Tunneling Spectroscopy of Two-Dimensional Materials Based on Via Contacts

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Abstract

We introduce a novel planar tunneling architecture for van der Waals heterostructures based on via contacts, namely metallic contacts embedded into through-holes in hexagonal boron nitride (hBN). We use the via-based tunneling method to study the single-particle density of states of two different two-dimensional (2D) materials, NbSe2 and graphene. In NbSe2 devices, we characterize the barrier strength and interface disorder for barrier thicknesses of 0, 1 and 2 layers of hBN and study the dependence on tunnel-contact area down to (44 ± 14)² nm². For 0-layer hBN devices, we demonstrate a crossover from diffusive to point contacts in the small-contact-area limit. In graphene, we show that reducing the tunnel barrier thickness and area can suppress effects due to phonon-assisted tunneling and defects in the hBN barrier. This via-based architecture overcomes limitations of other planar tunneling designs and produces high-quality, ultra-clean tunneling structures from a variety of 2D materials.

Keywords: tunneling spectroscopy, via contacts, niobium diselenide, superconductivity, graphene, two-dimensional materials

Tunneling spectroscopy is an indispensable experimental tool of modern condensed matter physics. Vertical planar tunneling, which uses a fixed-width tunnel barrier, offers advantages over other spectroscopic tools such as scanning tunneling microscopy (STM). One such advantage is the ability to tunnel in re-orientable and very large (≥ 40 T) magnetic fields at dilution refrigerator temperatures (≤ 30 mK), a capability that has application in, for example, determining the order parameter symmetry of novel two-dimensional superconductors [1]. Another advantage is that the materials used for both the tunneling electrodes and the barriers themselves can be varied, allowing for a variety of tunneling devices to be implemented, including magnetic tunnel junctions and Josephson junctions [2, 3]. The use of van der Waals materials in planar tunneling junctions offers the additional advantage that atomic precision of the tunneling distance can be achieved, and a variety of tunneling devices using van der Waals insulators, semiconductors, and magnetic insulators as tunneling barriers have been realized [4–13]. In the conventional design of planar tunneling junctions, however, it is difficult to control the area of a tunneling electrode that purely probes the bulk because such geometries necessarily include tunneling into the edge of a sample. To perform spectroscopy using planar tunneling junctions, it is advantageous to have tunneling contacts with a small area to limit the effects of sample inhomogeneity, thereby approximating the tunneling from an STM tip or, in the case of a transparent barrier, to perform point-contact spectroscopy where the effective radius of the tunneling electrode is smaller than the electron mean-free path.

In this work, we demonstrate planar tunneling junctions in van der Waals heterostructures whose size is limited in principle only by lithographic techniques. The junctions are based on metallic contacts that pass vertically through holes etched in exfoliated hBN flakes, named “via” contacts in analogy with their function in conventional circuits [14]. Figs. 1a and b depict side-view schematics of the two types of via-based tunneling structures studied. The “via” contacts are fabricated by reactive ion etching lithographically-patterned holes in 20-50 nm thick hBN flakes followed by e-beam deposition of a non-sticking metal such as Au, Pd, or Pt inside the holes (see Supplemental Information for details) [14]. The via-embedded hBN flakes are then used subsequently to pick up a thinner (hBN) tunnel barrier (typically ≤ 3 layers thick) and then the target material using the dry-polymer transfer technique [15]. This procedure can be performed entirely in an inert environment, ensuring good tunneling contact to air-sensitive materials. We study the dependence of via-based tunneling on the thickness of the hBN tunnel barrier, on the tunneling area, and on the role of defects in the tunnel barrier. This device geometry allows us to achieve junction areas as small as (44 ± 14)² nm², and in the limit of zero layers of hBN, we demonstrate the crossover from a diffusive contact to a ballistic point contact as the tunneling area of the device is reduced.

