

## Supporting Information

### The direct-current and alternating-current transport properties of hydrogenated bilayer $\beta_{12}$ - and $\chi_3$ -borophenes

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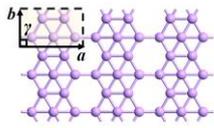
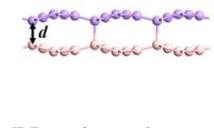
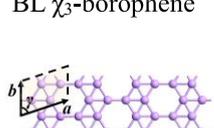
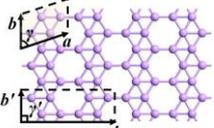
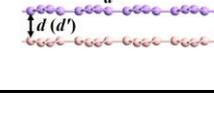
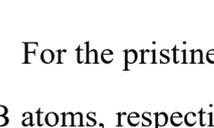
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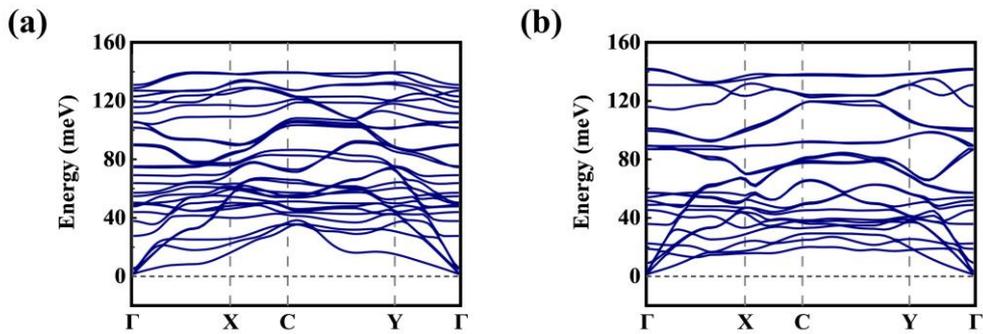
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**Table S1** The calculated lattice constants, intersection angles and interlayer distances of BL  $\beta_{12}$ - and  $\chi_3$ -borophenes.  $a$ ,  $b$ ,  $\gamma$ ,  $d$  are the results of the primitive cell, while  $a'$ ,  $b'$ ,  $\gamma'$ ,  $d'$  are the results of the conventional cell.

	$a$ (Å)	$b$ (Å)	$\gamma$ (°)	$d$ (Å)	$a'$ (Å)	$b'$ (Å)	$\gamma'$ (°)	$d'$ (Å)	
BL $\beta_{12}$ -borophene	<b>5.09</b>	<b>2.98</b>	<b>90</b>	<b>1.99</b>	—	—	—	—	<b>This work</b>
	5.09	2.98	90	1.99	—	—	—	—	Ref. 23
	5.06	2.94	90	2.00	—	—	—	—	Ref. 24
	4.98	2.93	90	1.96	—	—	—	—	Ref. 26
BL $\chi_3$ -borophene	<b>4.53</b>	<b>2.95</b>	<b>71</b>	<b>2.82</b>	<b>8.56</b>	<b>2.95</b>	<b>90</b>	<b>2.82</b>	<b>This work</b>
	—	—	—	—	8.56	2.95	90	2.82	Ref. 23
	—	—	—	—	8.41	2.90	90	2.80	Ref. 25
	—	—	—	—	8.35	2.89	90	2.84	Ref. 27

For the pristine BL  $\beta_{12}$ - and  $\chi_3$ -borophenes, the primitive cells contain ten and eight B atoms, respectively; the calculated  $a$ ,  $b$ ,  $\gamma$  and  $d$  are listed in Table S1. To facilitate comparisons with previous studies, we also give the calculated results for the conventional cell of BL  $\chi_3$ -borophene, *i.e.*,  $a'$ ,  $b'$ ,  $\gamma'$  and  $d'$ . They coincide well with the reported results.



