Lattice frustration in spin-orbit Mott insulator Sr$_3$Ir$_2$O$_7$ at high pressure

Jianbo Zhang, Dayu Yan, Sorb Yesudhas, Hongshan Deng, Hong Xiao, Bijuan Chen, Raimundas Sereika, Xia Yin, Ho-kwang Mao, Changjiang Yi, Youguo Shi, Zhenxian Liu, Ekaterina M. Pärschke, Cheng-Chien Chen, Jun Chang, Yang Ding and Ho-kwang Mao

The intertwined charge, spin, orbital, and lattice degrees of freedom could endow 5$d$ compounds with exotic properties. Current interest is focused on electromagnetic interactions in these materials, whereas the important role of lattice geometry remains to be fully recognized. For this sake, we investigate pressure-induced phase transitions in the spin-orbit Mott insulator Sr$_3$Ir$_2$O$_7$ with Raman, electrical resistance, and x-ray diffraction measurements. We reveal an interesting magnetic transition coinciding with a structural transition at 14.4 GPa, without a concurrent insulator-metal transition. The conventional correlation between magnetic and Mott insulating states is thereby absent. The observed softening of the one-magnon mode can be explained by a reduced tetragonal distortion, while the actual magnetic transition is associated with tilting of the IrO$_6$ octahedra. This work highlights the critical role of lattice frustration in determining the high-pressure phases of Sr$_3$Ir$_2$O$_7$. The ability to control electromagnetic properties via manipulating the crystal structure with pressure promises a new way to explore new quantum states in spin-orbit Mott insulators.

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INTRODUCTION

Exotic ground states in 5$d$ quantum materials, such as spin liquids, Weyl semimetals, topological insulators, and superconductors, are commonly believed to be driven by the competition between electron interaction $U$, spin-orbit coupling (SOC) $\xi$, and hopping amplitude $t$. In particular, for Ir$^{5+}$ atoms in octahedral crystal field, the 5$d$ orbital degeneracy is lifted by the strong on-site SOC, and the ground state is formed by effective $S = 1/2$ pseudospins. The lattice degree of freedom and its coupling to electron orbital angular momenta therefore have long been thought to play a trivial role. However, recently it has been recognized that pseudospin-lattice coupling may have strong impact on the low-energy physics of 5$d$ materials. In the single-layer perovskite Sr$_3$IrO$_6$, canted $S = 1/2$ pseudospins in the antiferromagnetic phase are shown to rigidly lock to IrO$_6$ octahedra due to strong SOC, and the pseudospin orientations rotate together with the octahedra under applied electric current. Moreover, pseudospin-lattice coupling can induce a tetragonal-to-orthorhombic structural transition and explain the in-plane magnon gaps of Sr$_2$IrO$_6$. Jahn–Teller effect also can explain some high energy features of different iridates in resonant inelastic x-ray scattering (RIXS) and the avoidance of metalization of Sr$_3$IrO$_4$ under pressure. These findings suggest that subtle structural changes may influence critically the low-energy Hamiltonian. While determining the exact role of lattice frustrations affect the electromagnetic properties and structural stability at high pressure.

RESULTS AND DISCUSSION

Figure 1 shows the temperature-dependent Raman spectra of Sr$_3$Ir$_2$O$_7$ at ambient pressure. Six Raman modes are identified, respectively, at 146, 181, 269, 392, 592, and 1360 cm$^{-1}$ based on the room-temperature spectrum. The frequencies are close to those reported in the literature. As temperature decreases, a new Raman peak appears at 780 ± 8 cm$^{-1}$ (96.7 ± 1.4 meV) at 270 K. We assign this new peak to the one-magnon mode, as its
2.3 cm
632.8 nm laser. The two-magnon peak increases slowly with excitations. Above \( T_N \) one-magnon peak exhibits an asymmetric Fano-like line shape, 1.9 GPa close to the 14.4 GPa are indicated by dots. Additional modes occurring at pressure behavior. Two additional phonon modes appearing at the one-magnon peak. Since the two-magnon mode merges with even at room temperature, suggesting a short-range spin