We study two different target 2D materials (< 25 nm thick NbSe2 and monolayer graphene, MLG), in order to investigate tunneling into a variety of electronic phases, such as into a 2D
superconductor and into a quantum Hall insulator. NbSe$_2$ is metallic and becomes superconducting below $T_c \approx 7$ K. MLG is a Dirac semimetal whose carrier density can be tuned by external gates and develops Landau levels (LLs) under perpendicular magnetic fields. In Fig. 1c, we show a transmission electron microscopy (TEM) image of the vertical structure of a normal-metal/insulator/superconductor (NIS) junction. In the vicinity of the Au/hBN/NbSe$_2$ junction region, the interface is unbroken and atomically flat, indicating our fabrication procedure produces ultra-clean junction interfaces. Fig. 1d shows a false-colored top-view scanning electron microscopy (SEM) image of a representative tunneling device.

The role of hBN as a barrier can be approximated by calculating the transmission coefficient using a potential barrier with a finite height and width. The height in this approximation is determined by the offset of the hBN conduction band and the Fermi energy of the tunneling electrode (determined by the alignment of their vacuum levels), and the width is set by the thickness of hBN. The transmission coefficient $T$ then becomes

$$T = \left(1 + \frac{V_0^2 \sinh^2 (t_{hBN}/\lambda)}{4E_F (eV_0 - E_F)}\right)^{-1},$$

where $t_{hBN}$ is the hBN thickness, $\lambda = \hbar/\sqrt{2m (eV_0 - E_F)}$ is the characteristic wavelength, $E_F$ is the Fermi energy of the electrons in Au, and $eV_0$ is the barrier height. Fig. 1e shows a summary of the measured tunneling resistance normalized by tunnel area, $R$ (in $\Omega \cdot \mu$m$^2$), as a function of the number of hBN layers $L$. Here, we compare the normal state tunneling resistance of the normal/insulating/normal (NIN) junctions in NbSe$_2$ at 10 K (above $T_c$) and the NIN junctions in graphene doped to approximately $-2 \times 10^{12}$ cm$^{-2}$ at 4 K. For $t_{hBN}/\lambda > 1$, the tunneling resistance versus hBN layer number approximates an exponential dependence versus barrier width, consistent with previous measurements on Au/hBN/Au tunnel junctions [5]. Our data fits the predicted $\sinh^2$ dependence for $L \leq 3$, with an extracted length scale of $\lambda \approx 0.10$ nm. The corresponding barrier height relative to the Au Fermi energy, from $\lambda = \hbar/\sqrt{2m (eV_0 - E_F)}$ and using the bare electron mass, is $eV_0 - E_F = 3.82$ eV, consistent with Schottky barrier calculations on metal/hBN interfaces [16]. For thicker tunnel barriers ($L = 7$), the tunneling resistance is reduced due to phonon-assisted tunneling in MLG [17], as will be discussed in detail in later sections.

We now turn to a detailed investigation of tunneling into the superconducting state of NbSe$_2$. In our analysis of the NIS tunneling, we employ the Blonder-Tinkham-Klapwijk (BTK) theory [18], in which the conductance of an NIS junction is primarily characterized by the barrier strength $Z = H/hv_F$, where $H$ models the strength of the delta-function potential at the normal-metal-superconductor interface, i.e. $V(z) = H\delta(z)$, and $v_F$ is the Fermi velocity. Large $Z$ corresponds to low-transparency junctions. Fig. 2a shows calculations of the tunnel conductance based on the BTK model for two tunneling limits, $Z = 0$ and $Z = 5$. Also included in these calculations is the role of energy broadening of the density of states, which can come from interface disorder and inhomogeneity, external noise sources [19], or inelastic scattering of
electrons at the junction interface [20]. Although not in the original BTK derivation [18], it can be included by introducing a finite quasiparticle scattering lifetime \(1/\Gamma\) and substituting \(E \rightarrow E + i\Gamma\). These two parameters, \(Z\) and \(\Gamma\), will be used to characterize via-based tunneling into NbSe\(_2\) through hBN tunnel barriers of various thicknesses (see Supplemental Information for details on extracting \(Z\) and \(\Gamma\)).

