The angular dependent magnetoresistance in \( \alpha-(BEDT-TTF)_2\text{KHg(SCN)}_4 \)

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PACS. 71.20.Rv – Polymers and organic compounds.
PACS. 72.15.Gd – Galvanomagnetic and other magnetotransport effects.
PACS. 71.45.Lr – Charge-density-wave systems.

Abstract. – In spite of extensive experimental studies of the angular dependent magnetoresistance (ADMR) of the low temperature phase (LTP) of \( \alpha-(BEDT-TTF)_2\text{KHg(SCN)}_4 \) about a decade ago, the nature of LTP remains elusive. Here we present a new study of ADMR in \( \alpha-(ET)_2 \) salts assuming that LTP is unconventional charge density wave (UCDW). In the presence of magnetic field the quasiparticle spectrum in UCDW is quantized, which gives rise to striking ADMR in UCDW. The present model appears to account for many existing ADMR data of \( \alpha-(BEDT-TTF)_2\text{KHg(SCN)}_4 \) remarkably well.

Introduction. – The series of the quasi-two-dimensional organic conductors \( \alpha-(BEDT-TTF)_2\text{M(Hg(SCN)}_4 \) (where BEDT-TTF is bis(ethylenedithio)tetrathiafulvalene and M=K, NH\(_4\), Rb and Tl) have attracted considerable attention in the last few years due to the richness of physical phenomena observed \[1\]. Whereas the M=NH\(_4\) compound becomes superconducting below 1 K, the other salts, at \( T_c = 8 – 10 \) K, undergo a phase transition into a specific low temperature phase (LTP), with associated numerous anomalies in magnetic field. LTP is thought to be caused by a density wave formation, but its nature appears still to be unsettled. We have proposed recently that unconventional charge density wave can account for a number of features of LTP in \( \alpha-(BEDT-TTF)_2\text{KHg(SCN)}_4 \) including the threshold electric field \[2\]. Recently, unconventional charge density wave (UCDW) and unconventional spin density wave (USDW) have been proposed by several people as possible
electronic ground states in quasi-one-dimensional and quasi-two-dimensional crystals \[5–9\]. Unlike conventional density wave \[10\] the order parameter in UCDW $\Delta(\mathbf{k})$ depends on the quasiparticle wavevector $\mathbf{k}$. In $\alpha$-(ET)$_2$ 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 $\Delta(\mathbf{k}) = \Delta \cos(c_k z)$ (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 \[11\]. It is known also that the thermodynamics of UCDW and USDW is identical to the one in d-wave superconductors \[8, 12\].

Also, in spite of the clear thermodynamic signal, the first order term in $\Delta(\mathbf{k})$ usually vanishes when averaged over the Fermi surface. This implies no clear X-ray or spin signal for UCDW and USDW. Due to this fact unconventional density wave (i.e. UCDW and USDW) is sometimes called the phase with hidden order parameter \[9\]. In a magnetic field the quasiparticle spectrum is quantized as first shown by Nersesyan et al. \[5, 6\]. This dramatic change in the quasiparticle spectrum is most readily seen in ADMR, as demonstrated recently for SDW plus USDW in (TMTSF)$_2$PF$_6$ below $T = T^* (~ 4K)$ \[13, 14\].

About a decade ago ADMR in LTP in $\alpha$-(ET)$_2$ salts has been studied intensively. However the origin of ADMR has been hotly debated \[15–18\]. At that time the Fermi surface reconstruction due to nesting has been accepted as the most likely solution \[19, 20\].

In the following we shall show that the quasiparticle spectrum in UCDW in $\alpha$-(ET)$_2$ salts is quantized in the presence of magnetic field. The small energy gap which depends on both the direction and the strength of the magnetic field can be seen through ADMR. Indeed we can describe most aspects of ADMR seen in LTP of $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$ consistently. Therefore we may conclude that ADMR in $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$ provides conclusive evidence that the LTP is UCDW.

**Landau quantization.** – We shall consider the configuration shown in Fig. \[1\], where a magnetic field $\mathbf{B}$ is applied within the a-b plane (i.e. $\phi = 0$), by angle $\theta$ tilted away from the b axis and in the plane with angle $\phi$ from the a axis. For simplicity we shall limit our analysis to $\phi = 0$. Here the conducting plane is the a-c plane and the quasi-one-dimensional Fermi surfaces lie perpendicular to the a axis. In addition there is a quasi-two-dimensional Fermi surface with elliptical cross-section in the a-c plane.

Therefore we assume that there are two conducting channels in this system: the quasi-two-dimensional one stays in the normal state while the quasi-one-dimensional one undergoes UCDW transition around $T = 8K$. Then the quasiparticle spectrum in the quasi-1D channel is given by

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

(1)

where $\xi \approx v_a k_a$, $\Delta(\mathbf{k}) = \Delta \cos(c_k z)$ and $\varepsilon_0$ is the parameter describing the imperfect nesting \[21, 22\]. Finally $b'$ is the vector lying outside of the a-c plane. In order to fit the dip in ADMR at $\theta = \theta_0$ it is necessary to tilt $b'$ from the b axis by an angle $\theta_0$ \[18\]. As is seen from Eq. (1), the quasiparticle spectrum in the absence of magnetic field is gapless. When the magnetic field tilted by an angle $\theta$ from the b axis is applied, the lowest Landau level above the Fermi surface is given by

$$E(B, \theta) = \sqrt{2v_a \Delta \varepsilon |B \cos \theta|} - \varepsilon_0 \exp \left[ -\frac{2\Delta b' e}{v_a} |B| \sin^2(\theta - \theta_0) \right].$$

(2)

The first term in Eq. (2) is obtained following Nersesyan et al. \[5, 6\], while the second term in Eq. (2) comes from the spatial average of the second term in Eq. (1) using the wavefunction of the Landau level at the Fermi surface ($\sim \exp(-v_a e |B \cos \theta| z^2/2\Delta)$).
Noting the fact that the system has two conducting channels (i.e., quasi-1D Fermi surface and quasi-2D Fermi surface) and that only the quasi-1D Fermi surface is affected by the formation of UCDW, the ADMR is written as

\[ R(B, \theta) = \frac{1}{4\sigma_1 + \sigma_2}, \]

(3)

\[ x = \beta E(B, \theta), \]

(4)

where only the thermal excitations to the lowest Landau level are taken into account explicitly, which is doubly degenerated.