**Fig. S1** The phononic energy bands of the pristine BL (a)  $\beta_{12}$ - and (b)  $\chi_3$ -borophenes.



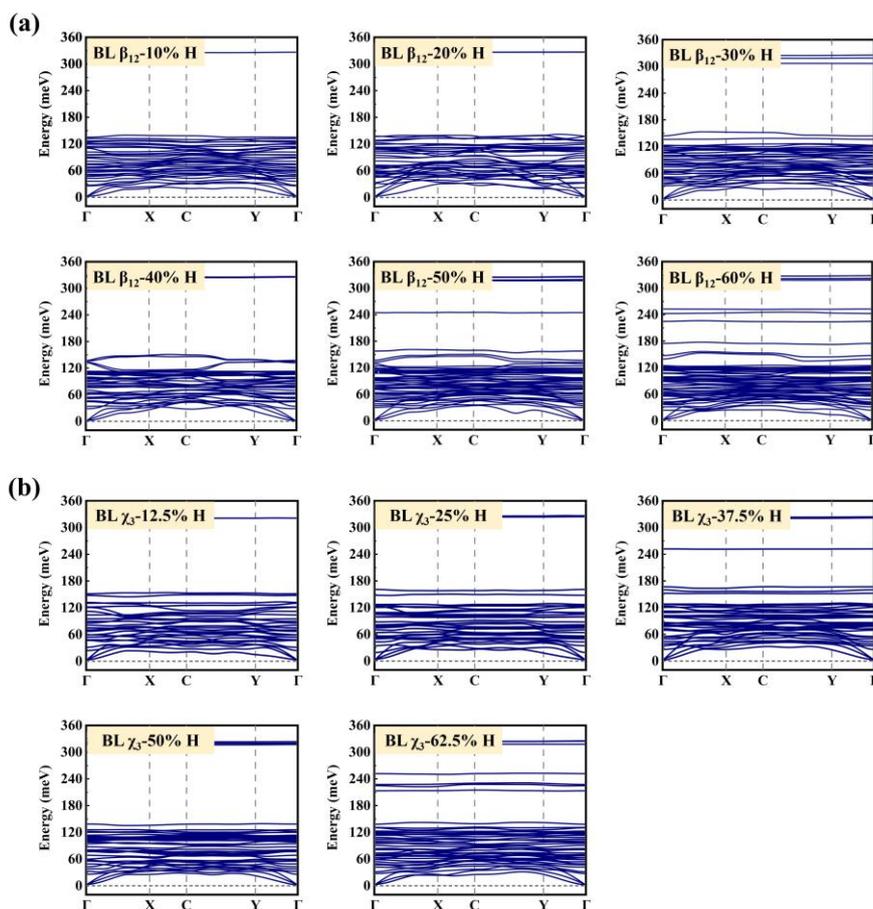
**Fig. S2** The examples of the initial structures and optimized structures of hydrogenated BL  $\beta_{12}$ -borophene under different H coverages.

To explore the hydrogenated BL structures, the design method and nomenclature of the initial structures are important. Based on BL  $\beta_{12}$ - and  $\chi_3$ -borophenes, we considered various possible initial hydrogenated BL structures.

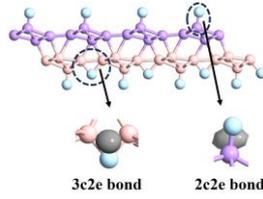
For BL  $\beta_{12}$ - and  $\chi_3$ -borophenes, their primitive cells contain ten and eight B atoms, respectively. Therefore, different H coverages were fully studied. Using BL  $\beta_{12}$ -borophene as an example, in Fig. S2 we give the initial structures and optimized structures as the H coverages are 10%, 20%, 30% and 100%. For a given H coverage, the optimized structures are ranked in descending order according to their formation energies (absolute values), and in the following discussion the symbols ‘I, II...’ are used

to indicate their stabilities. For example, when the H coverage is 10%, BL  $\beta_{12}$ -10% H-I represent the most stable hydrogenated BL structure, while BL  $\beta_{12}$ -10% H-II is the first metastable hydrogenated BL structure. In addition, all the optimized structures were verified by the calculated phononic energy bands. For BL  $\chi_3$ -borophene, the same nomenclature is adopted.

