PHYSICAL REVIEW B **71**, 060402(R) (2005)

Spin fluctuations and carrier mass enhancement in pyrochlore $Y_{2-x}Bi_xRu_2O_7$

Makoto Tachibana, Hitoshi Kawaji, and Tooru Atake
Materials and Structures Laboratory, Tokyo Institute of Technology, 4259 Nagatsuta-cho, Midori-ku, Yokohama, 226-8503, Japan
(Received 17 September 2004; published 8 February 2005)

We present magnetic susceptibility and heat capacity data for pyrochlore $Y_{2-x}Bi_xRu_2O_7$ ($0 \leqslant x \leqslant 2$). With decreasing x, the ground state of the system evolves from paramagnetic (PM) metal to spin-glass (SG)-like metal, and to an antiferromagnetic (AF) Mott insulator. The low-temperature heat capacity in the metallic phases can be represented as $\gamma T + \beta T^3 + \delta T^3 \ln T$, with significant enhancement of both γ and δ as the SG-like phase is approached in the PM phase. The $\delta T^3 \ln T$ term signals spin fluctuations in the nearly localized regime, possibly enhanced by the strong geometrical frustration. The heat capacity of the integer-spin ($S=1$) insulator $Y_2Ru_2O_7$ suggests the presence of gapped spin excitations in the cubic AF phase.

DOI: 10.1103/PhysRevB.71.060402 PACS number(s): 75.50.Ee, 71.30.+h, 71.28.+d, 65.40.Ba

The discovery of heavy-fermion behavior in spinel LiV_2O_4 (Ref. 1) has led to the recognition that the combination of strongly correlated metal and geometrical spin frustration results in complex and unusual properties. This is particularly significant because these two phenomena were mostly studied separately in the past, with both phenomena receiving considerable interest in recent years. On one hand, the understanding of electron correlation in Mott-Hubbard systems has made a great progress in the last two decades,[2,3] which was fueled by the discovery of high-T_c superconducting cuprates and colossal magnetoresistive manganites. Especially, there are now many systems with controlled parameters of the on-site Coulomb interaction U, bandwidth W, and band-filling n that make possible the access to various ground states. On the other hand, the study of geometrical frustration in localized moment systems has been equally fruitful.[4] In those spinel and pyrochlore oxides that have the corner-sharing tetrahedral network (pyrochlore lattice) of magnetic ions, such exotic ground states as spin liquid, spin glass, and spin ice have been discovered.[4] Therefore, it was immediately speculated[1] that the heavy-fermion behavior in LiV_2O_4, an example in d-electron system, might be ascribed to the intimate interplay between strong electron correlation and antiferromagnetic (AF) geometrical frustration. Recent experiments, using both bulk[5] and microscopic[6] techniques, corroborate this view.

An important experimental method in investigating the effect of electronic correlation, and metal-insulator transition (MIT) observed in some systems, is the systematic control of bandwidth or band filling via chemical substitution.[2] This is particularly useful in ternary or multinary transition metal oxides, where electronic properties can be varied while keeping the important metal-oxygen network intact. Using such techniques, important phenomena such as Brinkman-Rice mass enhancement, AF metal phase, and non-Fermi-liquid-like behavior have been studied in the vicinity of MIT.[2,3,7] Similar techniques have also been employed on the geometrically frustrated $Li_{1-x}Zn_xV_2O_4$,[5] where the ground state evolves from heavy-fermion to spin glass to AF Mott insulator with increasing x. Thus, in this case, one approaches the heavy-fermion state from a geometrically frustrated insulator. However, an important and challenging issue is to explore the other side of the (hypothetical) phase diagram, i.e., the emergence of the heavy-fermion state from a normal metal, with increasing electron correlation in a geometrically frustrated system. In an attempt to explore the role of electron correlation in geometrically frustrated system, we have studied the pyrochlore $Y_{2-x}Bi_xRu_2O_7$ with MIT at $x \sim 0.5$. Here the pyrochlore network of Ru ions provides the charge and spin degrees of freedom, while the bandwidth and electronic properties can be systematically controlled by changing x. In this paper, we report the observation of spin fluctuation contribution ($\delta T^3 \ln T$) to the low-temperature heat capacity in metallic $Y_{2-x}Bi_xRu_2O_7$. The $\delta T^3 \ln T$ term signals spin fluctuations in an almost localized Fermi liquid,[3,8] and is prominent in such strongly correlated systems as UAl_2 (Ref. 9) and the heavy-fermion UPt_3.[10] Moreover, we find that the coefficient δ scales with the enhancement of electronic heat capacity coefficient γ, establishing significant coupling between charge and spin degrees of freedom in such a strongly correlated and geometrically frustrated system.

