

SEARCH FOR THE QUANTUM SPIN LIQUID STATE
IN PYROCHLORE OXIDES*H. FUKAZAWA^{a†}, D. YANAGISHIMA^a, R. HIGASHINAKA^a
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We have investigated a pyrochlore Mott insulator $Y_2Ir_2O_7$ with an expectation that $5d^5$ electrons of Ir ions in the t_{2g} orbitals give rise to an $S = 1/2$ Heisenberg system and form a quantum spin-liquid state. However, it exhibits spin-glass ordering below about 170 K. Such spin-glass ordering is also observed in its isomorphs $Y_2Mo_2O_7$ and $Y_2Ru_2O_7$ with $S = 1$.

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Quantum spin-liquid (QSL) state is one of the important subjects of the research on the geometrically frustrated systems [1]. In this state all the spins form only spin-singlet pairs, which give rise to macroscopic degeneracy of the ground state, namely a residual entropy. The existence of the QSL state has been theoretically expected for the Heisenberg antiferromagnetic (AF) spins on the pyrochlore lattice [2]. However, there are few candidates of the real materials in which the QSL state is indeed realized.

Recently pyrochlore iridates, $R_2Ir_2O_7$ ($R = Y$ and lanthanides), have provided additional research interests in this respect [3–5]. These materials were reported as early as about 30 years ago but were little studied [6]. Kennedy *et al.* [7] studied the crystal structure of the materials, but did not report the low-temperature properties, which are essential for the characterization of the frustrated magnetic systems.

The magnetic frustration arises because of the pyrochlore structure in which R and Ir sub-lattices form individual networks of linked tetrahedra. Systematic variations of the physical properties have been investigated by

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changing the elements of the R site [3]. Furthermore, one may expect that each Ir ion has a quantum spin $S = 1/2$ if the five $5d$ electrons in the t_{2g} orbitals are localized in the presence of strong electronic correlation. Although the QSL state is theoretically expected for antiferromagnetic Heisenberg pyrochlore magnet [2], there have been few candidates of the actual materials studied up to now. These urge us to study the $S = 1/2$ pyrochlore system, pyrochlore iridates.

Of the pyrochlore iridates, $\text{Y}_2\text{Ir}_2\text{O}_7$ serves as a reference material, since it does not possess a magnetic rare-earth element on the R site and its physical properties are comparable with those of its isomorphs $\text{Y}_2\text{Mo}_2\text{O}_7$ [8] and $\text{Y}_2\text{Ru}_2\text{O}_7$ [9], based on $S = 1$.

$\text{Y}_2\text{Ir}_2\text{O}_7$ exhibits non-metallic behavior down to 4.2 K and exhibits quite a small ferromagnetic (FM) component below $T_m = 170$ K, though it has not been well understood whether such FM component is intrinsic or not [3, 4]. In spite of the non-metallic behavior, it has been reported by Taira *et al.* [4] that the γ is finite, $4.1(3)$ mJ/K²mol-Ir, suggesting the existence of the Fermi surface. We will discuss the origins of the FM component and the finite γ of $\text{Y}_2\text{Ir}_2\text{O}_7$.

We used polycrystals synthesized by the conventional solid-state-reaction method [3]. We measured the resistivity by a standard four-probe method below 300 K and the specific heat by a relaxation method between 1.8 and 300 K (Quantum Design, PPMS). We investigated the dc magnetization with a SQUID magnetometer (Quantum Design, MPMS55) between 1.8 and 350 K. The crystal structure is cubic with the lattice parameter at room temperature $a = 10.176(1)$ Å.

In Fig. 1, we plot the dc magnetic susceptibility $M(T)/H \equiv \chi(T)$ ($\mu_0 H = 1$ T). Very small FM component, amounting to 4×10^{-3} of the saturated moment of $S = 1/2$ spins, is observed. The T_m of $\text{Y}_2\text{Ir}_2\text{O}_7$ is about 170 K consistent with the value previously reported by us [3] and by Taira *et al.* [4]. We obtained the effective spin $S_{\text{eff}} = 0.07(1)$ from the Curie-Weiss fitting

$$\chi(T) = \chi_0 + \frac{4\mu_B^2 S_{\text{eff}}(S_{\text{eff}} + 1)}{3k_B(T - \theta_{\text{CW}})}$$

for $\text{Y}_2\text{Ir}_2\text{O}_7$ above T_m . It corresponds to only 14(2)% of the expected spin $S = 1/2$.

