Heat transport in magnetic fields by quantum spin liquid in the organic insulators \(\text{EtMe}_3\text{Sb}[\text{Pd(dmit)}_2]_2\) and \(\kappa-(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3\)

V. R. Shaginyan\(^1\), A. Z. Msezane\(^2\), K. G. Popov\(^3\), G. S. Japaridze\(^2\) and V. A. Khodel\(^4,5\)

\(^1\)Petersburg Nuclear Physics Institute - Gatchina, 188300, Russia
\(^2\)Clark Atlanta University - Atlanta, GA 30314, USA
\(^3\)Komi Science Center, Ural Division, RAS - Syktyvkar, 167982, Russia
\(^4\)Russian Research Center Kurchatov Institute - Moscow, 123182, Russia
\(^5\)McDonnell Center for the Space Sciences and Department of Physics, Washington University

St. Louis, MO 63130, USA

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Abstract – Measurements of the low-temperature thermal conductivity collected on insulators with geometrical frustration produce important experimental facts shedding light on the nature of quantum spin liquid composed of spinons. We employ a model of strongly correlated quantum spin liquid located near the fermion condensation phase transition to analyze the exciting measurements of the low-temperature thermal conductivity in magnetic fields collected on the organic insulators \(\text{EtMe}_3\text{Sb}[\text{Pd(dmit)}_2]_2\) and \(\kappa-(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3\). Our analysis of the conductivity allows us to reveal a strong dependence of the effective mass of spinons on magnetic fields, to detect a scaling behavior of the conductivity, and to relate it to both the spin-lattice relaxation rate and the magnetoresistivity. Our calculations and observations are in a good agreement with experimental data.

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obtained dependence at low temperatures resembles that of the spin-lattice relaxation rate \(1/T_1(T)\) at fixed temperature as a function of the magnetic field \(B\) at low fields is insensitive to \(B\), displaying a response to the increasing magnetic field \(B\). On the other hand, it is suggested that the observed \(B\)-dependence implies that some spin-gap-like excitations coupling to the magnetic field are also present at low temperatures \([4,5]\). As a result, we face a challenging problem of interpretation of the experimental data in a consistent way, including the \(B\)-dependence of the heat conductivity.

In this letter we explain the \(B\)-dependence of the low-temperature thermal conductivity \(\kappa\) in EtMe\(_3\)Sb[Pd(dmit)_2]_2 and \(\kappa\)-(BEDT-TTF)_2Cu_2(CN)_3. We employ a model of strongly correlated quantum spin liquid (SCQSL) located near the fermion condensation phase transition (FCQPT) to analyze the \(B\)-dependence of \(\kappa\) \([7–10]\). Our analysis allows us to detect a scaling behavior of \(\kappa(B,T)\), and relate it to the scaling behavior of the spin-lattice relaxation rate \(1/T_1\) measured on both the herbervitsmithite ZnCu\(_3\)(OH)\(_6\)Cl\(_2\) and the heavy-fermion (HF) metal YbCu\(_{5−x}\)Au\(_x\), and the magnetoresistivity measured on the HF metal YbRh\(_2\)Si\(_2\). Our calculations are in good agreement with experimental data.

Representing a special case of QSL, SCQSL is a quantum state of matter composed of spinons —chargeless fermionic spinons with spin 1/2 \([7,9,10]\). In insulating compounds, SCQSL can emerge when interactions among the magnetic components are incompatible with the underlying crystal geometry, leading to a geometric frustration generated by the triangular and kagome lattices of magnetic moments, as it is the case in the ZnCu\(_3\)(OH)\(_6\)Cl\(_2\), see, e.g., \([11–17]\). In the case of ideal two-dimensional (2D) lattice the frustration of the lattice leads to a dispersionless topologically protected branch of the spectrum with zero excitation energy known as the flat band \([18–20]\).