The cubic pyrochlore $Y_2Ru_2O_7$ is a Mott insulator[11,12] with the local moment $S=1$ for Ru^{4+}, and a large negative Weiss temperature of $\Theta_w \sim -1100$ K.[13] It undergoes an AF transition at $T_N = 77$ K, with a complex noncollinear spin configuration and the ordered moment of $1.36\mu_B$/Ru.[14] The transition is not accompanied by a change in the lattice structure.[14,15] On the other hand, the isostructural $Bi_2Ru_2O_7$ is a Pauli paramagnetic metal with AF spin interactions.[16] The metallic behavior in $Bi_2Ru_2O_7$ results from the greater overlap between the Ru-4d-t_{2g} and O-2p orbitals,[12] though some studies[17] have suggested that there may be some mixing of the Bi-6p state as well. The substitution of Bi for Y causes an insulator-to-metal transition at $x \sim 0.5$, as determined by transport[11,18] and spectroscopic[12] measurements. Therefore, $Y_{2-x}Bi_xRu_2O_7$ should be a desirable system to study systematically the effect of electron correlation on a geometrically frustrated lattice, and to explore the possible relationship with the heavy fermion behavior in LiV_2O_4.

Polycrystalline samples of $Y_{2-x}Bi_xRu_2O_7$ ($0 \leqslant x \leqslant 2$) were prepared from stoichiometric amounts of Y_2O_3, Bi_2O_3, and RuO_2 by solid-state reactions in air. Successively lower sintering temperatures were employed with increasing x, which varied from 1473 K for $x=0$ to 1223 K for $x=2$. Powder x-ray diffraction measurements indicated that the products are single phase. Magnetic susceptibility was measured on the heating direction in the range 2–300 K, using a supercon-

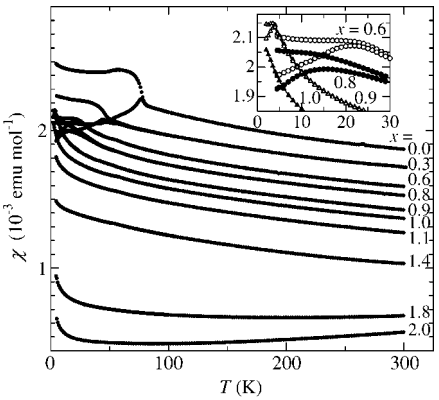

FIG. 1. The dc magnetic susceptibility of $Y_{2-x}Bi_xRu_2O_7$ taken under $H=10$ kOe, showing zero-field-cooled (lower) and field-cooled (upper) curves. For $x \geq 1.0$, these curves coincide within experimental accuracy. The inset shows the low-temperature data for $x=0.6$, 0.8, 0.9, and 1.0.

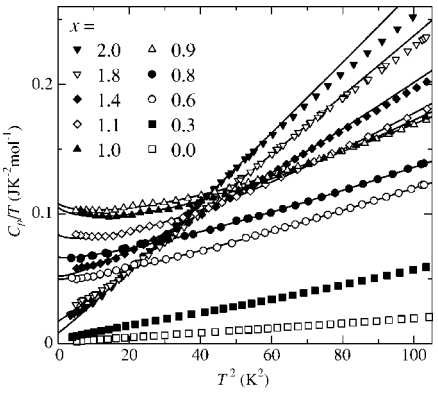

FIG. 2. The low-temperature heat capacity of $Y_{2-x}Bi_xRu_2O_7$ plotted as C_p/T vs T^2. The lines are the best fits by $C_p=\gamma T+\beta T^3+\delta T^3 \ln T$ as described in the text.

ducting quantum interference device (SQUID) magnetometer. Heat capacity (C_p) was measured in the range 1.8–300 K with a Quantum Design PPMS employing a heat-pulse relaxation technique. The C_p measurements were performed on both heating and cooling directions, and the results were indistinguishable within experimental accuracy.

