## An in-situ Annealing Study of Interlayer Conduction in Twisted Bilayer Graphene

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The interlayer transport between individual layers of 2D materials is an ideal subject for studying the fundamental aspects of charge conduction in layered materials [1-2]. We report on the experimental observation of incoherent conduction in twisted bilayer graphene and a significant reduction in the interlayer resistance by applying an in-situ annealing process. This process enables the reduction of wrinkles and improves homogeneities in the interlayer, thus lessen the distance between the bottom and top graphene layers.

The interlayer transport measurements of the graphene crossbar device were done using the four-point resistance technique. We apply the current between the electrodes of the bottom and top layers ( $V_s$  and  $V_D$ ) and measure the interlayer voltage drop across the other two branches ( $V_L$  and  $V_H$ ) as shown in the Fig. 1(a). As the fabricated device was heavily P-doped, the interlayer resistance before annealing was in the range of k $\Omega$  (Fig. 1(b)). Next, the device was annealed in vacuum for 2 hours at 200°C, followed by hydrogen annealing for 2.5 hours at 275°C (ambient pressure). Figure 1(c) shows the interlayer resistance after in-situ annealing, which shows clearly that the charge neutrality point is shifted to 0 volts. Furthermore, the clear temperature dependence is observed, as well as a resistance reduction by one order of magnitude. The interlayer resistance of the device before and after the in-situ annealing is shown as a function of temperature in Fig. 1(d). The metal-insulator-metal junction model can describe the interlayer transport between the bottom and top graphene layers. The interlayer tunneling occurs between graphene layers through a potential barrier of height *U* and width *w*. The barrier width *w* determines the tunneling probability between the Fermi surfaces of the bottom and top graphene layers. The effective barrier width *w*, i.e. the interlayer distance, is reduced by in-situ annealing. This is attributed to the effective reduction of wrinkles and inhomogeneities at the interlayer junction due to the in-situ annealing.



**Figure 1.** (a) Interlayer resistance measurement setup. (b) Interlayer resistance as function back gate (as a fabricated sample). (c) Interlayer resistance after in-situ annealing. (d) Interlayer resistance as a function of temperature.

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## References

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