

Supplemental Material for “Antiferroelectric topological insulators in orthorhombic $AMgBi$ compounds ($A = Li, Na, K$)”

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I. NaMgBi UNDER EPITAXIAL STRAIN AND HYDROSTATIC PRESSURE

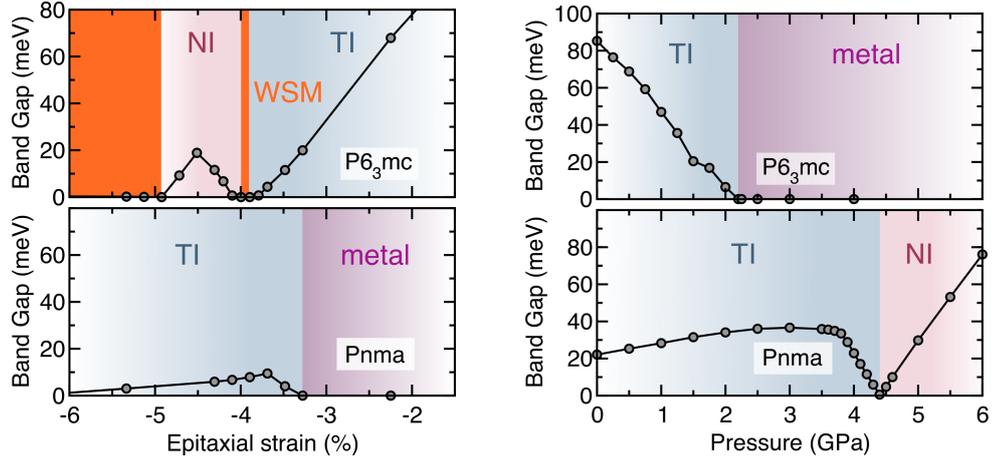


FIG. 1: Band gap of the NaMgBi compounds of polar $P6_3mc$ (top) and antipolar $Pnma$ (bottom) structures as a function of epitaxial strain (left) and hydrostatic pressure (right). The topological character of each phase is also indicated as normal insulator (NI), topological insulator (TI), Weyl semimetal (WSM), or metal.

II. KMgBi UNDER EPITAXIAL STRAIN AND HYDROSTATIC PRESSURE

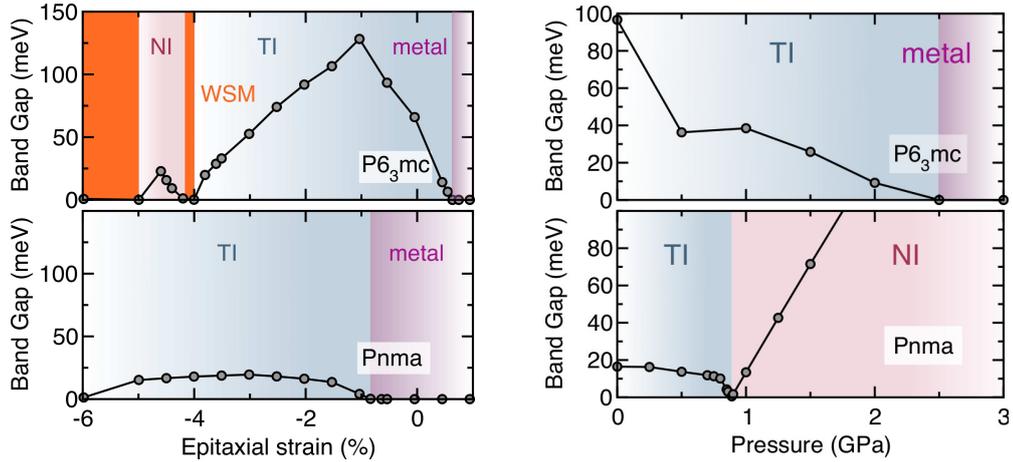


FIG. 2: Band gap of the KMgBi compounds of polar $P6_3mc$ (top) and antipolar $Pnma$ (bottom) structures as a function of epitaxial strain (left) and hydrostatic pressure (right). The topological character of each phase is also indicated as normal insulator (NI), topological insulator (TI), Weyl semimetal (WSM), or metal. The graph of hydrostatic pressure (right) is reproduced from the main manuscript for completeness.