The temperature (T) dependence of the magnetic susceptibility χ is shown in Fig. 1. The measurements were performed in a field of 10 kOe, both in zero-field-cooled (ZFC) and field-cooled (FC) conditions. All data shown have been corrected for core diamagnetism. The results are in good agreement with the previous study[11] and show the following effects as the Bi content x increases: (1) For $x \leqslant 0.9$, a peak with hysteresis is observed in the low-T region, the temperature of which decreases with increasing x. These peaks have been attributed to spin-glass (SG) transition,[11] though they are unusual in that the applied magnetic field of up to 10 kOe has a very small influence on the shape of the ZFC data.[11] Recent neutron diffraction measurements[14] observed long-range AF ordering for $x=0$, and as seen from Fig. 4 there is a corresponding anomaly in C_p for $x \leqslant 0.3$ but not for $x \geqslant 0.6$; therefore, we attribute the former as AF ordering and the latter as SG-like freezing. (2) At room temperature, χ decreases smoothly with increasing x and the behavior changes gradually from Curie-Weiss-like for $x \leqslant 1.4$ to Pauli-like for $x \geqslant 1.8$. These are consistent with the transitional behavior from the localized to itinerant character of the electron states. For $x \leqslant 1.4$, χ deviates slightly from the modified Curie-Weiss form $\chi_0+C/(T+\Theta_w)$ below 300 K, possibly due to the high negative Weiss temperatures.[13] There seems to be no obvious discontinuity near the MIT boundary of $x \sim 0.5$, reminiscent of such systems as $NiS_{2-x}Se_x$ and $La_{1-x}Sr_xVO_3$.[2,3] For $x=1.8$ and 2.0, upturns at low T are attributable to small amounts of extrinsic spins.

The low-T C_p data are shown in Fig. 2, which are plotted as C_p/T versus T^2. There are several interesting changes in C_p with the change in x. First, the slope or the lattice contribution increases rapidly with increasing x. Above 9 K, where the lattice contribution dominates over electronic contributions, C_p increases with increasing x. As discussed below, this can be attributed to the lattice softening induced by the static disorder from the Bi-$6s^2$ lone-pair electrons. For $x \leqslant 0.3$, C_p/T approaches a small value (<2 mJ K^{-2} mol^{-1}) in the limit of $T \to 0$ K, indicating the presence of a true gap in the electronic excitation spectrum. A small *downturn* is observed for $x=0.3$ below 3 K, and such a downturn has been considered as a signature of the barely localized regime.[7] Above this region, these data can adequately be fitted with $\gamma T+\beta T^3$.

For $x \geqslant 0.6$, which are in the metallic phase, C_p can be fitted accurately with $C_p=\gamma T+\beta T^3+\delta T^3 \ln T$ (Solid lines in Fig. 2). The last term signifies spin fluctuations in an almost localized Fermi liquid,[8,19] which are specified by the form $\delta T^3 \ln(T/T_0)$ with T_0 being the characteristic temperature. Therefore, the coefficient β contains both $-\delta \ln T_0$ and the usual lattice contribution β_{ph}. It should be noted that the upward curvatures seen for $x \geqslant 0.6$ are much broader than those upturns expected from magnetic impurities, and replacing the $\delta T^3 \ln T$ term with T^{-2} or T^{-1} does not fit the data well at all. These observations, together with the systematic variation of the $\delta T^3 \ln T$ contribution with x (discussed below; see Fig. 3), provide strong evidence for the spin fluctuation term in $Y_{2-x}Bi_xRu_2O_7$. For some compositions such as $x=1.8$ and 2.0, small positive deviations from the fit are observed below 3 K. These deviations can be accurately fitted with an addi-

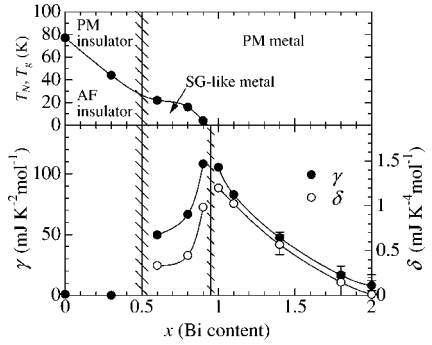

FIG. 3. (a) The phase diagram of $Y_{2-x}Bi_xRu_2O_7$. The antiferromagnetic (AF) or spin-glass (SG)-like transition temperature is determined from the magnetic susceptibility data. (b) The x dependence of the electronic heat capacity coefficient γ and the spin fluctuation coefficient δ. The lines are to guide the eye.

tional T^{-2} contribution, indicating that they are of magnetic impurities in origin.[20] This may be related to the low sintering temperatures necessary for the synthesis of single-phase samples of high x. Also, for $x \geqslant 1.4$, an additional $\beta_5 T^5$ term (with $\beta_5 < 0$) is needed to fit the negative curvature of C_p that starts below 10 K. This is consonant with the low Debye temperature implied from the large slope, and we have restricted the fitting to the temperature region where the lattice contribution obeys the T^3 behavior. For $x \leqslant 1.1$, the $\beta_5 T^5$ term is not needed for $T < 10$ K, and narrowing the fitting region does not change the parameters in any significant manner. Also, by estimating the lattice $\beta_{\rm ph} T^3$ contribution from the temperature region where the C_p/T versus T^2 plot becomes effectively linear, we obtain $T_0 \sim 13$–15 K for each x. We did not observe any significant systematic behavior of T_0 as a function of x.

