Supplementary material to:
Structural Variety beyond Appearance: High-pressure Phases of CrB$_4$ in comparison with FeB$_4$

Yunkun Zhang, a Lailei Wu, *a Biao Wan, a Yan Zhao, a Rui Gao,b Zhiping Li, b Jingwu Zhang*, a Huiyang Gou*cd and Ho-kwang Mao cd

a Key Laboratory of Metastable Materials Science and Technology, College of Material Science and Engineering, Yanshan University, Qinhuangdao 066004, China.
b Key Laboratory of Applied Chemistry, College of Environment and Chemical Engineering, Yanshan University, Qinhuangdao 066004, China.
c Center for High Pressure Science and Technology Advanced Research, Shanghai 201203, China
d Geophysical Laboratory, Carnegie Institution of Washington, 5251 Broad Branch Road NW, Washington, DC 20015, USA

Fig. S1 Total energy as a function of unit volume (per f.u.) curves for (a) CrB$_4$ and (b) FeB$_4$.

Fig. S2 Enthalpy difference (relative to $P_{nnm}$ structure) as a function of pressure for $I4_1/acd$-type and $P2_1 3$-type CrB$_4$ compared with the $mP10-\alpha$-CrB$_4$, $mP10-\beta$-CrB$_4$, and $oP10-\beta$-CrB$_4$. 
Fig. S3 Phonon dispersion curves of high pressure phases for CrB₄ and FeB₄. (a) mP10-α-CrB₄; (b) mP10-β-CrB₄; (c) oP10-β-CrB₄ at 0 GPa; (d) oP10-β-CrB₄ at 294 GPa; (e) tI40-FeB₄; (f) cP20-FeB₄. The oP10-β-CrB₄ is dynamically unstable at zero pressure with imaginary frequency in the Brillouin Zone. At pressure above 294 GPa, however, the oP10-β-CrB₄ becomes dynamically stable with all frequencies become positive. The other phases are dynamically stable at zero pressure with the absence of imaginary phonon frequencies throughout the Brillouin Zone. A supercell with 3×5×3 k-points for mP10-α-CrB₄, with 3×5×3 k-points for mP10-β-CrB₄, with 3×6×3 k-points for oP10-β-CrB₄, with 3×3×1 k-points for tI40-FeB₄, and with 4×4×4 k-points for cP20-FeB₄ were adopted.
Table SI Calculated elastic constants $C_{ij}$ (GPa) of the $mP10$-$\alpha$-CrB$_4$ and $mP10$-$\beta$-CrB$_4$.

<table>
<thead>
<tr>
<th>S.G.</th>
<th>$C_{11}$</th>
<th>$C_{22}$</th>
<th>$C_{33}$</th>
<th>$C_{44}$</th>
<th>$C_{55}$</th>
<th>$C_{66}$</th>
<th>$C_{12}$</th>
<th>$C_{13}$</th>
<th>$C_{15}$</th>
<th>$C_{25}$</th>
<th>$C_{35}$</th>
<th>$C_{46}$</th>
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<tbody>
<tr>
<td>$mP10$-$\alpha$-CrB$_4$</td>
<td>$P2/m$</td>
<td>592</td>
<td>490</td>
<td>477</td>
<td>186</td>
<td>225</td>
<td>224</td>
<td>125</td>
<td>179</td>
<td>173</td>
<td>67</td>
<td>-11</td>
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<tr>
<td>$mP10$-$\beta$-CrB$_4$</td>
<td>$Pm$</td>
<td>580</td>
<td>486</td>
<td>535</td>
<td>105</td>
<td>184</td>
<td>179</td>
<td>106</td>
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