Comment on "New Ground-State Crystal Structure of Elemental Boron"

In a recent Letter, density functional theory (DFT) calculations were carried out on a structure observed by high resolution transmission electron microscopy experiments and used to predict that τ-B is the ground state structure of elemental boron at atmospheric pressure [1]. We have performed DFT total energy calculations on the structures provided in the Supplemental Material (SM) of Ref. [1] using the same code (VASP v.5.3.5) and calculation conditions used in that work, and with two projector-augmented wave pseudopotentials distributed with the VASP code that have different core radius for the projection operator: RMAX = 2.583 (PS 1) and RMAX = 1.732 (PS 2) [2–5]. In Ref. [1], it was reported that the pseudopotential from Ref. [5] was used for all calculations. An additional set of calculations were performed using PWSCF v.5.2.0 [6] with a cutoff energy of 40 Ry using an ultrasoft pseudopotential [7] and with a well converged numbers of k points. The relative energies of our three independent calculations are all in agreement (within about 0.2 meV/atom) and indicate that τ-B106 is higher in energy than both α-B12 and β-B106 ($\delta E_{\tau-\alpha} = +12.9$, $\delta E_{\tau-\beta} = +11.8$ meV/atom); i.e., the ground state of elemental boron is not τ boron. To understand the source of discrepancy between our results and the results presented in Ref. [1], we compared the total energies of these systems calculated with VASP, the code that was used to obtain the results of Ref. [1], using the two different pseudopotentials PS 1 and PS 2 (see Fig. 1). It has turned out that our total energies of τ-B105 and τ-B106 calculated with PS 2 are identical to the corresponding values reported in Table S1 of the SM of Ref. [1], while our total energies of α-B12, β-B105, β-B106, and γ-B28 calculated with PS 1 are identical to the corresponding values in the SM for Ref. [1] (Fig. 1). This observation strongly suggests that the total energies provided in the SM of Ref. [1] were obtained with two different pseudopotentials. Finally, we comment on the revised conclusions provided in the Erratum [8] regarding relative stabilities of β boron and τ boron at high temperature. The claim that τ boron may be more stable than β boron for $T > 950$ K, is in contradiction with the experimental observation that liquid boron solidifies into β boron, not τ boron [9,10].

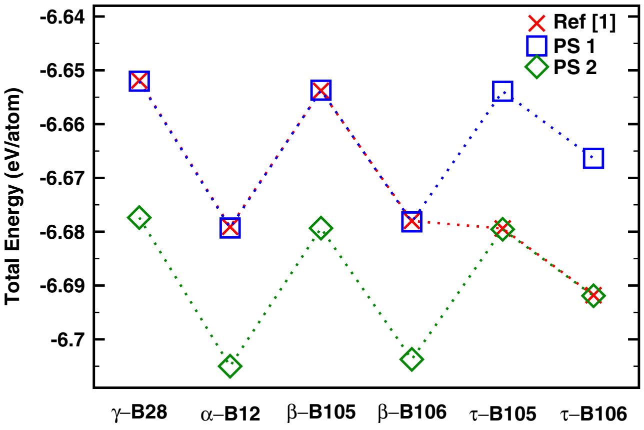

FIG. 1. Total energies of boron allotropes calculated with two different pseudopotentials compared to the data published in Ref. [1]. It appears that the published data in Ref. [1] were generated with two different pseudopotentials: PS 1 for α-B12, β-B105, β-B106 and γ-B28, and PS 2 for τ-B105 and τ-B106.

In summary, we have performed DFT total energy calculations of the boron allotrope structures published in Ref. [1] using two different *ab initio* codes, VASP and PWSCF, and two different pseudopotentials for VASP. None of our simulation results support the conclusion of Ref. [1], and instead indicate that τ boron is not the ground state. We have shown clear evidence that the total energies calculated with VASP and published in Ref. [1] were most likely obtained by the inconsistent use of pseudopotentials, which has led to an incorrect conclusion.

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