In addition, for BL  $\beta_{12}$ -borophene, we found many 2D semiconducting structures as the H coverage is 20%, 40% or 60%. The suffix ‘‘S’’ was added to denote their semiconducting properties. We also calculated the phononic energy bands to verify their dynamical stabilities. In the present study, three 2D semiconducting structures, *i.e.*, BL  $\beta_{12}$ -20% H-S, BL  $\beta_{12}$ -40% H-S and BL  $\beta_{12}$ -60% H-S, are discussed. Among the obtained 2D semiconducting structures, each of them has the largest formation energy (absolute value) under the given H coverage.



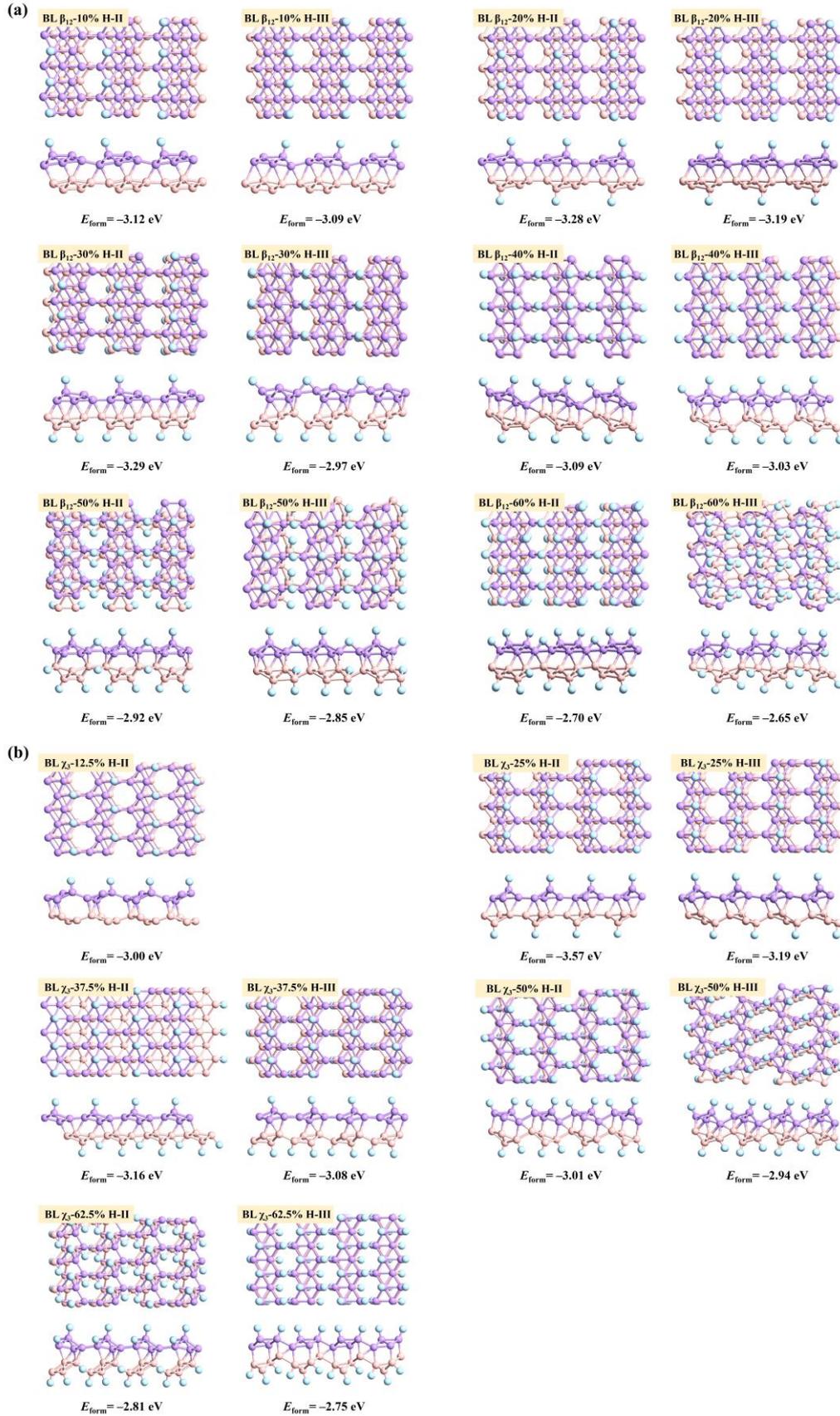
**Fig. S3** The phononic energy bands of the most stable hydrogenated BL (a)  $\beta_{12}$ - and (b)  $\chi_3$ -borophenes.