The systematic change in the value of γ and δ, together with the phase diagram as determined from the magnetic susceptibility data, are shown in Fig. 3 as a function of x. The systematic variation of γ with x is in fair agreement with Ref. 11, though the $\delta T^3 \ln T$ contribution and the actual C_p data were not reported in that study. The overall phase diagram and the corresponding changes in γ are remarkably similar to other Mott-Hubbard systems undergoing MIT, such as $NiS_{2-x}Se_x$ (Ref. 21) and $La_{1-x}Sr_xVO_3$.[22] In $Y_{2-x}Bi_xRu_2O_7$, the SG-like metal phase appears to be replacing the AF metal phase observed in other systems, which may be a consequence of the geometrical frustration. There is a significant enhancement of γ with decreasing x in the paramagnetic (PM) metal phase, which reaches a large value of 105 mJ K^{-2} mol^{-1} (53 mJ K^{-2} Ru mol^{-1}) for $x = 1.0$. Together with the enhancement of χ_0 with decreasing x,[11] this is the characteristic of Brinkman-Rice mass enhancement implying a high density of states at the Fermi level, and this behavior is observed in various Mott-Hubbard systems. However, $Y_{2-x}Bi_xRu_2O_7$ differs from the others in that there is an additional contribution from the $\delta T^3 \ln T$ term in C_p. It can be seen from Fig. 3 that the enhancement of δ closely tracks that of γ in the PM metal phase, reaching 1.2 mJ K^{-4} mol^{-1} (0.6 mJ K^{-4} Ru mol^{-1}) for $x = 1.0$.

The $\delta T^3 \ln T$ term describes the low-T C_p of a strongly correlated system where the energy spectrum is noticeably affected by spin fluctuations,[8] and is seen in such materials as UAl_2 (Ref. 9) and the heavy-fermion UPt_3.[10] In these f-electron systems, the Kondo coupling is effective in modifying the low-energy excitation spectrum, and the heavy-fermion behavior, as well as the $\delta T^3 \ln T$ term in C_p, result from such renormalization.[23] On the other hand, the $\delta T^3 \ln T$ term is rare among transition metal oxides (TMO). One example where this term has been reported is $LaNiO_3$, though the contribution is very small ($\delta = 0.12$ mJ K^{-4} mol^{-1}).[24] In TMO, spin fluctuations usually manifest themselves as the enhanced γ in C_p and/or the $T^{1.5}$ term in resistivity, which become especially conspicuous in the vicinity of MIT.[2,3] In this respect, it is interesting to note that the low-T C_p of LiV_2O_4 can be described well[1] with the spin-fluctuation term, with $\gamma = 428$ mJ K^{-2} mol^{-1}, $\delta = 1.6$ mJ K^{-4} mol^{-1}, and $T_0 = 25$ K. Therefore, LiV_2O_4 and $Y_{2-x}Bi_xRu_2O_7$ are unusual among TMO in that their low-T properties resemble those of strongly correlated f-electron systems. As these oxides are

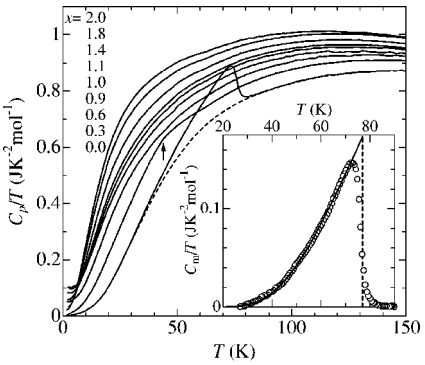

FIG. 4. Heat capacity of $Y_{2-x}Bi_xRu_2O_7$ (solid lines). The closely spaced experimental points are joined, and the data for $x = 0.8$ are omitted, for clarity. The arrow marks the magnetic anomaly for $x = 0.3$. The dashed line corresponds to the baseline for $Y_2Ru_2O_7$, which was used to obtain the magnetic heat capacity C_m (open circles in the inset). In the inset, the vertical dashed line marks T_N and the solid line is the fit to $C_m = A \exp(-\Delta/T)$ as described in the text.

