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Heavy fermion spin liquid in herbertsmithite

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A B S T R A C T

We analyze recent heat capacity measurements in herbertsmithite $ZnCu_3(OH)_6Cl_2$ single crystal samples subjected to strong magnetic fields. We show that the temperature dependence of specific heat C_{mag} formed by quantum spin liquid at different magnetic fields B resembles the electronic heat capacity C_{el} of the HF metal $YbRh_2Si_2$. We demonstrate that the spinon effective mass $M^*_{mag} \propto C_{mag}/T$ exhibits a scaling behavior like that of C_{el}/T. We also show that the recent measurements of C_{mag} are compatible with those obtained on powder samples. These observations allow us to conclude that $ZnCu_3(OH)_6Cl_2$ holds a stable strongly correlated quantum spin liquid, and a possible gap in the spectra of spinon excitations is absent even under the application of very high magnetic fields of 18 T.

Quantum spin liquids (QSLs) are promising new phases, where exotic quantum states of matter could be realized. Although much theoretical effort has been devoted to understand their physical nature, the question is still far from its complete clarification. Generally speaking, the QSL is a quantum state, formed with hypothetic particles like fermionic spinons carrying spin 1/2 and no charge. A number of QSLs with various types of ground states are proposed [1–18] but the lack of real materials possessing them obscure the underlying physical mechanism. On the other hand, one needs a real theory that plays important role in the understanding and interpreting accessible experimental facts [19–22]. Measurements on magnetic insulators with geometrical frustration produce important experimental data shedding light on the nature of spinon composing QSL. Recent measurements indicate that the insulator $ZnCu_3(OH)_6Cl_2$ (herbertsmithite) is very likely to be the first promising candidate to host a QSL in real bulk materials. In herbertsmithite, the dynamic magnetic susceptibility shows that at low temperatures quasiparticle excitations, or spinons, form a continuum, and populate an approximately flat band crossing the Fermi level [17,21]. At the same time, our analysis of herbertsmithite thermodynamic properties allows us to reveal their scaling behavior. The above results demonstrate that the properties of the herbertsmithite are similar to those of heavy-fermion (HF) metals. Thus, it can be viewed as a new type of strongly correlated HF electrical insulator exhibiting properties of HF metals but resisting electric current [20–22].

The development of this concept, however, faces fundamental problems still remaining to be resolved. The first one is that the experimental data are taken in measurements on herbertsmithite powder samples [4–6,20–22]. As a result, both out-of-plane magnetic defects and site disorder between the Cu and Zn ions can strongly change the behavior of the bulk susceptibility or even alter its low-temperature scaling at weak magnetic fields, see e.g. Ref. [16]. The second problem is based on experimental data showing that QSL in the herbertsmithite is unstable against the application of external magnetic fields. This instability is represented by the emergence of so-called spin-solid phase, which, in turn, is separated from the spin-liquid one by a gap induced by an applied magnetic field [23]. Thus, the experimental observations of QSL remain scattered and need regimentation. In our opinion, this can be partially achieved by presenting a reliable interpretation of the recent measurements of specific heat C in high magnetic fields performed on herbertsmithite single crystal samples [24,25].

In this Communication we analyze the recent heat capacity C measurements in strong magnetic fields on herbertsmithite. We interpret the above measurements in terms of QSL spinon contribution. To obtain the spinon-determined partial specific heat C_{mag}, we subtract the phonon and Schottky effect contributions from the total heat capacity C. It turns out that obtained C_{mag} as a function

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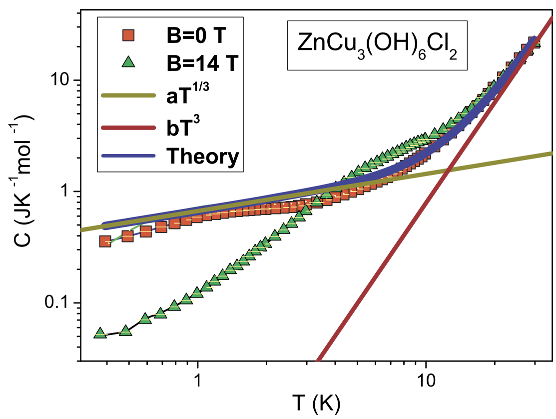

