Enhanced coupling of material properties offers new fundamental insights and routes to multifunctional devices. In this context 5\textit{d} oxides provide new paradigms of cooperative interactions that drive novel emergent behaviour. This is exemplified in osmates that host metal-insulator transitions where magnetic order appears intimately entwined. Here we consider such a material, the 5\textit{d} perovskite NaOsO\textsubscript{3}, and observe a coupling between spin and phonon manifested in a frequency shift of 40 cm\textsuperscript{-1}, the largest measured in any material. The anomalous modes are shown to involve solely Os–O interactions and magnetism is revealed as the driving microscopic mechanism for the phonon renormalization. The magnitude of the coupling in NaOsO\textsubscript{3} is primarily due to a property common to all 5\textit{d} materials: the large spatial extent of the ion. This allows magnetism to couple to phonons on an unprecedented scale and in general offers multiple new routes to enhanced coupled phenomena in 5\textit{d} materials.
Transition metal oxides with 3d ions host a remarkable variety of intriguing phenomena, such as unconventional superconductivity, multiferroic behaviour, colossal magnetoresistance and the Mott metal–insulator transition (MIT)\(^1\). These properties arise from the strong electron correlations and localized orbitals characteristic of 3d ions. Materials with 5d ions reside in an alternative regime of intermediate electron correlations, extended orbitals, enhanced spin–orbit coupling (SOC) and large crystalline electric field splitting. The competition and cooperation of these new sets of interactions can drive the emergence of novel behaviour beyond that possible in 3d-based materials\(^2,3\). This is manifested in the insulating states of iridates and osmates. In Sr\(_2\)IrO\(_4\) (ref. 4) and other iridates\(^2,4\), a Mott-like insulating state exists due to enhanced SOC creating a half-filled \(J_{\text{el}} = 1/2\) electronic band that can be split by even the reduced on-site Coulomb interactions of 5d ions. Conversely the neighbouring osmate NaOsO\(_3\), that we focus on here, is believed to host a Slater MIT with behaviour that falls outside the Mott–Hubbard paradigm successfully employed to describe 3d transition metal oxides\(^7\).\(^10\).

In the case of a Slater MIT it is the onset of magnetic order and the accompanying creation of a periodic potential that acts as a direct and continuous tuning parameter between metallic and insulating states. The interactions within the 5d\(^3\) ion in NaOsO\(_3\) appear central to the occurrence of the MIT, with the first proposed three-dimensional Slater candidate Cd\(_2\)Os\(_2\)O\(_7\) sharing the same 5d\(^3\) electronic ground state.

Here we access the collective excitations and fundamental interactions through the Slater MIT in NaOsO\(_3\) by probing the phonon modes. Our experimental and theoretical results reveal a high degree of cooperation between the magnetic structure, lattice and electronic conductivity that results in a spin-phonon-electronic coupled transition. The magnitude of the phonon coupling is anomalously large leading us to consider and contrast our results with 3d transition metal oxides, where investigating spin-phonon coupling has proven extremely useful in understanding multiferroics\(^11,12\), systems with the same perovskite structure as NaOsO\(_3\), as well as in a variety of other systems, including high temperature superconductors\(^13\). The largest reported phonon shift in a perovskite is found in (Sr,Ba)MnO\(_3\) with a value of \(\Delta\omega = 25\) cm\(^{-1}\) in the TO\(_1\) polar phonon\(^14,15\). While the investigations of phonon modes in the context of 5d materials is currently limited, it was recently found that the mixed 3d–5d half-metal double perovskite Ba\(_2\)FeReO\(_6\) hosts a dramatic spin-electron-phonon coupling as evidenced by a phonon shift of \(\Delta\omega = 30\) cm\(^{-1}\), (ref. 16) the largest ever reported prior to our present work on NaOsO\(_3\). The phonon shift in Ba\(_2\)FeReO\(_6\) is reported as being directly linked to the interaction between the 3d and 5d ions. Conversely, we show here that 5d ions alone can produce even larger spin-phonon shifts. By considering the various competing mechanisms in NaOsO\(_3\), including the electronic changes at the MIT and structurally driven charge disproportionation, we find the microscopic behaviour to be driven by the G-type magnetic structure that orders in the perovskite structure. The enhanced nature is promoted by the extended orbitals of the 5d ion that supports strong coupling between the magnetic superexchange and phonon vibrations. By contrasting our results with measurements on Cd\(_2\)Os\(_2\)O\(_7\), that show a much reduced spin-phonon shift, we consider the key ingredients required to achieve even larger spin-phonon coupling in general in further systems.