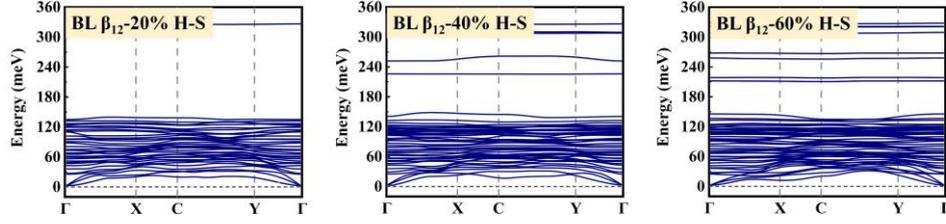
**Fig. S4** The result of SSAdNDP for BL  $\chi_3$ -37.5% H-I.

**Table S2** The electronic configurations of B atoms in the pristine and hydrogenated BL  $\chi_3$ -borophenes.

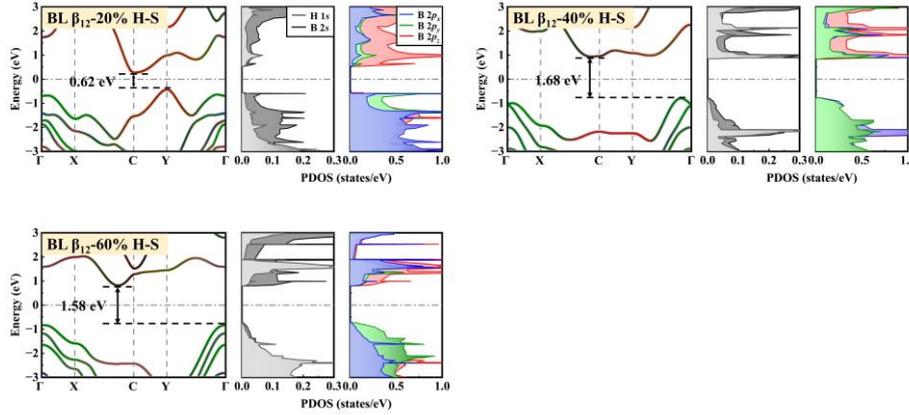
	The pristine BL $\chi_3$ -borophene	BL $\chi_3$ -12.5% H-I	BL $\chi_3$ -25% H-I
B <sub>1</sub>	$2s^{1.15}2p_x^{0.57}2p_y^{0.44}2p_z^{0.63}$	$2s^{1.16}2p_x^{0.67}2p_y^{0.51}2p_z^{0.63}$	$2s^{1.17}2p_x^{0.60}2p_y^{0.51}2p_z^{0.67}$
B <sub>2</sub>	$2s^{1.21}2p_x^{0.55}2p_y^{0.63}2p_z^{0.56}$	$2s^{1.20}2p_x^{0.56}2p_y^{0.56}2p_z^{0.54}$	$2s^{1.16}2p_x^{0.52}2p_y^{0.59}2p_z^{0.61}$
B <sub>3</sub>	$2s^{1.15}2p_x^{0.57}2p_y^{0.44}2p_z^{0.63}$	$2s^{1.12}2p_x^{0.56}2p_y^{0.48}2p_z^{0.47}$	$2s^{1.13}2p_x^{0.55}2p_y^{0.48}2p_z^{0.53}$
B <sub>4</sub>	$2s^{1.21}2p_x^{0.55}2p_y^{0.63}2p_z^{0.56}$	$2s^{1.23}2p_x^{0.52}2p_y^{0.56}2p_z^{0.59}$	$2s^{1.19}2p_x^{0.49}2p_y^{0.60}2p_z^{0.55}$
B <sub>5</sub>	$2s^{1.21}2p_x^{0.55}2p_y^{0.63}2p_z^{0.56}$	$2s^{1.18}2p_x^{0.54}2p_y^{0.46}2p_z^{0.70}$	$2s^{1.19}2p_x^{0.50}2p_y^{0.50}2p_z^{0.56}$
B <sub>6</sub>	$2s^{1.15}2p_x^{0.57}2p_y^{0.44}2p_z^{0.63}$	$2s^{1.09}2p_x^{0.62}2p_y^{0.50}2p_z^{0.50}$	$2s^{1.11}2p_x^{0.53}2p_y^{0.49}2p_z^{0.57}$
B <sub>7</sub>	$2s^{1.15}2p_x^{0.57}2p_y^{0.44}2p_z^{0.63}$	$2s^{1.17}2p_x^{0.59}2p_y^{0.52}2p_z^{0.62}$	$2s^{1.15}2p_x^{0.60}2p_y^{0.53}2p_z^{0.66}$
B <sub>8</sub>	$2s^{1.21}2p_x^{0.55}2p_y^{0.63}2p_z^{0.56}$	$2s^{1.22}2p_x^{0.58}2p_y^{0.60}2p_z^{0.51}$	$2s^{1.19}2p_x^{0.44}2p_y^{0.61}2p_z^{0.58}$
