Suppression of metal-insulator transition at high pressure and pressure-induced magnetic ordering in pyrochlore oxide $\text{Nd}_2\text{Ir}_2\text{O}_7$

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We report the pressure dependence of electrical resistance $R(T)$ of a frustrated pyrochlore oxide $\text{Nd}_2\text{Ir}_2\text{O}_7$, which shows a second-order metal-insulator transition (MIT) at 36 K. This MIT is monotonically suppressed by the application of pressure. The insulating phase disappears at around 10 GPa. Then, in the pressure-induced metallic state above 10 GPa, a new pressure-induced phase transition emerges at around 3 K, which is characterized by a resistance drop. The new phase transition is due to a ferromagnetic (FM) ordering, which is suggested to be an ordered spin ice (two-in two-out) state of the Nd moment via the RKKY interaction. We succeeded in observing the phase competition between the MIT and the FM ordering by the application of high pressure.

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Geometrical frustration in a magnetic material can lead to novel phenomena such as a macroscopic degeneracy in the ground state with no long-range ordering (LRO). Pyrochlore oxides $A_2B_2O_7$ have attracted great interest because of their structure, which includes corner-shared tetrahedra whose vertices are occupied by spins, may show a strong geometrical frustration such as spin ice and spin liquid states. Furthermore, in metallic pyrochlore oxides, a frustration originating from the pyrochlore lattice might also lead to novel types of electronic properties. In fact, $4d$ and $5d$ transition-metal pyrochlore oxides have been reported to show various transport properties such as superconductivity in $\text{Cd}_2\text{Re}_2\text{O}_7$ and $\text{AOS}_2\text{O}_6$ (where $A = \text{K}, \text{Rb}, \text{and Cs}$). The unconventional anomalou Hall effect in $\text{Nd}_2\text{Mo}_2\text{O}_7$ and $\text{Pr}_2\text{Ir}_2\text{O}_7$ and metal-insulator transition (MIT) in $\text{Cd}_2\text{Os}_2\text{O}_7$, $\text{Tl}_2\text{Ru}_2\text{O}_7$, and $\text{Hg}_2\text{Ru}_2\text{O}_7$ are reported in Ref. 22. Pressure was generated using a diamond anvil cell. Sodium chloride was used as a pressure medium; as a result the applied pressure was quasi-hydrostatic. Pressure
Rapid Communication. At 2 GPa, the lowest pressure in this study, the drop in $R(T)$ at an applied pressure greater than 8 GPa. The numerals on the $R(T)$ curves denote the value of pressure.

The MIT has been found to be continuous at ambient pressure\textsuperscript{19} and at pressures less than 9 GPa based on the absence of discontinuities and thermal hysteresis of the MIT. Once the pressure increases to approximately 11 GPa, no MIT is observed in the measured temperature region.

In a previous study, a minimum in resistivity of a polycrystalline sample at ambient pressure was not observed.\textsuperscript{19} Here, the minimum in $R(T)$ occurs in the metallic phase; at a temperature denoted as $T_{\text{min}}$ in Fig. 1(a). This minimum became observable by the suppression of the MIT under pressure. With increasing pressure, $T_{\text{min}}$ shifts toward high temperature up to 13 GPa. Above 13 GPa, $T_{\text{min}}$ decreases with increasing pressure. It should be noted that a minimum in $R(T)$ has also been observed in similar pyrochlore iridate Pr$_2$Ir$_2$O$_7$.\textsuperscript{23} The present result suggests that the resistance minimum is a feature common to Lr$_2$Ir$_2$O$_7$ in the metallic state. Therefore, there is a possibility that the resistance minimum of Nd$_2$Ir$_2$O$_7$ is caused by the Kondo effect, as is the case with Pr$_2$Ir$_2$O$_7$. However, we do not have additional information that would elucidate the exact reason for the resistance minimum. Therefore, a further discussion of $T_{\text{min}}$ is beyond the scope of this Rapid Communication.