Similarly we obtain

\[ \frac{\Delta R}{R(0,0)} = \frac{2\sigma_1(e^x - 1)}{[4\sigma_1 + \sigma_2(1 + e^x)]}. \]

(5)

Comparison with experiments. – First, we compare Eq. (3) with \( R(B, \theta) \) versus \( T \) and \( R(B, \theta) \) versus \( B \) in Figs. 2 and 3. The temperature dependence of the magnetoresistance, for \( B=5T \) perpendicular to the a-c plane, is presented in Fig 2. Solid line is the fit based on Eq. (3). At low temperatures the consideration of the lowest Landau level provides convincing agreement. But the higher the temperature the higher Landau levels should be taken into account, and close to \( T_c \) the thermal fluctuations play also an important role what we neglected here for simplicity. The strength of the two conducting channels was found to be \( \sigma_2/\sigma_1 = 0.372 \), and by assuming the weak coupling value of \( \Delta = 17K \) and using \( c = 9.778A/11 \), the Fermi velocity is obtained as \( v_a = 7 \times 10^6 \text{cm/s} \).

The magnetic field dependence of the magnetoresistance at \( T = 2.2 \text{ K} \) and \( T = 4.2 \text{ K} \) for magnetic field perpendicular to the a-c plane is shown in Fig. 3. Solid line is the fit based on the Eq. (3). At higher fields, where our simple approximation is valid, the agreement looks perfect again. Here \( \sigma_2/\sigma_1 = 0.24 \) and 0.48 for \( T = 2.2 \text{K} \) and 4.2K, respectively, and the
Fig. 2 – The temperature dependence of the magnetoresistance is plotted at $B = 5T$.

Fig. 3 – The magnetic field dependence of the magnetoresistance is shown at $T = 2.2K$ and $4.2K$.

extracted Fermi velocities (assuming again the weak coupling value of $\Delta$) are $v_a = 3 \times 10^6$ cm/s and $7 \times 10^6$ cm/s. Finally ADMR is shown in Fig. 4 as a function of angle $\theta$ at $T = 4.2K$, $B = 5T$. The solid line shows the fit to the theoretical model explained above. As is seen from the figure the global $\theta$ dependence is given by $x \approx \beta \sqrt{2v_a \Delta \alpha e|B \cos \theta|}$, since the data is taken at $T = 4.2K$ and $B = 5T$.

Fig. 4 – The angle dependent magnetoresistance is shown at $T = 4.2K$ and $B = 5T$. 
The ratio of the conductivities in the two channels is $\sigma_2/\sigma_1 = 0.36$. The different values of this ratio might arise from the fact that we considered only the explicit $B$ and $T$ dependence of the main mechanism coming from the UCDW condensate, which according to us is responsible for the general behaviour of the measured magnetoresistance, and we neglected the magnetic field and temperature dependence of the other possible conducting processes. The Fermi velocity is obtained as $v_f = 5 \times 10^6 \text{cm/s}$. From these data, the Fermi velocity turned out to be of the order of $10^6 \text{cm/s}$, and its uncertainty should also be affected by the exclusion of the other conducting mechanisms.

The dip structure in ADMR is described fairly well by assuming $\varepsilon_0 = 0.132K$ and $\theta_0 = 40^\circ$. This $\varepsilon_0$ value is an order of magnitude smaller than the one we needed to describe the threshold electric field observed in $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$ [2–4, 24]. However, compared to other similar data (see [18]), it appears that there are considerable variability in the magnitude of $\varepsilon_0$ and the angle $\theta_0$ in different crystals ($\theta$ varies between 35$^\circ$ and 50$^\circ$). Therefore, we may conclude that UCDW in a magnetic field describes a variety of features in ADMR in $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$ satisfactorily.

Concluding remarks. – From the analysis of the temperature dependence of the threshold electric field we have concluded earlier that the LTP in $\alpha$-(ET)$_2$ salts is most likely UCDW [3]. The present analysis of ADMR appears to confirm this identification. The quasiparticle spectrum in UCDW in magnetic field is quantized in general [5, 6]. This effect should be most readily seen by the angular dependent magnetoresistance as we have demonstrated in $(\text{TMTSF})_2\text{PF}_6$ [13, 14]. We believe that ADMR will provide a powerful technique to explore other possible UCDW or USDW states in transition metal trichalogenate [25] and URu$_2$Si$_2$ [26, 27] for example.

Acknowledgements. – We thank Peter Thalmeier, Amir Hamzić and Silvia Tomić for useful discussions on the related topics. One of the authors (B. D.) gratefully acknowledges the hospitality of the Max Planck Institute for the Physics of Complex Systems, Dresden, where part of this work was done. This work was supported by the Hungarian National Research Fund under grant numbers OTKA T032162 and T037451, and by the Ministry of Education under grant number FKFP 0029/1999.

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