Then, FCQPT can be considered as quantum critical point of SCQSL, composed of chargeless heavy spinons with \(S = 1/2\) and the effective mass \(M^*_{mag}\) occupying the corresponding Fermi sphere with the Fermi momentum \(p_F\). Therefore, the properties of insulating compounds coincide with those of heavy-fermion metals with one exception: it resists the flow of electric charge \([7,9,10]\). As we are dealing with compounds confining non-ideal triangular and kagome lattices, we have to bear in mind that the real magnetic interactions and possible distortion of the lattices can shift the SCQSL from the exact FCQPT, positioning it somewhere near FCQPT. Therefore, the actual location of the SCQSL phase in fig. 1 has to be established by analyzing the experimental data only.

In the vicinity of the FCQPT, pronounced deviations from the behavior of Landau Fermi liquid (LFL) with regard to physical properties are observed. These so-called non-Fermi-liquid (NFL) phenomena are related to the action of strong enhancement of the effective mass \(M^*_{mag}\) associated with the FCQPT. We note that there are different kinds of instabilities of LFL connected with several perturbations of the initial quasiparticle spectrum \(\varepsilon(p)\) and occupation numbers \(n(p),\) associated with strong enhancement of the effective mass and leading to the emergence of a multi-connected Fermi surface, see, e.g., \([8,21–23]\). Depending on the parameters and analytical properties of the Landau interaction, such instabilities lead to several possible types of restructuring of the initial LFL ground state. This restructuring generates topologically distinct phases. One of them is the fermion condensation associated with FCQPT, another one belongs to a class of topological phase transitions, where the sequence of rectangles \(n(p) = 0\) and \(n(p) = 1\) is realized at \(T = 0\). In fact, at elevated temperatures the systems located at these transitions exhibit a behavior typical to those located at FCQPT \([8]\). Therefore, we do not consider the specific properties of these topological transitions, and focus on the behavior of systems located near FCQPT.

We start with a brief outline of the effective mass dependence on magnetic field and temperature, \(M^*_{mag}(B,T)\). The key point of the formalism is the extended quasiparticle paradigm when the effective mass is no more constant but depends on temperature \(T\), magnetic field \(B\) and other external parameters such as pressure \(P\) \([8]\). To study the low-temperature transport properties, scaling behavior, and the effective mass \(M^*_{mag}(B,T)\) of SCQSL, we use the model of a homogeneous HF liquid. In that case, the model permits to avoid complications associated with the crystalline anisotropy of solids \([8]\), while the Landau equation, describing the effective mass \(M^*\) of HF liquid, reads \([8,24]\)

\[
\frac{1}{M^*_\sigma(B,T)} = \frac{1}{M} + \sum_\sigma \int \frac{dp}{p_F^\sigma} F_{\sigma,\sigma}(p_F,p) \times \frac{\partial n_{\sigma}(p,T,B)}{\partial p} \left(\frac{2\pi}{p}\right)^3, \tag{1}
\]

where \(M\) is the corresponding bare mass, \(F_{\sigma,\sigma}(p_F,p)\) is the Landau interaction, which depends on the Fermi momentum \(p_F\), momentum \(p\) and spin index \(\sigma\). The distribution function \(n\) can be expressed as

\[
n_{\sigma}(p,T) = \left\{1 + \exp \left(\frac{\varepsilon(p,T) - \mu_{\sigma}}{T}\right)\right\}^{-1}, \tag{2}
\]

where \(\varepsilon(p,T)\) is the single-particle spectrum. In our case, the chemical potential \(\mu\) depends on the spin due to Zeeman splitting \(\mu_{\sigma} = \mu \pm \mu_B B\), where \(\mu_B\) is the Bohr magneton.

In LFL theory, the single-particle spectrum is a variational derivative of the system energy \(E[n_{\sigma}(p,T)]\) with respect to the occupation number \(n, \varepsilon(p,T) = \delta E[n(p)]/\delta n\). The choice of the interaction and its parameters is dictated by the fact that the system has to be at FCQPT \([8,25,26]\). Thus, the sole role of the Landau interaction is to bring the system to the FCQPT point, where the Fermi surface alters its topology so that the effective mass acquires temperature and field dependence \([8,23,25]\). The variational
procedure, being applied to the functional $E[n_σ(p, T)]$, gives the following form for $ε_σ(p, T)$:

$$\frac{∂ε_σ(p, T)}{∂p} = \frac{p}{M} - \sum_{σ_1} \int \frac{∂F_{σ, σ_1}(p, p_1)}{∂p} n_{σ_1}(p_1, T) \frac{d^3p_1}{(2π)^3}.$$  

