Strongly-coupled quantum critical point in an all-in-all-out antiferromagnet

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Dimensionality and symmetry play deterministic roles in the laws of Nature. They are important tools to characterize and understand quantum phase transitions, especially in the limit of strong correlations between spin, orbit, charge, and structural degrees of freedom. Here, using newly-developed, high-pressure resonant X-ray magnetic and charge diffraction techniques, we have discovered a quantum critical point in Cd₂Os₂O₇ as the all-in-all-out antiferromagnetic order is continuously suppressed to zero temperature and, concomitantly, the cubic lattice structure continuously changes from space group Fd-3m to F-43m. Surrounded by three phases of different time reversal and spatial inversion symmetries, the quantum critical region anchors two phase lines of opposite curvature, with striking departures from a mean-field form at high pressure. As spin fluctuations, lattice breathing modes, and quasiparticle excitations interact in the quantum critical region, we argue that they present the necessary components for strongly-coupled quantum criticality in this three-dimensional compound.

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Fundamental symmetry constraints determine allowable collective states in solids. Of particular recent interest is broken inversion symmetry in non-centrosymmetric materials, which can lead to hidden topological order and odd-parity superconductivity. With the added consideration of broken time-reversal invariance, exotic magnetic states can emerge. A quantum critical point, where quantum fluctuations tie together competing ground states, is a fertile region to investigate and manipulate intertwined charge, spin, and structural degrees of freedom under different symmetry conditions. However, many experimental systems either manifest first-order quantum phase transitions without critical behavior, as exemplified by itinerant ferromagnets, or simply follow mean-field behavior. Strongly-coupled systems with pronounced spin-orbit interactions provide a potential means to move to non-trivial criticality in three dimensions. This scenario has been proposed for all-in-all-out (AIAO) antiferromagnetic order on a pyrochlore lattice, but remains to be identified experimentally.

The AIAO arrangement of spins on the pyrochlore lattice is an unusual form of magnetism that preserves the underlying cubic lattice symmetry. With strong spin-orbit coupling and low itinerant electron density, compounds with AIAO spin order are desirable candidates to explore non-trivial quantum critical behavior in three dimensions. AIAO spin order has been observed in FeF₃, Nd₂Zr₂O₇, A₂Ir₂O₇ (A = Sm, Eu, and Nd), and Cd₂Os₂O₇ (refs.7-12) and suggested for additional A₂Ir₂O₇ systems with A = Y, Lu, Gd, Tb, Dy, Ho, and Yb. For our purposes, Cd₂Os₂O₇ is most desirable. With the transition temperature (Tₘₙₐ = 227 K) roughly 60% higher than all other A₂Ir₂O₇ members, Cd₂Os₂O₇ should demonstrate the strongest correlation effects. Moreover, among spin-orbit coupled 5d compounds, only Cd₂Os₂O₇ and Nd₂Ir₂O₇ consistently manifest both an antiferromagnetic insulating phase and a metallic paramagnetic phase with dp/dT > 0 (refs.13-15). It remains unclear whether iridates such as Eu₂Ir₂O₇ and Sm₂Ir₂O₇ are metallic or insulating in the paramagnetic phase, presumably due to vacancies and site disorder from the 3+/4+ valence condition. By comparison, Cd₂Os₂O₇ exhibits less site disorder from its 2+/3+ valence condition, with no extraneous spin order arising from the A site of the A₂Ir₂O₇ structure. Both pressure and chemical tuning of the A site drive the insulating transition to lower temperature in the A₂Ir₂O₇ compounds, but little is known about the behavior of the AIAO magnetic order.

Here, to address this issue directly, we present results on the evolution of the spin, orbit, and lattice degrees of freedom, using polarization analysis of resonantly diffracted X-rays from Cd₂Os₂O₇ under diamond anvil cell pressures. This resonant diffraction technique is highly challenging with regard to both the single-crystal quality at cryogenic temperatures and the high flux X-ray source necessary to detect weak magnetic diffraction signals from miniature samples of 5 × 10⁻⁵ mm³ size. Nevertheless, AIAO magnetic order and 5d orbital order can be resolved directly and tracked to the highest pressures under resonant conditions in the polarization switching process of the forbidden lattice orders, respectively. The lattice symmetry and space group assignments were detected via the π-π' scattering channel.