**Results**

**Measurement of anomalous spin-phonon coupling in NaOsO\(_3\).** To follow the behaviour of collective excitations in NaOsO\(_3\) through the magnetic MIT we performed inelastic neutron scattering (INS) measurements. Figure 1a shows the key result of the temperature dependence of the phonon density of states (pDOS) whose peaks are related to the underlying phonon modes. We focus on the region around 700 cm\(^{-1}\) that covers the essential physics of interest. The full spectrum is shown in Supplementary Fig. 1. Three distinct resolution limited peaks in the pDOS are observed around 700 cm\(^{-1}\) and fitting these each to a Gaussian, as shown in Fig. 1a, allows the energy of the modes to be followed with temperature. The key result of a pronounced phonon frequency shift is observed in Fig. 1a,b. Moreover there is an anomalous and counterintuitive intensity increase with decreasing temperature through the MIT as shown inset Fig. 1a considering the entire range of 550–800 cm\(^{-1}\). The results are significant in several regards. First, the onset of the phonon mode shift is concurrent with the magnetic MIT in NaOsO\(_3\) at 410 K, indicating a coupling of the phonons to the magnetic and electronic transitions. Second, the phonons show a shift of \(\Delta\omega = 40\) cm\(^{-1}\), the largest measured in any material for a spin–phonon coupled transition.

To begin to understand the microscopic origin of the behaviour in NaOsO\(_3\) we consider the role of the MIT with complimentary neutron measurements on Cd\(_2\)Os\(_2\)O\(_7\), Cd\(_2\)Os\(_2\)O\(_7\) was chosen since it has the same 5d\(^3\) electronic configuration of the Os\(^{5+}\) ion and hosts a magnetic MIT that is very similar to NaOsO\(_3\), with current debate as to whether the mechanism is Slater or Lifshitz\(^17,18\). The inelastic neutron measurements through the magnetic MIT in Cd\(_2\)Os\(_2\)O\(_7\) are shown in Supplementary Fig. 2. While there is an apparent phonon shift at the magnetic transition the value of \(\Delta\omega = 4\) cm\(^{-1}\) is much reduced from NaOsO\(_3\). The disparate results for NaOsO\(_3\) and Cd\(_2\)Os\(_2\)O\(_7\) indicate that the underlying mechanism for the anomalously large behaviour in NaOsO\(_3\) cannot be attributed to the occurrence of the MIT, since both host similar MITs with similar energy scales. Instead, as we support with further results and calculations, the microscopic mechanism is related to the magnetic ordering and lattice topology of NaOsO\(_3\).

Considering a further pertinent material, the 3d–5d material Ba\(_3\)FeReO\(_6\) that showed a phonon shift of \(\Delta\omega = 30\) cm\(^{-1}\), we note this occurred concurrent with a structural symmetry change\(^16\). No symmetry change has been detected in NaOsO\(_3\) (refs 8,9). However, to explore this possibility further we performed detailed neutron pair density functional measurements through the Slater MIT, see Supplementary Figure 3, and found no local symmetry change. Hence, the enhanced spin-phonon coupling does not appear to arise due to static long or short range lattice distortions in NaOsO\(_3\).

**Theoretical demonstration of spin-phonon shift.** To gain a microscopic insight into the origin of the anomalous phonon mode behaviour in NaOsO\(_3\) and disentangle the myriad of competing interactions at the magnetic and electronic transition we performed detailed density functional theory (DFT) calculations. The DFT results show the same three phonon modes observed with INS between 600 to 900 cm\(^{-1}\), see Fig. 1c, and as expected for the orthorhombic structure in NaOsO\(_3\) these are themselves composed of three branches, unresolvable in the current powder INS measurement. The theoretical shift is in very close agreement with the measured value of \(\Delta\omega = 40\) cm\(^{-1}\). This indicates the calculations that probe only the Brillouin zone center accurately reproduce the essential physics of the system as measured by neutrons that probe the entire Brillouin zone. The thermal behaviour was captured in the calculations by increasing the magnetic moment \(<S_iS_j>\) to reproduce the onset of G-type antiferromagnetic order in NaOsO\(_3\) with the predicted magnetic
The Slater transition of phonon mode frequencies obtained from inelastic neutron scattering and (of phase) represent symmetric stretching breathing modes. Error bars throughout the figure represent the s.d. in the data fitting procedure. (shown with the directions indicated by the red arrows. Ag (in phase) and B3g (out of phase) correspond to asymmetric stretching. B1g (in phase) and B2g (out of phase) have frequencies than the asymmetric stretching, with B2g occurring at the highest frequency. (asymmetric Jahn–Teller stretching modes Ag (in phase) and B3g appear most pronounced at the highest frequency, which of Fig. 1a, is consistent with an increase in vibration with decreasing temperature, counter to usual thermal behaviour. This appears most pronounced at the highest frequency, which, corresponds to the breathing mode B2g and consequently appears central to the behaviour of NaOsO3.

