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Role of Frustration in Strongly Correlated Itinerant Electron Systems

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The role of geometric frustration in strongly correlated itinerant electron systems were investigated. It was argued that the frustration gives rise to a quantum spin liquid state even in itinerant electron systems by taking account of spin fluctuations. Paramagnetic neutron scattering and nuclear spin relaxation measurements have revealed that the ground state of paramagnetic YMn_2 can be regarded as the quantum spin liquid.

KEYWORDS: Frustration, Spin Fluctuations, Spin Liquid, Itinerant Electron, YMn_2

§1. Introduction

Frustration of magnetic interactions gives rise to various anomalous properties in the ground state magnetic structure and critical behavior at the transition point. Among them, the ground state of the so called fully frustrated (FFR) system, which has a macroscopic number of degenerate spin configurations and, therefore, can not take a unique magnetic structure, are attracting much attention. Anderson and coworkers[1] introduced the concept of quantum spin liquid (QSL) as a nonmagnetic ground state of the FFR system, which is characterized by strong spin fluctuations with antiferromagnetic pair correlation. So far, candidate for QSL have been searched in ionic compounds with quasi-two dimensional lattices. Recently, attempts are also extended to three dimensional lattice, in particular, pyrochlore compounds.[2] However, in most cases, a magnetically ordered phase is formed at low temperatures. On the other hand, metallic magnetic systems have not been intensively studied from the view point of frustration. Taking account of the spin fluctuation (SF) theory, there is no reason why the frustration plays an important role and a QSL state is realized in the strongly correlated itinerant systems with antiferromagnetic correlation. Figure 1 illustrates magnetic states and the magnitude of local magnetic moment as a function of t/U, where U is the intra-atomic correlation energy and t the transfer integral (We use t/U as the horizontal axis to avoid divergence of U/t in the local moment limit but discuss U/t in the text). Before the SF theory, itinerant electron and local moment systems are separated each other as shown in Fig. 1(a). In the itinerant electron side, an ordered state appears if the Stoner (Lidiard) condition is fulfilled ($U/t > 1$) and T_C (or T_N), and the spontaneous (sublattice) moment increase with increasing U/t. For $U/t < 1$, the ground state is a merely (exchange enhanced) Pauli paramagnet. It is difficult to take into consideration of the role of frustration in this picture. The SF theory has greatly modified this scheme[3] as shown in Fig. 1(b). The critical value of U/t and the critical temperature (T_C, T_N)