**Results**

The magnetic quantum phase transition. We summarize in Fig. 1 the symmetry evolution under pressure. At ambient pressure, Cd₂Os₂O₇ has the pyrochlore structure of the Fd-3m space group (No. 227). Optical Raman scattering maps out a unique Fd-3m phase up to P = 29 GPa and between T = 10 and 300 K. The phonon modes characteristic of that space group develop smoothly over the entire range (Fig. 2). The overall cubic symmetry is further verified by X-ray diffraction up to 41 GPa at 4 K (Fig. 3), as lineshapes of the (1, 1, 1), (2, 2, 2), (0, 2, 2), (0, 4, 4), and (4, 0, 0) diffraction orders remain single peak. The cubic lattice constant decreases smoothly under pressure, without discernable discontinuity at the quantum critical point Pₘₚ = 35.8 GPa (Fig. 1, discussed below). While the Fd-3m space group is uniquely determined by the unit cell’s lattice constant and one free coordinate x for oxygen position on 48f sites that characterizes the trigonal distortion of the OsO₆ cluster, diffraction intensities at (1, 1, 1) and (0, 2, 2) show a continuous evolution through the quantum phase transition, with x increasing by a small amount from 0.319 at P = 0 (ref.14) to 0.325 at Pₘₚ (Fig. 3b, c). Neither the AIAO antiferromagnetic order nor the continuous space group evolution within the cubic symmetry (Fig. 1) would be detectable by macroscopic approaches such as electrical transport. Instead, we address these issues using resonant single crystal diffraction with polarization analysis at the Os L₂ edge.

Resonant diffraction at both (6, 0, 0) and (4, 2, 0) in the π-σ and π-π' polarization channels are displayed in Fig. 4 for pressures that traverse the quantum phase boundary. The magnetic diffraction intensity Iₙₜ (6, 0, 0) in the π-σ channel at E = 12.387 keV, which scales to the ordered staggered moment <mₗ> as Iₙₜ <mₗ>⁻², decreases continuously with increasing pressure (Fig. 5a). The scatter in the data makes it difficult to identify definitively the quantum critical region, but a phenomenological fit of the intensity data as Iₙₜ <mₗ>⁻²(Pₘₚ-P)² over the whole pressure range gives a critical pressure Pₘₚ = 35.8 ± 0.7 GPa and an...
The space group $F\bar{4}3m$ breaks lattice inversion symmetry, opening possibilities for a number of exotic states. We did not detect further symmetry breaking such as the small tetragonal distortion in $Cd_2Re_2O_7$ that would remove the three-fold rotational symmetry$^{23}$. The broken inversion symmetry derives from differently sized adjacent tetrahedra of the Os and Cd sublattices, which are no longer centrosymmetric and can be regarded as fully-softened breathing modes$^3$ with spontaneous symmetry breaking. These breathing modes disappear above $T_c$ ~ $K|\Delta_{Os}|^{12}$, with the lower $F\bar{4}3m$ symmetry replaced by the higher Fd-3m symmetry with adjacent tetrahedra of equal size. Here $K$ is the vibrational elastic constant and $\Delta_{Os}$ is the amplitude of the Os tetrahedron breathing mode. We expect $T_c$ ~ $L$, given that the measured X-ray diffraction intensities (Fig. 5b) depend on these lattice distortions as $I ~ f_{Cd}\Delta_{Cd} + f_{Os}\Delta_{Os}$, where $f_{Cd,Os}$ are X-ray atomic form factors. Hence $T_c$ increases two orders of magnitude in a short pressure interval: from approximately 5 K at 37 GPa to ~500 K at 41 GPa, leading to the striking concave high-pressure phase boundary in Fig. 1.