### Octahedral B2g breathing mode and charge disproportionation

The calculations reveal all of the phonon modes and are shown in Fig. 1d. They all correspond to Os–O vibrations, specifically breathing modes B1g (in phase) and B2g (out of phase) and two asymmetric Jahn–Teller stretching modes Ag (in phase) and B3g (out of phase). To reveal the role of these modes in NaOsO3, we begin by first considering the static behaviour of the octahedra and propensity towards Jahn–Teller distortion. This can be quantified by introducing parameters Q2 and Q3, which are shown schematically in Fig. 2a, and defined as $Q_2 = (x_1 - x_4 - y_2 + y_3)/\sqrt{2}$ and $Q_3 = (2z_3 - 2z_4 - x_1 + x_4 - y_2 + y_3)/\sqrt{6}$, where x, y and z are the oxygen positions. Thereby the values of Q2 and Q3 reveals the degree of static octahedral anisotropy, with the larger the value the more distorted the octahedra. Calculations from experimentally determined atomic parameters for NaOsO3 (ref. 9) reveal Q2 and Q3 to be small at all temperatures, but counter intuitively decrease through the Slater MIT. Specifically, at 500 K $Q_2 = 0.0114(15)$ a.u. and at 300 K $Q_2 = 0.0035(11)$ a.u. While at 500 K $Q_3 = 0.0171(18)$ a.u and at 300 K $Q_3 = 0.0114(14)$ a.u. Therefore this reveals that in NaOsO3 the octahedra actually become more isotropic in three dimensions within the low temperature insulating regime. This behaviour is at odds to the normal Jahn–Teller distortions of increased anisotropy and does not favour the asymmetric stretching modes Ag and B3g.

Instead the increased static octahedral isotropy is more conducive to the symmetric breathing distortions B1g and B2g. Indeed the abnormal behaviour of the intensity increase of the pDOS, in inset of Fig. 1a, is consistent with an increase in vibration with decreasing temperature, counter to usual thermal behaviour. This appears most pronounced at the highest frequency, which, corresponds to the breathing mode B2g and consequently appears central to the behaviour of NaOsO3.

Considering the B2g mode further we find that sufficiently large breathing distortions of the octahedra, much larger than accessed in our measurements, offers a potential route to opening the insulating gap in the paramagnetic regime, see Fig. 2b. Our frozen DFT results show that in the perovskite structure of NaOsO3, the gap opening can occur since the octahedral breathing causes neighbouring octahedra to expand/contract that in turn creates a periodic charge disproportionation on the Os ion. In addition there is apparent isosymmetric ordering and coupling between the G-type antiferromagnet and octahedral B2g mode ordering (behaviour shown schematically in Fig. 2d). We note that no other phonon distortion produces similar periodic ordering or routes to open a gap. For the B2g mode to create a gap the minimum required oxygen displacement is $u = 0.2 \text{ Å}$, see Fig. 2b. This corresponds to 10% of the actual Os–O bond distance and therefore it is too large to allow this mechanism to drive the MIT in NaOsO3. However, statically, while not opening a gap the periodic octahedral breathing ordering creates a strong charge disproportionation of $\Delta\delta/\Delta u = 7.0 \text{ e\text{Å}^{-1}}$ in the lattice due to the change of the electronic potential around the Os ion and places...
the system on the verge of a MIT. For example considering a nominal Os–O phonon vibration displacements of the order 0.01 Å as occurring in NaOsO₃ then the dynamic charge disproportionation will be ~0.01e. This indicates that suitable control of the octahedra via pressure or strain is a potential route to tune the MIT in NaOsO₃. We stress that, as shown in frozen DFT calculations, the breathing distortion can open a band gap as revealed in the DOS from DFT calculations. (c) We stress by reproducing published results⁸ that it is solely the onset of G-type magnetic ordering that opens the gap via the Slater mechanism in NaOsO₃. No oxygen displacement, u = 0 Å, (black line) shows no gap in the DOS whereas an insulating gap is created for G-type antiferromagnet (AFM) order (blue line). This ordering is indicated by the blue arrows. (d) Although the required u displacement is too large to drive the MIT in NaOsO₃, it creates charge disproportionation (δ(e)) on the Os ion. As shown schematically the G-type antiferromagnetic ordering (blue arrows) and periodic expansion/contraction of the B₂g breathing mode ordering are isosymmetric in NaOsO₃. A consequence of the static ordering of the octahedra, as shown in frozen DFT calculations, is the creation of charge disproportionation, indicated by the creation of +δ and −δ ordering (magenta sphere). The predicted value in NaOsO₃, δ(e), is indicated by the blue shaded region.

