

To the editor – In the article “Substrate-induced bandgap opening in epitaxial graphene” [1], Zhou *et al.* assert that chemical bonds to the substrate break the “A-B” symmetry of the graphene lattice, opening a gap in the bands near the Dirac energy E_D . This contradicts our observation of a kink at E_D related to electron-plasmon scattering [2], a conclusion supported by doping studies and by theory [3, 4]; it also contradicts STM measurements and theory which find no such gap [5, 6]. Zhou also asserts that gaps observed in multilayer graphene are dominated by this same substrate effect, and not by the electric field across the film as proposed by Ohta *et al.* [7, 8]. In advancing these claims, Zhou *et al.* have misrepresented our MDC self-energy analysis [2] as naively including artifacts from tails of far-away EDC peaks. Actually, our MDC and EDC peaks always coincide as a consequence of our self-consistent treatment [9] and careful alignment.

We already presented a substantial case against substrate-induced gaps in graphene [9] for our samples [2, 9], which were characterized by electron microscopy and STM studies to have large, uniform graphene terraces. In particular, the strong intensity anisotropy of the Fermi surface (Fig. 1a, upper) imposes a strict limit on the gap due to A-B symmetry breaking (from any source) to a value much smaller than Zhou’s observation. To this we add that the weak asymmetry of the satellite bands at E_D offered by Zhou in support of their symmetry breaking is absent in our samples (see Ref. [9] Fig. 4).

In our view, Zhou’s results are not intrinsic to single-layer graphene and have a simple, intuitive explanation. We find that Zhou’s bandstructure (Fig. 1b) has much less in common with that of our optimized samples (Fig. 1a) than an islanded graphene sample readily prepared by underannealing (Fig. 1c), displaying not only the “gapped” EDC spectrum at K and ~ 50 meV shift of E_D reported by Zhou but also a significant broadening and an elevated diffuse background in the Fermi level MDCs (Fig 1d). That such samples are islanded is supported by the enhanced diffuse hexagonal emission pattern at E_D —attributed to exposed regions of the carbon-rich interface layer [9]—in Zhou’s (Ref. [1], Fig. 4b) and in our islanded (Fig. 1c) samples compared to our optimized samples (Ref. [9], Fig. 4) as well as by electron microscopy showing an inhomogeneous distribution of small irregular graphene islands. There may also be defects within the graphene islands: our ideal samples have very few defects visible in STM [5] unlike samples studied by Rutter *et al.* [10], suggesting that such defect formation is sensitive to sample preparation.

These features can easily explain Zhou’s data: the edges of irregular islands and presence of point defects naturally break the A-B atom symmetry, increase the diffuse scattering background, cause spectral broadening, alter the doping level, and induce an inhomogeneously broadened gap at E_D . More subtle properties such as the intrinsic self-energy due to electron-plasmon or electron-phonon coupling [2] and the layer-dependent charge and out-of-plane screening length [8] cannot easily be determined from such samples.

We agree that a gap was induced in Zhou’s graphene samples, but not by substrate bonding as claimed, but instead by modulation of its lateral structure, a more promising route to new devices.

