

Cooperative elastic fluctuations provide tuning of the metal-insulator transition

G. G. Guzmán-Verri^{1,2,3a}, R. T. Brierley⁴, P. B. Littlewood^{3,5b}

¹*Centro de Investigación en Ciencia e Ingeniería de Materiales (CICIMA),
Universidad de Costa Rica, San José, Costa Rica 11501,*

²*Escuela de Física, Universidad de Costa Rica, San José, Costa Rica 11501,*

³*Materials Science Division, Argonne National Laboratory, Argonne, Illinois, USA 60439,*

⁴*Department of Physics, Yale University,*

New Haven, Connecticut 06511, USA, and

⁵*James Franck Institute, University of Chicago,*

929 E 57 St, Chicago, Illinois, USA 60637.

(Dated: August 27, 2019)

^a gian.guzman@ucr.ac.cr

^b pblittlewood@anl.gov

Metal to insulator transitions¹ (MITs) driven by strong electronic correlations are common in condensed matter systems, and are associated with some of the most remarkable collective phenomena in solids, including superconductivity and magnetism. Tuning and control of the transition holds the promise of novel, low power, ultrafast electronics², but the relative roles of doping, chemistry, elastic strain and other applied fields has made systematic understanding difficult to obtain. Here we point out that existing data on tuning of the MIT in perovskite transition metal oxides through ionic size effects provides evidence of systematic and large effects on the phase transition due to dynamical fluctuations of the elastic strain, which have been usually neglected. This is illustrated by a simple yet quantitative statistical mechanical calculation in a model that incorporates cooperative lattice distortions coupled to the electronic degrees of freedom. We reproduce the observed dependence of the transition temperature on cation radius in the well-studied manganite³ and nickelate⁴ materials. Since the elastic couplings are generically quite strong, these conclusions will broadly generalize to all MITs that couple to a change in lattice symmetry.

MITs driven by electronic correlations have energy scales in the electron volts, yet it is common to find that these phase transitions happen at temperatures corresponding to much lower energies¹. In the absence of a mechanism of fine tuning the coupling constants, it is natural to look for entropic rather than enthalpic contributions 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 to the origin of these interactions, a large number of transition metal oxides (TMO) with the ABO_3 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)⁵⁻⁷. This size effect can shift the transition temperature T_{MI} by hundreds of Kelvin, and the widely accepted explanation is that it 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 variations⁸⁻¹¹. Moreover, it seems remarkable that a critical value of the ratio of interaction strength to bandwidth can be crossed in every $3d$ TMO, solely by varying the counterion¹².

Instead, we propose here that even when the transition is quite clearly driven by local electronic correlations, anisotropic long range forces induced by elastic compatibility conditions produce

enormous entropic contributions to the free energy, which we show are crucial to describe the trends of the MIT with cation size. We illustrate this with a model of highly fluctuating cooperative lattice distortions that competes with a low temperature phase of constant free energy, i.e., a ferromagnetic metal (FM) for the manganites and a paramagnetic insulator (PMI) for the nickelates. We do not aim to capture the complex charge, orbital, and magnetic orderings of these materials, but rather their high temperature melted version where the entropy is dominated by the cooperative distortions. Our view is that the natural experiments in the manganite and nickelate series broadly implicate elastic interactions as being important in a wide class of MITs, not only in the perovskites.

In building our model, we account for the electronic degrees of freedom by assuming we can separate the energy into components that can be calculated locally while keeping the long-range physics explicit. At zero Kelvin, state-of-the-art first-principles calculations can give such local free energy containing implicitly electron-phonon coupling on a unit cell as well as band structure energy and Coulomb correlation. We have not performed such calculations here. Instead, we have assumed that there is a simple functional outcome that can be parametrized, is the same across each material series, and is independent of the long-range piece.

Our approach has of course its limitations: not every TMO is electronically the same, e.g., the bandwidth is not the only indicator and/or key parameter of structural changes in the electronic structure when varying the rare-earth ion nor the local electronic correlations are independent of the tolerance factor^{13–15}. These are idealizations which can only describe real materials approximately. Nonetheless, it allows us to illustrate that the non-trivial and surprisingly subtle effects from long range elastic interactions mediated between local degrees of freedom cannot be ignored when it comes to determine the structural trends of MITs that couple to symmetry breaking distortions.

