Metal-to-insulator transitions (MITs) driven by electronic correlations have energy scales of a few electronvolts, yet it is common to find that these phase transitions happen at temperatures corresponding to much lower energies. In the absence of a mechanism for fine-tuning the coupling constants, it is natural to look for entropic rather than enthalpic contributions with which to describe these transitions. Since all observed MITs couple to the lattice, one is then driven to look for phononic entropic contributions. As a hint at the origin of these interactions, a large number of transition-metal oxides with the ABO₃ perovskite crystal structure allow tuning of the MIT by not only by the choice and average valence of the electronically active B ion (usually a 3d transition metal) but also by the size of the electronically inactive A ion (usually a rare earth or alkaline earth ion). This size effect can shift the transition temperature $T_{\text{MIT}}$ by hundreds of kelvin, and the widely accepted explanation is that the shift is due to a reduction in the electron bandwidth as the bond bending induced by ionic size changes the orbital overlap. However, the changes in bandwidth are not sufficiently large to explain such temperature variations. Moreover, it seems remarkable that a critical value of the ratio of interaction strength to bandwidth can be reached in every 3d transition-metal oxide, solely by varying the ionic size.

Instead, we propose here that even when the transition is clearly driven by the coupling constants, it is not only by the choice and average valence of the electronically active B ion but also by the size of the electronically inactive A ion that determines the transition temperature. This size effect can shift the transition temperature $T_{\text{MIT}}$ by hundreds of kelvin, and the widely accepted explanation is that the shift is due to a reduction in the electron bandwidth as the bond bending induced by ionic size changes the orbital overlap. However, the changes in bandwidth are not sufficiently large to explain such temperature variations. Moreover, it seems remarkable that a critical value of the ratio of interaction strength to bandwidth can be reached in every 3d transition-metal oxide, solely by varying the ionic size.

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be ignored when it comes to determining the structural trends of MITs that couple to symmetry-breaking distortions.

The crystal structure of a perovskite transition-metal oxide consists of corner-sharing oxygen octahedra surrounding the B transition-metal ion, as shown in Fig. 1a. In general, the octahedra are tilted relative to their neighbours in an alternating pattern, and the tilt angle $\phi_0$ increases with smaller A-site cation radius $r_A$. The large changes in functional behaviour of perovskites when varying $\phi_0$ have led to proposals to engineer material properties by using a combination of strain, doping and pressure. In addition to variations of the atomic size, doping with A-site cations also introduces disorder in the cation size; careful distinction of the effects of doping and disorder for the manganites have demonstrated that disorder reduces the $T_{\text{MIT}}$ as effectively as varying $r_A$ (ref. 5).

Although purely electronic mechanisms to describe transition-metal oxides are appealing in their theoretical simplicity, it is known that the strong electron–phonon coupling means that the effects of lattice distortions cannot be neglected, and this is particularly well studied in manganites and nickelates. An electron that is localized by correlation effects in a unit cell will lower its energy further by the creation of a lattice distortion, which may have different symmetry in different materials. In the nickelates this is a simple breathing distortion, and in the manganites it is a so-called Jahn–Teller distortion, which lowers the cubic symmetry of the octahedron, as shown in Fig. 2a. The competition between this potential energy gain and the kinetic energy gained by delocalization to form a metal gives rise to the complex MIT phenomena in these materials.

The corner-sharing constraint on the octahedra introduces compatibility conditions between distortions at different lattice sites; when integrating out the phonon degrees of freedom these yield highly anisotropic, long-range interactions. Previous studies of phonon cooperativity in the manganites have explained the complex charge-ordered phases and mesoscopic structures that have been observed in the manganites and studied some effects of cooperative coupling on the transition. However, these studies did not consider the effect of octahedral tilting on the long-range interaction of the distortions. The purpose of this work is to study such effects, and in doing so, to construct a complete theory for cooperative elastic effects at a phase transition.