(3)

Equations (2) and (3) constitute the closed set for the self-consistent determination of $ε_σ(p, T)$ and $n_σ(p, T)$ and the effective mass, $p_F/M_{max} = \partialε(p)/∂p|_{p=p_F}$. We emphasize here that in our approach the entire temperature and magnetic-field dependence of the effective mass is brought to us by dependences of $ε_σ(p)$ and $n_σ(p)$ on $T$ and $B$. At $B = 0$, the effective mass strongly depends on $T$ demonstrating the NFL behavior [8,25]

$$M^*(T) \simeq a_2T^{-2/3}.$$  

(4)

At finite $T$, the application of the magnetic field $B$ drives the system to the LFL region with

$$M^*(B) \simeq a_B B^{-2/3}.$$  

(5)

A deeper insight into the behavior of $M^*(B, T)$ can be achieved using some “internal” (or natural) scales. Namely, near FCQPT the solutions of eq. (1) exhibit a behavior so that $M^*(B, T)$ reaches its maximum value $M_{max}$ at some temperature $T_M \propto B$ [8]. It is convenient to introduce the internal scales $M_{max}$ and $T_M$ to measure the effective mass and temperature. Thus, we divide the effective mass $M^*$ and the temperature $T$ by the values, $M_{max}$ and $T_M$, respectively. This generates the normalized effective mass $M^*_N = M^*/M_{max}$ and the normalized temperature $T_N = T/T_M$. Near FCQPT the normalized solution of eq. (1) $M^*_N(T_N)$ can be well approximated by a simple universal function of $T_N$ [8]. The interpolation occurs between the LFL andNFL regimes and represents the universal scaling behavior of $M^*_N$ [8]

$$M^*_N(y) \simeq c_0 \frac{1 + c_2y^2}{1 + c_2y^{8/3}}.$$  

(6)

Here, $y = T_N = T/T_M$, $c_0 = (1 + c_2)/(1 + c_1)$, $c_1, c_2$ are fitting parameters. The magnetic field $B$ enters eq. (1) only in the combination $μ_B B/T$, making $T_M \sim μ_B B$. It follows from eq. (6) that

$$T_M \simeq a_1 μ_B B,$$  

(7)

where $a_1$ is a dimensionless factor. Thus, in the presence of fixed magnetic field the variable $y$ becomes $y = T/T_M \sim T/μ_B B$. Taking into account eq. (7), we conclude that eq. (6) describes the scaling behavior of the effective mass as a function of $T$ vs. $B$ — the curves $M^*_N$ at different magnetic fields $B$ merge into a single one in terms of the normalized variable $y = T/T_M$. Since the variables $T$ and $B$ enter symmetrically, eq. (6) describes the scaling behavior of $M^*_N(B, T)$ as a function of $B$ vs. $T$. The normalization procedure deserves a remark here. Namely, since the magnetic-field dependence of $M^*_N(B, T)$ at fixed $T$ does not have a maximum, the normalization is performed at its inflection point, occurring at $B = B_{inf}$. As a result, we have $y = B/B_{inf}$ and $M^*_N = M^*(B, T)/M^*(B_{inf}, T)$. In other words, the curves $M^*_N$ at different $T$ merge into a single one in terms of the normalized variable $y = B/B_{inf}$, while eq. (7) transforms into the equation

$$μ_B B_{inf} \simeq a_2 T,$$  

(8)

with $a_2$ a dimensionless factor.

Now we construct the schematic phase diagram of SCQSL of the organic insulators EtMg(Sb[Pd(dmit)])$_2$ and κ-(BEDT-TTF)$_3$Cu$_2$(CN)$_3$. The phase diagram is reported in fig. 1. We assume that at $T = 0$ and $B = 0$ the system is approximately located at FCQPT without tuning. Both magnetic field $B$ and temperature $T$ play the role of the control parameters, shifting the system from FCQPT and driving it from the NFL to LFL regions as shown by the vertical and horizontal arrows. At fixed temperatures the increase of $B$ drives the system along the horizontal arrow from the NFL region to the LFL one. On the contrary, at fixed magnetic field and increasing temperatures, the system transits along the vertical arrow from the LFL region to the NFL one. The hatched area denoting the transition region separates the NFL state from the weakly polarized LFL state and contains the solid line tracing the transition region: $T^* = T_M(B)$. Referring to eq. (7), this line is defined by the function $T^* \propto μ_B B$, and the width $W(B)$ of the NFL state is seen to be proportional to $T$. In the same way, it can be shown that the width $W^*(B)$ of the transition region is also proportional to $T$. 

