ASPECTS OF UNCONVENTIONAL DENSITY WAVES

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**Abstract**

Recently many people discuss unconventional density waves (i.e. unconventional charge density waves (UCDW) and unconventional spin density waves (USDW)). Unlike in conventional density waves, the quasiparticle spectrum in these systems is gapless. Also these systems remain metallic. Indeed it appears that there are many candidates for UDW. The low temperature phase of \(\alpha\)-(BEDT-TTF)\(_2\)KHg(SCN)\(_4\), the antiferromagnetic phase in UR\(_{2}\)Si\(_2\), the CDW in transition metal dichalcogenite NbSe\(_2\), the pseudogap phase in high \(T_c\) cuprate superconductors, the glassy phase in organic superconductor \(\kappa\)-(BEDT-TTF)\(_2\)Cu[N(CN)\(_2\)]Br. After a brief introduction on UCDW and USDW, we shall discuss some of the above systems, where we believe we have evidence for unconventional density waves.

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1 Introduction

As is well known quasi-one dimensional electron systems have four canonical ground states: s-wave (spin-singlet) superconductor, p-wave (spin-triplet) superconductor, charge density wave (CDW) and spin density wave (SDW) [1, 2, 3, 4]. All of these states have quasiparticle (QP) energy gap $\Delta$ and their QP density decreases exponentially at low temperatures ($T \ll \Delta$). Also the thermodynamics of these states is practically described by the BCS theory of s-wave superconductors[5]. Indeed except for p-wave superconductors these ground states have been found and their properties are actively pursued even today. As to p-wave superconductors it is most likely realized in quasi-one dimensional superconductor Bechgaard salts or (TMTSF)$_2$X with X=PF$_6$ and ClO$_4$[6]. The thermal conductivity measurement show the presence of energy gap, and most recent NMR study indicates the triplet pairing[6].

However, since 1979 a new class of superconductors have appeared on the scene: heavy fermion superconductors (1979), organic superconductors (1980), high $T_c$ cuprate superconductors (1986), Sr$_2$RuO$_4$ (1994) and rare earth transition metal borocarbides (1994). Now most of these new superconductors look like unconventional and/or nodal[7, 8]. However only recently $d_{x^2-y^2}$-symmetry of both hole and electron doped superconductors have been established[9]. Also the superconductivity in Sr$_2$RuO$_4$ is f-wave[10, 11, 12] and the one in YNi$_2$B$_2$C and LnNi$_2$B$_2$C is s+g-wave[13, 14]. Therefore unconventional superconductivity has taken center stage in the 21st century physics.

Parallel to these developments many people consider unconventional and/or nodal density waves (UDW)[15, 16, 17, 18]. We believe now that the low-temperature phase (LTP) of $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$[19, 20, 21, 22], the antiferromagnetic phase in URu$_2$Si$_2$[23, 24], the CDW in 2H-NbSe$_2$[25], the pseudogap phase in high $T_c$ cuprates[16, 18, 26] and the glassy phase in $\kappa$-(ET)$_2$ salts[27] belong to UDW.

2 Physical properties of UCDW and USDW

First of all the thermodynamic properties of UDW are very well described in terms of mean field theory like the BCS one. In fact the thermodynamics of most of UDW is described in terms of the BCS theory for d-wave superconductors[8, 28]. Qualitatively the thermodynamics of d-wave superconductors is not much different from the one for s-wave superconductors. In particular a clear jump in the specific heat at $T = T_c$ (transition temperature) is observable in both cases. On the other hand at low temperatures, unlike in conventional DW, there are nodal excitations, giving rise to the power law specific heat like $C \sim T^2$. Also due to the nodal excitations UCDW and USDW are metallic down to $T = 0K$. Further unlike conventional density wave there is no clear x-ray or spin signal indicating the phase transition, since $\langle \Delta(k) \rangle = 0$. Here $\langle \ldots \rangle$ means average over the Fermi surface. For this reason UDW is an important candidate for states with hidden order parameter.
For the existence of UDW we need higher dimensionality and competing interactions. Therefore we can see here clearly the paradigm shift from quasi-one dimensional systems to quasi-two dimensional and three dimensional systems. Also in order to study UDW experimentally we need more subtle and delicate technique. In this context the angular dependent magnetoresistance provides a unique window to study UDW.

