TABLE III: Calculated elastic constants, various moduli, and Poisson's ratio υ for thorium hydrides.

Compound	C_{11} (GPa)	C_{12} (GPa)	C_{44} (GPa)	C_{13} (GPa)	C_{33} (GPa)	C_{66} (GPa)	B (GPa)	$B^{'}$ (GPa)	G (GPa)	E (GPa)	υ
bct ThH_2	131	100	29	78	89	5	91	3.3	16	44	0.419
fcc ThH_2	46	119	57				95 (99)[a]	3.8			
Th_4H_{15}	121	66	59				85	2.2	43	111	0.282

[a] Reference[5].

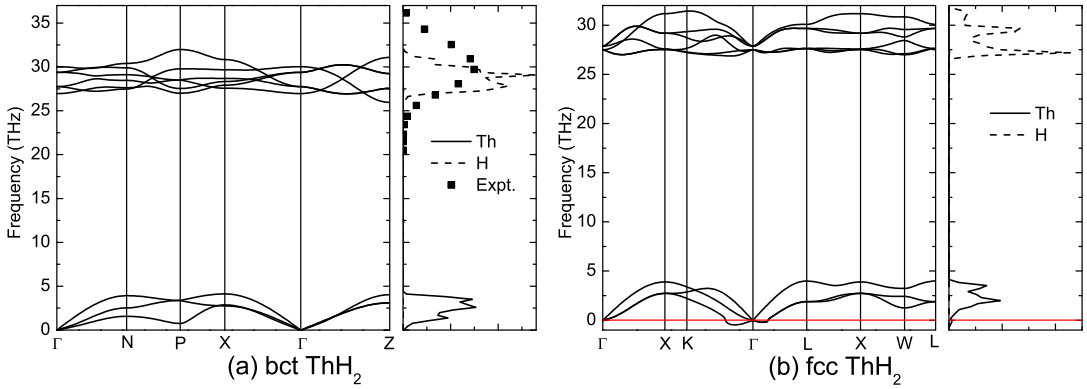

FIG. 6: (Color online) Calculated phonon dispersion curves and corresponding PDOS for (a) bct ThH_2 and (b) fcc ThH_2. For comparison, the experimental data in Ref. [4] are also shown (squares).

$\Gamma-N-P-X-\Gamma-Z$ directions for bct ThH_2 and along the $\Gamma-X-K-\Gamma-L-X-W-L$ directions for fcc ThH_2 are displayed in Fig. 6(a) and Fig. 6(b), respectively.

For both bct and fcc ThH_2, there are only three atoms in their primitive cells. Therefore, nine phonon modes exist in the dispersion relations. Due to the fact that thorium is much heavier than hydrogen atom, the vibration frequency of thorium atom is apparently lower than that of hydrogen atom. Therefore, evident gap between the optic modes and the acoustic branches exits and the phonon DOS of bct (fcc) ThH_2 can be viewed as two parts. One is the part lower than 4.4 (4.1) THz where the main contribution comes from the thorium sublattice, while the other part range from 26.0 to 32.1 (26.4 to 31.8) THz is dominated by the dynamics of the light hydrogen atoms. In experimental measurements, Dietrich *et al.* reported that the acoustic branches of ThH_2 are lower than 4.8 THz and optic modes range from 23.4 to 36.2 THz[4]. Their measured data of optic modes are presented in Fig. 6(a) for comparison and the acoustic-phonon data are unable obtained from their time-of-flight spectrum. Obviously, our calculated results are on the whole consistent with the experimental data. In addition, while all frequencies are positive for bct ThH_2, which assures a dynamical stability of the bct phase against mechanical perturbations. For fcc ThH_2, one can see from Fig. 6 (b) that the transverse acoustic (TA) mode close to Γ point becomes imaginary along the $\Gamma-K$ and $\Gamma-L$ directions. The minimum of the TA branch occurs along the $\Gamma-K$ direction. This indicates instability of fcc phase of ThH_2 compared to stable bct phase, which is well consistent with our previous mechanical stability analysis of ThH_2. Moreover, there exists LO-TO splitting at Γ point in bct phase. But in fcc phase there is no LO-TO splitting.

IV. CONCLUSION

In summary, we have used the first-principles DFT-GGA method to calculate the structural, electronic, mechanical, and thermodynamic properties of ThH_2 in its stable bct phase and metastable fcc phase and Th_4H_{15} in bcc phase. Our optimized structural parameters are well consistent with experiments. The occupation characters of electronic orbitals also accord well with experiments and previous calculations. Through Bader analysis, we have found that the Th−H bonds in all thorium hydrides exhibit weak covalent character, but the ionic property for ThH_2 and Th_4H_{15} are different. While ∼1.5 electrons transfer from each Th atom to H in ThH_2, in Th_4H_{15} about 2.1 electrons deviate from each Th atom. In addition, our calculated phonon curves of fcc ThH_2 have shown that the TA mode becomes imaginary close to Γ point and the subsequent instability of this phase.

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