Supplementary Material for

Spectroscopic evidence for electron correlations in epitaxial bilayer graphene with interface-reconstructed superlattice potentials

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SUPPLEMENTARY TEXT

I. Discussion About Mott Correlated Phase as the Candidate Origin of Detected Spectral-Weight Transfer

In the Mott–Hubbard model for *d*-electron systems, the interplay of on-site Coulomb energy U, kinetic energy U_t, bandwidth W, and filling factor v cooperatively drives the Mott-insulator transition [1]. The strong electron correlations are set by the high ratio of the on-site Coulomb interaction U to the bandwidth W of the flat band. In the Mott–Hubbard scenario, the strong on-site Coulomb repulsion forbids neighboring electron hopping; further at half-filling, Mott insulator occurs as the ground state with a correlated charge gap between the 'splitted' upper and lower Hubbard bands derived from the *d*-electron band [1].

Experimentally, in the doped Mott phase of cuprates, as increasing the doping level, the spectral weight transfers from Hubbard bands to charge-transfer gap near E_F as in-gap excitations [2,3]. These excitation modes 'straddle' the simultaneously formed sharp V-shaped gap at E_F. With further carrier doping towards the underdoped superconducting (SC) regime, the V-shaped gap can finally evolve into the SC gap [3]. The resemblance between our result and such spectral-weight transfer in a doped Mott insulator indicates the Mott correlation as a likely explanation.

In the candidate Mott picture for our BLG, the Dirac-type conductance dip, where the CNP is located and where the LB/UB overlap, should be a correlated gap separating the Hubbard bands by U in essence. Physically, the relative spectral weights of LB and UB flanking the correlated gap sensitively depend on the local carrier density, naturally reconciling the observed spectral-weight transfer behavior [Fig. 3(d)].

II. Electron Correlations Arising from Electron–Plasmon Coupling

We now discuss the possible concrete mechanism responsible for the detected electron correlations. Theoretically, plasmarons have been proposed in 2D electron gas defined as the resonantly coupled electron–plasmon composite excitations [4]. In the spectral function $A(\boldsymbol{k},E)$ and the LDOS spectra, the signal of the plasmaron appears as a satellite peak aside from the most tightly bounded conventional quasiparticle peak (main peak) [4,5]. By increasing chemical doping and/or disorder, the plamaron signature is expected to be suppressed [4,5].

For graphene, previous study shows that the carrier decaying via plasmon scattering is a well-defined quasiparticle only outside the region of kinematically allowed electron–hole excitations in $(E,\boldsymbol{k})$ phase diagram [6]. This constraint causes that the states near Dirac point interact strongly with the plasmons [Fig. 3(d)], meaning that the plasmarons can have a large influence on the many-body effect around $E\sim E_D$ [6,7]. Consistently, in experiments by angle-resolved photoemission spectroscopy (ARPES), the lower-energy plasmaron satellite band and the higher-energy main band straddling the Dirac point far below E_F (~−0.5 eV) have been detected in the spectral function of quasi-free-standing graphene, where the plasmaron band appears suppressed by higher chemical doping [8]. Furthermore, while the interactions between quasiparticles and plasmons are stronger in the 2D massless Dirac system than in an ordinary 2D parabolic-band system [7], the plasmaron in a 2D massive-electron system (e.g. GaAs quantum well) is still observed by tunneling spectroscopy [9]. All these arguments suggest that, the LB near Dirac energy in our BLG, which behaves as the satellite band of UB and is suppressed as increasing electron doping (more negative eV_D) [Figs. 3(a)–3(c)], can be

interpreted as the plamaron band. Equivalently speaking, the electron correlations probably arise because of the plasmon emissions for electrons near the Dirac point [Fig. 3(d)], i.e., the plasmarons consisting of resonantly bounded electrons and plasmons.

Compared with the plasmaron peak in quasi-free-standing graphene directly grown epitaxially on SiC [8], the plasmaron band in ordinary epitaxial graphene on buffer layer/SiC is normally not well resolvable in the ARPES-measured spectral function due to the high dielectric screening (with effective dielectric constant ε 3–4 times higher), which is likely further damped by the disorder in the sample. Thus, the spectral-weight transfer indicated by STS provides an alternative method to effectively reveal the many-body effect induced by the electron–plasmon coupling in ordinary epitaxial graphene.

Note that the plasmon emission occurs for the states near E_D [6]. The gap opening at E_D for epitaxial BLG would weaken the plasmaron response. Furthermore, the plasmaron band usually occurs at an energy roughly proportional to E_F or carrier density [8,10]. Yet, the energies of LB and UB here appear approximately unchanged with doping as shifting E_F (such absence of rigid following for LB and UB is also an indication of the existence of electron correlations) [Fig. 3(a)], which is left for future explorations for a full understanding within the plasmaron scenario.