to the stretching of Sr atoms against the IrO\(_6\) octahedra. 24 The \( A_{1g} \) mode at 181 cm\(^{-1}\) involves displacements of Sr atoms along the c-axis with antiphase motion of adjacent layers and in-plane rotations of O atoms. 21 The \( A_{1g} \) mode at 269 cm\(^{-1}\) is attributed to the bending of the Ir–O–Ir bond 21,24 due to IrO\(_6\) rotation, and the \( B_{2g} \) mode at 392 cm\(^{-1}\) is associated with some out-of-plane atomic displacement of in-plane oxygens. 21 Finally, the \( A_{1g} \) mode at 592 cm\(^{-1}\) originates from the vibration of apical oxygen atoms in IrO\(_6\) octahedra. 25 These modes are illustrated in Fig. 1a. When cooled down to 10 K, these Raman modes shift to higher frequencies as shown in Fig. 1b.

Figure 1b shows the pressure-dependent Raman spectra of Sr\(_3\)Ir\(_2\)O\(_7\) at 10 K. First, we note that two weak phonon modes emerge close to the \( A_{1g} \) mode (187 cm\(^{-1}\)) and \( B_{2g} \) mode (403 cm\(^{-1}\)) at 1.9 GPa. These two emergent modes, indicated by arrow and asterisk, respectively, in Fig. 1b, have been discovered previously 24 and are visible below \( T_N \) owing to magnetic interaction. Second, the one-magnon peak softens continuously and broadens with increasing pressure; the peak eventually disappears around 14.4 GPa, evincing a magnetic transition. In addition, two new modes appear below 152 cm\(^{-1}\) and the above \( A_{1g} \) mode weakens at around 14.4 GPa. Similarly, the \( A_{1g} \) mode at 288 cm\(^{-1}\) (the bending of the Ir–O–Ir bond) and 595 cm\(^{-1}\) (the vibration of apical oxygens in IrO\(_6\) octahedra) harden with pressure prior to almost vanishing around 14.4 GPa, which indicates increased IrO\(_6\) rotation and tilt angles at high pressure. These changes in phonon modes suggest a structural transition concurring with the magnetic transition around 14.4 GPa. We also performed room temperature and high-pressure Raman measurements to provide further evidence of the structural transition at 23.2 GPa. The results are given in the Supplementary Material in Fig. S2.

The pressure-induced structural phase transition is also confirmed by X-ray diffraction (XRD) at room temperature. The XRD measurements are performed on single crystals, and the results confirm that the sample is in an \( I4/mmm \) phase up to 21.2 GPa at room temperature, while the new phase was fitted with space group \( C2 \) (Fig. 2b). The integrated XRD patterns of Sr\(_3\)Ir\(_2\)O\(_7\) up to 33.2 GPa are presented in the Supplementary Material in Fig. S3a. At 33.2 GPa, the X-ray diffraction pattern fitting using the symmetry of \( I4/mmm \) starts to fail (see Fig. S3b in the Supplementary Material), suggesting that the structures of the new phase should adopt a lower symmetry, i.e., \( C2 \). Detailed XRD analysis of the Sr\(_3\)Ir\(_2\)O\(_7\) crystal performed to exclude possible admixture of Sr\(_2\)IrO\(_4\) is also shown in the Supplementary Material in Fig. S4.

Our discovery of magnetic and structural transitions at 14.4 GPa now could explain the mysterious origin of a second-order phase transition reported by Zhao et al. 25 In their work, the second-order transition derived from the P-V data was attributed to an insulator-metal like transition at \( \sim 13 \) GPa, 26 but they also suspected that