**Figure 2 | Charge disproportionation created by B₂g breathing mode.** (a) The static octahedral distortion can be quantified with parameters Q₂ and Q₃ that represent the degree of octahedral anisotropy defined as Q₂ = (x₁ − x₄ − y₂ + y₃)/√2 and Q₃ = (2z₋₂z₆ − x₁ + x₄ − y₂ + y₃)/√6, where x, y and z are the oxygen positions labelled 1–6. The red arrows indicate the directions of Os–O distortions. Unusually Q₂ and Q₃ both show reduced values below the Slater MIT indicating the octahedra become more isotropic at lower temperature. (b) This counterintuitive behaviour is compatible with the symmetric three-dimensional B₂g distortion, depicted by red arrows, increasing in the low temperature insulating regime. The solid lines are DOS calculations for oxygen displacements (u), within the paramagnetic regime, of u = 0 Å (black line), u = 0.1 Å (magenta line) and u = 0.2 Å (red line). For the large oxygen displacement of u = 0.2 Å, much beyond that accessed in our measurements, the breathing distortion can open a band gap as revealed in the DOS from DFT calculations. (c) We stress by reproducing published results⁸ that it is solely the onset of G-type magnetic ordering that opens the gap via the Slater mechanism in NaOsO₃. No oxygen displacement, u = 0 Å, (black line) shows no gap in the DOS whereas an insulating gap is created for G-type antiferromagnet (AFM) order (blue line). This ordering is indicated by the blue arrows. (d) Although the required u displacement is too large to drive the MIT in NaOsO₃, it creates charge disproportionation (δ(e)) on the Os ion. As shown schematically the G-type antiferromagnetic ordering (blue arrows) and periodic expansion/contraction of the B₂g breathing mode ordering are isosymmetric in NaOsO₃. A consequence of the static ordering of the octahedra, as shown in frozen DFT calculations, is the creation of charge disproportionation, indicated by the creation of +δ and −δ ordering (magenta sphere). The predicted value in NaOsO₃, δ(e), is indicated by the blue shaded region.

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**Suppressed role of SOC in NaOsO₃.** While SOC is often attributed to anomalous behaviour of 5d materials, in NaOsO₃ the 5d² t₂₂-degenerate ground state will suppress the effective SOC²⁰. We nevertheless addressed the role of SOC with X-ray absorption near edge spectroscopy that allows a quantitative comparison with SOC enhanced iridates. As expected our results indicate SOC does not play a dominant role in the behaviour of NaOsO₃ as discussed in the Supplementary Material (Supplementary Fig. 4 and Supplementary Note 1). Our first-principle results additionally show the large coupling without SOC.

**Coupling of lattice, magnetic order and MIT.** The coupled properties in NaOsO₃ are illustrated in Fig. 3 where experimentally there is a direct scaling of the structural anomaly of the lattice constants, phonon mode shift, magnetic moment, with the MIT qualitatively following a similar trend. This reveals a high degree of cooperation in NaOsO₃ via spin-phonon-electronic coupling. While the realization of numerous overlapping phenomena is currently rare it is likely that additional 5d materials will host similar rich phase diagrams with the prospect of enhanced magnitudes.

**Discussion** We have presented both experimental and theoretical results that show an enhanced phonon shift in NaOsO₃ which along with the concurrent magnetic MIT creates a spin-phonon-electronic transition above room temperature. Considering the collective
spin-phonon-electronic coupling. The lattice parameters have been
ref. 9) show a direct scaling with temperature through the Slater MIT due
to stretching/shrinking between the oxygen and osmium ions of
diamond) and

c

Figure 3 | Phonon, lattice and magnetic degrees of freedom couple
through the Slater MIT in NaOsO3. The measured temperature
dependence of the phonon mode frequency shift (blue square), the [110]
magnetic Bragg reflection intensity9 (white square), and the α (black
diamond) and c (red triangle) lattice constants in NaOsO3 (altered from
ref. 9) show a direct scaling with temperature through the Slater MIT due
to spin-phonon-electronic coupling. The lattice parameters have been
corrected by removing a constant sloping thermal background. Error bars
throughout the figure represent the s.d. in the data fitting procedure. The
shaded regions distinguish the low temperature magnetic-insulating and
high temperature non-magnetic metallic phases in NaOsO3.

results we argue that the occurrence of the anomalous spin-
phonon behaviour is a direct consequence of the extended 5d
orbitals coupling to the magnetic structure via the Os–O–Os
superexchange interactions on an unprecedented scale and is not
driven by the MIT or structural anomalies. The central role of
magnetism is emphasized in our DFT calculations that require
the inclusion of magnetic order to reproduce the experimental
phonon shift. However, as the reduced spin-phonon shift in
Cd2Os2O7 attests, the presence of 5d magnetic order alone is not
sufficient to induce enhanced coupled phenomena. Instead,
within 5d systems, it is not just the onset of magnetic ordering
but the specific type of magnetic structure and the lattice it resides
on that supports enhanced spin-phonon coupling.