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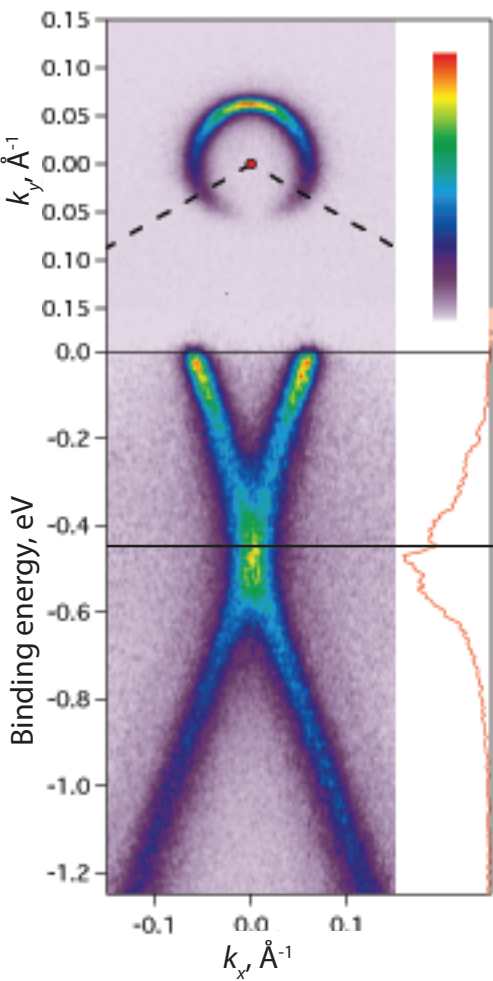
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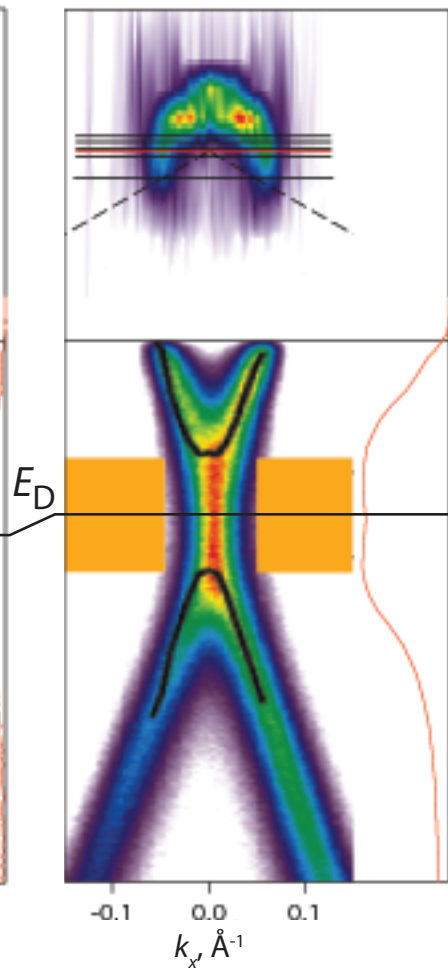
Figure 1. (a,b) Comparison of Fermi surfaces (at $E=E_F$), band structures (at $k_y=0$) and EDC energy spectra (red curves at K, i.e. $k_x=k_y=0$) of graphene near K by Bostwick [2, 9] and Zhou [1], taken in the same experimental geometry. As the images are plotted with the same color scale, differences in linewidth are readily apparent. The orange region in (b) represents the energy gap reported by Zhou. (c) Momentum distribution map at $E=E_D$, the associated bandstructure (along the double yellow arrow through K), and the EDC at K for an underannealed, islanded sample showing spectral features similar to Zhou's. (The difference in intensity of the two arms arises from the different detector geometry used.) (d) Comparison of MDC spectra at $E=E_F$ (open symbols) and their decomposition into gaussian-broadened lorentzians (for the main peaks, which correspond to the Fermi level crossings) and gaussians (for the central, diffuse peak which fills in the Dirac cone) from Bostwick[2, 9] and Zhou[1], resp. The spectra are both normalized to unit area, and reveal the increased broadening and diffuse intensity attributable to defect scattering in Zhou's samples.

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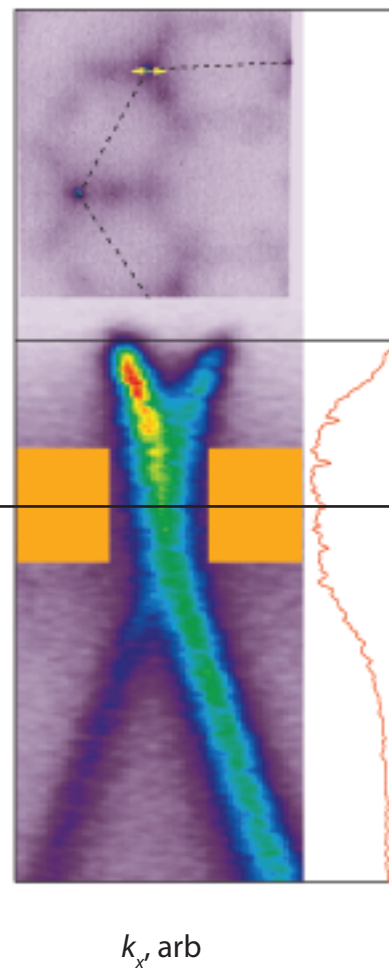
(a) *Bostwick et al.*



(b) *Zhou et al.*



(c) *Bostwick et al. islanded*



(d)