	<b>BL <math>\chi_3</math>-37.5% H-I</b>	<b>BL <math>\chi_3</math>-50% H-I</b>	<b>BL <math>\chi_3</math>-62.5% H-I</b>
B <sub>1</sub>	$2s^{1.11}2p_x^{0.57}2p_y^{0.52}2p_z^{0.54}$	$2s^{1.15}2p_x^{0.54}2p_y^{0.49}2p_z^{0.69}$	$2s^{1.12}2p_x^{0.60}2p_y^{0.47}2p_z^{0.50}$
B <sub>2</sub>	$2s^{1.20}2p_x^{0.45}2p_y^{0.62}2p_z^{0.55}$	$2s^{1.15}2p_x^{0.53}2p_y^{0.59}2p_z^{0.63}$	$2s^{1.19}2p_x^{0.59}2p_y^{0.51}2p_z^{0.65}$
B <sub>3</sub>	$2s^{1.15}2p_x^{0.63}2p_y^{0.54}2p_z^{0.67}$	$2s^{1.12}2p_x^{0.51}2p_y^{0.41}2p_z^{0.63}$	$2s^{1.14}2p_x^{0.61}2p_y^{0.58}2p_z^{0.66}$
B <sub>4</sub>	$2s^{1.19}2p_x^{0.49}2p_y^{0.54}2p_z^{0.60}$	$2s^{1.14}2p_x^{0.44}2p_y^{0.65}2p_z^{0.52}$	$2s^{1.15}2p_x^{0.59}2p_y^{0.61}2p_z^{0.66}$
B <sub>5</sub>	$2s^{1.10}2p_x^{0.35}2p_y^{0.70}2p_z^{0.56}$	$2s^{1.15}2p_x^{0.48}2p_y^{0.57}2p_z^{0.55}$	$2s^{1.15}2p_x^{0.58}2p_y^{0.63}2p_z^{0.68}$
B <sub>6</sub>	$2s^{1.14}2p_x^{0.64}2p_y^{0.53}2p_z^{0.68}$	$2s^{1.11}2p_x^{0.50}2p_y^{0.47}2p_z^{0.61}$	$2s^{1.12}2p_x^{0.64}2p_y^{0.49}2p_z^{0.69}$
B <sub>7</sub>	$2s^{1.13}2p_x^{0.41}2p_y^{0.64}2p_z^{0.55}$	$2s^{1.15}2p_x^{0.60}2p_y^{0.51}2p_z^{0.66}$	$2s^{1.10}2p_x^{0.62}2p_y^{0.50}2p_z^{0.55}$
B <sub>8</sub>	$2s^{1.11}2p_x^{0.53}2p_y^{0.47}2p_z^{0.65}$	$2s^{1.15}2p_x^{0.52}2p_y^{0.60}2p_z^{0.61}$	$2s^{1.12}2p_x^{0.51}2p_y^{0.62}2p_z^{0.59}$



**Fig. S5** The structures of the metastable hydrogenated BL (a)  $\beta_{12}$ - and (b)  $\chi_3$ -borophenes. The formation energies are also given.



