

Supporting Information for

Interface Coupling and Charge Doping in Graphene on Ferroelectric

$\text{BiAlO}_3(0001)$ Polar Surfaces

Jin Yuan, Jian-Qing Dai,* Cheng Ke, and Zi-Cheng Wei

Faculty of Materials Science and Engineering, Kunming University of Science and
Technology, Kunming 650093, P. R. China

*Corresponding author. Fax: +86 871 65107922.

E-mail address: djqkust@sina.com (J.-Q. Dai).

ORCID: 0000-0003-4352-0789 (Jian-Qing Dai)

1. Effect of tensile strain of the $\text{BAO}(0001)$ surfaces

For the Gr/BAO(0001) heterostructures, the BAO(0001) surfaces suffer from about 6% tensile strain. The effect of this tensile strain on BAO(0001) surface electronic structure and electrostatic charge doping in the graphene channel should be addressed. The calculated band structure of clean $\sqrt{3}\times\sqrt{3}$ BAO(0001) Z^\pm surfaces without strain and with 6% tensile strain are shown in Fig. S1. For the BAO Z^+ and Z^- surface, we find that the Fermi level is located at VBM and CBM, respectively. Obviously, this result is consistent with our previous prediction in Ref. [J. Phys. Chem. C, 122 (2018) 29220-29227], i.e., the BAO Z^+ surface shows insulating feature while the BAO Z^- surface is characterized by a weak n -type surface metallization. Due to the tensile strain, the band gap of the BAO Z^+ surface is reduced from 2.24 eV to 1.87 eV (Fig. S1a and S1b). In view the fact that the electrostatic doping in Gr/BAO Z^+ is due to electrons transferring from graphene to the VBM of BAO Z^+ surface, so we expect the reduction of band gap has negligible influence on the charge doping behavior. As regard to the BAO Z^- surface, the effect of the tensile strain on the band gap is negligible (Fig. S1c and S1d). In addition, as pointed out in Ref. [J. Phys. Chem. C 122 (2018) 29220-29227], both the clean BAO Z^+ and Z^- surfaces show almost vanishing surface charges due to the combination of

different polarity compensation mechanisms.

Another important aspect to identify the effect of tensile strain on charge doping in the graphene channel is the band offset between clean BAO(0001) surfaces and the pristine graphene. As shown in Fig. S2, we find that the influence of tensile strain of the BAO(0001) surfaces on the band offset with respect to pristine graphene is very small. The change of band offset for BAO Z+ and Z- surface is only 0.08 eV and 0.02 eV, respectively. Therefore, the influence of this tiny change in band offset on the charge doping behavior is also expected to be negligible. In other words, although a tensile strain of $\sim 6\%$ is employed in the BAO(0001) surfaces for the Gr/BAO(0001) heterostructures, all the results and conclusions remain valid even if for the BAO(0001) surfaces without any strain.

