beta E_1) + \cosh(\beta E_1^1)} + \frac{\exp(-\beta E_1) + \cosh(\beta E_1^2)}{\cosh(\beta E_1) + \cosh(\beta E_1^2)} \right) + \sigma_2. \qquad (7)$$

Here σ_1 and σ_2 are the conductivities of the $n = 1$ Landau level and quasi-two-dimensional channels, in which the contribution of the $n = 0$ Landau level was melted, respectively. The same expressions were found for $\Delta(\mathbf{k}) = \Delta \sin(ck_z)$.

To compare the ADMR predicted by Eq. (7) with the experimentally observed behavior of α-(BEDT-TTF)$_2$KHg(SCN)$_4$, we have measured the interlayer resistance of a single crystal of this compound as a function of temperature, magnetic field strength, and orientation. The measurements were performed, using the standard four-probe ac technique ($f = 330$ Hz, $I_{\text{sample}} = 10\ \mu$A) in the temperature interval 1.4–20 K under magnetic field up to 15 T. The obtained ADMR data are consistent with the previous reports [18–20].

In Figs. 1 and 2 we compare the B dependence of the magnetoresistance at $T = 1.4$ and 4.14 K and the T dependence of the magnetoresistance for $B = 15$ T, for $\theta = 0°$. In fitting the temperature dependence of the resistivity, we assumed $\Delta(T)/\Delta(0) = \sqrt{1 - (T/T_c)^3}$, which was found to be very close to the exact solution of $\Delta(T)$ [11].

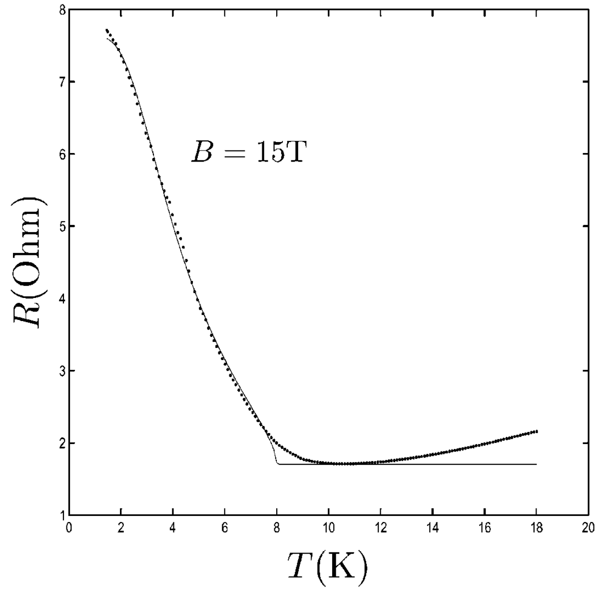

FIG. 2. The temperature dependent magnetoresistance is shown at $B = 15$ T. The dots are the experimental data, and the solid line is our fit.

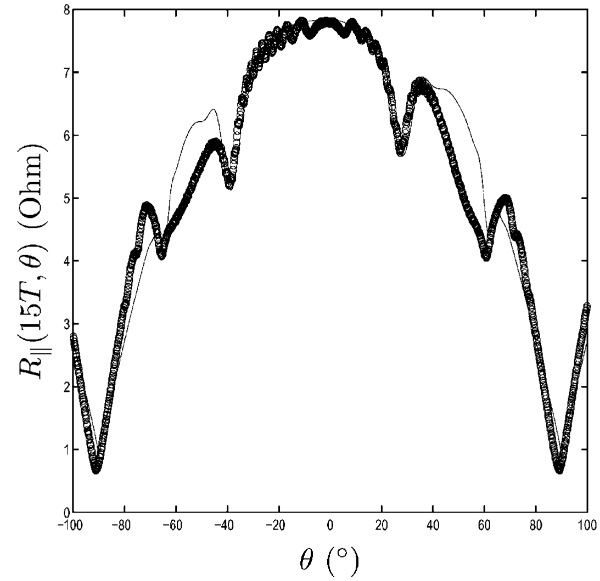

FIG. 3. The angular dependent magnetoresistance is shown for current parallel to the a-c plane at $T = 1.4$ K, $B = 15$ T. The open circles belong to the experimental data, and the solid line is our fit based on Eq. (7).

The influence of imperfect nesting terms in these cases is negligible, since they contribute only close to $\theta = \theta_n$.

Clearly the fitting becomes better as T decreases and/or B increases. Also for $T = 1.4$ K Shubnikov–de Haas oscillation becomes visible around $B = 10$ T, then the fitting starts breaking away. Clearly in this high field region the quantization of Fermi surface itself starts interfering with the quantization described above. In this region, the explicit B and T dependence of σ_1 and σ_2 should be taken into account for what we neglected here for simplicity. Also the deviaton of the theoretical curve from the experimental one above T_c in Fig. 2 is originated from this neglect. Here we concentrated on the dominant conduction mechanism, that is, thermally excited quasiparticles across the magnetic field induced gap. From these fittings we obtain $\sigma_2/\sigma_1 \sim 0.1$ and 0.3, and by assuming the mean field value of Δ (17 K), we get $v_a \sim 6 \times 10^6$ cm/s. In Figs. 3 and 4 we show the experimental data of ADMR as a function of θ for current parallel and perpendicular to the conducting plane for $T = 1.4$ K, $B = 15$ T, and $\phi = 45°$. As is readily seen, the fittings are excellent. From this we deduce $\sigma_2/\sigma_1 \sim 0.1$, $b' \sim 30$ Å, $\varepsilon_0 \sim 3$ K, which is much smaller than the Fermi temperature (~ 3000 K). Also from the exponential decay of ε_n, the spatial extension of the Wannier functions is estimated as $2b' \approx 60$ Å. Finally, we show in Fig. 5 R versus θ for different ϕ and compare with the experimental data side by side. Perhaps there are still differences in some details, but the overall agreement is very striking. These differences might arise from the fact that similar to the neglect of magnetic field and temperature dependence of σ_1 and σ_2, we also assumed them to be independent of ϕ and θ. The present model can describe a similar figure found in Ref. [20] rather well.