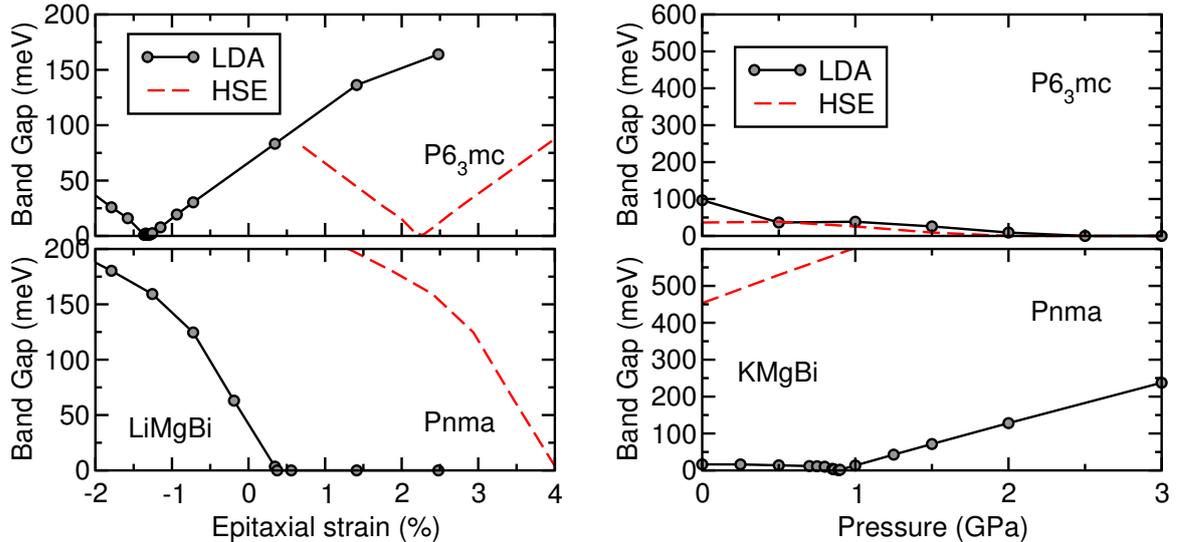


FIG. 3: Minimum band gap of the LiMgBi (left) and KMgBi (right) compounds of polar $P6_3mc$ (top) and antipolar $Pnma$ (bottom) structures as a function of epitaxial strain and hydrostatic pressure, respectively. The HSE results for the minimum band gap are extrapolated from the band gap at the Γ point as described in the text.

III. HYBRID FUNCTIONAL CALCULATIONS

The band gap results presented in the main text have been calculated using the local density approximation (LDA) to the exchange-correlation functional [1, 2]. It is well-known that band gaps evaluated using semilocal functionals such as the LDA tend to underestimate the magnitude of the band gap for normal insulators. Topological properties are insensitive to the precise magnitude of the band gap, and can only change when the band gap closes. For this reason, we expect that the phase diagrams presented in the main text are qualitatively independent of the approximation to the exchange-correlation functional used, and only the precise location of the phase boundaries might change with different levels of theory.

To confirm this, we have performed selected band gap calculations using the hybrid HSE functional [3, 4], and the results are shown in Fig. 3 for LiMgBi under epitaxial strain (left) and KMgBi under hydrostatic pressure (right) for the polar $P6_3mc$ (top) and antipolar $Pnma$ (bottom) structures. Due to the high computational cost of hybrid functional calculations, we have adopted the following strategy to reduce the number of calculations required, which we exemplify with the $P6_3mc$ structure of LiMgBi. We have performed HSE calculations at strains between -1.5% and $+4.0\%$, but rather than attempting to locate the minimum band gap at each strain, we have instead only calculated the band gap at the Γ point. Comparing the minimum and Γ -point band gaps at the LDA level suggests that the Γ -point band gap is a good proxy for the behaviour of the minimum band gap. Indeed, the HSE Γ -point band gap exhibits a minimum at a strain of $+2.3\%$. The Γ -point band gap at this minimum has a value of 0.12 eV, and the character of the bands switches between the valence and conduction bands across this minimum. The same behaviour is observed for LDA calculations, but the minimum Γ -point band gap then occurs at strains around -1.3% , which coincides with the strain-induced topological phase transition. The difference in the Γ -point band gaps between the LDA and the HSE calculations is constant over the entire strain range, and therefore, for clarity, in Fig. 3 we show the HSE results as a dashed line which corresponds to the shifted LDA results by the appropriate amount. A similar procedure is used to obtain the other diagrams shown in Fig. 3. We note that the $P6_3mc$ structure of KMgBi shows a smaller band gap shift than the rest in going from LDA to HSE, and this is caused by a reordering of the valence bands when changing the level of theory used.

In LiMgBi (Fig. 3a), the HSE band gap inversion for the polar $P6_3mc$ structure occurs at an epitaxial strain of about $+2.3\%$, compared to -1.3% for LDA, which corresponds to a shift in epitaxial strain of about 3.6% . Similarly, the epitaxial strain at which the antipolar $Pnma$ structure becomes metallic increases from the LDA $+0.4\%$ value to about $+4.0\%$ for HSE, a similar increase of 3.6% in epitaxial strain. These results imply that LiMgBi exhibits an AFTI phase of type I at strains in the range $+2.3$ to $+4.0\%$ in the HSE description.

In KMgBi (Fig. 3b), we predict that for the ground state antipolar $Pnma$ structure, the material is in the normal phase already at 0 GPa, and the topological phase is pushed to *negative* pressures. For the polar $P6_3mc$ structure,

the transition to the metallic phase occurs at pressures about 0.5 GPa lower using the HSE functional compared to the LDA functional, and the material stays in the topological phase for pressures below about 2 GPa. These results imply that KMgBi is an AFTI of type I under ambient conditions, and remains so at moderate pressures.

Overall, the use of a hybrid functional leads to quantitative differences in the phase diagrams of LiMgBi under strain and KMgBi under pressure. But the general structure of the phase diagrams remains unchanged. In particular, the existence of an AFTI of type I in both materials is robust with respect to the choice of exchange-correlation functional.

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