By considering the specific modes in NaOsO3, the B2g
breathing mode emerges as central to the behaviour. The mode is
characterized by expansion/contraction of neighbouring octahedra
that in NaOsO3 can statically order in a periodic way, isosymmetric with the magnetic spins (G-type antiferromagnet),
with every nearest neighbouring octahedra oppositely expanded/
contracted (see Fig. 2). The breathing mode ordering causes
charge disproportionation that grows as the B2g vibrations
increase within the low temperature insulating phase and can
promote the opening of a charge gap for sufficiently large vibrations. However, the distortion under the experimental
conditions we measured falls well short of the value required to
drive a MIT via this alternative non-magnetic mechanism in
NaOsO3. Instead, once the insulating gap develops via solely the
magnetic Slater mechanism, the breathing mode becomes
favourable and the structure subtly alters allowing the octahedra
to become more isotropic as a route to increase the symmetric
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to become more isotropic as a route to increase the symmetric

Methods
Synthesis. Polycrystalline samples of NaOsO3 were prepared using a high pressure solid state synthesis with pressures of 6 GPa, as described in ref. 8. Polycrystalline Cd2Os2O7 was prepared with isotopic 114Cd for neutron measurements to negate
the extremely high neutron absorption of standard Cd using sold state techniques
from 114CdO and OsO2 powders.

Inelastic neutron scattering. Inelastic neutron scattering measurements were
performed on the ARCS and SEQUOIA spectrometers at the spallation neutron
source on a 5-g polycrystalline sample of NaOsO3 and 7-g polycrystalline sample of
Cd2Os2O7, respectively. The NaOsO3 sample was loaded into a vanadium can and
measurements performed between 300 and 500 K using an incident energy of
120 meV. The Cd2Os2O7 sample was measured in an Al can from 150 to 250 K
using an incident energy of 100 meV. Corrections for the Bose factor, where
appropriate, were performed using the DAVE software21.

DFT. First-principles calculations were performed using density functional
theory within the generalized gradient approximation GGA + U method with the
Perdew–Becke–Erzenhof parameterization as implemented in the Vienna ab initio
Simulation Package (VASP 5.3)22. Theoretical details for spin-phonon coupling
are described in ref. 23. We use the Dudarev24 implementation with on-site Coulomb
interaction $U = 1.7$ eV and on-site exchange interaction $J_{soc} = 1$ eV, so $U_{eff} = 0.7$ eV
to treat the localized d electron states in Os. Within GGA + U, this small $U$ gives
excellent agreement between the experimental Neel temperature ($T_N = 411$ K)
and calculated one ($T_{N,MIT} = 415$ K) in mean-field approximation. The projector augmented wave potentials25 explicitly include 9 valence electrons for Na (2
$p^1$), 14 for Os (5$p^6 5d^{10} 6s^2$) and for oxygen (2$p^6 2p^4$). To capture spin-phonon coupling with respect to temperature we employed the method successfully used for various magnetic perovskites26,27.

Neutron pair density function. Neutron pair density functional measurements
were performed on the Nanoscale-Ordered Materials Diffractionometer (NOMAD)
beamline at the spallation neutron source on a powder sample of NaOsO3 from 370
to 460 K. The data were analyzed and modelled with pdfgui26.

X-ray absorption near edge spectroscopy. X-ray absorption measurements were
performed at the advanced photon source on sector 4-ID-D. Spectra were collected
at room temperature on a powder sample (~100 mg) in transmission mode
through the Os L3 and L2 edges. Analysis was performed with the Athena
software27.
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Author contributions

S.C., M.B.S., M.D.L., A.D.C. and M.F. conceived and performed the neutron experiments. J.C.L. performed the X-ray measurement. Y.G.S., Y.S.S., Y.T., K.Y. prepared the NaOsO3 sample and J.Q.Y. and Z.Z. prepared the Cd3Os2O7 sample. J.H.L. performed the DFT calculations. S.C. and J.H.L. led the manuscript preparation with contributions from all authors.

Additional information

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