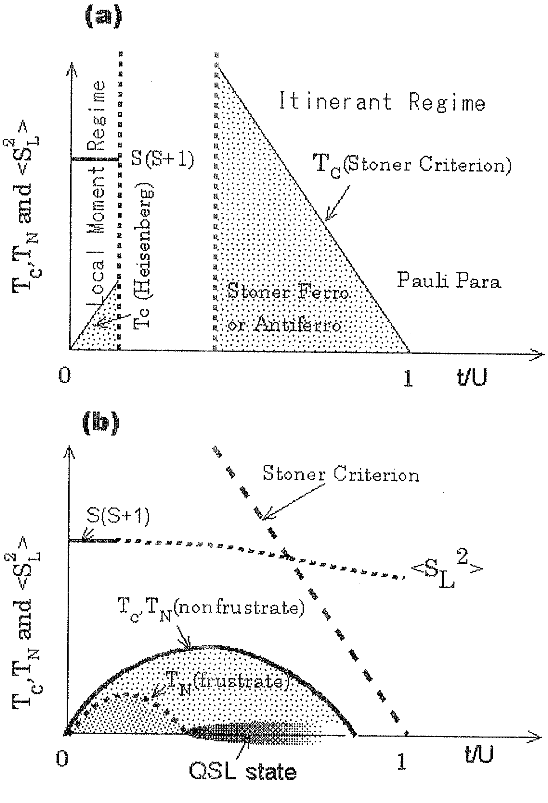

Fig. 1. Schematic diagram of magnetic phase and the magnitude of local moment $\langle S_L^2 \rangle$ as functions of the inverse relative correlation energy to transfer integral t/U. a) Classical model: The Heisenberg model for a localized moment system. (t corresponds to the inter-atomic exchange integral and $\langle S_L^2 \rangle$ to the Hunt rule value $S(S+1)$); the Stoner (Lidiard) model for an itinerant system. The phase boundary is determined by the Stoner criterion. b) Unified model: The ordered phase is considerably reduced from the Stoner criterion by taking account of SF. For an ideal fully frustrated system, no ordered state appears and the ground state is QSL. The frustration in the real system remarkably reduces the ordered region. The ground state of nonmagnetic region, near the boundary at least, can be regarded as also QSL state. $\langle S_L^2 \rangle_{total}$ would continuous to $S(S+1)$ value of the local moment limit.

are suppressed from the Stoner criterion. For the system without frustration, the paramagnetic phase near the boundary is characterized by the existence of long wave length spin fluctuations, even at 0 K as zero point fluctuations and, probably, non fermi liquid characters are observed. With increasing U/t, the amplitude of SF increases and the spatial distribution becomes local moment-like. Thus, the gap between itinerant and local moment systems disappears. In the real 3d electron system, the fluctuations become static and a magnetically ordered state is formed at low temperatures. What is the role of frustration in this picture? The resonating valence bond (RVB) theory[1] was originally proposed as the ground state of FFR system in the local moment system. In the present picture, RVB state or QSL state is also expected in the itinerant system with geometrical frustration, at least in large U/t region. Starting from the weak correlation limit, the frustration suppress the formation of the magnetically ordered state and so the critical t/U for an ordered state shifts the far left. Then, we can expect giant and spatially localized zero point antiferromagnetic SF for the paramagnetic ground state near to the phase boundary. This is nothing but QSL state. We should say that the transfer integral t helps the formation of QSL state and so that the non-magnetic ground state of frustrated itinerant systems should have more or less spin liquid characters. In this context, we have intensively studied the magnetic properties of Sc or Lu doped YMn_2 which can be regarded as a typical itinerant frustrated system with the QSL ground state.

§2. Spin Liquid Characters of Y(Sc,Lu)Mn_2

2.1 *Macroscopic Properties*

YMn_2 has the C15 Laves phase structure where Mn atoms forms a network of corner-sharing tetrahedra. Mn-Mn interaction is antiferromagnetic and, therefore, it can be regarded as a fully frustrated system. At ambient pressure, it becomes antiferromagnetic below 100 K with fairly large Mn moment of $3\mu_B$. The spin structure is not a simple collinear but helically modulated with an extremely long period of 400 Å.[4] The complex spin structure may be due to frustration. The antiferromagnetic state is unstable and highly sensitive to pressure or impurity. By substituting 3% Sc for Y, which reduces its lattice parameter a little, a paramagnetic state is stabilized down to the lowest temperature.[5] The paramagnetic state is not a simple Pauli paramagnet but exhibits several anomalies such as an extraordinary large low temperature specific heat of $\gamma = 150$ mJK^{-2} mol^{-1}, being comparable to a heavy fermion system.[6] The enhancement of the γ value implies the existence of large amplitude of spin fluctuations even at low temperatures.

2.2 *Neutron scattering of Y(Sc)Mn_2*

In order to detect the spin fluctuations, we have performed paramagnetic neutron scattering experiments using polarized neutron beam. Figure 2 shows the quasi-elastic Q-scan spectra with energy window of $2\Gamma_{res}$ = 20 meV for $Y_{0.97}Sc_{0.03}Mn_2$ at 10 K and 290 K. A large scattering was observed centered around 1.6 Å^{-1}. The corresponding wave length is approximately twice of the inter-atomic distance, indicating antiferromagnetic correlation of spin fluctuations. It should be noted that large scattering is observed even at 1.4 K, suggesting the existence of strong zero-point fluctuations with antiferromagnetic correlation. This interpretation is confirmed by the energy spectrum , which is shown in Fig. 3. At 10 K, scattering was observed only on the neutron energy-loss side ($\omega > 0$), implying the absence of thermally excited fluctuations. The width of the spectra is approximately 20meV (200K), indicating the characteristic frequency of