The crystal structure of perovskite TMOs consists of corner-sharing oxygen octahedra surrounding the B transition metal ion, as shown in Fig. 1(a). In general, the octahedra are tilted relative to their neighbors in an alternating pattern, and the tilt angle ϕ_0 increases with smaller A-site cation radius r_A . The dramatic changes in the functional behavior of perovskites when varying ϕ_0 have led to proposals¹⁶ 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 demonstrated that disorder reduces the T_{MI} as effectively as varying r_A .¹⁷

Although purely electronic mechanisms to describe TMOs 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^{18,19}.

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 be of different symmetry in different materials. In the nickelates this is a simple breathing distortion, and in the manganites a so-called Jahn-Teller (JT) distortion that lowers the cubic symmetry of the octahedon, as shown in Fig. 2 (a). 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 interactions²⁰. Previous studies²¹⁻²³ of phonon cooperativity in the manganites have demonstrated that they can explain the complex charge ordered phases and mesoscopic structures that have been observed in the manganites, and studied some effects of cooperative coupling on the transition²⁴. 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 by squares, as shown in Fig. 1(b). Although the physics of bulk perovskites is three-dimensional, two-dimensional models^{21,23} of elastic interactions capture their anisotropy and long-range decay (they fall-off as r^{-D} for $D = 2$, and 3 dimensions) which in turn have been shown to generate structural inhomogeneity over a wide range of length scales. This is the most relevant aspect to our work and one of the most salient features that have been experimentally seen in TMOs. At a lattice site \mathbf{r} , the squares can undergo the distortions shown in Fig. 2 (a): deviatoric/JT modes $T_{\mathbf{r}}$, dilatation/breathing modes $D_{\mathbf{r}}$, shear modes $S_{\mathbf{r}}$, and small rotations $R_{\mathbf{r}}$ of the squares from an initial equilibrium antiferrodistortive rotation ϕ_0 , i.e., $\phi_{\mathbf{r}} = (-1)^{|\mathbf{r}|}\phi_0 + R_{\mathbf{r}}$. Assuming a harmonic energy penalty for creating distortions from an equilibrium configuration,

$$H = \sum_{\mathbf{r}} a_T T_{\mathbf{r}}^2 + a_D D_{\mathbf{r}}^2 + a_S S_{\mathbf{r}}^2, \quad (1)$$

combined with the corner-sharing constraint, we can find an effective interaction $V_{\mathbf{r}\mathbf{r}'}(\phi_0)$ between different types of distortion which gives rise to lattice cooperativity (see Supplementary Note 1). a_T , a_D , and a_S are, respectively, the stiffness of the JT, breathing, and shear distortions in a single, free octahedron and are independent of \mathbf{r} .

Fig. 3 shows that the interaction strength is reduced by an increase tilt angle for JT 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 JT distortion with and without rotations are shown in Fig. 2 (b).

Both manganites³ and nickelates⁴ 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. When the distortion interaction $V_{\mathbf{r}\mathbf{r}'}(\phi_0)$ is reduced by changes in ϕ_0 , the high-temperature phase is favoured by a reduction in the polaron formation energy²⁴. To study this behaviour, we use $V_{\mathbf{r}\mathbf{r}'}(\phi_0)$ to form a statistical mechanical model for the distortions in this high-temperature phase, with a Hamiltonian,

$$H = \sum_{\mathbf{r}} \left[\frac{1}{2} \Pi_{\mathbf{r}}^2 - \frac{\kappa}{2} Q_{\mathbf{r}}^2 + \frac{\gamma}{4} Q_{\mathbf{r}}^4 \right] + \sum_{\mathbf{r}\mathbf{r}'} V_{\mathbf{r}\mathbf{r}'}(\phi_0) Q_{\mathbf{r}} Q_{\mathbf{r}'} - \sum_{\mathbf{r}} h_{\mathbf{r}} Q_{\mathbf{r}}, \quad (2)$$

where $Q_{\mathbf{r}}$ is a JT (breathing) distortion for the manganites (nickelates) and $\Pi_{\mathbf{r}}$ 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 $Q_{\mathbf{r}}$ to a local quenched random distortion $h_{\mathbf{r}}$. We choose the $h_{\mathbf{r}}$'s to be normally distributed with mean $\bar{h}_{\mathbf{r}} = 0$ and variance Δ^2 . The negative sign of the $Q_{\mathbf{r}}^2$ term describes the local tendency towards distortion due to the presence of electrons.