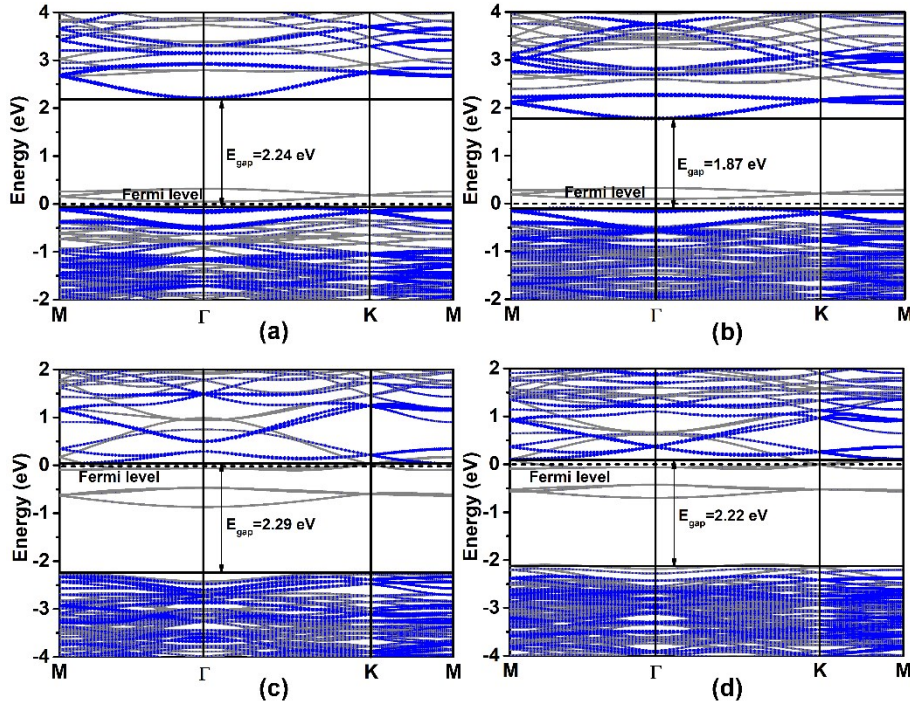


Fig. S1 Band structure along high-symmetry directions of the supercell Brillouin zone for (a, c) clean BAO Z+ and Z- surfaces with zero strain, and (b, d) clean BAO Z+ and Z- surfaces with $\sim 6\%$ tensile strain. The blue circles represent projected band of the surface termination plus one underlying Al-O₃-Bi trilayer.

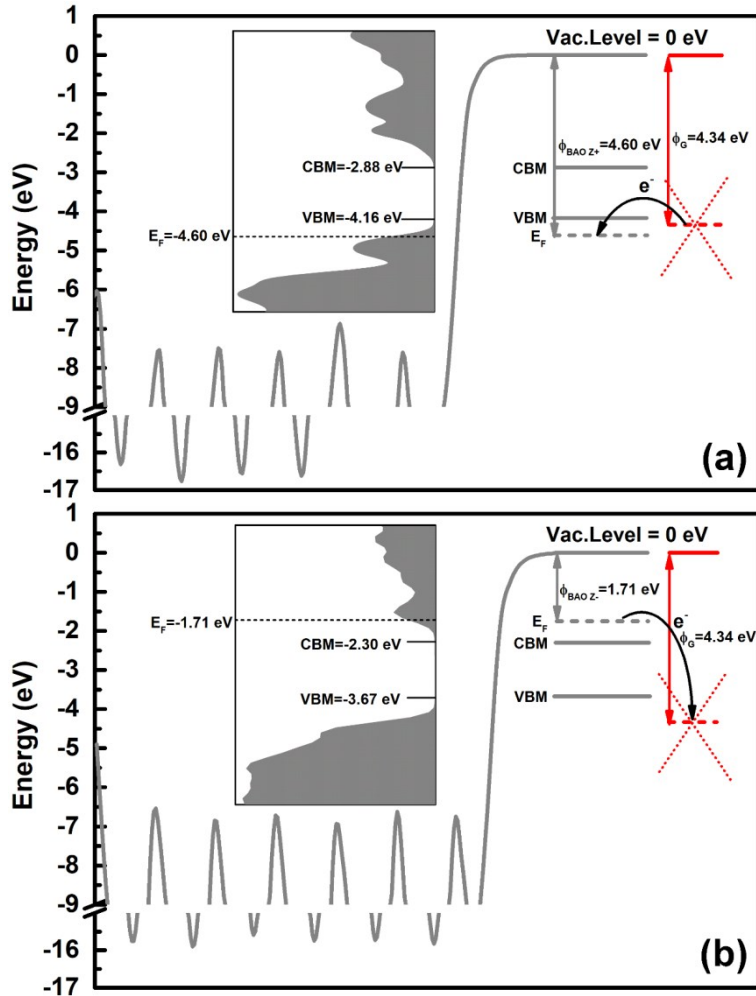


Fig. S2 The plane-averaged electrostatic potential and band arrangement between graphene and the clean (a) BAO Z⁺ surface and (b) BAO Z⁻ surface with zero strain. The gray curves denote the BAO Z[±] surface while the red short dash line indicates the Fermi level of the pristine graphene. Insets in Fig. S2a and S2b show surface DOS of the clean BAO(0001) slab. The vacuum level is set to 0 eV. The gray solid lines represent the CBM and the VBM. The work functions of pristine graphene (ϕ_G), clean BAO Z⁺ ($\phi_{\text{BAO Z}^+}$) and Z⁻ ($\phi_{\text{BAO Z}^-}$) surfaces are also shown.