**Fig. S6** The phononic energy bands of the semiconducting hydrogenated BL  $\beta_{12}$ -borophenes.



**Fig. S7** The electronic energy bands and PDOSs of the semiconducting hydrogenated BL  $\beta_{12}$ -borophenes calculated using the HSE06 method.

### Experimental feasible pathway

In fact, BL  $\alpha$ - and  $\beta_{12}$ -borophenes and hydrogenated ML  $\beta_{12}$ -borophene have been synthesized experimentally (see refs. 17, 18 and 36 in the manuscript). Here, we proposed a possible experimentally feasible pathway for achieving hydrogenated BL borophenes through a two-step approach, involving B deposition and hydrogenation under ultrahigh-vacuum conditions.

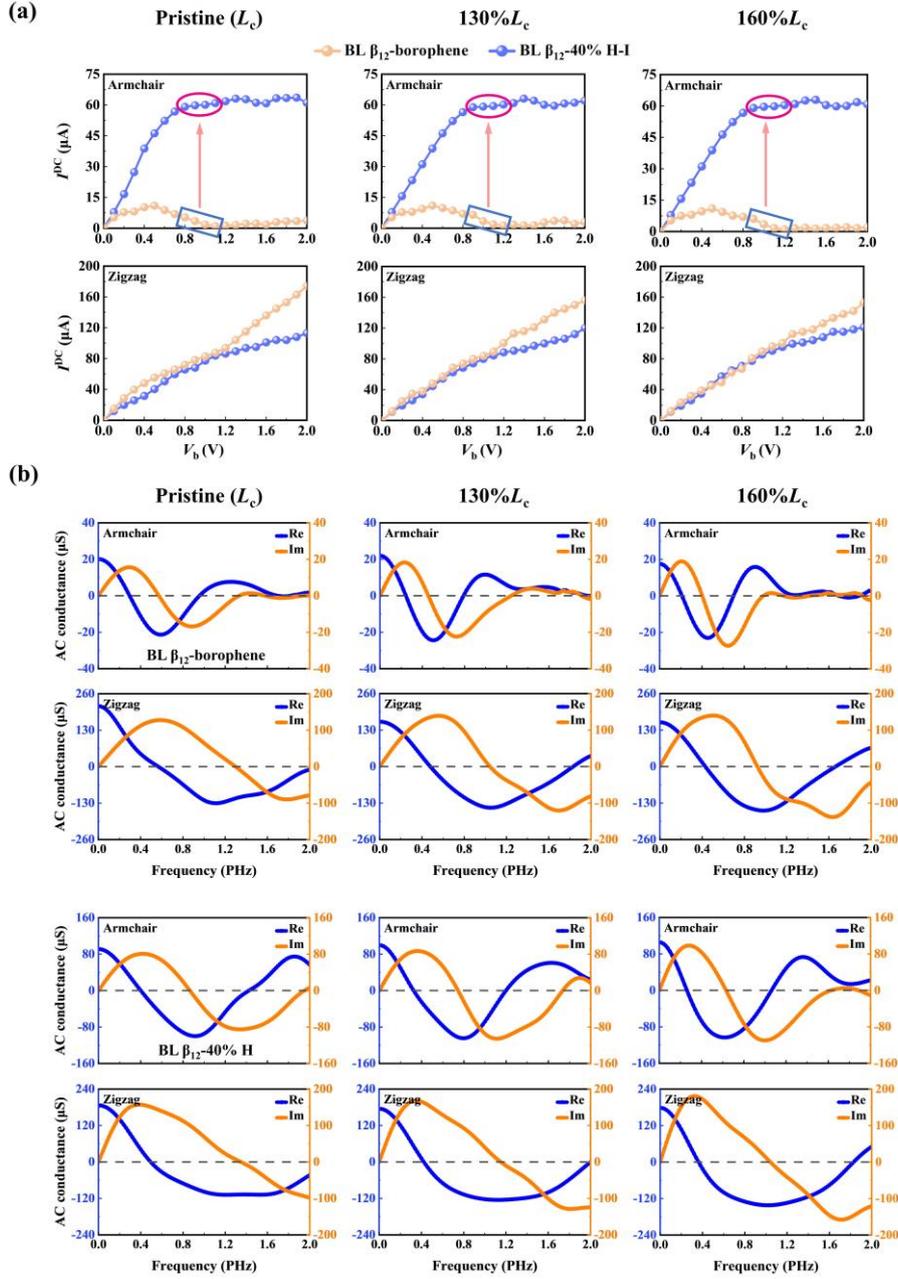
First, BL borophene can grow on Ag(111) surface using ultrahigh-vacuum molecular beam epitaxy technology, where high-purity B is evaporated onto a clean Ag(111) surface via electron beam evaporation. During deposition, the temperature should be maintained within an optimal range, and the substrate must be sufficiently large and flat to provide adequate electrons for stabilizing the BL structures, preventing excess B from aggregating into clusters.