As described in Methods and Supplementary Note 2, we use a variational approach to calculate the temperature, tilt angle and disorder dependence of the free energy $F_{\text{lattice}}(T, \phi_0, \Delta)$ of Hamiltonian (2); and we identify the location of T_{MI} by comparing $F_{\text{lattice}}(T, \phi_0, \Delta)$ to a free energy $F_{\text{low } T}$ of the low temperature FM (PMI) 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. 4(a)), as its magnetic ordering is different from that of the insulating phase above it. Similarly for the manganites, at low enough temperatures the PMM phase becomes either charge-ordered or glassy, beyond our approximations.

In this paper 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 which 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 titanates⁷, high temperature superconductors²⁵, ferroelectrics²⁶, and molecular fullerides²⁷.

At low enough temperatures one should surely take care of 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, e.g. a variational scheme such as the Lang-Firsov transformation. Nonetheless, the model we employ does generate a quantum critical point on account of 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 manganites⁹ and nickelates²⁸, hydrostatic compression decreases ϕ_0 . According to our model, this should result in an increase of T_{MI} promoted by the enhancement of the elastic interaction in the manganites, and viceversa for the nickelates. These are indeed the trends that have been observed in these materials.^{28,29} We believe a similar mechanism is at play when the transition is tuned with tensile and compressive stresses³⁰.

The idea that cooperative phonon-phonon couplings tune the MIT is supported by a recent ab-initio calculation¹⁹. By using density functional theory (DFT), it has been found that the tilts of the NiO_6 units in the nickelates destabilize their breathing distortions, which in turn are associated with the phase transition, thus providing a mechanism for tuning T_{MI} . However, DFT treats the elastic interactions only in average and it cannot produce finite temperature properties, thus T_{MI} was obtained by fitting it 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_{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 the energy could be separated into a relatively simple local free energy plus a complex long range piece, could provide a basis for a fully computational methodology that could be applied relatively simply to very complex oxides in general.

Acknowledgements. We acknowledge insightful discussions with Gil Lonzarich, Hyowon Park and Fabián Ballar-Trigueros. Work at Argonne National Laboratory is supported by the U.S. Department of Energy, Materials Science Division, Office of Basic Energy Sciences under contract no. DE-AC02-06CH11357. G.G.G.V. acknowledges support from the Vice-rectory for Research (project no. 816-B7-601), and the Office of International Affairs at the University of Costa Rica, the Royal Society International Exchanges programme (IES\R3\170025), Churchill College (Cambridge), and thanks the Department of Materials Science and Metallurgy and the Cavendish Laboratory at the University of Cambridge for hospitality where part of this work was done. R.T.B. acknowledges support from the Yale Prize Postdoctoral Fellowship and Homerton College (Cambridge).

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.

Methods

Statistical mechanical solution. We use a variational pair-distribution function that incorporates mean-field behavior, Gaussian corrections to the thermal and quantum fluctuations, and averaging over compositional disorder at the level of the replica method^{31,32}. Details are provided in Supplementary Note 2.

Model parameters. Our model has six parameters ($\kappa, \gamma, a_D, a_S, a_T$, and $F_{\text{low}T}$), which are reduced to five as a_T (a_D) is combined with κ for the manganites (nickelates). We begin by choosing a set of physically reasonable parameters which give phonon frequencies that are in order-of-magnitude agreement with the observed relevant modes^{33,34}. We then take the resulting set of parameters and fine tune them to fit the observed dependence of T_{MI} with the tolerance factor and compositional disorder: $F_{\text{low}T}$ is a parameter of the model assumed to be independent of T, ϕ_0 , and Δ , fixed by the observed onset of the MIT, i.e., $F_{\text{lattice}}(T = 0 \text{ K}, \phi_0 = \phi_{\text{onset}}, \Delta = 0) = F_{\text{low}T}$, where $\phi_{\text{onset}} \simeq 11.5^\circ$ (12.5°) for the manganites (nickelates). The dependence of T_{MI} on ϕ_0 shown in Figs. 4(a) and 4(b) is given by $F_{\text{lattice}}(T_{\text{MI}}, \phi_0, \Delta = 0) = F_{\text{low}T}$, while the dependence of T_{MI} on Δ shown in Fig. 4(c) is given by $F_{\text{lattice}}(T_{\text{MI}}, \phi_0 = 8.0^\circ, \Delta) = F_{\text{low}T}$ and by rescaling Δ by a constant factor ($4.71 \text{ \AA}/\text{meV}$) to match the units of cation variance. The resulting values are given in Extended Data Table I.

