Moore *et al.* Reply: In the preceding Comment [1] on our recent Letter [2], Simak and Ruban argue that is unclear how a first-principles code could produce the difference in bonding force as our calculations showed [2]. Simak and Ruban [1] argue that similar crystallographic directions could never exhibit different force responses. They do agree, however, that our results are possible to obtain if the calculations are "done with such a setup which destroys the symmetry of the system." Indeed, contrary to most first-principles calculations, we did not enforce explicit symmetrization of the crystal structure by removing all crystal-symmetry constraints. This idea of removing crystal-symmetry constraints in electronic-structure calculations has been used for Mn [3], another metal with complex electronic and magnetic structure.

Mn and Pu lie on the transition between magnetic and superconductive valence electron behavior, both exhibiting crystallographic phase instability. A rearranged Periodic Table is shown in Fig. 1(a) with the $4f$, $5f$, $3d$, $4d$, and $5d$ metals [4]. At ground state, metals in the blue area exhibit superconductivity and metals in the red area exhibit magnetism. The white band is a transition between localized (magnetic) and itinerant (conductive) valence electron behavior. This diagram is extended in Fig. 1(b) [5] to reveal the crystallographic phase instability of the metals that lie on this transition by using gray scale for the number of solid allotropic phases. Each metal on the transition has frustrated valence electrons, behaving unlike "normal" metals, and it is clear that both Mn and Pu exhibit numerous phases, meaning the use of unorthodox approaches to elucidate the electronic structure of each metal is justified.

Hobbs, Hafner, and Spišák [3] showed this in great detail for α-Mn, a metal with a complex body centered cubic unit cell consisting of four crystallographically distinct Mn atoms. Their calculations [3] were performed "without any symmetry constraint. This is in contrast to most *ab initio* crystallographic optimization which are performed under the constraint of conservation of the space-group symmetry and where charge densities, potentials, and forces are symmetrized after each iteration." Only when Hobbs, Hafner, and Spišák removed all symmetry constraints posed by the crystal geometry, thus allowing the possibility of a symmetry-breaking state, were their calculations able to correctly describe the ground state of Mn. They found that groups of atoms, which were related through symmetry operations, and thus were crystallographically equivalent, split into subclasses with different magnetic and electronic structure.

Hobbs, Hafner, and Spišák [3] did go on to say that "The drawback [of this approach] is that without explicit symmetrization of charge and spin densities and of the interatomic forces and magnetic torques, a higher level of convergence must be achieved to avoid spurious symmetry breaking due to numerical fluctuations." We fully understand this and took great strides to ensure that the con-
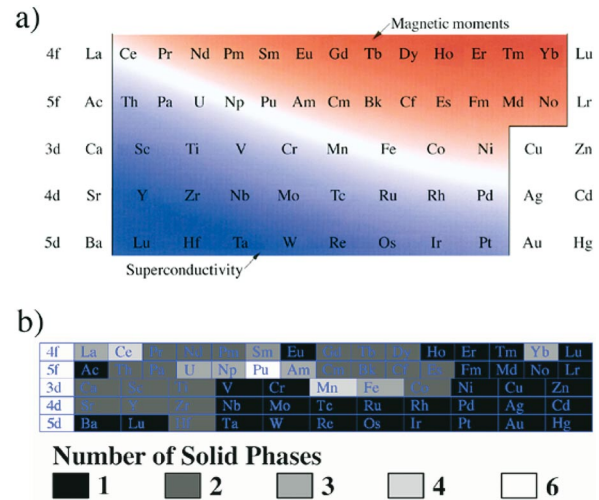

FIG. 1 (color). (a) Rearranged Periodic Table of the $4f$, $5f$, and $3d$ to $5d$ metal series (after [4]). (b) Version of (a) showing the number of solid allotropic phases for each metal [5].

vergence of our calculations was both robust and repeatable. Indeed, we even tested our approach on Al [2], a well-known isotropic metal that is also face-centered-cubic, and found the same bonding force between the 12 nearest neighbors, ruling out any systematic computational error.

The exact mechanism for the broken-symmetry state found in δ-Pu is not known. Subtle shifts in the computed total energy are responsible, but not easily analyzed within the framework of highly complex electronic-structure calculations. The result is likely related to the properties of the $5f$ electrons, which are known to have a strong preference for anisotropy giving rise to low symmetry crystal structures in Pu and the actinides in general.

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[1] S. I. Simak and A. V. Ruban, preceding Comment, Phys. Rev. Lett. **99,** 019703 (2007).
[2] K. T. Moore, P. Söderlind, D. E. Laughlin, and A. J. Schwartz, Phys. Rev. Lett. **96,** 206402 (2006).
[3] D. Hobbs, J. Hafner, and D. Spišák, Phys. Rev. B **68,** 014407 (2003).
[4] J. L Smith and E. A. Kmetko, J. Less-Common Met. **90,** 83 (1983).
[5] K. T. Moore *et al.*, Philos. Mag. **84,** 1039 (2004).