peak intensity decreases significantly with increasing temperature, and its energy is also consistent with the zone-center magnon excitation (~90 meV) reported in RIXS studies of Sr\(_3\)Ir\(_2\)O\(_7\). 22,23 The one-magnon peak exhibits an asymmetric Fano-like line shape, which could result from interaction with a continuum of excitations. Above \( T_N \sim 285 \) K, the magnon peak disappears. The Raman spectra also reveal a two-magnon mode at 1360 ± 2.3 cm\(^{-1}\) (168.62 ± 0.3 meV) [Fig. 1a], which is different from the previously reported energy ~1500 cm\(^{-1}\) (~185 meV) measured by 632.8 nm laser. 21 The two-magnon peak increases slowly with decreasing temperature, and its intensity remains appreciable even at room temperature, suggesting a short-range spin-correlation character. In contrast, the previously reported two-magnon mode was strongly suppressed with temperature and vanished at \( T_N = 285 \) K. 21 Given that the two-magnon mode in our study also could be excited by 488 nm laser, different excitation source is probably not the reason for the observed discrepancy. Rather, the discrepancy could originate from subtle variation in the compositions of different samples.

Apart from the assigned magnon modes at 780 and 1360 cm\(^{-1}\), the other five Raman peaks are assigned to phonon modes, which could be indexed according to the \( I4/mmm \) space group of tetragonal symmetry. 18 The fourteen Raman active modes expected from group theory include \( \Gamma_{Raman} = 5A_{1g} + 2B_{1g} + 1B_{2g} + 6E_g \). 24 However, only one \( B_{2g} \) mode at 392 cm\(^{-1}\) and four \( A_{1g} \) phonon modes are observable in our Raman experiments, whereas the rest of the modes are absent probably due to their weak Raman scattering cross sections.

As previously reported, the \( A_{1g} \) mode at 146 cm\(^{-1}\) corresponds to the stretching of Sr atoms against the IrO\(_6\) octahedra. 24 The \( A_{1g} \) mode at 181 cm\(^{-1}\) involves displacements of Sr atoms along the c-axis with antiphase motion of adjacent layers and in-plane rotations of O atoms. 21 The \( A_{1g} \) mode at 269 cm\(^{-1}\) is attributed to the bending of the Ir–O–Ir bond 21,24 due to IrO\(_6\) rotation, and the \( B_{2g} \) mode at 392 cm\(^{-1}\) is associated with some out-of-plane atomic displacement of in-plane oxygens. 21 Finally, the \( A_{1g} \) mode at 592 cm\(^{-1}\) originates from the vibration of apical oxygen atoms in IrO\(_6\) octahedra. 25 These modes are illustrated in Fig. 1a. When cooled down to 10 K, these Raman modes shift to higher frequencies as shown in Fig. 1b.

Fig. 1a Temperature-dependent Raman spectra of Sr\(_3\)Ir\(_2\)O\(_7\) at ambient pressure. The direction of decreasing temperature is indicated by the dashed arrow line. Five major vibrational modes are identified, and their atomic motions in the polyhedral are represented at the top of the figure. B Raman spectra of Sr\(_3\)Ir\(_2\)O\(_7\) for selected pressures at 10 K. The dotted lines are guides to the eye, showing the evolution of three \( A_{1g} \) modes, respectively, at 152, 288 and 595 cm\(^{-1}\). The dashed curve tracks the pressure dependence of the one-magnon peak. Since the two-magnon mode merges with the first order Raman mode of diamond, we cannot study its high-pressure behavior. Two additional phonon modes occurring at 14.4 GPa are indicated by dots. Additional modes occurring at 1.9 GPa close to the \( A_{1g} \) (187 cm\(^{-1}\)) and \( B_{2g} \) (403 cm\(^{-1}\)) modes are indicated by arrow and asterisk, respectively.
magnetic transition might be a possible cause. To investigate if there is a concurring insulator-metal transition (similar to that observed by in pump-probe experiment at ambient conditions), we also perform electrical transport measurement on a Sr$_3$Ir$_2$O$_7$ single crystal. The results are plotted in Fig. 3a.

The electrical resistance within the $a$-$b$ plane follows an activation law $R_{a-b}(T) = \exp(\Delta/2k_BT)$, where $\Delta$ is the charge gap and $k_B$ is the Boltzmann’s constant. We obtain the value of $\Delta$ at each pressure point from linear fitting of $\ln R(T)$ vs. $1/T$. The gap energy (black square) as a function of pressure is plotted in Fig. 3b.