3 Angular dependent magnetoresistance in α-(BEDT-TTF)$_2$KHg(SCN)$_4$

The LTP in α-(BEDT-TTF)$_2$MHg(SCN)$_4$ with M=K, Tl, Rb is still controversial. This compound is quasi-two dimensional system with 1D like and 2D like Fermi surfaces as shown in Fig. 3[29]. From the magnetic phase diagram in a magnetic field $H \parallel b^*$, it is believed that the LTP is not SDW but a kind of CDW[30]. We have proposed recently that UCDW can account for a number of features in LTP of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ including the threshold electric field[19, 20, 31, 32]. More recently we have discovered that the angular dependent magnetoresistance (ADMR) observed in LTP can be interpreted in terms of Landau quantization of the quasiparticle spectrum in UCDW[15, 22, 33]. First let us assume that the QP spectrum in

![Figure 1: The Fermi surface of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ is shown in the left panel. In the right one the geometrical configuration of the magnetic field with respect to the conducting plane is plotted.](image)

UCDW is given by[22, 33]

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

(1)

where $\xi \approx v_a(k_a - k_F)$, $v_a$ is the Fermi velocity, $\Delta(k) = \Delta \cos(ck_z)$, $b'$ is the vector lying outside of the $a - c$ plane and $\varepsilon_0$ is the parameter describing the imperfect nesting[34, 35, 36, 21]. In fitting the experimental data we discovered that 1. Eq. (1) gives only one single dip in ADMR, 2. therefore the imperfect nesting term has to be generalized as

$$\varepsilon_0 \cos(2b'k) \longrightarrow \sum_n \varepsilon_n \cos(2b'_n k),$$  

(2)
where \( b' \) = \( b' \hat{r}_b + \tan(\theta_n)(\hat{r}_a \cos \phi_0 + \hat{r}_c \sin \phi_0) \), \( \varepsilon_n = \varepsilon_0 2^{-n} \), \( \tan(\theta_n) = \tan(\theta_0) + nd_0 \). Indeed ADMR has a broad peak at \( H \parallel b^* \) (or \( \theta = 0 \)) and exhibits a number of dips at \( \theta = \theta_n \) (see Fig. 3!)

\[
\tan(\theta_n) \cos(\phi - \phi_0) = \tan(\theta_0) + nd_0,
\]

where \( \tan \theta_0 \approx 0.5 \), \( d_0 \approx 1.25 \), \( \phi_0 \approx 27^\circ \) and \( n = 0, \pm 1, \pm 2 \ldots \) [37, 38]. Now in the presence of magnetic field \( H \) with the orientation described by \( \theta \) and \( \phi \) (see Fig. 3), the QP spectrum changes to

\[
E_n = \pm \sqrt{2nv_a \Delta c \cos \theta \mid B},
\]

where \( n = 0, 1, 2 \ldots \) This is readily obtained following Ref. [15]. The contribution from the imperfect nesting term is considered as a perturbation and the lowest order corrections to the energy spectrum are given by:

\[
E^1_0 = E^1_1 = - \sum_m \varepsilon_m \exp(-y_m),
\]

\[
E^2_1 = - \sum_m \varepsilon_m (1 - 2y_m) \exp(-y_m),
\]

where \( y_m = v_a b'^2 \mid B \cos(\theta) \mid [\tan(\theta) \cos(\phi - \phi_0) - \tan(\theta_m)]^2 / \Delta c \). The \( n = 1 \) level was twofold degenerate, but the imperfect nesting term splits the degeneracy by \( E^1_1 \) and \( E^2_1 \). 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 2 conducting channels and only the quasi-one dimensional one is affected by the appearance of UCDW, the ADMR is written as

\[
R(B, \theta, \phi)^{-1} = 2\sigma_1 \left( \frac{\exp(-\beta E_1) + \cosh(\beta E^1_1)}{\cosh(\beta E_1) + \cosh(\beta E^1_1)} + \frac{\exp(-\beta E_1) + \cosh(\beta E^2_1)}{\cosh(\beta E_1) + \cosh(\beta E^2_1)} \right) + \sigma_2
\]

