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Large phonon entropy drives the metallization of vanadium dioxide (VO2)[1]

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Vanadium dioxide (VO_2) exhibits a first-order metal-insulator transition (MIT) near room temperature, where conductivity is suppressed and the lattice changes from tetragonal to monoclinic on cooling. This MIT in VO_2 has attracted intense interest from both fundamental and technological perspectives. However, most studies performed in the past 50 years have focused on the electronic structure and energetics of the transition, ignoring the role of phonons and their entropic contribution to the phase stability. Much of the reason is that the standard tool of neutron scattering does not yield coherent scattering from V nuclei, and first-principles methods with harmonic approximation cannot capture the stable phonons for the rutile phase. We close this gap by using a combination of ab initio molecular dynamics calculations and neutron/x-ray scattering to establish that the entropy driving the MIT is dominated by soft, anharmonic phonons of the metallic phase.[2] The MIT results from the competition between lower electronic energy in insulating M1 phase due to the Peierls instability, and the higher entropy of the metallic rutile phase resulting from soft anharmonic phonons. This understanding of the role of lattice dynamics and their relationship to electronic structure provides a critical component for developing more complete physical models of phase competition in functional transition metal oxides.

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[2]J. D. Budai*, J. Hong*, M. E. Manley, E. D. Specht, C. W. Li, J. Z. Tischler, D. L. Abernathy, A. H. Said, B. M. Leu, L. A. Boatner, R. J. McQueeney and O. Delaire, Nature, (2014), doi:10.1038/nature13865