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First principles study of the BeF_2 phase diagram: A critical comparison of outcomes using LDA, GGA and van der Waals corrected DFT
ANDREW CHIZMESHYA, GEORGE WOLF, NARGES MASOUMI, Arizona State University — Beryllium fluoride continues to draw attention because of its industrial and scientific applications, and because of its structural analogy with silica, but its detailed phase diagram has yet to be established experimentally. Recently the ordering of a dozen BeF_2 crystalline polymorphs was studied using GGA-based DFT static lattice enthalpies (Nelson et al, PRB **95** 054118, 2017). For pressures below 8 GPa this approach predicts the phase sequence [α-cristobalite $\rightarrow$ α-quartz$\rightarrow$coesite$\rightarrow$moganite] while severely overestimating the volume of the ambient pressure α-quartz phase by ˜10%. Since lattice expansion arising from thermal corrections would further increase this error we undertook a critical reinvestigation of the structures, relative stabilities and compression behavior of candidate phases using thermally corrected LDA and van der Waals corrected (DFT-D3) functionals. We find that the static lattice LDA description correctly predicts the observed α-quartz ground state, and a phase ordering [α-quartz$\rightarrow$coesite$\rightarrow$moganite] and that inclusion of thermal corrections at 300K do not significantly alter this ordering. In contrast, while the dispersion corrected PBE-GGA functional (DFT-D3) reproduces the observed 300K equilibrium volume only in the static lattice approximation, thermal corrections worsen the agreement. A more severe deficiency of DFT-D3 is that compressibility is severely underestimated (by about 50%) making it unsuitable for EOS studies. A rudimentary model of the BeF_2 phase diagram based on quasi-harmonic free-energy (LDA) will be discussed in detail.

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