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Scaling in dynamic susceptibility of herbertsmithite and heavy-fermion metals

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ABSTRACT

We present a theory of the dynamic magnetic susceptibility of quantum spin liquid. The obtained results are in good agreement with experimental facts collected on herbertsmithite $ZnCu_3(OH)_6Cl_2$ and on heavy-fermion metals, and allow us to predict a new scaling in magnetic fields in the dynamic susceptibility. Under the application of strong magnetic fields quantum spin liquid becomes completely polarized. We show that this polarization can be viewed as a manifestation of gapped excitations when investigating the spin-lattice relaxation rate.

1. Introduction

Landau Fermi liquid (LFL) theory is highly successful in the condensed matter physics. The key point of this theory is the existence of fermionic quasiparticles defining the thermodynamic, relaxation and dynamic properties of the material. However, strongly correlated Fermi systems encompassing a variety of systems that display behavior not easily understood within the Fermi liquid theory and called non-Fermi liquid (NFL) behavior. A paradigmatic example of the NFL behavior is represented by heavy-fermion (HF) metals, where a quantum phase transition (QPT) induces a transition between LFL and NFL [1,2]. QPT can be tuned by different parameters, such as the chemical composition, the pressure, and the magnetic field. Magnetic materials, in particular copper oxides and organic insulators, are interesting subjects of study due to a quantum spin liquid (QSL) that can emerge when they approach QPT and are cooled to low temperature T. Exotic QSL is formed with such hypothetic particles as fermionic spinons carrying spin 1/2 and no charge. A search for the materials is a challenge for condensed matter physics [3]. The experimental studies of herbertsmithite $ZnCu_3(OH)_6Cl_2$ and the organic insulator $EtMe_3Sb[Pd(dmit)_2]_2$ have discovered gapless excitations, analogous to excitations near the Fermi surface in HF metals, indicating that $ZnCu_3(OH)_6Cl_2$ and $EtMe_3Sb[Pd(dmit)_2]_2$ are the promising systems to investigate their QPTs and QSLs [4–14]. The observed behavior of the thermodynamic properties of $ZnCu_3(OH)_6Cl_2$ strongly resembles that in HF metals since a simple kagome lattice being strongly frustrated has a dispersionless topologically protected branch of the spectrum with zero excitation energy [14–17]. This indicates that QSL formed by the ideal kagome lattice is located on the ordered side of the fermion condensation quantum phase transition (FCQPT) that is characterized by the presence of the spectrum with zero excitation energy [2]. This observation allows us to establish a close connection between QSL and HF metals whose HF systems are located near FCQPT and, therefore, exhibiting an universal scaling behavior [2,14,17]. As we are dealing with the real 3D compound $ZnCu_3(OH)_6Cl_2$ rather than with the ideal 2D kagome lattice, we have to bear in mind that the magnetic interactions and the presence of layers of nonmagnetic Zn^{2+} ions separating magnetic kagome planes in the substance can shift the QSL from the initial point, positioning it in front of or behind FCQPT. Therefore, the actual location has to be established by analyzing the experimental data. As a result, the location coincides with that of HF metals, and turns out to be at FCQPT [14,17], as it is shown in Fig. 1. Thus, FCQPT can be considered as QPT of $ZnCu_3(OH)_6Cl_2$ QSL and both herbertsmithite and HF metals can be treated in the same framework, so that QSL is composed of fermions and these with zero charge and spin $\sigma = \pm 1/2$ occupy

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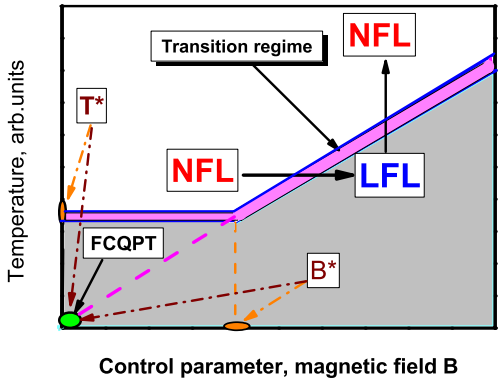

Fig. 1. (Color online.) T–B phase diagram of QSL and HF liquid. The vertical and horizontal arrows, crossing the transition region depicted by the thick lines, show LFL–NFL and NFL–LFL transitions at fixed B and T, respectively. At temperatures $T < T^*$ and magnetic field $B < B^*$ shown by the dash-dot arrows the effective mass $M^* \simeq const$ and the system in the LFL region. The dash line continuing the thick line represents the transition region provided the system were located at FCQPT shown by the arrow.

the corresponding Fermi sphere with the Fermi momentum p_F [2,14,17]. The ground state energy $E(n)$ is given by the Landau functional depending on the quasiparticle distribution function $n_\sigma(\mathbf{p})$, where $\mathbf{p}$ is the momentum. In spite of numerous experimental facts collected in measurements of inelastic neutron scattering spectrum and spin-lattice relaxation rates on herbertsmithite, a theoretical understanding of how the dynamical spin susceptibility of QSL behaves on approaching QPT and how it is affected by external parameters, such as the magnetic field, is still missing.

In this Letter we employ the Landau transport equation to construct the dynamical spin susceptibility. We elucidate how the calculated susceptibility is affected by magnetic field and describe experimental facts collected on herbertsmithite and heavy-fermion metals. The obtained results are in good agreement with the facts and allow us to predict a new scaling emerging under the application of magnetic field in the dynamic susceptibility. Taking into account that QSL becomes completely polarized in strong magnetic fields, we show that this polarization can be seen as the presence of gapped excitations when investigating the spin-lattice relaxation rate.