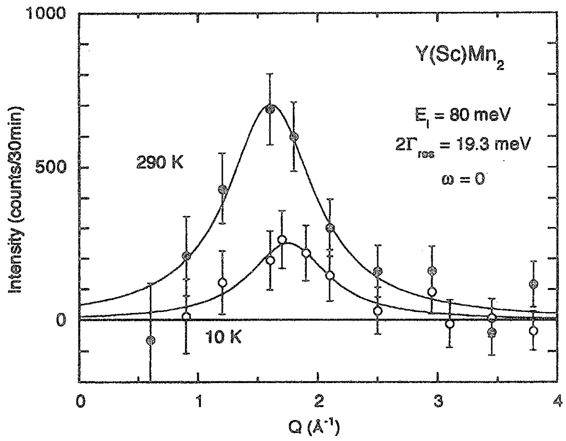

Fig. 2. Magnetic neutron scattering rate of Y(Sc)Mn_2 as a function of wave vector Q at zero energy transfer with energy resolution of $2\Gamma_{res}$ = 19.3 meV at 10 K(○) and 290 K (●).

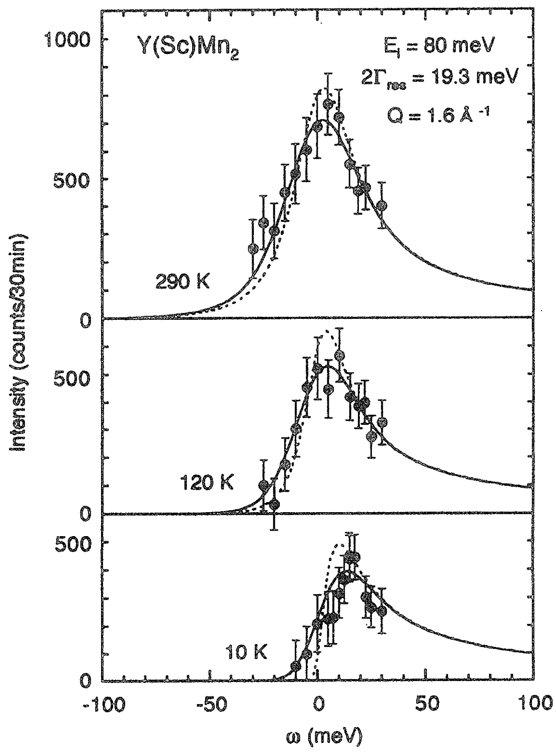

Fig. 3. Magnetic neutron scattering rate of Y(Sc)Mn_2 as a function of energy transfer measured for a momentum transfer of Q = 1.6 Å. Dashed curves indicate deconvoluted values.

zero point fluctuations of 5 x 10^{12} Hz. At high temperatures, the scattering amplitude increases on both side, indicating an increase of thermally excited spin fluctuations. From the integrated intensity of scattering, we have roughly estimated the local amplitude of SF as 1.2 μ_B at 10 K and 1.5 μ_B at 290 K. Figure 4 represents the schematic feature of temperature variation of SF. The total amplitude of local SF, $\langle S_L^2 \rangle$, stays almost constant against temperature. At 0K, the fluctuations are purely quantum ones and in this sense, we may regard the ground state being in the QSL state. With increasing temperature, low lying modes of SF are easily excited thermally and so the amplitude of thermal SF, $\langle S_L^2 \rangle_{th}$ increases rapidly, giving rise to the enhancement of low temperature specific heat. The heavy fermion characters of the present system may be thus explained.