Discussion

The reduction of lattice symmetry from Fd-3m to Fd-3m in $Cd_2Os_2O_7$ has direct implications for phonon coupling$^2$ to spin fluctuations at the quantum critical point. Although spin-phonon coupling is known to generate unequal bond lengths in spin-Peierls dimers and antiferromagnetic superlattices$^{20}$, the breathing phonons are not fully softened in the AIAO phase and, at least in the static limit, AIAO order only induces an external magnetostriction with a homogenous expansion$^{22}$. In the high-pressure phase, a breathing lattice could in principle still permit AIAO spin configurations on different sized tetrahedra, despite a loss of site inversion symmetry in the $F\bar{4}3m$ space group. Nevertheless, as no long-range commensurate antiferromagnetic order was observed experimentally (Fig. 4c), the magnetic ground state at high pressure is likely spin-disordered, as ferromagnetically interacting Ising moments along local <1,1,1> axes would generate a high level of frustration$^7$. Tuning by either chemical doping or pressure drives the ratio of the magnetic interaction strength to the hopping integral smaller in $A_2(\text{Os,Ir})_2O_7$. With an increasing electron density under 15% volume reduction by pressure, and moving away from the strong interaction strength limit$^{13}$, one would not naturally expect a ferromagnetic ground state. Furthermore, ferromagnetic quantum phase transitions, as well as commensurate-incommensurate antiferromagnetic transitions, are first order, which would contrast with the continuous AIAO quantum phase transition observed in our experiment.

The increased bandwidth under pressure suggests that the electronic properties of $Cd_2Os_2O_7$ in the high-pressure $F\bar{4}3m$ state become more metallic, potentially even superconducting in analogy to superconducting $Cd_2Re_2O_7$ with its broken inversion symmetry (ref.$^{25}$). The relationship between the spin and charge transitions is also intriguing. AIAO magnetic order and the metal-insulator transition respond similarly to compression across the $P$-$T$ phase diagram. Our projected magnetic phase boundary in Fig. 1 gives $dT_N/dP ~ -5.0$ K GPa$^{-1}$ at $P = 0$, which is consistent with $dT_{M_P}/dP = -4$ K GPa$^{-1}$ in $Cd_2Os_2O_7$. The discussion of the electronic properties of $Cd_2Os_2O_7$ in the high-pressure $F\bar{4}3m$ state becomes more metallic, potentially even superconducting in analogy to superconducting $Cd_2Re_2O_7$ with its broken inversion symmetry (ref.$^{25}$). The relationship between the spin and charge transitions is also intriguing. AIAO magnetic order and the metal-insulator transition respond similarly to compression across the $P$-$T$ phase diagram. Our projected magnetic phase boundary in Fig. 1 gives $dT_N/dP ~ -5.0$ K GPa$^{-1}$ at $P = 0$, which is consistent with $dT_{M_P}/dP = -4$ K GPa$^{-1}$ in $Cd_2Os_2O_7$. The discussion of the electronic properties of $Cd_2Os_2O_7$ in the high-pressure $F\bar{4}3m$ state becomes more metallic, potentially even superconducting in analogy to superconducting $Cd_2Re_2O_7$ with its broken inversion symmetry (ref.$^{25}$). The relationship between the spin and charge transitions is also intriguing. AIAO magnetic order and the metal-insulator transition respond similarly to compression across the $P$-$T$ phase diagram. Our projected magnetic phase boundary in Fig. 1 gives $dT_N/dP ~ -5.0$ K GPa$^{-1}$ at $P = 0$, which is consistent with $dT_{M_P}/dP = -4$ K GPa$^{-1}$ in $Cd_2Os_2O_7$. The discussion of the electronic properties of $Cd_2Os_2O_7$ in the high-pressure $F\bar{4}3m$ state becomes more metallic, potentially even superconducting in analogy to superconducting $Cd_2Re_2O_7$ with its broken inversion symmetry (ref.$^{25}$). The relationship between the spin and charge transitions is also intriguing. AIAO magnetic order and the metal-insulator transition respond similarly to compression across the $P$-$T$ phase diagram. Our projected magnetic phase boundary in Fig. 1 gives $dT_N/dP ~ -5.0$ K GPa$^{-1}$ at $P = 0$, which is consistent with $dT_{M_P}/dP = -4$ K GPa$^{-1}$ in $Cd_2Os_2O_7$.
measured over the first 2 GPa range\textsuperscript{14}. Comparing the two 5\textit{d} AIAO ordered compounds with clear high-temperature metallic states, we find an average \( dT_0/dP \sim -6.5 \) K GPa\textsuperscript{-1} over the whole AIAO phase in Cd\textsubscript{2}Os\textsubscript{2}O\textsubscript{7} and a \( dT_{\text{MIT}}/dP \sim -5.8 \) K GPa\textsuperscript{-1} in Nd\textsubscript{3}Ir\textsubscript{2}O\textsubscript{7}\textsuperscript{17,18}. This comparison holds true despite large differences in \( T_0 \) of 227 K and 33.5 K, respectively.