2. Dynamic spin susceptibility of quantum spin liquid and heavy-fermion metals

To construct the dynamic spin susceptibility $\chi(\mathbf{q},\omega,T) = \chi'(\mathbf{q},\omega,T) + i\chi''(\mathbf{q},\omega,T)$ as a function of momentum q, frequency ω and temperature T, we use the model of homogeneous HF liquid located near FCQPT [2]. To deal with the dynamic properties of Fermi systems, one can use the transport equation describing a slowly varying disturbance $\delta n_\sigma(\mathbf{q},\omega)$ of the quasiparticle distribution function $n_0(\mathbf{p})$, and $n = \delta n + n_0$. We consider the case when the disturbance is induced by the application of external magnetic field $B = B_0 + \lambda B_1(\mathbf{q},\omega)$ with B_0 being a static field and λB_1 a ω-dependent field with $\lambda \to 0$. As long as the transferred energy $\omega < qp_F/M^* \ll \mu$, where M^* is the effective mass and μ is the chemical potential, the quasiparticle distribution function $n(\mathbf{q},\omega)$ satisfies the transport equation [18]

$$(\mathbf{qv_p} - \omega)\delta n_\sigma - \mathbf{qv_p}\frac{\partial n_0}{\partial \varepsilon_p}\sum_{\sigma_1\mathbf{p}_1} f_{\sigma,\sigma_1}(\mathbf{pp}_1)\delta n_{\sigma_1}(\mathbf{p}_1)$$

$$= \mathbf{qv_p}\frac{\partial n_0}{\partial \varepsilon_p}\sigma\mu_B(B_0 + \lambda B_1). \tag{1}$$

Here μ_B is the Bohr magneton and ε_p is the single-particle spectrum. We assume that B_0 is finite but not so strong to lead to the full polarization of the corresponding quasiparticle band. In the field B_0, the two Fermi surfaces are displaced by opposite amounts, $\pm B_0\mu_B$, and the magnetization $\mathcal{M} = \mu_B(\delta n_+ - \delta n_-)$, where the two spin orientations with respect to the magnetic field are denoted by $\pm$, and $\delta n_\pm = \sum_p \delta n_\pm(\mathbf{p})$. The spin susceptibility χ is given by $\chi = \partial\mathcal{M}/\partial B_{|B=B_0}$. In fact, the transport equation (1) is reduced to two equations which can be solved for each direction $\pm$ and allows one to calculate $\delta n_\pm$ and the magnetization. The response to the application of $\lambda B_1(\mathbf{q},\omega)$ can be found by expanding the solution of Eq. (1) in a power series with respect to $M^*\omega/qp_F$. As a result, we obtain the imaginary part of the spin susceptibility

$$\chi''(\mathbf{q},\omega) = \mu_B^2\frac{\omega(M^*)^2}{2\pi q}\frac{1}{(1+F_0^a)^2}, \tag{2}$$

where F_0^a is the dimensionless spin antisymmetric quasiparticle interaction [18]. The interaction F_0^a is found to saturate at $F_0^a \simeq -0.8$ [19,20] so that $(1+F_0^a)$ is positive. It is seen from Eq. (2) that the second term is an odd function of ω. Therefore, it does not contribute to the real part χ' and forms the imaginary part χ''. Taking into account that at relatively high frequencies $\omega \geqslant qp_F/M^* \ll \mu$ in the hydrodynamic approximation $\chi' \propto 1/\omega^2$ [21], we conclude that the equation

$$\chi(\mathbf{q},\omega) = \frac{\mu_B^2}{\pi^2(1+F_0^a)}\frac{M^*p_F}{1 + i\pi\frac{M^*\omega}{qp_F(1+F_0^a)}}, \tag{3}$$

produces the simple approximation for the susceptibility χ and satisfies the Kramers–Kronig relation connecting the real and imaginary parts of χ.

To understand how can χ'' and χ given by Eqs. (2) and (3), respectively, depend on temperature T and magnetic field B, we recall that near FCQPT point the effective mass M^* depends on T and B, and is given by the Landau equation (LE) [2,22]. The interaction function F of LE is completely defined by the fact that the system has to be at FCQPT. The sole role of F is to bring the system to FCQPT, where the Fermi surface alters its topology so that M^* acquires T and B dependencies [2,23,24]. At FCQPT, LE can be solved analytically: At $B = 0$, the effective mass depends on T

$$M^*(T) \simeq a_T T^{-2/3}. \tag{4}$$

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

$$M^*(B) \simeq a_B B^{-2/3}. \tag{5}$$

Here a_T and a_B are constants. At finite B and T near FCQPT, the solutions of LE can be well approximated by a simple universal interpolating function. The interpolation occurs between the LFL ($M^*(T) \propto const$) and NFL ($M^*(T) \propto T^{-2/3}$) regions. It is convenient to introduce the normalized effective mass M_N^* and the normalized temperature T_N dividing the effective mass M^* 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 [2]

$$M_N^*(y) \approx c_0\frac{1 + c_1y^2}{1 + c_2y^{8/3}}. \tag{6}$$

Here $c_0 = (1+c_2)/(1+c_1)$, c_1 and c_2 are fitting parameters, making $M_N^*(y=1) = 1$. Magnetic field B enters LE only in the combination $\mu_B B/k_B T$, making $k_B T_{\max} \simeq \mu_B B$ where k_B is the