Comment on "Symmetry and Stability of δ Plutonium: The Influence of Electronic Structure"

In a recent Letter [1], Moore *et al.* offer a new explanation for the complex structural behavior of plutonium, based on the different bonding strength between an atom and its 12 nearest neighbors in the face-centered cubic (fcc) δ-Pu. The bonding strength is estimated in Ref. [1] through the calculations of the energy response of a small ($\sim$2%) displacement of an atom along each of the 12 nearest-neighbor directions. The authors of Ref. [1] report that this response is asymmetric, for instance it is different for the displacements of an atom in the supercell $3 \times 3 \times 3$ in $[0, 1, 1]$ and $[0, -1, -1]$ directions by 1.6 mRy/Å.

Unfortunately it is not clear from Ref. [1] how such a result can be obtained by any of the standard first-principles codes (including the one used by Moore *et al.* [2]). The arrangement of atoms in real space dictates the symmetry of the electronic structure and bonding. This is the basis for the whole solid state physics. Without it the Bloch theorem [3] would not be valid and electronic structure calculations would not be possible. It is up to someone's taste to consider all the atoms as inequivalent in the perfectly arranged ferromagnetic fcc lattice [1]; however, the self-consistent electron density of every atom must obey the proper translational and rotational symmetry. Since the electronic structure uniquely determines the total energy of the system, the equivalent from the symmetry point of view displacements must produce the same response. Figure 1 illustrates the actual problem of Ref. [1] in two dimensions. Note that from Ref. [1] it follows that energies of the equivalent structures in Figs. 1(b) and 1(d) are different.

There is the basic principle of the symmetry of physical laws, so if the calculations for some reason produce the results, which violate it, they must be erroneous, or done with such a setup that destroys the symmetry of the system. Certainly, the numerical calculations have finite accuracy, since the computers can handle only rational numbers. However, if the calculations are properly done, the error should be negligible.

In order to estimate typical numerical noise we have repeated the calculations of Ref. [1] using exactly the same setup ($3 \times 3 \times 3$ 27 atom fcc-based supercell, ferromagnetic spin alignment, all the atoms treated as inequivalent, no spin-orbit coupling, etc.) within a similar first-principles technique, the projector augmented wave (PAW) method [4], as implemented in the Vienna *ab initio* simulation package [5]. We find that the difference in the energy response for the displacements in the equivalent directions toward the nearest neighbors in δ-Pu is of order 0.001 mRy/Å. This value is up to 3 orders of magnitude less than those found in Ref. [1] and such a noise cannot be considered as the basis for new revolutionary ideas.

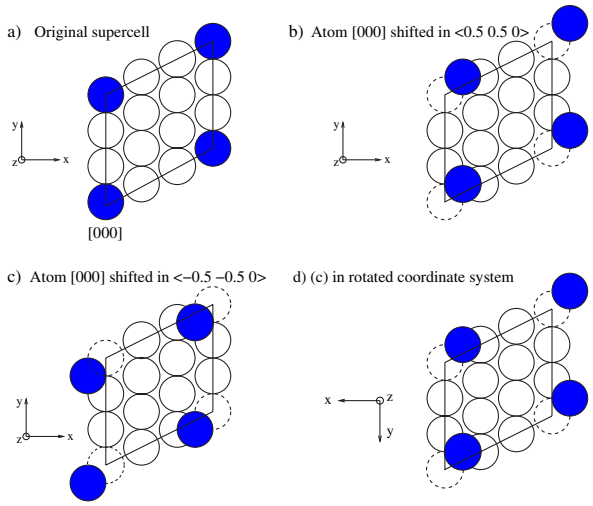

FIG. 1 (color online). The actual problem of Ref. [1], illustrated in two dimensions. The supercell is shown with solid lines. Note that it is periodically repeated in the *xy* plane (not shown). Atoms are shown with circles. Closed circles correspond to atom [000] and its periodic images, dashed circles to their initial positions. According to Ref. [1], total energies of (b) and (c) may be different. However, both are ferromagnetic configurations without spin-orbit coupling [1]. Therefore, there is no preferred direction and (c) is equivalent to (d), which is identical to (b). Therefore, energies of (b) and (c) must be the same.

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