For illustration, we use a two-dimensional model of a perovskite, where we replace the octahedra with squares, as shown in Fig. 1b. Although the physics of bulk perovskites is three-dimensional, two-dimensional models of elastic interactions capture their anisotropy and long-range decay, which in turn have been shown to generate structural inhomogeneity over a wide range of length scales, which has been experimentally seen in transition-metal oxides and is relevant to our work. At a lattice site $r$, the squares can undergo the distortions shown in Fig. 2a: deviatoric/Jahn–Teller modes $T_r$, dilatation/breathing modes $D_r$, shear modes $S_r$, and small rotations $R_r$ of the squares from an initial equilibrium antiferrodistortive rotation $\phi_0$, that is, $\phi_r = (-1)^r \phi_0 + R_r$. Assuming a harmonic energy penalty for creating distortions from an equilibrium configuration:

$$H = \sum_r a_1 T_r^2 + a_2 D_r^2 + a_3 S_r^2$$

(1)

combined with the corner-sharing constraint, we can find an effective interaction $V_{\text{eff}}(\phi_0)$ between different types of distortion, which gives rise to lattice cooperativity (see Supplementary Note 1). $a_1$, $a_2$ and $a_3$ are, respectively, the stiffness of the Jahn–Teller, breathing and shear distortions in a single, free octahedron and are independent of $r$.

Figure 3 shows that the interaction strength is reduced by an increase of the tilt angle for Jahn–Teller distortions. This occurs because in the tilted configuration it is possible for the distortion to be accommodated by additional rotations to the neighbouring sites, rather than changes in the shape. Characteristic strain responses of the lattice to a local Jahn–Teller distortion with and without rotations are shown in Fig. 2b.

Both manganites and nickelates undergo first-order transitions from a characteristic low temperature phase to a high-temperature polaronic phase. This suggests that the motion of conduction electrons through the lattice is associated with the creation of local structural distortions that lead to a bad metal (highly resistive; $>10^{-5} \Omega \text{m}$). When the distortion interaction $V_{\text{eff}}(\phi_0)$ is reduced by changes in $\phi_0$, the high-temperature phase is favoured by a reduction in the polaron formation energy. To study this behaviour, we use $V_{\text{eff}}(\phi_0)$ to form a statistical mechanical model for the distortions in this high-temperature phase, with a Hamiltonian:

$$H = \sum_r \left[ \frac{1}{2} N_r^2 + \frac{K}{2} Q_r^2 + \frac{V}{4} Q_r^4 \right] + \sum_r V_{\text{eff}}(\phi_0) Q_r Q_r - \sum_r \hbar_r Q_r$$

(2)

where $Q_r$ is a Jahn–Teller (breathing) distortion for the manganites (nickelates) and $\hbar_r$, its conjugate momentum. To model the compositional disorder that arises in the manganites from chemical substitution of the alkaline earth element at the $A$ site of the perovskite structure, we consider a linear coupling of the lattice distortions $Q_r$ to a local quenched random distortion $\hbar_r$. We choose the $\hbar_r$ values to be normally distributed.
with mean $\bar{\rho}_z = 0$ and variance $\bar{\rho}_z^2 = \Delta^2$. The negative sign of the $Q^2$ term describes the local tendency towards distortion due to the presence of electrons.

As described in the Methods and Supplementary Note 2, we use a variational approach to calculate the temperature, tilt angle and disorder dependence of the free energy $F_{\text{lat}}(T, \phi_0, \Delta)$ of Hamiltonian (2); and we identify the location of $T_\text{MI}$ by comparing $F_{\text{lat}}(T, \phi_0, \Delta)$ to the free energy $F_{\text{net}}$ of the low-temperature ferromagnetic metal (paramagnetic insulator) phase of the manganites (nickelates). The results are shown in Fig. 4. Despite the over-simplicity of the model, the relationship between tilt angle, disorder and transition temperature is well reproduced. We do not attempt to describe the effects of the strain interactions on the MIT of the nickelates at low temperatures (see green region in Fig. 4a), because its magnetic ordering is different from that of the insulating phase above it. Similarly for the manganites, at low enough temperatures the polaronic, paramagnetic bad metal phase becomes either charge-ordered or glassy, beyond our approximations.