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

-
- ¹ M. Imada, A. Fujimori, and Y. Tokura, “Metal-insulator transitions,” *Rev. Mod. Phys.* **70**, 1039–1263 (1998).
- ² Z. Yang, C. Ko, and S. Ramanathan, “Oxide electronics utilizing ultrafast metal-insulator transitions,” *Ann. Rev. Mater. Res.* **41**, 337–367 (2011).
- ³ Y. Tokura, “Critical features of colossal magnetoresistive manganites,” *Rep. Prog. Phys.* **69**, 797–851 (2006).
- ⁴ S. Catalano, M. Gibert, J. Fowlie, J. Íñiguez, J.-M. Triscone, and J. Kreisel, “Rare-earth nickelates $RNiO_3$: thin films and heterostructures,” *Rep. Prog. Phys.* **81**, 046501 (2018).
- ⁵ J. B. Torrance, P. Lacorre, A. I. Nazzari, E. J. Ansaldo, and Ch. Niedermayer, “Systematic study of insulator-metal transitions in perovskites $RNiO_3$ ($R=Pr, Nd, Sm, Eu$) due to closing of charge-transfer gap,” *Phys. Rev. B* **45**, 8209–8212 (1992).
- ⁶ H. Y. Hwang, S-W. Cheong, P. G. Radaelli, M. Marezio, and B. Batlogg, “Lattice effects on the magnetoresistance in doped $LaMnO_3$,” *Phys. Rev. Lett.* **75**, 914–917 (1995).
- ⁷ T. Katsufuji, Y. Taguchi, and Y. Tokura, “Transport and magnetic properties of a Mott-Hubbard system whose bandwidth and band filling are both controllable: $R_{1-x}Ca_xTiO_{3+y/2}$,” *Phys. Rev. B* **56**, 10145–10153 (1997).
- ⁸ D. D. Sarma, N. Shanthi, and P. Mahadevan, “Electronic structure and the metal-insulator transition in $LnNiO_3$ ($Ln=La, Pr, Nd, Sm$ and Ho): bandstructure results,” *J. Cond. Matt. Phys.* **6**, 10467–10474 (1994).
- ⁹ P. G. Radaelli, G. Iannone, M. Marezio, H. Y. Hwang, S-W. Cheong, J. D. Jorgensen, and D. N. Argyriou, “Structural effects on the magnetic and transport properties of perovskite $A_{1-x}A'_xMnO_3$ ($x = 0.25, 0.30$),” *Phys. Rev. B* **56**, 8265–8276 (1997).
- ¹⁰ M. Medarde, P. Lacorre, K. Conder, F. Fauth, and A. Furrer, “Giant ^{16}O - ^{18}O isotope effect on the metal-insulator transition of $RNiO_3$ perovskites ($R=rare\ earth$),” *Phys. Rev. Lett.* **80**, 2397–2400 (1998).
- ¹¹ J. Varignon, M. N. Grisolia, J. Íñiguez, A. Barthélémy, and M. Bibes, “Complete phase diagram of rare-earth nickelates from first-principles,” *npj Quantum Materials* **2**, 21 (2017).
- ¹² A. Fujimori, “Electronic structure of metallic oxides: Band-gap closure and valence control,” *J. Phys. Chem. Solids* **53**, 1595–1602 (1992).
- ¹³ E. Pavarini, A. Yamasaki, J. Nuss, and O. K. Andersen, “How chemistry controls electron localization in $3d^1$ perovskites: a Wannier-function study,” *New J. Phys.* **7**, 188 (2005).
- ¹⁴ E. Gorelov, M. Karolak, T. O. Wehling, F. Lechermann, A. I. Lichtenstein, and E. Pavarini, “Nature of the Mott transition in Ca_2RuO_4 ,” *Phys. Rev. Lett.* **104**, 226401 (2010).
- ¹⁵ Q. Han and A. Millis, “Lattice energetics and correlation-driven metal-insulator transitions: the case of Ca_2RuO_4 ,” *Phys. Rev. Lett.* **121**, 067601 (2018).
- ¹⁶ J. M. Rondinelli, S. J. May, and J. W. Freeland, “Control of octahedral connectivity in perovskite oxide