As shown in Fig. 1(b), at a pressure of 10 GPa, $R(T)$ drops at around 3.5 K and increases slightly at 3 K. This slight increase corresponds to the MIT observed at lower pressures. With increasing pressure, the MIT is suppressed completely but the drop in $R(T)$ remained up to around 15 GPa. This drop indicates a new pressure-induced phase transition. Figure 2 shows $R(T)$ at 11 GPa under several magnetic fields. At a pressure of 11 GPa, a drop of 4% in $R(T)$ at 0 T is observed for $T_{\text{O}} = 3.2$ K, where $T_{\text{O}}$ is the onset temperature of the resistance drop. With increasing magnetic field, the drop in $R(T)$ broadens and $R(T)$ starts to decrease at higher temperature. Furthermore, the magnetoresistance is negative. The magnetic field dependence indicates that the drop in $R(T)$ is caused by a ferromagnetic (FM) transition.

Next, we will discuss the pressure-induced FM ordering in a pyrochlore lattice in the metallic state. As the origin of the FM ordering, there are three possibilities: ordering of the Ir moment, the Nd moment, or both Ir and Nd moments. First, it is important to consider the degree of freedom of the localized Nd moment. The crystalline electric field with $D_{3d}$ symmetry splits the ground state $J = 9/2$ multiplet in Nd$^{3+}$ into
five Kramers doublets. In this case, the Kramers ground-state doublet has a magnetic degree of freedom. The ground-state entropy of NdIn2 has to be relieved down to 0 K. It is known that the Nd moment in pyrochlore oxide, Nd2Mo2O7, has a local (111) Ising anisotropy.10 Our preliminary magnetization measurement of Nd2Ir2O7 indicates that the Nd moment also has a local (111) Ising anisotropy.24

A local (111) Ising model on a pyrochlore lattice interacting via the long-range RKKY interaction has been studied theoretically.25 According to this theory, LRO of localized moment with local (111) Ising anisotropy is realized in the metallic state. In the present results, it should be noted that only a single phase transition due to the FM ordering is observed above 11 GPa; in particular, no additional phase transition is observed at 13 GPa down to 100 mK. If the FM ordering originates from only the Ir moment, the ground-state entropy of the Nd moment is not relieved down to near 0 K in spite of a metallic state. Obviously, this scenario is contradictory to the aforementioned theoretical result. Therefore, it is highly possible that the FM ordering is mainly driven by the Nd moment although further measurements are required to elucidate the origin of the FM ordering. Furthermore, in general, the resistivity of localized f-electron systems decreases by a FM transition, because the c-f exchange scattering is suppressed below the Curie temperature.26 The obtained results are consistent with this fact. Therefore, it is quite reasonable to say that the FM ordering originates from the Nd moment in the metallic state.

Next, we discuss the ordered state. According to the aforementioned theoretical study, in the case of an FM RKKY interaction, the theoretical result shows that the FM ordering of “two-in two-out” with the wave vector $q = (0,0,0)$ or $(0,0,2\pi/a)$ has been realized; this is an “ordered spin ice” state in a metallic state.25 Therefore, we propose that the pressure-induced transition of Nd2Ir2O7 at 3 K is the FM ordering of “two-in two-out” with $q = (0,0,0)$ or $(0,0,2\pi/a)$.

Figure 3 shows the pressure-temperature phase diagram obtained from the measurement of electrical resistance. $T_{MI}$ is considered as the temperature at which $R(T)$ starts to increase abruptly, as shown in Fig. 1(a). $T_O$ emerges suddenly at 10 GPa, which is close to the pressure at which the insulating phase disappears. At pressures lower than 10 GPa, no resistance drop assigned to the magnetic LRO is observed in the plot of temperature dependence of resistance; therefore, there is no LRO of the Nd moment at least in the measured temperature region. Thus, the boundary pressure of the insulating phase should be around 11 GPa.