Here we have outlined a systematic theory for the incorporation of long-range elastic couplings into a simplified statistical mechanical theory of Mott-like phase transitions, where the electronic contributions to the free energy are incorporated at the level of Landau theory. That these elastic interactions are explicitly relevant for the manganites and the nickelates is confirmed by the ability of such a theory to systematically explain size effects or tolerance factor variations that have already been documented. However, the couplings, including their rough order of magnitude, are generic, and the ideas presented here will surely be relevant to other classes of materials such as the titanates, high-temperature superconductors, and molecular fullerides.

At low enough temperatures we also ought to consider other low-energy degrees of freedom such as spin fluctuations and electronic quantum fluctuations, which our model does not take into account. Doing so requires explicitly adding them to our model Hamiltonian and to our statistical mechanical solution through, for example, a variational scheme such as the Landau–Firsov transformation. Nonetheless, the model we employ does generate a quantum critical point attributable to elastic interactions alone. Moreover, the long range and anisotropy of these elastic couplings will modify the critical dynamics away from that arising from short-range models generated by purely electronic couplings.

We also note that our simple model provides an explanation for the observed tuning of the MIT under applied pressures. In both the manganites and nickelates, hydrostatic compression decreases $\phi_{\text{MI}}$. According to our model, this should result in an increase of $T_\text{MI}$ promoted by the enhancement of the elastic interaction in the manganites, and vice versa for the nickelates. These are indeed the trends that have been observed in these materials. We believe a similar mechanism is at play when the transition is tuned with tensile and compressive stresses.

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**Fig. 3** | Effective elastic energy. a, b. Effective elastic energy for Jahn–Teller distortions in momentum space for $\phi_0 = 0$ (a) and $\phi_0 = 15^\circ$ (b). Rotations of the BO$_6$ octahedron allowed by the reduction of the A-cation size decrease the elastic energy. The characteristic ‘butterfly’ pattern is a consequence of the anisotropy and long-range nature of strain forces, which in turn can generate the salient nano- and meso-scale structural inhomogeneities, similar to those that have been observed in the manganites such as domain patterns in the form of stripes and tweeds formed by interwoven incommensurate structures. c, d. Effective elastic energy for breathing distortions in momentum space for $\phi_0 = 0$ (c) and $\phi_0 = 15^\circ$ (d). Tilts of the NiO$_6$ octahedron increase the effective elastic energy. The butterfly pattern is similar to that of the manganites, which produces structural inhomogeneity. $(k_x, k_y)$ is a wavevector in the reciprocal space of the two-dimensional lattice of lattice constant $a$ (shown in Fig. 1b).

**Fig. 4** | Comparison to experiments. a. Comparison of the nickelates for the transition temperature as a function of octahedral tilt angle. Filled and open circles are experimental transition temperatures for the paramagnetic insulator (PMI) and antiferromagnetic insulator (AFI) phases respectively. The extension of the green shading beyond the blue dashed line is an extrapolation. The open square (lower left) denotes LaNiO$_3$, which is a polaronic, paramagnetic metal (PMM) at all temperatures. For the manganites, the comparison is made with results (red circles) that separate the effect of tilt angle (b) and compositional disorder (c) on the transition from the paramagnetic, polaronic bad metal phase (PMM) to the ferromagnetic metal (FM) phase. Model parameters are fitted as described in the Methods. In Supplementary Note 3, we explore reasonable variations of the model parameters to demonstrate its generality.
The idea that cooperative phonon–phonon couplings tune the MIT is supported by a recent ab initio calculation. By using density functional theory, it has been found that the tilt of the NiO₆ units in the nickelates destabilize their breathing distortions, which in turn are associated with the phase transition, thus providing a mechanism for tuning $T_{\text{MI}}$. However, density functional theory treats the elastic interactions only on average and it cannot produce finite temperature properties; $T_{\text{MI}}$ has previously been obtained from fits to experiments with a Landau theory that has multiple sets of values for the model parameters depending on the tolerance factor. By contrast, we have calculated $T_{\text{MI}}$ from a single set of model parameters, and the MIT is driven by entropic effects that result from elastic couplings, thus providing a physical interpretation of the ab initio results.