In summary, we have succeeded in describing the salient feature of ADMR observed in LTP in α-(BEDT-TTF)$_2$KHg(SCN)$_4$ in terms of UCDW with the Landau quantization of the quasiparticle spectrum. Very similar ADMR have been seen in $M =$ Rb and Tl compounds as well. Therefore we conclude that LTP in α-(BEDT-TTF)$_2M$Hg(SCN)$_4$ salts should be UCDW. Also we believe that ADMR provides a clear signature for the presence of UCDW and USDW. Therefore this

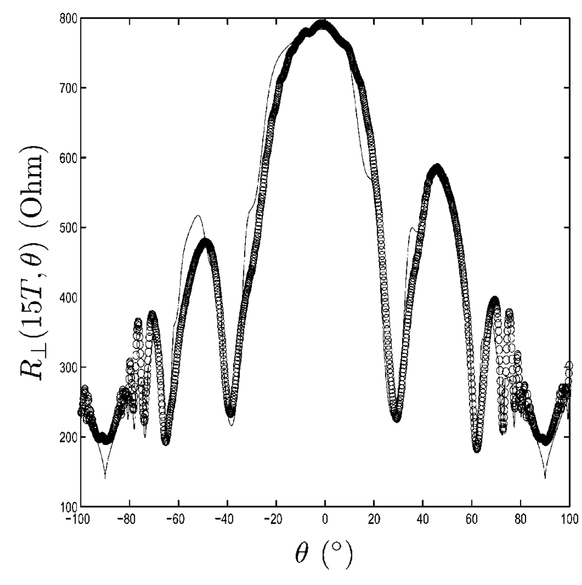

FIG. 4. The angular dependent magnetoresistance is shown for current perpendicular to the a-c plane at $T = 1.4$ K, $B = 15$ T. The open circles belong to the experimental data, and the solid line is our fit from Eq. (7).

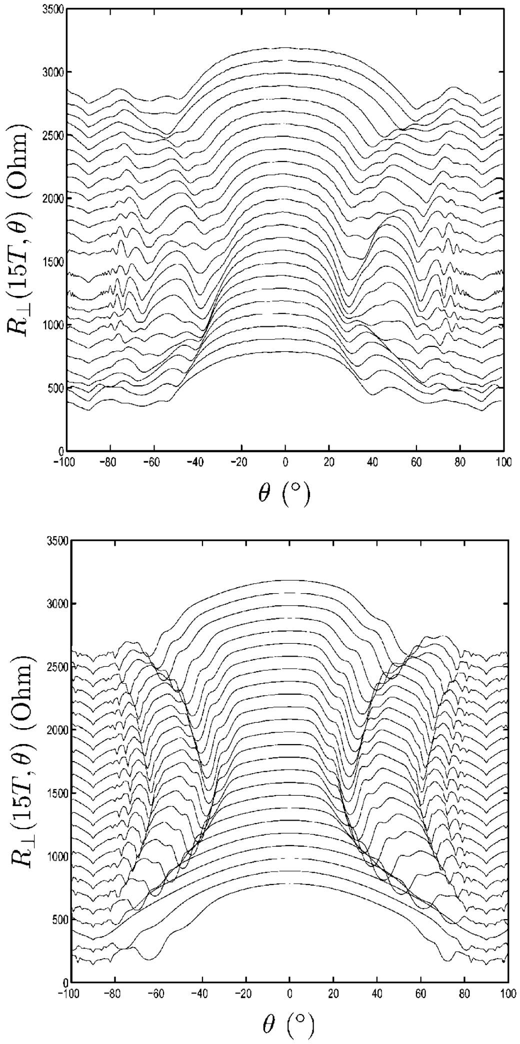

FIG. 5. ADMR is shown for current perpendicular to the *a*-*c* plane at $T = 1.4$ K and $B = 15$ T for $\phi = -77°$, $-70°$, $-62.5°$, $-55°$, $-47°$, $-39°$, $-30.5°$, $-22°$, $-14°$, $-6°$, $2°$, $10°$, $23°$, $33°$, $41°$, $48.5°$, $56°$, $61°$, $64°$, $67°$, $73°$, $80°$, $88.5°$, $92°$ and $96°$ from bottom to top. The top (bottom) panel shows experimental (theoretical) curves, which are shifted from their original position along the vertical axis by $n \times 100\ \Omega$, $n = 0$ for $\phi = -77°$, $n = 1$ for $\phi = -70°$, ...

technique can be exploited for other possible candidates of UDW.

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