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As was mentioned above, SCQSL plays a role of HF liquid. Thus, we expect that SCQSL in organic insulators behaves like the electronic HF liquid in HF metals, provided that the charge of an electron is zero. In that case, the thermal resistivity $w$ of SCQSL is related to the thermal conductivity $\kappa$

$$w = \frac{L_0 T}{\kappa} = w_0 + A_w T^2.$$  

(9)

In magnetic fields, the resistivity $w$ behaves like the electrical magnetoresistivity $\rho_B = \rho_0 + A_\rho T^2$ of the electronic liquid, since $A_w$ represents the contribution of spinon-spinon scattering to the thermal transport, being analogous to the contribution $A_\rho$ to the charge transport, defined by electron-electron scattering. Here, $L_0$ is the Lorenz number, $\rho_0$ and $w_0$ are residual resistivity of electronic liquid and QSL, respectively, and the coefficients $A_w \propto (M_{mag}^*)^2$ and $A_\rho \propto (M^*)^2$ [8]. Thus, in the LFL region the coefficient $A_w$ of the thermal resistivity of SCQSL under the application of magnetic fields at fixed temperature behaves like the spin-lattice relaxation rate shown in fig. 2, $A_w(B) \propto A_\rho \propto 1/T_1(T) B \propto (M^*(B)_{mag})^2$ [26]. In accordance with eq. (5) as seen from fig. 2, panel A, the magnetic field $B$ progressively reduces $1/T_1$ [27,28], and $1/T_1$ as a function of $B$ possesses an inflection point at some $B = B_{inf}$ shown by the arrow. The same behavior is seen from fig. 2, panel B: The magnetic field $B$ diminishes the longitudinal magnetoresistivity [29], and as a function of $B$ it possesses an inflection point shown by the arrow. This behavior is consistent with the phase diagram displayed in fig. 1: At growing magnetic fields the NFL behavior first converts into the transition one and then transforms into the LFL behavior.

Figure 2, panels A and B, displays the normalized spin-lattice relaxation rates $(1/T_1)_{N}$ and the longitudinal magnetoresistivity $\rho_B$ at fixed temperature vs. the normalized magnetic field $B_N$, respectively. To clarify the universal scaling behavior of the herbertsmithite and the magnetic field $YbCu_{0.5-x}Au_x$, we normalize both the functions $1/T_1(T)$ and $\rho_B - \rho_0$ and the magnetic field. Namely, we normalize the functions by their values at the inflection point, and the magnetic field is normalized by $B_{inf}$, $B_N = B/B_{inf}$. Since $(1/T_1)_{N} = \rho_B - \rho_0 = (M^*_N(B))^2$ [8,26], we expect that the different strongly correlated Fermi systems located near FCQPT exhibit the same behavior of the effective mass, as is seen from fig. 2, panels A and B. We shall see below that the heat conductivity of the organic insulators exhibits the same behavior.

The study of the thermal resistivity $w$ given by eq. (9) allows one to reveal spinons as itinerant excitations. It is important that $w$ is not contaminated by contributions coming from localized excitations. The temperature dependence of the thermal resistivity $w$ represented by the finite term $w_0$ directly shows that the behavior of SCQSL is similar to that of metals, and there is a finite residual term $\kappa/T$ in the zero-temperature limit of $\kappa$. The presence of this term immediately proves that there are gapless excitation associated with the property of normal and HF metals, in which gapless electrons govern the heat and charge transport, revealing a connection between the classical physics and quantum criticality [30]. The finite $w_0$ means that in QSL both $k/T$ and $C_{mag}/T \propto M_{mag}$ remain nonzero at $T \to 0$. Therefore, gapless spinons, forming the Fermi surface, govern the specific heat and the transport. Key information on the nature of spinons is further provided by the $B$-dependence of the coefficient $A_w$. The specific $B$-dependence of $(1/T_1)_{N} \propto (M_{mag}^*)^2$, shown in fig. 2, panel A, and given by eq. (5), establishes the behavior of QSL as SCQSL. We note that the heat transport is polluted by the phonon contribution. On the other hand, the phonon contribution is hardly influenced by the magnetic field $B$. Therefore, we expect the $B$-dependence of the heat conductivity to be governed by $A_w(B,T)$. Consider the approximate relation