To further address how lattice frustration affects the magnetic order, we use spin-wave theory to study the magnon excitations. In particular, we adopt the magnetic exchange Hamiltonian from the reference, which describes well the spin-wave dispersion of Sr$_3$Ir$_2$O$_7$ in RIXS measurements. The pressure evolution of magnon dispersion could originate from a change in three microscopic parameters: the ratio of Hund’s coupling to the on-site Coulomb interaction $\eta = J_H/U$, the IrO$_6$ rotation angle $\alpha$, and the effective tetragonal distortion $\theta$ that parametrizes the tetragonal splitting of $t_{2g}$ levels. It is important to understand which parameter dominates the softening of single-magnon energy under pressure.

First, pressure could enhance $\eta$ via screening the Hubbard $U$, while leaving $J_H$ nearly unchanged. Since we do not observe any metallization at 14.4 GPa, the impact of pressure on $\eta$ should be small. Second, it has been shown that pressure can increase the rotation angle $\alpha$ by a few degrees. However, our numerical calculations indicate that such a small change in $\alpha$ only lowers the magnon frequency by a few percent (see Fig. 4), which is not enough to account for the experimentally observed magnon softening. On the other hand, decreasing $\theta$ can significantly reduce the zone-center magnon energy and lift the degeneracy of the magnon branches (see Fig. S1 in the Supplementary Material). The evidence for reduced tetragonal distortion at high pressure is indeed found in the RIXS experiment by Ding et al., in which the peak width of spin-orbiton excitation at ~0.5 eV reduces from 0.56 eV at 0.98 GPa to 0.48 eV at 12.4 GPa. Such a reduction strongly suggests a reduced tetragonal distortion, as in principle the peak width should have increased under pressure due to the broadening of $J_{\text{eff}} = 1/2$ and $\Delta_{\text{eff}} = 3/2$ bands. We thus conclude that one major effect of pressure on the material is to reduce the anisotropy arising from tetragonal distortion of IrO$_6$ octahedra (see Fig. 4).

Although the model could explain the magnon softening, it is unable to illustrate the disappearance of magnon. To explain this effect, the IrO$_6$ tilting angle $\beta$ should be considered. It has already been reported that the actual symmetry of Sr$_3$Ir$_2$O$_7$ should be $C2/c$ with a tilting angle $\beta$ less than 1 degree. Such a small tilting usually has been regarded as having a trivial effect. In our high-
pressure study, however, the tilting appears to be important. In fact, the observed blue shift and disappearance of the 152 cm
\(^{-1}\) and 595 cm
\(^{-1}\) Raman modes suggest that pressure enhances both \(a\) and \(\beta\), which could induce buckling or disordering of IrO
\(_6\) octahedra along the \(c\)-axis and results in a local symmetry breaking (structural transition). The antiferromagnetic order (that gives rise to the magnon dispersion) in Sr
\(_3\)Ir
\(_2\)O
\(_7\) stems from the interlayer exchange coupling \(J_e\) between Ir atoms mediated through apical oxygens.\(^{30}\) A strong IrO
\(_6\) tilting could suppress \(J_e\) and result in the disappearance of magnon excitation. In principle, full suppression of the magnon peak also could arise from other effects, such as staggered distortions where the intralayer interactions change differently in two neighboring layers. Spin-phonon coupling mediated by single-ion anisotropy also can induce a small in the spin gap, although the phonon renormalization after magnetic transition in Sr
\(_3\)Ir
\(_2\)O
\(_7\) appears weaker compared with that in other 5d compounds, such as Cd
\(_2\)Os
\(_2\)O
\(_7\).\(^{34-38}\) A future comprehensive theoretical investigation of these effects is greatly needed.