In the inset of Fig. 1, we show specific heat of $\text{Y}_2\text{Ir}_2\text{O}_7$ between 100 and 200 K. No peak was observed at around T_m , though the slope of C vs T changes at around 150 K. The linear fittings between 100 and 145 K and between 170 and 200 K, represented by the two solid lines in the inset, cross at 150 K. This temperature reasonably corresponds to the T_m in $\chi(T)$. The

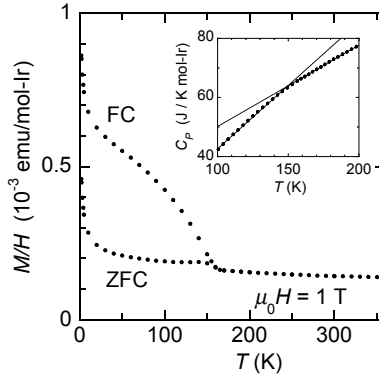


Fig. 1. Magnetic susceptibility of $\text{Y}_2\text{Ir}_2\text{O}_7$. Data for both zero-field-cooling (ZFC) and field-cooling (FC) sequences are shown. Specific heat of $\text{Y}_2\text{Ir}_2\text{O}_7$ are shown in the inset. Two solid lines denote the linear fittings between 100 and 145 K and between 170 and 200 K.

small magnetic moment of $\text{Y}_2\text{Ir}_2\text{O}_7$ below T_m may be due to either spin-glass ordering or canted antiferromagnetic ordering. Since only the slope change but no peak was observed in the specific heat, the FM component is ascribable to spin-glass ordering as previously reported in other pyrochlores, such as $\text{Y}_2\text{Mo}_2\text{O}_7$ [8] and $\text{Y}_2\text{Ru}_2\text{O}_7$ [9].

We should note that an additional step increase of magnetization was observed with decreasing temperature below about 15 K. Since no change in $\rho(T)$ was observed below this temperature, the increase is attributable to magnetic impurities or localized Ir spins at grain boundaries.

In Fig. 2, we plot the specific heat divided by temperature, $C_P(T)/T$, against T^2 . Solid line denotes the quadratic fitting, $C_P/T = \gamma + \beta T^2$, between 14 and 20 K. Below 14 K, it is difficult to perform a valid quadratic

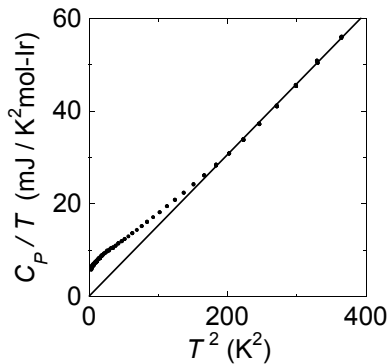


Fig. 2. Specific heat of $\text{Y}_2\text{Ir}_2\text{O}_7$. Solid line denotes the quadratic fitting between 14 and 20 K.

fitting because of additional increase of C_P/T with decreasing T [10]. This increase cannot be explained in terms of the localization effect, since in such a case C_P/T should exhibit linear behavior (the Debye T^3 term) with the same slope and the same finite intercept γ at least up to 20 K. We speculate that the origin of the increase is attributable to a magnetic contribution, since it appears to correspond to the steep increase of $\chi(T)$ below 15 K.

Although C_P/T of $Y_2Ir_2O_7$ at 1.8 K is 5.8(2) mJ/K²mol-Ir, we may consider that the γ , which is equal to 0.0(5) mJ/K²mol-Ir, is the intrinsic electronic specific heat coefficient. This value strongly suggests that $Y_2Ir_2O_7$ is a Mott insulator.

In summary, we have revealed that $Y_2Ir_2O_7$ is a Mott insulator. Although it does not exhibit the QSL behavior, spin-glass ordering occurs below 170 K reflecting the geometrical frustration of the pyrochlore structure as previously reported in $Y_2Mo_2O_7$ and $Y_2Ru_2O_7$.

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