$$1 - \frac{A_w(B,T)}{A_w(0,T)} = 1 - \left(\frac{M^*(B,T)_{mag}}{M^*(0,T)_{mag}}\right)^2 \approx a(T) \frac{\kappa(B,T) - \kappa(0,T)}{\kappa(0,T)} \equiv a(T) I(B,T),$$  

(10)

where the coefficient $a(T)$ is $B$-independent. To derive (10), we employ eq. (9), and obtain

$$\frac{\kappa}{L_0 T} = \frac{1}{w_0 + A_w T^2 + b T^2}. \quad \text{(11)}$$

Here, the term $b T^2$ describes the phonon contribution to the heat transport. Upon carrying out simple algebra and

Fig. 2: (Color online) Panel A: the relaxation properties of the herbertsmithite vs. those of HF metals. The normalized spin-lattice relaxation rate $1/(T_1)N$ at fixed temperature as a function of the magnetic field: squares correspond to data on $(1/T_1)N$ extracted from measurements on ZnCu$_3$(OH)$_6$Cl$_2$ [27], while the triangles correspond to those extracted from measurements on YbCu$_{0.5}$Au$_x$ with $x = 0.4$ [28]. The inflection point, representing the transition region, where the normalization is taken is shown by the arrow. Our calculations based on eqs. (1) and eq. (6) are depicted by the solid curve, tracing the scaling behavior of $(M_N^*)^2$ and representing the $B$-dependence of the thermal resistivity $w$, see main text and eq. (9). Panel B: the normalized longitudinal magnetoresistivity $\rho_B$ vs. $B_N$, $\rho_0$ is extracted from measurements on YbRh$_2$Si$_2$ at different temperatures [29] listed in the legend. The solid curve represents our calculations of $(M_N^*)^2$.
assuming that \( 1 - A_{0}(B, T)/A_{0}(0, T) \) \( \leq 1 \), we arrive at eq. (10). It is seen from fig. 2, both panels, that the effective mass \( M^*_{N}(B) \propto M_{\text{mag}}(B) \) is a diminishing function of the magnetic field \( B \). Then, it follows from eqs. (5), (6) and (10) that the function \( I(B, T) = \kappa(B, T) - \kappa(0, T) \) increases at elevated field \( B \) in the LFL region, while \( I(B, T) \geq 0 \) in the NFL region, for the function is approximately independent of \( B \) in that case.