The experimentally observed coincidence of magnetic and structural phase transitions at \( P_C \sim 35.8 \) GPa, along with the similarities in the pressure evolution of the spin and charge degrees of freedom, point to one critical point within our experimental resolution (~1 GPa in the region around \( P_C \)) between the insulating AIAO order and the spin-disordered metallic phase. As Landau’s phase transition theory would dictate the [con]currence of magnetic and structural phase transitions within our experimental resolution, and the clear deviation from mean-field behavior of the high-pressure phase, stimulate a discussion of strong coupling in the quantum critical region.

From the band structure perspective, the Os 5\textit{d} \( t_{2g} \) band in Cd\textsubscript{2}Os\textsubscript{2}O\textsubscript{7} is neither degenerate and forming a \( S = 3/2 \) state under Hund’s rule, as indicated by the reduced staggered moment \( < m > = 0.59 \mu_B \) Os\textsuperscript{-1}, nor cleanly separated into several narrow bands as demonstrated for a perfect OsO\textsubscript{6} octahedron\textsuperscript{14,25,26}. Instead, the \( t_{2g} \) orbitals in Cd\textsubscript{2}Os\textsubscript{2}O\textsubscript{7} extend continuously over a spectral width of order 2 eV from the combined effect of \( U \) (~1 eV)\textsuperscript{27}, spin-orbit coupling (~0.35 eV)\textsuperscript{26}, and trigonal distortion (~0.3 eV) on the OsO\textsubscript{6} octahedron\textsuperscript{19}. Through the continuous quantum phase transition, the overall stability of the empty \( t_{2g} \) band is verified by the constant charge resonance profile at \((4, 2, 0)\), with a coarse energy resolution slightly above 1 eV. From a metallic paramagnetic state, the formation of antiferromagnetic order would influence the oscillating dynamic component of the quasi-particle self-energy\textsuperscript{28}, and in turn introduce a spin-order dependent repulsion between empty and filled states that could account for the insulating state below \( P_C \). This
above the magnetic phase boundary, with a small leakage into the energy scan at (5.98, 0, 0) (gray line, shape remains, which comes from X-ray The azimuthal angel intensity decreases continuously with pressure. Beyond 36 GPa (c), the sharp resonance disappears, and a background similar to the Os L₂ absorption edge shape remains, which comes from X-ray fluorescence that passed through the polarization analyzer. Its origin is demonstrated by a comparison with an energy scan at (5.98, 0, 0) (gray line, c), where no magnetic diffraction is expected, and also with an Os L₂ absorption spectrum at 39.3 GPa (black line, c). The fluorescence part of the background intensity is dependent on the detector slit size. At (4, 2, 0), the π–σ channel reveals the orbital ordering via the anisotropic tensor susceptibility resonance. For both (6, 0, 0) and (4, 2, 0) orders, the π–σ charge diffraction intensities rise dramatically at pressures above the magnetic phase boundary, with a small leakage into the π–σ channel becoming apparent in f through the polarization analyzer (Methods).

The essentially concurrent, continuous quantum phase transitions of antiferromagnetism, structure, and (apparently) charge in Cd₂Os₂O₇ provides the necessary ingredients for a generic approach to strongly-coupled, non-mean-field quantum criticality in three-dimensions⁵. With the Fermi surface fully gapped, quasiparticle fluctuations would involve all itinerant states in reducing the screening on Coulomb U, and the increased interaction range would then help stabilize a continuous quantum phase transition⁶. Indeed, at the ambient-pressure metal-insulator transition in Cd₂Os₂O₇, an increase in U from 0.8 to 1.5 eV in the theoretical modeling is consistent with the observed spectral weight shift in infrared conductivity over the broad range of 0–4 eV (ref.30). Above the quantum critical point and in the spin-disordered phase space of the Fd-3m space group, soft AIAO spin fluctuations and lattice breathing modes could exist and compete, and further couple to quasiparticle fluctuations. The competition between spin and lattice degrees of freedom might explain the remarkable concave-shaped phase line at high pressure, as Tₙ scales to pressure with a non-trivial exponent much larger than one, a characteristic of strongly-coupled quantum criticality⁵. We note as well that the quantum critical region is asymmetric in P–T phase space, as the magnetic and structural phase lines approach Tₙ with different asymptotic behavior.

