Supporting Information

Resistivity of Rotated Graphite-Graphene Contacts

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S1. Metal-graphite contact resistance

It is important for the practical use of graphite-graphene contacts that the corresponding metal-graphite contact resistance be negligibly small in comparison. To evaluate metal-graphite contact resistance, we employ the conventional transmission length method (TLM) to extract the metal-graphite contact resistance. The graphite is exfoliated on 285-nm SiO_2 on Si substrates and then etched into a 2-µm-wide bar. E-beam lithography is used to define the contact regions with 1, 2, 4, 8, and 12 µm spacing. We define this spacing as the channel length, L_{ch}. Furthermore, we fabricate two sets of TLM contacts, one with a contact length (L_C) of 1 µm and another with a L_C of 2 µm. Prior to metallization, the sample is exposed to an O_2 plasma for 30s to remove any resist residue. We then evaporate 1 nm/50 nm Cr/Au and liftoff in acetone for at least 8 hours.

Electrically, we make two-terminal resistance (R_{2P}) measurements between adjacent contacts. The two-terminal resistance scales linearly with the channel length:

$$R_{2P} = \frac{L_{ch}}{W}\rho_G + 2R_C$$

Fig. S1 shows a plot of R_{2P} as a function of channel length for both contact lengths. Fitting a line to this data yields an extracted R_C of 3.3 Ω and 2.6 Ω for an L_C of 2 µm and 1 µm, respectively. The lack of contact length scaling in this data suggests that both contacts are longer than the transfer length of the metal-graphite contact. The contact resistivity of only 5 – 6 Ω-µm is significantly less than the 100 Ω-µm to 400 Ω-µm graphite-graphene contact resistances shown in Fig. 2.

With respect to the crystal orientation measurement shown in Fig. 3, we can also use the graphite resistivity extracted from the linear fits of the TLM data to estimate the resistance contribution of the graphite across the narrow 300 nm gap. Depending on the angle of the graphite contact, this contribution will vary from1 to 6 Ω, due to geometrical factors.

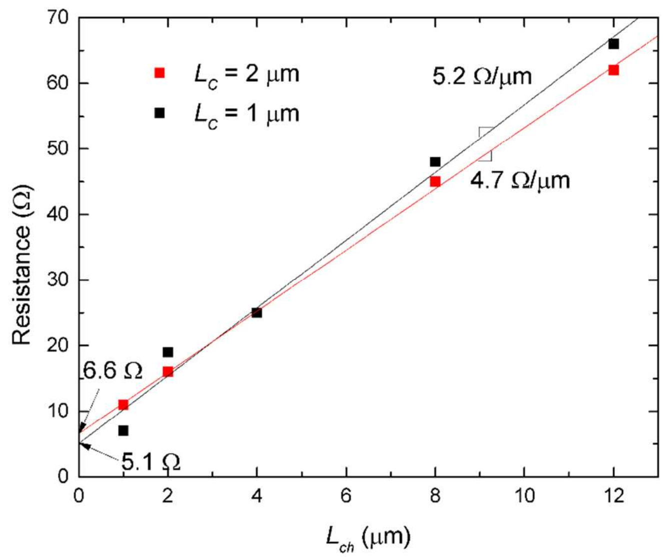

Fig. S1. Transmission length method of metal-graphite contacts. Two contact lengths were measured, 1 μm (black squares) and 2 μm (red squares). Linear fits show a similar contact resistance for both contact lengths of 5.1 Ω and 6.6 Ω for an L_C of 1 μm and 2 μm, respectively. The bulk graphite sheet resistance can be extracted to be 9.9 Ω/□ ± 0.5 Ω/□.

S2. Device Fabrication

Bulk graphite and monolayer graphene crystals are mechanically exfoliated onto SiO_2 substrates. The crystals are initially characterized by atomic force microscopy (AFM) (Fig. S2) to ensure there are no defects (such as cracks, wrinkles, or residues) and no step edges. By using clean crystals and using our previously developed fabrication techniques (see reference 5 in the main text), we ensure that the crystals are atomically smooth and that the graphene is in contact with the same atomic planes of the graphite in each contact (as in Fig. 2) and at each rotation angle (as in Fig. 3). Consequently, the devices in Fig. 2 exhibit identical crystal orientations since they are all fabricated from the same constituent crystals.