both strongly correlated and geometrically frustrated, strong spin fluctuations arising from the combination of these two phenomena may be invoked to be the origin of the unusual properties. It should be noted that $(Y_{0.97}Sc_{0.03})Mn_2$, a geometrically frustrated metal with the pyrochlore lattice, also shows significant spin fluctuations and strongly enhanced γ.[25] However, there is no evidence of the $\delta T^3 \ln T$ term in C_p, which may be correlated with the weaker electron correlation expected for this system.

The $\delta T^3 \ln T$ term persists in the SG-like metal phase, but compared to the PM metal phase, the contribution is significantly reduced relative to the value of γ. Such coexistence of spin fluctuations and SG-like freezing may be attributed to a local character of the fluctuations, as previously discussed for Y-doped UAl_2.[26] In the SG-like metal phase, both γ and δ decrease on the approach to the insulating phase. Previous Hall coefficient measurements[11] showed a significant T dependence and decrease in the carrier density when x falls below $x = 1.0$, in contrast to the T-independent metallic behavior for $x \geqslant 1.0$. Also, the electric resistivity, which increases with decreasing x, starts to show an upturn below ~ 40 K for $x < 1.0$.[11] Therefore, these transport quantities and the C_p results indicate that MIT in $Y_{2-x}Bi_xRu_2O_7$ is associated with a depletion of itinerant carriers in the SG-like metal phase.

Finally, we remark on the high-T C_p, which is shown in Fig. 4. As already seen in Fig. 2, there is a significant enhancement of C_p with increasing x. This trend is seen up to 300 K, where the C_p/T values fall between 0.71–0.75 J K^{-2} mol^{-1}. Although this is qualitatively expected since Bi is heavier than Y, the magnitude of the enhancement is too large to be accounted by the mass effect alone.[27] A simple analysis[28] indicates that the enhancement in lattice C_p originates mostly from the reduction in the characteristic temperatures of Einstein modes. This observation correlates well with the reported static disorder of the Bi-6s lone-pair electrons,[29] as such disorder is expected to produce incoherent localized vibrations.

For $x = 0.0$ ($Y_2Ru_2O_7$), the AF transition is accompanied by a peak in C_p at 72 K, which is lower than $T_N = 77$ K

determined from the peak in χ. The magnetic heat capacity C_m was obtained by subtracting a smooth polynomial background, which was established by interpolating the C_p values of 10–30 K and 90–150 K, and the result is shown in the inset of Fig. 4.[30] This background is comparable to the lattice C_p obtained in the above-mentioned analysis. Interestingly, the shape of C_m resembles a mean-field, BCS-type transition, and the low-T side can be expressed with the exponential form of $C_m = A\exp(-\Delta/T)$, with $\Delta = 262\ \mathrm{K} = 3.4T_N$. Thus, the result is consistent with a second-order transition that involves the formation of an energy gap of the order of 262 K = 23 meV in the magnetic excitation spectrum. We could not resolve this contribution in the low-T (<10 K) C_p, as it diminishes exponentially with decreasing T. The entropy under the peak is determined to be 3.1 $\mathrm{J\,K^{-1}\,mol^{-1}}$, which is only 17% of the full value for $S = 1$ spins. The difference between the entropy under the measured specific heat curve and that under the BCS-type fit is nearly equal, which further substantiates the BCS-type analysis. It must be mentioned that similar gapped excitations have recently been reported on the cubic spinel $GeNi_2O_4$.[31] In $GeNi_2O_4$ with $S = 1$, the AF transition at 12 K is not accompanied by any change in the lattice structure, and the magnetic entropy is much smaller than the expected value, as in $Y_2Ru_2O_7$. This is in marked contrast with the case of half-integer spins on the pyrochlore lattice, which shows the structural distortions associated with AF transitions.[31] Accordingly, the authors[31] proposed that the unusual properties of $GeNi_2O_4$ stem from the integer spins, which may also be the case for $Y_2Ru_2O_7$. Inelastic neutron scattering measurements should provide important information on the magnetic excitations.

We acknowledge valuable discussions with A. P. Ramirez, Y. Nakazawa, and R. Stevens, and thank R. A. Fisher for helpful discussions and a critical reading of the manuscript.

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