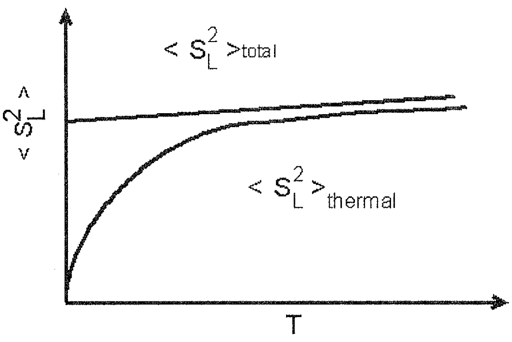

Fig. 4. Schematic diagram of the temperature dependence of local amplitude of spin fluctuations for a strongly correlated frustrated itinerant system.

2.3 Nuclear Spin Relaxation in Y(Lu)Mn$_2$

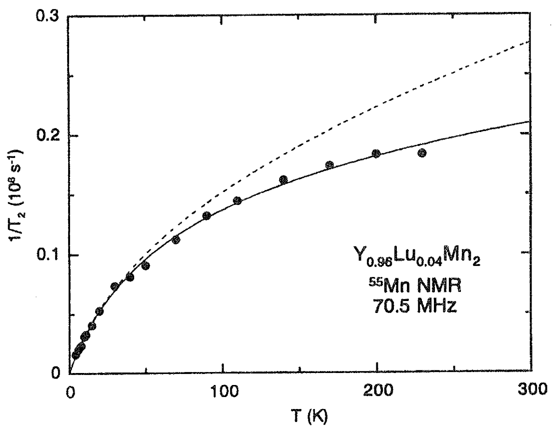

Fig. 5. Temperature dependence of ^{55}Mn spin-spin relaxation rate, $1/T_2$ of $Y_{0.96}Lu_{0.04}Mn_2$. The solid curve indicates fitting by eq. (2.3), dotted line by $T/(T+\theta)^{1/2}$, which is applicable to a nearly antiferromagnetic itinerant system.

Recently, Ballou *at al.*[7] have carried out neutron scattering experiments using a single crystal of Y(Sc)Mn$_2$ and revealed that SF exhibit anisotropic distribution in the reciprocal space. Canals and Lacroix[8] have shown that these anisotropic SF are characteristic of the pyrochlore structure with spin liquid characters and that can be expressed by the generalized susceptibility as

$$\chi^{-1}(Q+q,\omega) = \chi_Q^{-1} + a_1(q_x^2+q_y^2) + a_2q_z^4 - i\omega/\Gamma \quad (2.1)$$

For these anisotropic SF, Lacroix *et al.*[9] calculated the temperature dependence of the nuclear spin relaxation time, T_1 and obtained an expression as

$$1/T_1 \propto T\cdot\chi(Q)^{3/4} \quad (2.2)$$

Using the Curie-Weiss relation for the staggered susceptibility, we have

$$1/T_1 \propto T/(T+\Theta)^{3/4} \quad (2.3)$$

Except at very low temperatures, the spin-spin relaxation time, T_2, is also mediated by SF, then it is plausible the T_2 is proportional to T_1. In order to confirm this prediction, we have carefully measured the temperature dependence of T_2 of $Y_{0.96}Lu_{0.04}Mn_2$ (We doped Lu to stabilize the paramagnetic state because Lu has no NMR isotope). The result is shown in Fig. 5. As seen in the figure, we have fairly good fitting by eq. (2.3), supporting the spin-liquid character of SF in this system.

§3. Concluding Remarks

We argued that the geometrical spin frustration, which obstructs the formation of a magnetically ordered state, gives rise to the spin liquid state even (or rather more easily) in the strongly correlated itinerant electron system. By measuring paramagnetic neutron scattering of Y(Sc)Mn$_2$, we have demonstrated that the ground state of paramagnetic YMn$_2$ can be regarded as a quantum spin liquid. The temperature dependence of T_2 of ^{55}Mn nucleus in Y(Lu)Mn$_2$ was carefully measured and interpreted in terms of anisotropic SF expected for the 3D frustrated system.

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