In Fig. 2b, we show the tunneling spectra of NbSe\(_2\), acquired by varying the thickness of hBN. In each panel we plot the differential conductance normalized to the normal-state resistance versus voltage bias above and below the superconducting transition temperature of NbSe\(_2\). With \(L = 2\) layers of hBN, we observe a canonical superconducting spectra for \(T < T_C\) characterized by vanishing conductance below the superconducting gap, \(|eV_F| < \Delta = 1.3\) meV, and distinct quasiparticle peaks appearing at the edge of the superconducting gap. For tunneling spectra acquired under similar conditions but with the tunnel barrier reduced to a single monolayer of hBN \((L = 1)\), we now observe a finite conductance at zero bias and the quasiparticle peaks are almost non-existent. The observation of finite conductance below the gap is consistent with the expectation that decreasing layers of hBN increases the transparency of the junction \((Z = 3.0\) compared to \(Z = 14.0\) in the \(L = 2\) junction). The smeared-out quasiparticle peaks suggest an enhancement of interface broadening \((\Gamma < 0.1\Delta\) compared to \(\Gamma < 0.1\Delta\) in the \(L = 2\) junction). This is confirmed in the junctions fabricated with no layers of hBN. The spectrum is completely bias independent as the interface broadening dominates the observed spectra \((\Gamma \approx 3\Delta)\). This is not surprising considering our junctions are well outside the ballistic regime [20] since the cross-sectional interface area is much larger than the mean free path of Au squared, \(A \sim 1\) \(\mu m^2\) \(\gg l^2_{mfp} \approx l^2_{mfp} \approx 37\) nm is the mean free path of the Au. This allows for inelastic scattering near the interface, broadening the energy of the incident electrons [18, 20]. In all spectra, for \(T > T_C\), the differential conductance is independent of voltage bias, as expected for the normal state [5]. In Fig. 2c, we summarize the extracted barrier strength \(Z\) versus the number of hBN layers \(L\), where \(Z\) was calculated assuming \(\Gamma \approx 0\) (see Fig. S7 for details). We find \(Z \sim \sinh (L/D)\) well describes our data, with a fitting parameter \(D\). The extracted value is \(D = 0.52\), with a corresponding barrier height relative to the Au Fermi energy of \(eV_0 = E_F = 1.43\) eV.

One interesting observation in the hBN layer dependence is that adding a single layer of hBN (with a low tunnel resistivity of \(\approx 10^2 \Omega \cdot \mu m^2\)) decreases the energy broadening from \(\Gamma \approx 3.0\Delta\) to \(\Gamma < 0.1\Delta\). This implies that one could utilize monolayers of hBN to engineer homogeneous interfaces between disparate materials without significantly impacting the overall contact resistance. This effect has been observed in other systems [21, 22], where it was demonstrated that adding single layers of hBN between metal contacts and semiconductors prevents Fermi level pinning and band bending due to interfacial defects. A similar effect occurs here in the NIS junctions, in which the hBN barrier reduces inhomogeneity of the energy landscape caused by disorder and spatial inhomogeneities introduced during the fabrication process.

For the relatively large-area 0L hBN device shown in Fig. 2b \((\approx 0.6 \mu m^2)\), the effect of interfacial disorder, within the BTK model, was significant: the spectrum showed negligible bias dependence and thus a complete absence of spectroscopic information. Next, we investigate the role of contact area on tunneling spectra and present evidence that coherent tunneling spectra can be recovered by reducing the interfacial contact size (see Fig. S8 for fabrication details). Fig. 3a shows tunneling spectra for four 0L hBN devices with decreasing tunneling area, including the large-area device from Fig. 2b.

As we reduce the contact size to \((71 \pm 14)^2\) \(\mu m^2\), we observe the emergence of distinct quasiparticle peaks and a suppression of conductance for \(|eV_T| < \Delta\). Decreasing the contact area...


\[
T = 1.6 \text{ K} \\
R_N = 180 \Omega \\
R_N = 1.4 \text{ k}\Omega \\
R_N = 5.1 \text{ k}\Omega \\
R_N = 90 \text{ k}\Omega \\
(610 \pm 15)^2 \text{ nm}^2 \\
(71 \pm 14)^2 \text{ nm}^2 \\
(56 \pm 14)^2 \text{ nm}^2 \\
(44 \pm 14)^2 \text{ nm}^2 \\
\]