SUPPLEMENTARY FIGURES

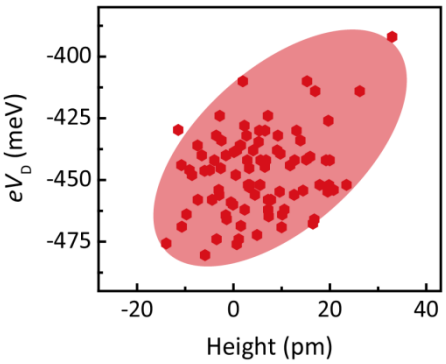

FIG. S1. Statistical correlation between eV_D and height. The data point are extracted from Figs. 2(b), 2(d) and 2(f), appearing positively correlated more clearly in statistics. The shaded ellipse is the guide to the eyes of the statistical positive correlations between eV_D and height.

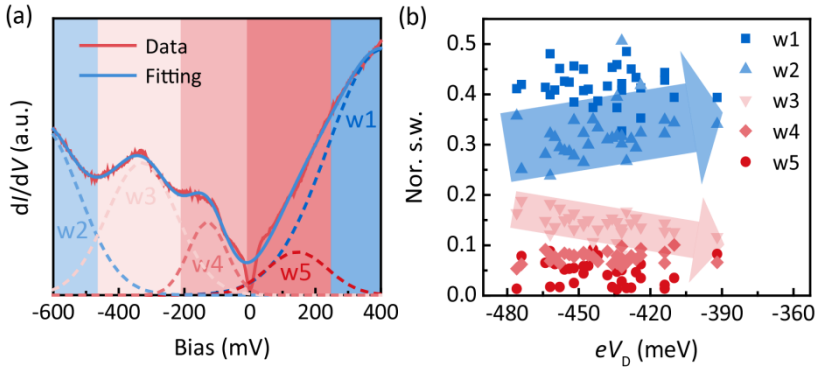

FIG. S2. Full information of the multi-Gaussian fit. (a) Exemplifying multi-Gaussian fit (solid blue curve) to the tunneling spectrum (solid red curve). The 'phonon' gap is ignored in the fit procedure. The involved five Gaussian components with integrated spectral weights w1–w5 are shown in dashed curves. Note that w2 (w3) is equal to w_{LB} (w_{UB}) as defined in main text. (c) Normalized spectral weights (s.w.) w1–w5 extracted from the spectra in (a) plotted vs. eV_D. Clearly, while w2 and w3 show positive and negative correlations with eV_D, respectively, w1, w4, and w5 behave eV_D-independent statistically, highlighting the truly existent spectral-weight transfer between w2 and w3.

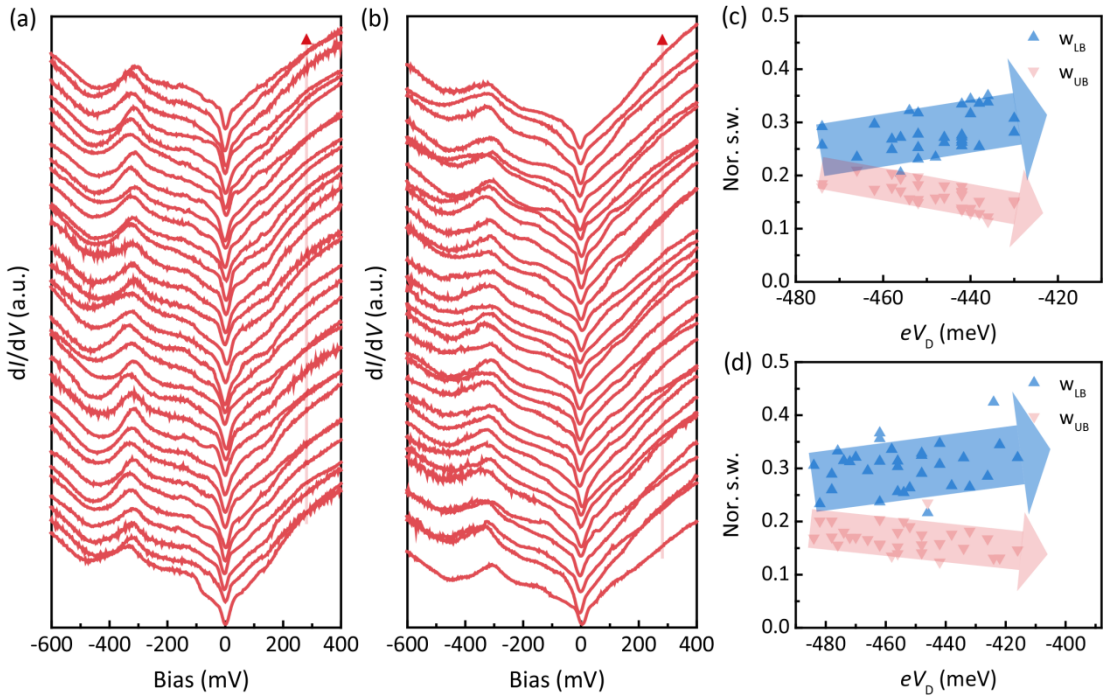

FIG. S3. Reproducibility of the spectral-weight transfer in BLG. (a,b) Tunneling spectra (vertically offset for clarity) taken along the arrows in Figs. 2(a) and 2(e), respectively. (c,d) Normalized spectral weights w_{LB} and w_{UB} extracted from the spectra in (a,b), respectively, plotted vs. eV_D.

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