Finally, we summarize our findings in a phase diagram shown in Fig. 5. After the magnetic transition at 14.4 GPa and 10 K, Sr
\(_3\)Ir
\(_2\)O
\(_7\) is still an insulator. It remains challenging to characterize the magnetic structure and the underlying mechanism of phase transition. Based on similar phenomena observed in doping experiments, the insulating phase above 14.4 GPa could be paramagnetic, spin frustrated,\(^{31}\) or even a totally new quantum state that has never been reported. Sr
\(_3\)Ir
\(_2\)O
\(_7\) at ambient conditions is located approximately at the center of the phase diagram plotted as functions of \(U/t\) and \(\xi/t\).\(^9\) When pressure increases, both \(U/t\) and \(\xi/t\) could decrease, and the system could eventually reach the metallic region. We have reported previously an insulator-metal transition in Sr
\(_3\)Ir
\(_2\)O
\(_7\) around 55–59.5 GPa,\(^{28}\) which is consistent with other studies.\(^{26,29}\) According to Fig. 5, the pathway for such a pressure evolution could possibly pass through the axion insulator phase\(^3\) before it reaches the metallic regime. Therefore, it is not impossible that the insulating phase discovered here might be an axion state. We speculate that applying pressure could be a promising way to search for various predicted phases in 5d spin-orbit materials.\(^9\)

In summary, we have revealed an interesting magnetic transition that concurs with a structural transition around 14.4 GPa by Raman measurements on Sr
\(_3\)Ir
\(_2\)O
\(_7\). We attribute the origin of these transitions to pressure-enhanced rotation and tilt angles of IrO
\(_6\) octahedra. The magnetic transition is shown to decouple from insulator-metal transition. The absence of usual correlation between magnetic and insulating phases in Sr
\(_3\)Ir
\(_2\)O
\(_7\) is similar to that in Sr
\(_2\)IrO
\(_4\).\(^{39,40}\) Our high-pressure study together with previous discoveries manifests the critical role of lattice frustration in determining the ground state properties of Sr
\(_3\)Ir
\(_2\)O
\(_7\), and maybe generically of 5d materials. Our work calls for more theoretical studies to unravel the interplay of intertwined degrees of freedom in spin-orbit systems and the exact mechanism of their phase transitions.

**METHODS**

**Sample synthesis**

Sr
\(_3\)Ir
\(_2\)O
\(_7\) single crystal was grown from flux method. High-purity SrCO
\(_3\), IrO
\(_2\) and SrCl
\(_2\)·6H
\(_2\)O powders were mixed together and placed in platinum crucible. The molar ratios of the source materials was 2:1:20. The crucible was heated to 1573 K and dwelt for 10 h and then slowly cooled down to room temperature. After that the single crystals were separated from the flux by washing with deionized water. The obtained single crystal had typical dimensions 0.8 × 0.8 × 0.3 mm
\(^3\). The experimental XRD pattern of Sr
\(_3\)Ir
\(_2\)O
\(_7\) and calculated pattern based on the standard ICSD (075587) date are shown in the Supplementary Material in Fig. S4a. This result indicated that our sample is tetragonal phase (space group I4/mmm) at ambient condition and has good quality.

**High-pressure Raman measurements**

Raman spectra were collected on a single-crystal Sr
\(_3\)Ir
\(_2\)O
\(_7\) at beamline 22-IR-1 of the National Synchrotron Light Source II, Brookhaven National laboratory. The sample was loaded inside a symmetric-type diamond anvil cell (DAC) and the DAC was placed in a microscopy cryostat system (Cryo Industries of America, Inc.). A pair of ultralow fluorescence type II diamonds with cuet size ~300 μm were used. A Spectra-Physics Excelsior solid-state laser with a wavelength of 532 nm was used in the Raman measurement. Potassium bromide (KBr) was used as the pressure transmitting medium, and the pressure inside the cell was determined by the shift of the ruby fluorescence line. The laser power was less than 1 mW. In our measurements, 300 grooves/mm grating and ~1–3 μm beam spot were applied.
High-pressure transport measurements

Electrical resistance was measured with a standard four-probe-electrode circuit on a single-crystal Sr$_3$Ir$_2$O$_7$. A T301 stainless steel gasket with cubic boron nitride/epoxy mixture powder inserts was used. Si oil was used as a pressure medium and the pressure was determined using ruby fluorescence technique. Four thin gold probes were attached to the samples with silver glue to measure the resistance.