We conclude by noting that the good agreement we found in these two systems suggests that our fundamental assumption that energy can be separated into a relatively simple local free energy plus a complex long-range energetic contribution could provide a basis for a fully computational methodology that could be applied relatively simply to very complex oxides in general.

Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41586-019-1824-9.
Methods

Statistical mechanical solution
We use a variational pair-distribution function that incorporates mean-field behaviour, Gaussian corrections to the thermal and quantum fluctuations, and averaging over compositional disorder at the level of the replica method\textsuperscript{31}. Details are provided in Supplementary Note 2.

Model parameters
Our model has six parameters ($\kappa$, $\gamma$, $a_0$, $a_4$, $a_5$ and $F_{\text{low}}$), which are reduced to five because $a_4$ ($a_5$) is combined with $\kappa$ for the manganites (nickelates). We begin by choosing a set of physically reasonable parameters that give phonon frequencies that are in order-of-magnitude agreement with the observed relevant modes\textsuperscript{32,33}. We then take the resulting set of parameters and fine-tune them to fit the observed dependence of $T_{\text{MI}}$ with the tolerance factor and compositional disorder: $F_{\text{low}}$ is a parameter of the model assumed to be independent of $T$, $\phi_0$ and $\Delta$, fixed by the observed onset of the MIT, that is, $F_{\text{lattice}}(T=0\, \text{K}, \phi_0 = \phi_{\text{onset}}, \Delta = 0) = F_{\text{low}}$, where $\phi_{\text{onset}} \approx 11.5^\circ$ ($12.5^\circ$) for the manganites (nickelates). The dependence of $T_{\text{MI}}$ on $\phi_0$, shown in Fig. 4a, b is given by $F_{\text{lattice}}(T_{\text{MI}}, \phi_0, \Delta = 0) = F_{\text{low}}$, while the dependence of $T_{\text{MI}}$ on $\Delta$ shown in Fig. 4c is given by $F_{\text{lattice}}(T_{\text{MI}}, \phi_0 = 80^\circ, \Delta) = F_{\text{low}}$ and by rescaling $\Delta$ by a constant factor ($\alpha$) to match the units of cation variance. The resulting values are given in Extended Data Table 1.

Data availability
Requests for materials should be addressed to G.G.G.-V., and P.B.L.

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Author contributions
P.B.L. conceived the study. G.G.G.-V. and R.T.B. performed the calculations. All authors constructed the model, wrote the manuscript, discussed the results and implications at all stages.

Competing interests
R.T.B. is currently an editor at Nature Communications.

Additional information
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Correspondence and requests for materials should be addressed to G.G.G.-V. or P.B.L.
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Extended Data Table 1 | Model parameters

|                  | κ [meV²] | γ [meV³] | a₁D [meV²] | a₁S [meV²] | a₁T [meV²] | F₁low [meV] | α [Å meV⁻¹] |
|------------------|----------|----------|------------|------------|------------|-------------|-------------|
| (R₀.₃M₀.₇)MnO₃  | 3.2 × 10³ | 1.8 × 10⁵ | 2.8 × 10⁴  | 9.5 × 10³  | combined with κ | 38          | 4.71        |
| RNiO₃           | 12.1 × 10³| 13.3 × 10⁵| combined with κ | 48.3 × 10³ | 1.21 × 10⁴ | 64          | -           |

R is a rare earth element such as La, Pr, Nd, and Sm; and M is an alkaline earth metal such as Ca, Sr and Ba.