Fig. 1. (Color online.) The heat capacity measurements on single crystal of $ZnCu_3(OH)_6Cl_2$ at $B = 0$ (squares) and $B = 14$ T (triangles) [24,25]. The thick blue line corresponds to our theoretical approximation (5) with fitting parameters a and b. The lines represent $C = aT^{1/3}$ and $C = bT^3$, see legend.

of temperature T at fixed magnetic field B behaves very similarly to the electronic specific heat C_{el} of the HF metal $YbRh_2Si_2$. This observation allows us to conclude that spinons form the Fermi sphere with the Fermi momentum p_F, while the gap in the spectra of spinon excitations and the spin-solid phase are absent. We demonstrate that the effective mass of spinon $M^*_{mag} \propto C_{mag}/T$ exhibits a scaling behavior resembling that of C_{el}/T. We also show that new measurements of C_{mag} on single crystal samples are compatible with those obtained on powder samples.

The $S = 1/2$ spins of the Cu^{+2} ions occupy the positions in a highly symmetric kagome lattice. As spins on the above lattice reside in a highly symmetric structure of corner-sharing triangles, the ground state energy does not depend on the spins configuration. As a result, spins located at the kagome hexagon, composed of the two triangles, form a (frustrated) pattern that is even more frustrated than the triangular lattice considered by Anderson [26]. As a result, the kagome lattice has a flat topologically protected branch of the spectrum with zero excitation energy [27,28]. Therefore, the fermion condensation quantum phase transition (FCQPT) can be considered as a quantum critical point (QCP) of the $ZnCu_3(OH)_6Cl_2$ QSL [20,30]. In that case, we expect that at low temperatures the heat capacity C of herbertsmithite becomes a function of the applied magnetic field, as it is seen from Fig. 1. We propose that mentioned QSL is composed of chargeless fermions called spinons with the effective mass M^*_{mag}. Latter fermions with spin $\sigma = 1/2$ occupy the corresponding Fermi sphere with the Fermi momentum p_F, and form the excitation spectrum typical for HF liquid located near FCQPT, while spinons represent HF quasiparticles of deconfined QSL. Thus, the ground state energy $E(n)$ is given by the Landau functional depending on the spinon distribution function $n_\sigma(\mathbf{p})$, where $\mathbf{p}$ is the momentum. Near FCQPT point, the effective mass M^*_{mag} is governed by the Landau equation [29–31]

$$\frac{1}{M^*_{mag}(T,B)} = \frac{1}{M^*_{mag}(T=0,B=0)} + \frac{1}{p_F^2}\sum_{\sigma_1}\int \frac{\mathbf{p}_F\mathbf{p}_1}{p_F} F_{\sigma,\sigma_1}(\mathbf{p_F},\mathbf{p}_1)\frac{\partial \delta n_{\sigma_1}(\mathbf{p}_1)}{\partial p_1}\frac{d\mathbf{p}_1}{(2\pi)^3}. \quad (1)$$

Here we have rewritten the spinon distribution function as $\delta n_\sigma(\mathbf{p}) \equiv n_\sigma(\mathbf{p},T,B) - n_\sigma(\mathbf{p},T=0,B=0)$. The Landau interaction F is defined by the fact that the system has to be at FCQPT. Thus, this interaction brings the system to FCQPT point, where Fermi surface alters its topology so that the effective mass acquires temperature and field dependence [30,32,33]. At this point, the term $1/M^*_{mag}(T=0,B=0)$ vanishes and Eq. (1) becomes homogeneous. It can then be solved analytically. At $B = 0$, the effective mass depends on T demonstrating the NFL (non-Fermi liquid) behavior