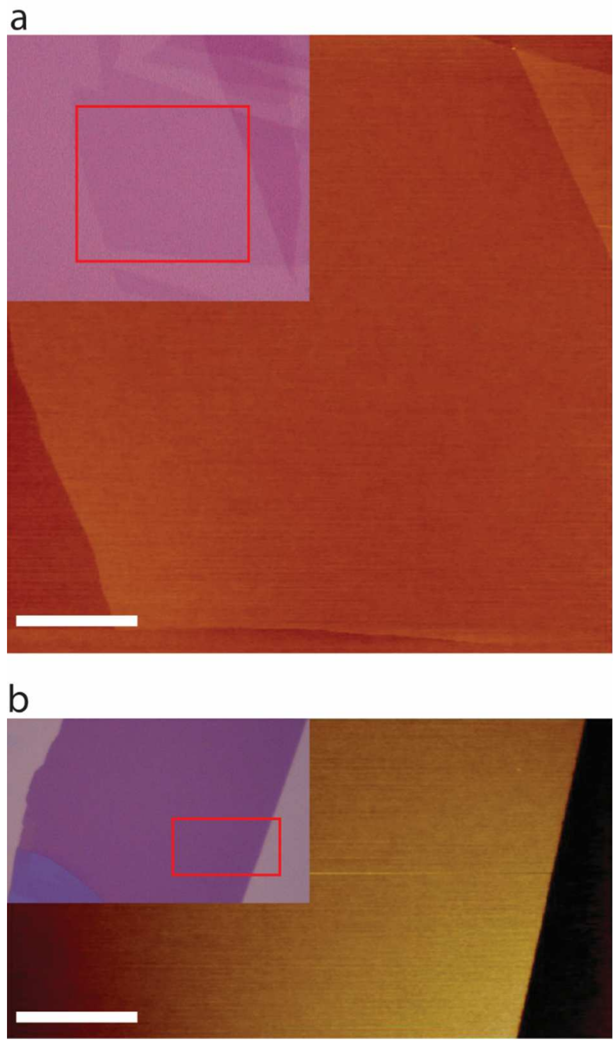

Figure S2 Typical AFM images of graphene (a) and graphite (b) crystals from mechanical exfoliation. The scale bar of each AFM image is 4 μm. Inset: Optical image of the respective crystals with an outline of the area measured by AFM.

S3. Measuring the relative crystal orientation

To measure the relative angle between the graphite contact and the graphene strips, we measure the angle of each (relative to the AFM image) and calculate the difference. For example, Fig. S3 shows an AFM image of the 60° rotation. We first measure the angle of the graphene, θ_g, which in this case is 24°. The angle of the graphite contact, θ_G, is measured by drawing a line between the mid points of two opposite extremities of the cross (as shown in Fig. S3). The same two points are used after every rotation to ensure a consistent calculation. In this example, the measured graphite angle is 84°. We then calculate the difference: $\theta = \theta_G - \theta_g$ where θ is the relative angle.

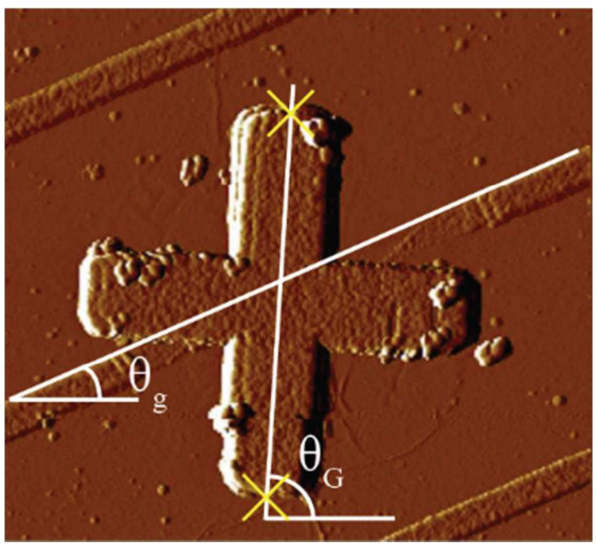

Fig. S3: Angle measurements. First we measure θ_g directly from the AFM image. Then we draw a line between the same two points (yellow crosses) on the graphite contact and measure θ_G. The relative angle, θ, is the difference between these angles. We always use the same two points on the graphite cross to measure θ_G.

S4. Rotating the graphite contact

The AFM used in this experiment is the Dimension Icon from Bruker with OTESPA probes. We employ a software package from Bruker called Nanoman in which imaging is performed in tapping mode and the physical manipulation of the contact is performed in contact mode. First, a tapping mode image is made. Then, we draw a linear vector on the tapping mode image to indicate where and in what direction the AFM probe will move. The AFM will then engage in contact mode and move the probe precisely along the drawn vector. As a result, the probe will become blunted over several rotations resulting in a poor resolution tapping mode image at which time the AFM probe is replaced. The metallic cap on the graphite is also critical as the AFM probe will create cracks, wrinkles, and folds in the graphite if it is unprotected.

The value of the applied force is unknown. However, it is correlated to the amplitude set-point parameter. If the value of the amplitude set point results in movement of the cross then we know the set point value is greater than the minimum value required to induce movement. If the value of the amplitude set point does not induce movement of the cross, then its magnitude is increased until the cross does moves. Although we cannot precisely quantify the minimum set point to induce movement of the cross for every angle, we qualitatively observe that the set-point magnitude had to be increased around at 0° and 60° angles (which correspond to angles of crystal alignment). Also, a quantitative study of the force as a function of angle has already been published for bulk graphite[S1].

Supplementary References

(S1) Koren, E.; Lortscher, E.; Rawlings, C.; Knoll, A. W.; Duerig, U. *Science 348* (6235), **2015** 679–683.