- heterostructures: an emerging route to multifunctional materials discovery,” *MRS Bulletin* **37**, 261–270 (2012).
- ¹⁷ L. M. Rodríguez-Martínez and J. P. Attfield, “Cation disorder and size effects in magnetoresistive manganese oxide perovskites,” *Phys. Rev. B* **54**, R15622–R15625 (1996).
 - ¹⁸ A. J. Millis, P. B. Littlewood, and B. I. Shraiman, “Double exchange alone does not explain the resistivity of $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$,” *Phys. Rev. Lett.* **74**, 5144–5147 (1995).
 - ¹⁹ A. Mercy, J. Bieder, J. Íñiguez, and P. Ghosez, “Structurally triggered metal-insulator transition in rare-earth nickelates,” *Nat. Comm.* **8**, 1 (2017).
 - ²⁰ S. Kartha, J. A. Krumhansl, J. P. Sethna, and L. K. Wickham, “Disorder-driven pretransitional tweed pattern in martensitic transformations,” *Phys. Rev. B* **52**, 803 (1995).
 - ²¹ K. H. Ahn, T. Lookman, and A. R. Bishop, “Strain-induced metal-insulator phase coexistence in perovskite manganites,” *Nature* **428**, 401–404 (2004).
 - ²² D. I. Khomskii and K. I. Kugel, “Elastic interactions and superstructures in manganites and other Jahn-Teller systems,” *Phys. Rev. B* **67**, 134401 (2003).
 - ²³ K. H. Ahn, T. F. Seman, T. Lookman, and A. R. Bishop, “Role of complex energy landscapes and strains in multiscale inhomogeneities in perovskite manganites,” *Phys. Rev. B* **88**, 144415 (2013).
 - ²⁴ A. J. Millis, “Cooperative Jahn-Teller effect and electron-phonon coupling in $\text{La}_{1-x}\text{A}_x\text{MnO}_3$,” *Phys. Rev. B* **53**, 8434–8441 (1996).
 - ²⁵ J. P. Attfield, A. L. Kharlanov, and J. A. McAllister, “Cation effects in doped La_2CuO_4 superconductors,” *Nature* **394**, 157–159 (1998).
 - ²⁶ P. V. Balachandran, S. R. Broderick, and K. Rajan, “Identifying the inorganic gene for high-temperature piezoelectric perovskites through statistical learning,” *Proc. R. Soc. A* **467**, 2271–2290 (2011).
 - ²⁷ R. H. Zadik, Y. Takabayashi, G. Klupp, R. H. Colman, A. Y. Ganin, A. Potočník, P. Jeglič, D. Arçon, P. Matus, K. Kamarás, Y. Kasahara, Y. Iwasa, A. N. Fitch, Y. Ohishi, G. Garbarino, K. Kato, M. J. Rosseinsky, and K. Prassides, “Optimized unconventional superconductivity in a molecular Jahn-Teller metal,” *Sci. Adv.* **1**, e1500059 (2015).
 - ²⁸ X. Obradors, L. M. Paulius, M. B. Maple, J. B. Torrance, A. I. Nazzal, J. Fontcuberta, and X. Granados, “Pressure dependence of the metal-insulator transition in the charge-transfer oxides RNiO_3 ($\text{R}=\text{Pr}, \text{Nd}, \text{Nd}_{0.7}\text{La}_{0.3}$),” *Phys. Rev. B* **47**, 12353–12356 (1993).
 - ²⁹ J. Fontcuberta, V. Laukhin, and X. Obradors, “Local disorder effects on the pressure dependence of the metal-insulator transition in manganese perovskites,” *Appl. Phys. Lett.* **72**, 2607–2609 (1998).
 - ³⁰ J. Liu, M. Kargarian, M. Kareev, B. Gray, P. J. Ryan, A. Cruz, N. Tahir, Y.-D. Chuang, J. Guo, J. M. Rondinelli, J. W. Freeland, G. A. Fiete, and J. Chakhalian, “Heterointerface engineered electronic and magnetic phases of NdNiO_3 thin films,” *Nat. Comm.* **4**, 2714 (2013).
 - ³¹ G. G. Guzmán-Verri, P. B. Littlewood, and C. M. Varma, “Pараelectric and ferroelectric states in a model for relaxor ferroelectrics,” *Phys. Rev. B* **88**, 134106 (2013).
 - ³² J. R. Arce-Gamboa and G. G. Guzmán-Verri, “Random electric field instabilities of relaxor ferroelectrics,”