Eq. (7) is compared to the ADMR data taken from a single crystal of \( \alpha-(BEDT-TTF)_2 KHg(SCN)_4 \) for the temperature interval 1.4-20 K under magnetic field up to 15 T[33]. The ADMR data are consistent with the previous reports [38, 39, 40]. In Fig. 2 we compare the \( B \) dependence of the magnetoresistance at \( T = 1.4 \) K and \( T = 4.14 \) K and the \( T \) dependence of the magnetoresistance for \( B = 15 \) T, for \( \theta = 0^\circ \). 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 \( \Delta(T) \) [17]. 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
Figure 2: The temperature dependent magnetoresistance is shown at $B = 15T$ in the left panel. The dots are the experimental data, the solid line is our fit. In the right panel, the magnetoresistance is plotted for $T = 1.4K$ and $4.14K$ as a function of magnetic field. The thick solid line is the experimental data, the thin one denotes our fit based on Eq. (7).

away. Clearly in this high field region the quantization of Fermi surface itself starts interfering with the quantization described above. Also the deviation of the theoretical curve from the experimental one above $T_c$ in Fig. 2 is due to the fact that the higher Landau levels contribute in this high temperature regime. From these fittings we can deduce $\sigma_2/\sigma_1 \sim 0.1$ and 0.3, and by assuming the mean field value of $\Delta$ (17 K), we get $v_a \sim 6 \times 10^6$ cm/s. In Fig. 3 we show the experimental data of ADMR as a function of $\theta$ for current parallel and perpendicular to the conducting plane for $T = 1.4K$, $B = 15T$ and $\phi = 45^\circ$. 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. This $b'$ is comparable to the lattice constant $b = 20.56$ Å. Finally we show in Fig. 4 $R$ versus $\theta$ for different $\phi$ and compare with the experimental data side by side. Perhaps there are still differences in some details but the overall agreement is very striking. The present model can describe a similar figure found in Ref. [40] rather well.

In summary the Landau quantization of the QP spectrum of UDW as proposed by Nersesyan et al.[15] can account for the striking ADMR found in LTP of $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$. Very similar ADMR have been seen also in M=Rb and Tl compounds. Therefore we conclude that LTP in $\alpha$-(BEDT-TTF)$_2$MHg(SCN)$_4$ salts should be UCDW. Also we believe that ADMR provides clear signature for the presence of UCDW and USDW. Therefore this technique can be exploited for other possible candidates of UDW.

4 Pseudogap phase in high $T_c$ cuprates

We believe that the most important legacy of high $T_c$ cuprates is that the mean field theory like the Landau theory of Fermi liquid[41, 42, 43] and the BCS theory of superconductivity[5]
Figure 3: The angular dependent magnetoresistance is shown for current parallel (left panel) and perpendicular (right panel) to the $a-c$ plane at $T = 1.4$K, $B = 15T$, $\phi = 45^\circ$. The open circles belong to the experimental data, the solid line is our fit based on Eq. (7).

Figure 4: ADMR is shown for current perpendicular to the $a-c$ plane at $T = 1.4$K and $B = 15T$ for $\phi = -77^\circ$, $-70^\circ$, $-62.5^\circ$, $-55^\circ$, $-47^\circ$, $-39^\circ$, $-30.5^\circ$, $-22^\circ$, $-14^\circ$, $-6^\circ$, $2^\circ$, $10^\circ$, $23^\circ$, $33^\circ$, $41^\circ$, $48.5^\circ$, $56^\circ$, $61^\circ$, $64^\circ$, $67^\circ$, $73^\circ$, $80^\circ$, $88.5^\circ$, $92^\circ$ and $96^\circ$ from bottom to top. The left (right) panel shows experimental (theoretical) curves, which are shifted from their original position along the vertical axis by $n \times 100$Ohm, $n = 0$ for $\phi = -77^\circ$, $n = 1$ for $\phi = -70^\circ$, \ldots.