2. Other supporting results

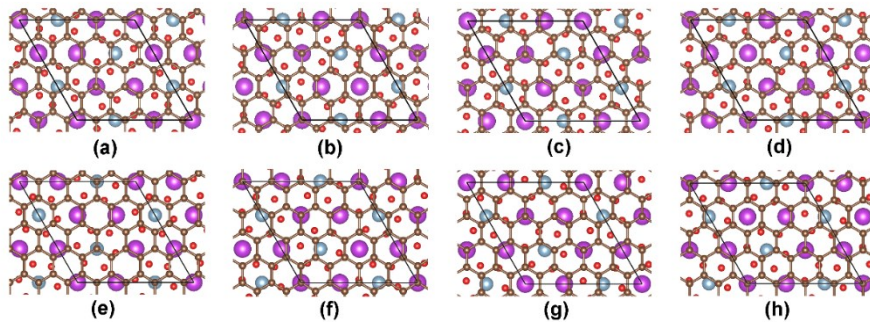


Fig. S3 Four representative interfacial configurations for (a-d) Gr/BAO Z⁺ and (e-h) Gr/BAO Z⁻ hybrid systems. For clarity, only graphene and the outer one -Al-O₃-Bi trilayer plus surface

termination are shown. The symbols of Bi, Al, O and C atoms are shown in Fig. 1.

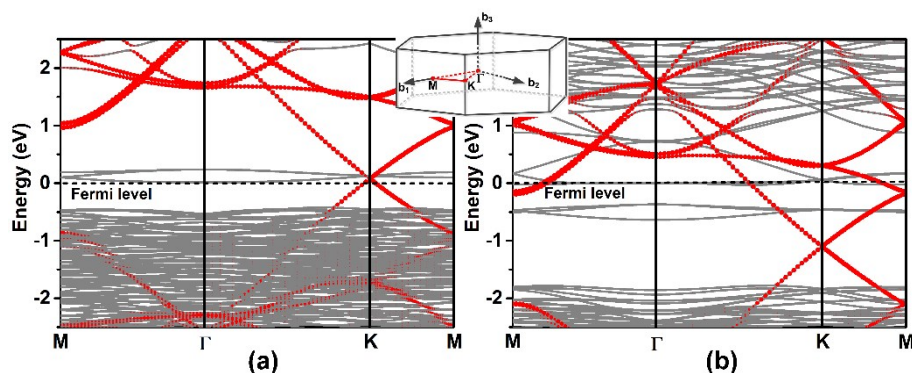


Fig. S4 Band structure along high-symmetry directions of the supercell Brillouin zone for (a, b) Gr/BAO Z+ and Z- hybrid systems. The red circles represent projected band of C $2p_z$ orbitals in the graphene layer. Inset shows the First Brillouin zone of the hexagonal supercell.

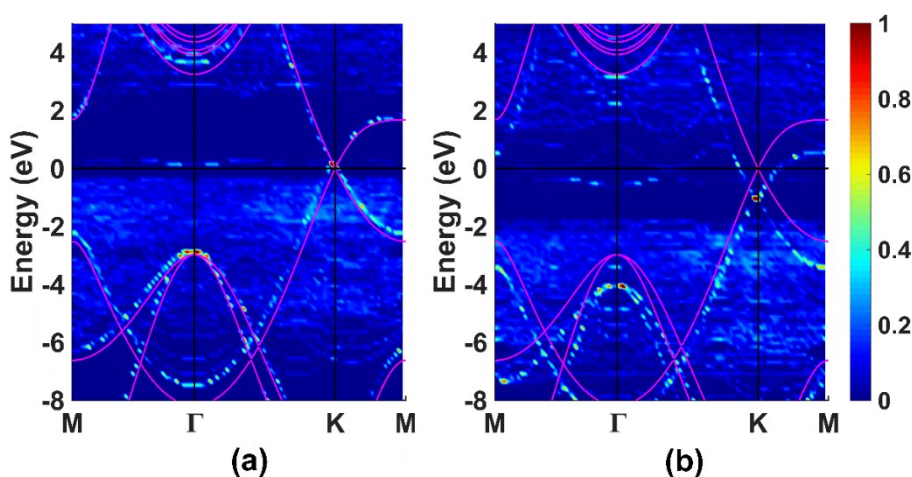


Fig. S5 Unfolded EBS of graphene along high-symmetry direction of the primitive-cell Brillouin zone for (a) Gr/BAO Z+ and (b) Gr/BAO Z- hybrid systems. The color scale corresponds to the number of unfolded primitive-cell bands. Note that the band structure of pristine graphene is plotted by the purple curves.