[npj Quantum Materials](#) **2**, 28 (2017).

- ³³ M. Zaghrioui, A. Bulou, P. Lacorre, and P. Laffez, “Electron diffraction and Raman scattering evidence of a symmetry breaking at the metal-insulator transition of NdNiO₃,” [Phys. Rev. B](#) **64**, 081102 (2001).
- ³⁴ L. Martín-Carrón, A. de Andrés, M. J. Martínez-Lope, M. T. Casais, and J. A. Alonso, “Raman phonons as a probe of disorder, fluctuations, and local structure in doped and undoped orthorhombic and rhombohedral manganites,” [Phys. Rev. B](#) **66**, 174303 (2002).

FIGURES

Figure 1: Pervoskite lattices. (a) 3D perovskite lattice showing the tilts of the BO_6 (B=Mn,Ni) octahedra. R is a rare earth element such as La, Pr, Nd, and Sm; and M is an alkaline earth metal such as Ca, Sr, Ba. (b) 2D representation of the tilts used in our model where ϕ_0 is an initial equilibrium antiferrodistortive rotation.

Figure 2: Lattice distortions and strain responses. (a) Lattice distortions considered in our model. (b) Strain responses of a lattice to a local JT distortion as a result of rotations. The color of each square indicates the strain energy associated with the local distortions of that square. The grey parallelogram at the centre has a JT distortion of fixed amplitude. The strain fields weaken by allowing the BO_6 to tilt, as the energy is more effectively absorbed locally. Additional distortions on this site, such as shear, are allowed. Top: lattice with $\phi_0 = 0$. Bottom: lattice with $\phi_0 = 15^\circ$.

Figure 3: Effective elastic energy. Effective elastic energy for JT distortions in momentum space for (a) $\phi_0 = 0$ and (b) $\phi_0 = 15^\circ$. 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^{21,23} such as domain patterns in the form of stripes and tweeds formed by interwoven incommensurate structures. Effective elastic energy for breathing distortions in momentum space for (c) $\phi_0 = 0$ and (d) $\phi_0 = 20^\circ$. 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.

Figure 4: Comparison to experiments. (a) Comparison for the nickelates for the transition temperature as a function of octahedral tilt angle. Filled and open circles are experimental transition temperatures⁴ 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 denotes LaNiO_3 , which is always in the high temperature polaronic, paramagnetic metal (PMM) phase. For the manganites, the comparison is made with results¹⁷ (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 Methods. In Supplementary Note 3, we explore reasonable variations of the model parameters to demonstrate its generality.

TABLES

Extended Data Table I. Model parameters. R is a rare earth element such as La, Pr, Nd, and Sm; and M is an alkaline earth metal such as Ca, Sr, Ba.

	κ [meV ²]	γ [meV ³]	a_D [meV ²]	a_S [meV ²]	a_T [meV ²]	$F_{\text{low}T}$ [meV]
(R _{0.3} M _{0.7})MnO ₃	3.2×10^3	1.8×10^5	2.8×10^4	9.5×10^3	combined with κ	38
RNiO ₃	12.1×10^3	13.3×10^5	combined with κ	48.3×10^3	1.21×10^4	64