The quantum phase transition in Cd₂Os₂O₇, with its interwoven spin, orbit, lattice, and charge degrees of freedom, contrasts sharply with systems that have a partially gapped Fermi surface, exemplified by itinerant spin density waves where persistent carriers screen spin fluctuations and lead to mean-field behavior⁴–⁶,31. The cubic AIAO antiferromagnet also differs from itinerant ferromagnets, where strong spin and charge mode coupling at wave vector q = 0 categorically induces first-order quantum phase transitions⁴. Spin-orbit coupling in 5d systems is regarded as intermediate-strong⁵, and pressure drives U/t smaller with increasing kinetic energy t, away from the strong-correlation limit. Pressure tuning thus likely induces a continuous quantum phase transition while still preserving non-trivial quantum criticality in this 5d antiferromagnet. By Luttinger’s
other diffraction orders such as (1, 1, 1), (4, 0, 0), (0, 2, 2), and (4, 2, 0) within the confined diamond anvil cell geometry, with the forbidden order (4, 2, 0) providing access to the charge-based anisotropic tensor susceptibility resonance. Several plates were polished down to 13 μm thickness, equivalent to one absorption length in Cd$_2$Os$_2$O$_7$ for X-rays at the Os L$_2$ edge (E = 12.387 keV). Unlike the iridates, resonance behavior at both the L$_2$ and L$_3$ edges are present in osmate. We chose the resonance at the L$_2$ edge at high P because the higher energy X-rays had a longer X-ray penetration length through both the pressure environment and the sample.

We used the low-temperature, high-pressure diffraction setup at beamline 4-ID-D of the Advanced Photon Source. To reduce absorption and enhance the signal-to-background ratio, a pair of wide-angle perforated Bohler diamond anvils (SYNTEK Co. LTD., Japan) were used, with culet size varying from 800 to 550 μm. A methanol/ethanol 4:1 mixture was used as the pressure medium inside rhenium gaskets. Pressure was calibrated by a Ag manometer in situ at 4.0 ± 0.5 K using a two-parameter Birck equation of state, with $B_0 = 108.83$ GPa and $B' = dB/dP = 5.7$ over the large pressure range. For X-ray polarization analysis, a highly oriented pyrolytic graphite (HOPG) plate of 5 mm thickness and 0.35° FWHM mosaic was used as the polarization analyzer. The (0, 0, 10) diffraction order of graphite at the Os L$_2$ edge of 12.387 keV introduces a leakage of approximately 1.3% of the intensity from the π-σ channel to the π-σ and π-π channel, and vice versa. Data presented here were collected from a total of 8 samples under pressure for spin (7) and charge (4) resonances. The absence of high-pressure commensurate antiferromagnetic order was verified on two crystals.

Optical Raman scattering. Shards of single crystal Cd$_2$Os$_2$O$_7$ with original growth surfaces were individually loaded with a Neon pressure medium in a diamond anvil cell, and subsequently thermally cycled in a liquid helium cryostat. Optical Raman scattering was performed using a Horiba LabRam HR Evolution system in the MRSEC facilities at the University of Chicago, equipped with a 633 nm wavelength laser for excitation.

Data availability. The data that support the findings of this study are available from the corresponding authors upon request.

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Author contributions

Y.F. and T.F.R. conceived of the research. D.M. provided single crystal samples. Y.W., Y.F., A.P., Y.R. and J.-W. K. performed the measurements. Y.W., Y.F. and T.F.R. analyzed the data and prepared the manuscript. All authors commented on the manuscript.

Additional information

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