Recent measurements of \( \kappa(B) \) on the organic insulators EtMe_{3}Sb[Pd(dmit)\_2] \text{ and } \kappa-(BEDT-TTF)_{2}Cu_{2}(CN)_{3} \text{ [4,5] are displayed in figs. 3 and 4, panels A. The measurements show that the heat is carried by phonons and SCQSL for the heat conductivity is well fitted by \( \kappa(T) = b_{1} + b_{2}T^{2} \), where \( b_{1} \) and \( b_{2} \) are constants. The finite \( b_{1} \) term implies that spinon excitations are gapless in EtMe_{3}Sb[Pd(dmit)\_2] \text{, while in } \kappa-(BEDT-TTF)_{2}Cu_{2}(CN)_{3} \text{ gapless excitations are under debate [5]. A simple estimation indicates that the ballistic propagation of spinons seems to be realized in the case of EtMe_{3}Sb[Pd(dmit)\_2] \text{ [4,5]. It is seen from figs. 3 and 4, panels A, that } I(B, T) = \kappa(B, T) - \kappa(B = 0, T) \text{ demonstrates a strong } B\text{-dependence, namely the field dependence shows an increase of the thermal conductivity for rising fields } B \text{. Such a behavior is in agreement with eq. (5) and fig. 2 which demonstrate that } (M_{\text{mag}}(B))^{2} \text{ is a diminishing function of } B \text{. As a result, it follows from eq. (10) that } I(B, T) \text{ is an increasing function of } B \text{. Our calculations based on eqs. (3) and (10) are depicted by geometrical symbols in figs. 3 and 4, panels A. Since we cannot calculate the parameter } a(T) \text{ entering eq. (10) we use it as a fitting parameter. Temperature } T \text{ was also used to fit the data at temperatures shown in the legend in figs. 3 and 4. It is seen from figs. 3 and 4, panels A, that } I(B, T) \text{ as a function of } B \text{ possesses an inflection point at some } B = B_{\text{inf}}. \text{ To reveal the scaling behavior of the heat conductivity of the organic insulators, we normalize both the function } I(B, T) \text{ and the magnetic field by their values at the inflection points, as was done in the case of } (1/T_{N})_{T_{N}} \text{, see fig. 2. The normalized heat conductivity } I_{N}(B_{N}, T) \text{ does not depend on the factor } a(T), \text{ entering

Fig. 3: (Color online) Panel A: magnetic-field dependence of the thermal conductivity \( I(B, T) \) measured on the organic insulator EtMe_{3}Sb[Pd(dmit)\_2] \text{ and standardized by the zero-field value } \kappa, \text{ that } I(B, T) = \kappa(B, T) - \kappa(B = 0, T) \text{ at temperatures shown in the legend [4,5]. Our calculations are based on eq. (10) and shown by hexagons and stars. Panel B: the normalized thermal conductivity \( I_{N}(B_{N}, T) \text{ vs. } B_{N} \text{ shown by geometrical symbols is extracted from the data shown in panel A. The inflection point is shown by the arrow. The magnetic-field dependence of the function } (1 - 1/T_{N})_{T_{N}} \text{ is extracted from measurements of } (1/T_{N})_{T} \text{ shown in fig. 2, panel A. The solid curve is obtained from the theoretical curve in fig. 2.)
eq. (10), and its calculations do not have any fitting parameters. It is seen from figs. 3 and 4, panels B, that in accordance with eq. (6) \( I_N(B_N, T) \) exhibits the scaling behavior and becomes a function of a single variable \( B_N \). It is instructive to compare the normalized values of the function \[ 1 - \frac{M^*(B, T)}{M^*(B = 0, T)} \] extracted from measurements of \( (1/T_1)N \) shown in fig. 2, both panels A, with \( I_N(B_N, T) \). The extracted values are normalized by their values at the inflection points and the magnetic field is normalized by \( B_{inf} \), as is done in the case of \( (1/T_1)N \). It is seen from figs. 3 and 4, panels B, that \( I_N(B_N, T) \) and \( I_N(B_N, T) \) are in good overall agreement with the solid curve depicting the theoretical function \[ 1 - \frac{M^*(B, T)}{M^*(B = 0, T)} \], received from our calculations represented by the solid curve in fig. 2, both panels. It is seen that this function demonstrates a flat dependence at low \( B_N \), for at \( B_N < 1 \) the system is in its NFL state and the \( B \)-dependence is weak. Thus, there is no need to introduce additional quasiparticles activated by the application of a magnetic field in order to explain the growth of \( I(B, T) \) at elevated \( B \) [4,5]. It is also seen from both figs. 3 and 4, panels B, that the organic insulators demonstrate the same behavior as \( \text{ZnCu}_3(\text{OH})_6\text{Cl}_2 \), \( \text{YbCu}_{1-x}\text{Au}_x \), and \( \text{YbRh}_2\text{Si}_2 \).

In summary, for the first time, we have explained the magnetic-field dependence of the low-temperature thermal conductivity \( \kappa \) in the organic insulators \( \text{EtMe}_3\text{Sb}[\text{Pd(dmit)}_2]_2 \) and \( \kappa-(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3 \). Our analysis allows us to detect SCQSL in these organic insulators, exhibiting the universal scaling behavior.

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