works in the quasi-two dimensional system with strong electron correlations\[44, 45\]. Of course the Fermi liquid theory as formulated by Landau for a spherical Fermi surface cannot be applied directly to high $T_c$ cuprates. In particular the quasi-two dimensionality and the resulting nesting feature of the Fermi surface has to be considered. But this feature is readily handled in terms of the renormalization group theory of two dimensional Fermi liquid\[46, 47, 48\]. Also as to the superconductivity the one band Hubbard model will give the simplest starting point. Then as discussed by Scalapino and others, $d_{x^2-y^2}$ superconductivity follows immediately\[49, 50\]. Further if
one limits oneself to single crystals of optimally doped high $T_c$ cuprates, one can do quantitative test of the BCS theory of d-wave superconductor in the weak coupling limit[28, 45, 51]. Unfortunately until now only three kinds of single crystals of high $T_c$ cuprates are available: LSCO, YBCO and Bi2212. If you compare $\Delta(0)/k_B T_c = 2.14$, the weak coupling theory prediction[28] for d-wave superconductor ($\Delta(0)$ is the maximum of the energy gap) to the one obtained for the optimally doped single crystals, we obtain 2.14, 2.8, 5 for LSCO, YBCO and Bi2212 respectively. This means that the superconductivity in LSCO is very close to the weak coupling limit, the one in YBCO is moderately in the strong coupling limit, while Bi2212 is definitely in the strong coupling limit[8]. In Fig. 5 a generic phase diagram of the hole doped high $T_c$ cuprates is shown. It is still controversial where $T^*$ line hits the superconducting transition temperature curve $T_c$. But from the validity of the mean field theory at optimal doping we assume that it hits somewhat in the underdoped side. Then it is possible that the extension of this line continues to $T = 0$ K at $x = 0.15$ at the quantum critical point. On this point we may refer to an earlier resistivity measurement in high magnetic field though it is limited unfortunately to only LSCO system[52]. Therefore the d-wave superconductivity in high $T_c$ cuprates is well understood in terms of two dimensional one band Hubbard model except one caveat: what means $T^*$? Earlier it was believed that $T^*$ is a crossover temperature where either superconducting or antiferromagnetic fluctuations becomes important[53]. More recently possible phase transition to d-wave density wave at $T = T^*$ has been proposed[16, 18, 26]. The most serious objection to this model is that no jump in the specific heat at $T = T^*$ has been observed until now, though many physical quantities like nuclear spin lattice relaxation rate $T_1^{-1}$, magnetic susceptibility, electric conductivity exhibit kinks at $T^*$[54]. The d-wave nature of density wave has been established by angular dependent photo-electron spectrum study[55]. Another less indirect signature of d-wave is the surprising relation $\Delta(0)/T^* = 2.14$ (the weak coupling result for d-wave density wave as well as for d-wave superconductor[17, 28]) established by STM study of $\Delta(0)$ (the energy gap in the density of states at $T = 0$ K) in LSCO, YBCO and Bi2212[56]. Therefore the only remaining question is if it is UCDW or USDW. We have proposed recently that USDW can interpret very readily two crucial experiments observed in the pseudogap phase in high $T_c$ cuprates YBCO and Bi2212: the weak antiferromagnetism[57] and the optical dishroism in ARPES[58]. Sidis et al. observed the appearance of the weak antiferromagnetism at $T = T^*$. This feature is qualitatively very similar to the weak antiferromagnetism observed in URu$_2$Si$_2$[24]. Unfortunately the temperature dependence of the intensity of the AF amplitude is rather different from the one in URu$_2$Si$_2$. But there are a few more possible contributions what we have neglected. In this picture the spin configuration of USDW is given by $S^\pm = S_x \pm i S_y$ lying in the $a - b$ plane. There are many attempts to describe this feature in terms of orbital angular momentum, but these models look too artificial. Perhaps the optical dishroism observed in the pseudogap phase in Bi2212 is still more controversial[58]. Indeed this is predicted by Chandra Varma based on a three band Hubbard model with a complicated order parameter[59]. There are many works
Figure 5: Left panel: The schematic phase diagram of high $T_c$ cuprates. Right panel: The angular dependent magnetoresistance is shown as a function of $\theta$ for current parallel to the $a$-axis for $\phi = 0^\circ$ (dashed line) and $\phi = 45^\circ$ (solid line).

trying to reinterpret this feature in terms of orbital currents associated with d-wave density wave[60].

One of the natural consequence of d-wave SDW with spin component $S^\pm$ is the optical dichroism as observed by Kaminski et al.[58]. The fact that the spin component lies in the $a-b$ plane is consistent with neutron scattering experiment[57]. Making use of the standard procedure to calculate ARPES, we find

$$I_\pm \sim 1 \pm \frac{\Delta(k)}{E(k)}$$

or

$$P = \frac{I_+ - I_-}{I_+ + I_-} = \frac{\Delta(k)}{E(k)},$$

where $\Delta(k) = \Delta \cos(2\phi)$ and $E(k)$ is the QP energy. Eq. (8) tells that the optical dichroism is proportional to $\cos(2\phi)$. In particular $P = 0$ for $k$ in the nodal directions while $P$ takes the maximum value at the antinodal directions. These facts are consistent with experiment[58]. We expect also in a uniform ground state the 100 % dichroism. But small dichroism is mostly due to the nonuniform ground state. We further expect the spin polarization of the outcoming electrons parallel to the photon polarization.

