A. S1. Role of metal Fermi surfaces in momentum conserved tunneling

In-plane momentum is treated as a conserved quantity in the tunneling processes described in the main text. Here we discuss the silver and chromium Fermi surfaces (FS) and their contributions to the tunnel current. For simple metals, the FS can be treated as a sphere, which works reasonably well for the polycrystalline Ag tunnel electrodes. Fig.S1a shows a top view of the Brillouin zone (BZ) of graphene, where the tiny red circles at the corner \( K \) and \( K' \)-points indicate the FS for a nominal density of \( 5 \times 10^{12} \text{cm}^{-2} \). The \( \Gamma - K \) distance for graphene is \( 1.70 \times 10^{10} \text{m}^{-1} \). The FS of Ag is shown as a sphere centered at the \( \Gamma \)-point (\( k_{F,Ag} \approx 1.2 \times 10^{10} \text{m}^{-1} \)). At low temperatures, the electrons that contribute to the tunnel current lie close to the Fermi surfaces of the metal and the graphene. Under normal circumstances, we observe that the Fermi surfaces have no overlap in momentum and hence elastic tunneling from the metal to the graphene layer is prohibited. Hence, only phonons or defect states in the heterostructure can contribute to inelastic tunneling processes, which are discussed in the following section.

Fig S 1 – (a) Top-view of the heterostructure elements as seen in the momentum space. The circles at the \( K/K' \) points are the Fermi surfaces of graphene for a density of \( n = 5 \times 10^{12} \text{cm}^{-2} \). The nearly spherical metal Fermi surface is centered at the \( \Gamma \)-point. (b) Chromium Fermi surface cross section in the (100) plane. The green square is the first BZ. The electron and hole octahedra are denoted by blue and red lines respectively. The black dashed line shows the graphene BZ and the tiny black dots indicate the graphene FS at the \( K \) and \( K' \)-points. In this orientation a \( K \) and a \( K' \) point overlap the Cr FS. Note that this is just one amongst the many orientations that are possible with a polycrystalline Cr top electrode.
In contrast, the FS of chromium is far from a simple sphere. The BZ of chromium is a dodecahedron and the FS has multiple electron and hole pockets within it\(^2\). Fig. S1b shows a schematic of the FS of chromium\(^2\). At the zone center, there exists a non-spherical electron pocket shown in blue. Hole pockets (red lines) are seen at the face-centers (\(N\)-points) and corner points (\(H\)-points). The polycrystallinity of the top-Cr tunnel electrode results in an uncertainty in the orientation of this FS with respect to the graphene BZ (dotted black lines). Assuming for simplicity that the (100) surface is normal to the graphene BZ, we observe that the BZ of Cr is a square (green) superimposed on the hexagonal graphene BZ as seen in Fig. S1b. Clearly, portions of the Cr FS do fall outside the graphene BZ. The in-plane orientation of the two Fermi surfaces depends on the orientations of the Cr crystallites. This presents a complicated phase space for momentum-conserved tunneling as the overlap of the two Fermi surfaces is governed by the relative orientations between the Cr crystallites and the graphene layer.

B. S2. Phonon-assisted tunneling

We now discuss the phonon-assisted tunneling phenomena assuming a spherical metal FS for simplicity. The electrons on the boundary of the spherical metal FS and the graphene FS contribute to tunneling. In Fig. S2, we schematically depict some of the possible phonon contributions to tunneling considering ‘in-plane’ momentum conservation. An electron from the north or south pole of the metal Fermi surface (blue disks) can use its out-of-plane momentum (blue dotted line) which is not conserved and a \(K\)-point phonon mode (blue solid line) to tunnel on to the graphene FS. In addition an \(M\)-phonon mode (yellow solid line) may also contribute to the tunnel current since it can be shifted across the Brillouin zone to the \(K\) or \(K'\)-point (green line). If there exists an electron in the Fermi surface that has the right \(z\)-momentum (green dashed line), this transition can be aided. This suggests that several phonons can contribute to tunneling by supplying appropriate \(z\)-momenta to the electrons on the surface of the metal FS.

Both graphene/graphite and h-BN have very similar phonon characteristics due to their closely matched crystal structure\(^3\text{--}^5\). Although the number of atoms in a unit cell is two for graphene and four for graphite (or hBN), many modes are doubly degenerate in graphite (or hBN) and hence the phonon dispersions are nearly identical with three optical (TO, LO or ZO) and three acoustic (TA, LA and ZA) branches. The phonons with the maximum density of states (with a flat band or a crossing in the phonon dispersion curve) will show the maximum contribution.
Interestingly, the above picture suggests that a relative orientation between the graphene and hBN layers will be inconsequential to the observed phonon modes as long as there are enough electrons in the Fermi surface that have corresponding $z$-momenta. The first peak in $d^2I/dV^2$ at $A = 36$ meV can be attributed to the flat band in hBN in the $M - K$ direction arising from the ZA mode\(^4\). We observe that the contributions to the two strongest peaks (at $B = 61$ meV and $C = 74$ meV respectively) come mainly from the ZA and ZO modes of the graphene and hBN\(^3,4\). This is in agreement with previous observations that the coupling of the out of plane modes to the graphene wavefunctions aid tunneling significantly\(^6\). More specifically, hBN has a flat band around $\sim 75$ meV in the $M - K$ direction arising from its ZO mode\(^4\). Graphene has a flat band at the $M$–point arising from the ZA mode, a crossing at the $K$–point arising from its ZA/ZO modes at $\sim 65$ meV and a ZA/ZO/TA band crossing at the $M$–point at $\sim 80$ meV\(^3\). Around the range $\sim 150 - 170$ meV, both hBN and graphene have many flat bands and crossings from LO and TO modes, which possibly lead to the broad peak that we observe at $D = 166$ meV.

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