High-pressure synchrotron diffraction measurements

The in situ high-pressure XRD measurements were carried out on a single-crystal Sr$_3$Ir$_2$O$_7$ at beamline 16-BM-D of the Advanced Photon Source (APS), Argonne National Laboratory. A symmetric type DAC with culet size of 300 μm was used for the measurement. Neen was used as a pressure medium and the pressure was determined using ruby fluorescence technique. The incident monochromatic x-ray beam energy was set to 30 keV (λ = 0.1413 Å). Diffraction patterns were recorded on a MAR345 image plate. We aligned the sample to the vertical-rotation axis and collected diffraction patterns in a step-scan method (1.5 s/step) with 1.0° step over the range from -20° to 20° up to 33.2 GPa, similar to the “rotation method” used in conventional single crystal crystallography.

Model Hamiltonian and spin-wave theory

We describe the pressure evolution of magnon excitation using the spin model reported in the ref. 22 Below we review the interaction terms in the model, which includes both intralayer (H$_{\text{in}}$) and interlayer (H$_{\text{int}}$) Hamiltonians:

$$H_{\text{in}} = \sum_{<ij>,<ij>,} [J_S S_i \cdot S_j + g S_i \cdot \vec{S}_j \cdot \vec{S}_j + D(S_i^x S_j^y - S_i^y S_j^x)] + \sum_{<ij>,} J_{ij} S_i \cdot S_j$$

$$H_{\text{int}} = \sum_i [J_{ij} S_i \cdot S_{i+1} + J_{ij} S_i \cdot S_{i-1} + D_i (S_i^x S_{i+1}^y - S_i^y S_{i+1}^x)]$$

where <ij>, and <ij>, denote the first, second, and third nearest neighbors within the a-b plane. J$_{ij}$ and J$_{ij}$ represent the isotropic coupling constants. The anisotropic coupling term $\Gamma$ stems from Hund's exchange interaction and staggered rotations of octahedra. The latter also results in a Dzyaloshinsky-Moriya (DM) interaction, characterized by the constant $D$. For the nearest-interlayer interactions, J$_{ij}$, $\Gamma_i$, and $D_i$ were adopted for the similar coupling constants along the c-axis, while J$_{ij}$ stands for the next-nearest-neighbor interlayer coupling. All these isotropic and anisotropic exchange coupling constants (except for the long-range interactions J$_{ij}$ and J$_{ij}$) can be expressed in terms of the three microscopic parameters: the Ir-O$_2$ rotation angle $\theta$, the effective tetragonal distortion $\eta$, and the ratio of Hund's coupling to onsite Coulomb interaction $\eta = J_{\text{in}}/U$.

Within linear spin-wave theory (applicable in the antiferromagnetic phase), the single-magnon excitation energy in the Brillouin zone center (i.e., that observed by Raman measurements) is expressed by

$$\omega = 1/2 \sqrt{\left(4F+1/3(6J_2+12J_1+4J_1+1)-4D+D_2\right)^2}.$$ 

Our calculations reveal that the major influence of pressure is to reduce the anisotropic coupling $\Gamma$, e.g., from 4.4 meV at ambient pressure to 2.0 meV at around 20 GPa. Also, both J$_{ij}$ and D$_i$ are enhanced, while other coupling constants are only slightly affected. We notice that tilting of the Ir-O$_2$ octahedra along the c-axis can reduce the interlayer coupling J$_{ij}$ and thus soften the magnon energy, although overall J$_{ij}$ is enhanced due to a pressure-enhanced bandwidth.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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AUTHOR CONTRIBUTIONS

Y.D. and J.Z. designed the project. J.Z., H.D., and Z.L. performed the Raman measurements. J.Z., S.Y., and H.D. carried out the XRD and transport measurements. C.Y., D.Y., and Y.S. synthesized the single crystals. J.C. performed the theoretical calculations. J.Z., Y.D., and J.C. analyzed the data. Y.D. supervised the project. All the authors helped with the project and read and commented on the manuscript.

ADDITIONAL INFORMATION

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