Also we propose that the angular dependent magnetoresistance will be a powerful method to investigate the d-wave density wave in high $T_c$ cuprates[61]. In a magnetic field $\mathbf{H}$ applied as shown in Fig. 3 (after replacing $a \rightarrow b, b \rightarrow c, c \rightarrow a$), the QP spectrum in d-wave density wave
changes to
\[ E_\pm = \sqrt{2\sqrt{2} e|H| \Delta (v_F a|\cos(\theta)| + v_c c \sin(\theta)\sin(\phi \pm \pi/4))}, \]
where \( v_F \) and \( v_c \) are the Fermi (in-plane) and perpendicular velocity, respectively. Again we followed Ref. [15] and neglected the imperfect nesting terms for simplicity. Therefore the magnetoresistance is given by
\[ \rho(H, \theta, \phi) - \rho(0, \theta, \phi) = \frac{e^{\beta(E_+ + E_-)} - 1}{e^{\beta E_+} + e^{\beta E_-} + 2} \]
A typical \( \theta \) dependence is shown in Fig 5.

Although this cannot distinguish between d-wave CDW and SDW, at least this will provide a unique test of the UDW proposed in high \( T_c \) cuprates.

Very recently d-wave symmetry of the superconductivity in heavy fermion layered compound CeCoIn\(_5\) and in the organic superconductor \( \kappa-(\text{BEDT-TTF})_2\text{Cu(NCS)}_2 \) have been established[62, 63]. Further both of these superconductors lie in the vicinity of a kind of antiferromagnetic state (most likely a kind of SDW)[27, 64]. Perhaps the most surprising phenomenon is the dependence of superconductivity in \( \kappa-(\text{BEDT-TTF})_2\text{Cu[N(CN)}_2\text{]Br} \) on the cooling rate[27]. For clarity we consider two extreme cases: the well annealed crystals are kept at liquid \( \text{N}_2 \) temperature for three days before final slow cooling to the temperature region around 10 K, while the quenched crystals were cooled down to the liquid He temperature within one hour. Surprisingly the superconducting transition temperature is little affected by the different cooling procedure. But from the diamagnetic response it is shown that the superfluid density in the quenched sample is less than 1% of the annealed sample. Further the temperature dependence of the superfluid density of the annealed sample is consistent with the one in d-wave superconductor, while the one for the quenched sample can be interpreted in terms of the one in s-wave superconductor. Therefore we suspect that the origin of the controversy over d-wave versus s-wave superconductivity lies in the question of the cooling rate. It is well understood that disorder in the ethylene groups attached to the BEDT-TTF molecule is destructive to superconductivity, though we do not know how. The slow cooling through the glassy transition temperature (100 K-70 K) where the ethylene group disorder sets in, helps to form more ordered ethylene groups[64]. Also it is very likely that disorder in the ethylene group is more disastrous to superconductivity than to SDW. Then a natural question is if this kind of SDW is USDW or not. Unfortunately, there is no experimental data on the characterization of this antiferromagnetic order parameter. Therefore we are sure that ADMR will be very useful to clarify this question.

Also can the weak superconductivity or gossamer superconductivity[65] found in the quenched sample be described in terms of coexisting d-wave superconductivity and d-wave SDW? We believe this is one of the most interesting questions in organic superconductors.
5 Concluding remarks

We have seen that UCDW and USDW are very likely realized in organic conductors, in heavy fermion systems and in the pseudogap phase in high $T_c$ cuprates. Also we have proposed that the angular dependent magnetoresistance will provide a unique probe to discover UDW. In particular we have identified successfully UCDW in $\alpha$-(BEDT-TTF)$_2$KHg(SCN)$_4$. Also we have pointed out that there are many similarities among the pseudogap phase in high $T_c$ cuprates, the glassy phase in organic superconductor $\kappa$-(BEDT-TTF)$_2$X and the 115 compounds in heavy fermion systems including CeCoIn$_5$ and PuCoGa$_5$[66]. The latter system with superconducting transition temperature $T_c = 18$ K is of great interest. Also as unconventional superconductivity becomes the superconductivity of the 21st century, we are confident that UCDW and USDW will be the density wave in this new century.

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