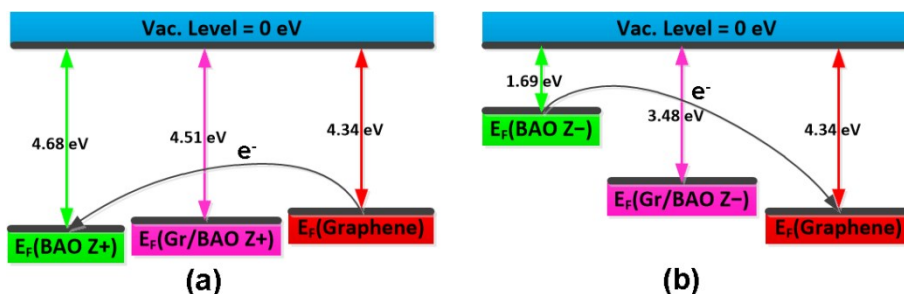


Fig. S6 Comparison of the Fermi level among the clean BAO(0001) surface, the pristine graphene, and the Gr/BAO(0001) hybrid system. (a) Gr/BAO Z+ system; (b) Gr/BAO Z- systems. Note that the vacuum level is set to 0 eV.

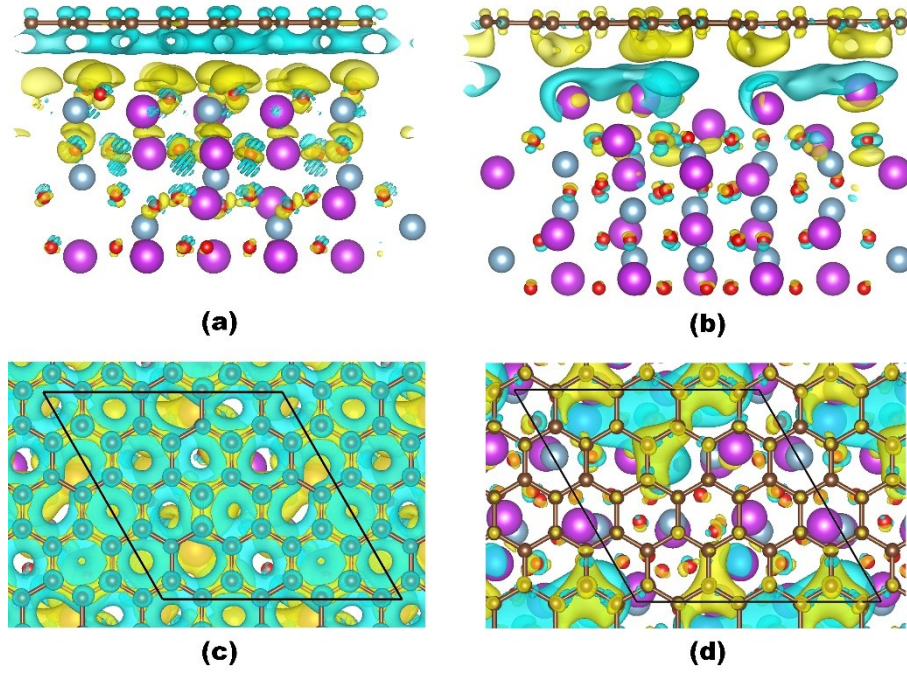


Fig. S7 The differential charge density of polarization-reversed Gr/BAO(0001) systems, where pale blue and yellow indicates the gain of holes and electrons, respectively. (a, b) Side-views of Gr/BAO $Z+\downarrow$ and Gr/BAO $Z-\uparrow$ interfaces, respectively. (c, d) Top-views of Gr/BAO $Z+\downarrow$ and Gr/BAO $Z-\uparrow$ interfaces, respectively. Maps are plotted for isosurfaces of $5 \times 10^{-4} e/\text{\AA}^3$. The symbols of Bi, Al, O and C atoms are shown in Fig. 1.