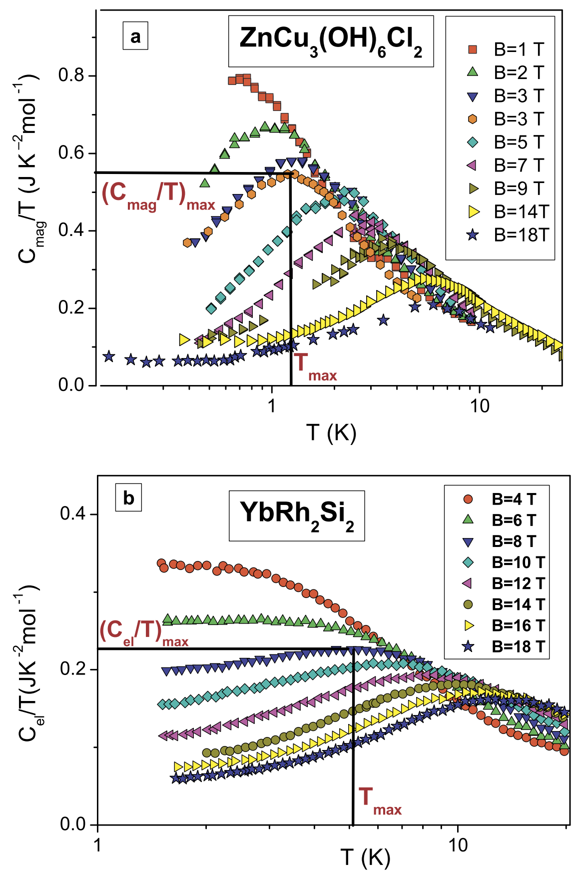

Fig. 2. (Color online.) Panel a: The specific heat C_{mag}/T of $ZnCu_3(OH)_6Cl_2$ measured on powder [5] and single crystal samples [24,25]. (C_{mag}/T) is displayed as a function of temperature T for fields B shown in the legends. Panel b: The specific heat C_{el}/T extracted from measurements on the HF metal $YbRh_2Si_2$ [34] exhibits the same behavior as C_{mag}/T. The illustrative values of the maxima $(C_{mag}/T)_{max}$ and $(C_{el}/T)_{max}$ of both C_{mag}/T and C_{el}/T and the corresponding T_{max} are also shown in panels a and b.

$$M^*_{mag}(T) \simeq a_T T^{-2/3}. \quad (2)$$

At finite T, the application of magnetic field B drives the system to Landau Fermi liquid (LFL) region with

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

At finite B and T near FCQPT, the solutions of Eq. (1) $M^*_{mag}(B,T)$ can be well approximated by a simple universal interpolating function. The interpolation occurs between the LFL ($M^*(T) \propto const$) and NFL ($M^*_{mag}(T) \propto T^{-2/3}$) regions. As it is seen from Eq. (3), under the application of magnetic field M^*_{mag} becomes finite, and at low temperatures the system demonstrates the LFL behavior $C_{mag}(T)/T \propto M^*_{mag}(T) \simeq M^*_{mag}(T=0) + a_1 T^2$. Then, as it is seen from Fig. 2, at increasing temperatures $M^*_{mag} \propto C_{mag}/T$ grows, and enters a transition regime, reaching its maximum $M^*_{max} \propto (C_{mag}(T)/T)_{max}$ at $T = T_{max}$, with subsequent diminishing given by Eq. (2).

To reveal a scaling behavior, we introduce the normalized effective mass M^*_N and the normalized temperature T_N dividing the effective mass M^*_{mag} by its maximal values, M^*_{max}, and temperature T by T_{max} at which the maximum occurs. The normalized effective mass $M^*_N = M^*/M^*_{max}$ as a function of the normalized temperature $y = T_N = T/T_{max}$ is given by the interpolating function that approximates the solution of Eq. (1) [30,31]