The MIT in Nd2Ir2O7 is monotonically suppressed by applying pressure. The pressure dependence of $T_{MI}$ is negative—approximately $-4$ K/GPa up to 6 GPa. This is similar to the values observed in other pyrochlore oxides, Cd2Os2O7 ($-4$ K/GPa)12 and Hg2Ru2O7 ($-5.4$ K/GPa).18 The application of pressure leads to structural deformation with a decrease in the Ir-O-Ir bond angle. However, we confirmed that this pressure effect is contradictory to the supposition made on the basis of the ionic radius dependence of $T_{MI}$ in Ln2Ir2O7. Consequently, the result indicates that the decrease in the Ir-O-Ir bond length by the application of pressure strongly affects the electrical conductivity of Ln2Ir2O7.

From the phase diagram, we can say that the insulating phase below the MIT suppresses the emergence of the magnetic ordered phase. The pressure-induced FM ordered phase competes with the insulating phase. The MIT involves magnetic ordering with a weak FM component from 5$d$ electrons. At ambient pressure, the magnetic susceptibility measurement of a polycrystalline Nd2Ir2O7 sample provides no evidence for magnetic LRO of the Nd moment down to 2 K.24 Because the internal field generated by a magnetic ordering due to 5$d$ electrons interacts with the Nd moments, the magnetic ordering of the Nd moment is considered to be suppressed or destroyed. The suppression of the MIT by the application of pressure leads to the magnetic ordering of the Nd moment via the RKKY interaction in the metallic state. Then, as Nd2Ir2O7 becomes more metallic due to pressure application, the RKKY interaction becomes stronger. Therefore, the appearance of the magnetic LRO phase at 3.5 K and 10 GPa is probably due to the strengthening of the RKKY interaction by pressure application. Furthermore, because the magnetic LRO of the Nd moment at 3.5 K and 10 GPa makes no opening of band gap, the MIT can occur below $T_O$.

The localized Nd moments in the metallic phase interact with itinerant 5$d$ electrons via c-f hybridization. We may consider that Nd2Ir2O7 in the metallic phase is a frustrated Kondo lattice system, as pointed out about Pr2Ir2O7 in Ref. 23. RKKY interaction stabilizes a magnetic LRO. On the other hand, the Kondo effect screens localized moments by a spin polarization cloud of conduction electrons. Consequently, a Fermi liquid state is formed by perfect screening of the magnetic moment. The magnetic phase diagram resulting from their competition is usually described by a model proposed by Doniach.27 The ground state depends on the strength of c-f hybridization. In Nd2Ir2O7, the application of pressure induces the magnetic LRO by the RKKY interaction. On the other hand, the pressure dependence of $T_O$ was slightly negative at pressures higher than 11 GPa. As one possible explanation for the suppression of magnetic LRO, we may consider that the Kondo effect becomes dominant by the application of higher pressure. Then, a magnetic quantum critical point (QCP) is expected to appear at pressures greater than 15 GPa. Around the magnetic QCP, we have expected that the magnetic moment reduced by the Kondo effect strongly fluctuates on the
frustrated pyrochlore lattice. A new physics on the frustrated pyrochlore lattice is underlying around QCP. In summary, we have observed that the MIT in frustrated pyrochlore oxides Nd$_2$Ir$_2$O$_7$ is suppressed by the application of pressure. Furthermore, we have found a new pressure-induced transition at pressures higher than 10 GPa. We have proposed that this transition is due to the FM ordering of the Nd moment, which is theoretically considered to be an “ordered spin ice” state in a metallic state. We have observed the phase competition between the MIT and the pressure-induced FM ordering. The pressure-induced magnetic LRO phase is suppressed slightly by applying further pressure but it remains at pressures of up to 15 GPa. Further studies are required in order to elucidate the details of the new magnetic LRO phase and to understand a physical property around magnetic QCP.

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