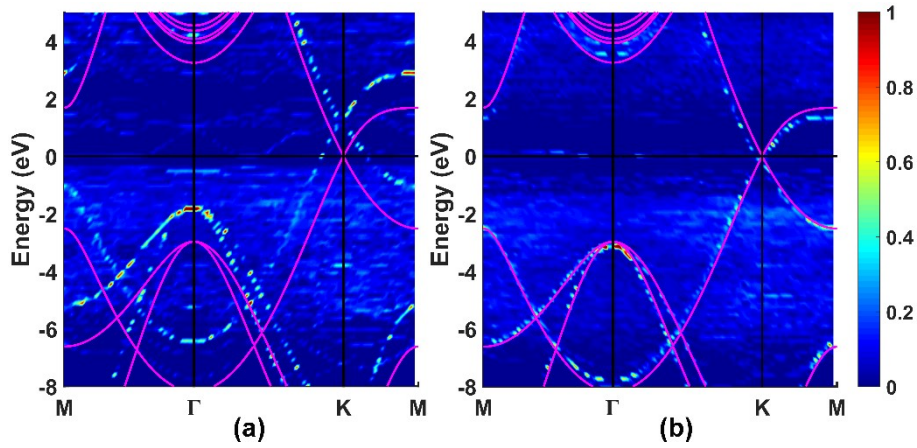


Fig. S8 Unfolded EBS of graphene along high-symmetry direction of the primitive-cell Brillouin zone for (a) Gr/BAO $Z+\downarrow$ and (b) Gr/BAO $Z-\uparrow$ hybrid systems. The color scale corresponds to the number of unfolded primitive-cell bands. Note that the band structure of pristine graphene is plotted by the purple curves.

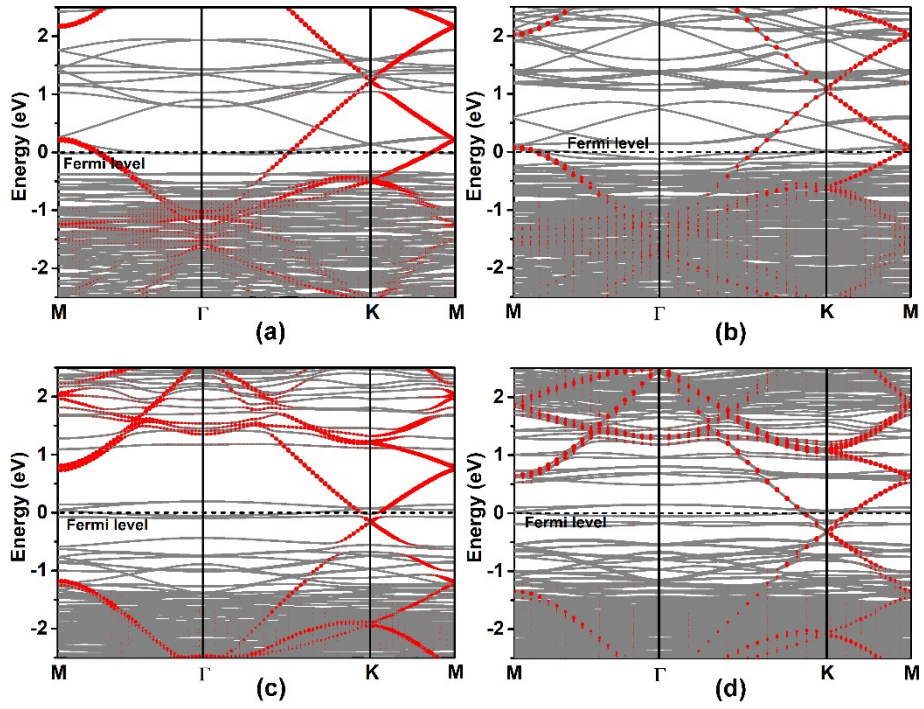


Fig. S9 Effect of SOC on the band structure in supercell Brillouin zone for the polarization-reversed Gr/BAO(0001) systems. (a, c) Gr/BAO $Z+\downarrow$ and $Z-\uparrow$ hybrid systems without the SOC; (b, d) Gr/BAO $Z+\downarrow$ and $Z-\uparrow$ hybrid systems with the SOC. The red circles represent projected band of C $2p_z$ orbitals in the graphene layer.