Second, subsequent hydrogenation can achieve by exposing the BL borophene

samples to H atoms, which can be generated by dissociating H<sub>2</sub> molecules with a hot tungsten filament. The hydrogenation process should incorporate independent flow controllers to regulate the H content, enabling continuous and precise control of H content to achieve different H coverages. Additionally, the exposure time of the BL borophene samples should be extended to ensure sufficient H doping. The hydrogenation temperature must be carefully controlled to facilitate in-situ hydrogenation while avoiding H<sub>2</sub> release caused by excessive temperature. Of course, specific experimental parameters should be carefully adjusted according to actual conditions.

**Table S3** The critical points of the pristine and hydrogenated BL structures. The unit is PHz.

Armchair/Zigzag	1st	2nd	3rd	4th	5th
BL $\beta_{12}$ -borophene	0.57/1.29	1.33/—	1.58/—	1.96/	—/—
BL $\beta_{12}$ -10% H-I	0.57/0.68	—/1.43	—/—	—/—	—/—
BL $\beta_{12}$ -20% H-I	0.87/0.98	—/—	—/—	—/—	—/—
BL $\beta_{12}$ -30% H-I	0.55/1.31	1.62/—	—/—	—/—	—/—
BL $\beta_{12}$ -40% H-I	0.87/1.35	1.93/—	—/—	—/—	—/—
BL $\beta_{12}$ -50% H-I	0.79/1.36	1.93/—	—/—	—/—	—/—
BL $\beta_{12}$ -60% H-I	0.72/0.58	—/1.71	—/—	—/—	—/—
BL $\chi_3$ -borophene	1.13/1.15	—/—	—/—	—/—	—/—
BL $\chi_3$ -12.5% H-I	0.82/1.38	1.82/—	—/—	—/—	—/—
BL $\chi_3$ -25.0% H-I	0.36/1.23	0.83/—	1.00/—	1.34/—	1.71/—
BL $\chi_3$ -37.5% H-I	0.63/0.87	0.84/—	1.89/—	—/—	—/—
BL $\chi_3$ -50.0% H-I	1.13/1.05	1.63/—	1.83/—	1.94/—	—/—
BL $\chi_3$ -62.5% H-I	1.02/0.85	1.69/1.86	1.75/—	—/—	—/—
BL $\beta_{12}$ -20% H-S	0.23/0.73	0.44/1.11	0.83/1.51	0.94/1.78	1.19/1.88
BL $\beta_{12}$ -40% H-S	0.21/0.22	0.42/0.42	0.63/0.96	0.79/1.29	0.91/1.57
BL $\beta_{12}$ -60% H-S	0.23/0.22	0.44/0.44	1.03/0.57	1.09/0.86	1.54/1.07



**Fig. S8** (a) The  $I^{DC}$ - $V_b$  curves and (b) AC conductances of the nanodevice models based on BL  $\beta_{12}$ -borophene and BL  $\beta_{12}$ -40% H-I with different lengths.

To check the length effect, using BL  $\beta_{12}$ -borophene and BL  $\beta_{12}$ -40% H-I as two examples, we expanded the corresponding lengths ( $L_c$ ) of the nanodevices to  $130\%L_c$  and  $160\%L_c$ . The  $I^{DC}$ - $V_b$  curves and AC conductances of the pristine ( $L_c$ ),  $130\%L_c$  and  $160\%L_c$  structures are given in Fig. S8. From the figure one can see that the influences of lengths of the nanodevice are limited and the DC and AC transport phenomena (*e.g.*, SDCC effect) are robust.