**Fig. 3. Crossover to point-contact spectroscopy in NbSe\textsubscript{2} devices.**

a) Tunnel conductance versus junction bias normalized to the normal-state conductance for various contact cross-sectional areas for 0 layer hBN tunnel barriers. The solid black line is the spectrum in Fig. 2b, given for reference. b) Contributions to the current in a point contact [20]. (Left panel) In the “thermal regime” where \( a = \sqrt{A} \gg \ell_{mfp} \), electrons undergo inelastic scattering in the contact region and transport through the junction is similar to normal transport between two conductors. (Right panel) In the “ballistic regime”, electrons within \( \ell_{mfp} \) are accelerated through the contact without scattering and gain energy \( eV \), where \( V \) is the applied voltage. In this regime, it is possible to obtain spectroscopic information about the target material.

The spectrum of D1 at \( B = 0 \) (Fig. 4a) exhibits a prominent gap-like feature at low tunneling energy \(|E_T| \leq E_{ph} = 63 \text{ meV}\), which arises from the intrinsic electron-phonon coupling in MLG due to the out-of-plane acoustic phonon modes near the K/K' points [25] and has been reported in STM measurements [17, 26]. Conductance is suppressed when \(|E_T| \leq E_{ph}\) due to the thick tunnel barrier (\( L = 7 \)) and the absence of phonon-assisted tunneling, whereas above \( E_{ph} \) it adds an additional conductance channel which appears as a sharp edge in the conductance maps. Such phonon-assisted tunneling process enhances the amplitude of \( G_0 \) despite the usage of a thick tunnel barrier (\( L = 7 \)). Owing to energy loss during this inelastic scattering, locations of Dirac point (minima of \( G_0 \) in \( E_T \) at given \( n_0 \) values, indicated by light blue circles), \( V_D \), are offset by the phonon energy \( E_{ph} \) from their true energy locations, \( E_D \), giving

\[
|eV_D| = E_{ph} = E_D = \hbar v_F \sqrt{\pi |n_0|},
\]

where \( v_F \) is the Fermi velocity of MLG. The inset in Fig. 4a shows the fit to Eq. (3), resulting in \( v_F \approx 1.13 \times 10^6 \text{ m s}^{-1} \).
Fig. 4. Via-based tunneling into monolayer graphene. a, b Zero-field spectra of D1 (L = 7, A = 3 μm²) and D2 (L = 3, A = 0.07 μm²), respectively. a Light blue circles mark the locations of Dirac point. Light green dashed lines (also in panel c) mark the boundaries of the phonon gap, with $E_{ph} \approx 63$ meV. The inset shows the fit to Eq. (3), giving $v_F \approx 1.13 \times 10^6$ m/s, with data and extracted curve indicated by the light blue circles and black dashed curve, respectively. Yellow dashed lines indicate the suppression of tunneling conductance $G_0$ near the charge neutrality point ($n_0 = 0$) due to the series resistance from in-plane transport. b The light blue arrow marks the feature of a representative defect from hBN tunnel barrier observed in the spectrum. c, d $G - G_0$ map of D1 and D2 measured at 12.5 T, respectively. Trace of each LLN (N = -2, -1, 0 and 1) is indicated by unique color. The white dashed line is taken at $n'_0 = -4.5 \times 10^{12}$ cm$^{-2}$. The inset shows the fit to Eq. (4) at $n'_0$ with the LL$-1$ and LL$-2$ points (indicated by pink and green dots), giving $v_F \approx 1.04 \times 10^6$ m/s. The black dashed line indicates the fitted line. The triangles for LL0 and LL1 are taken from the split peaks, and the dots are their corresponding midpoints.

m/s, consistent with the previous transport and STM studies [17, 27, 28]. Fig. 4b shows the spectrum of D2 at $B = 0$. With a thinner tunnel barrier ($L = 3$), we now observe the emergence of sharp peaks (indicated by the light blue arrow) in $G_0$ associated with single electron charging of the defects in hBN, which is suppressed in D1 due to its larger barrier thickness, consistent with our conclusions from NbSe$_2$ devices. The enhancement of tunneling current also leads to the disappearance of a clearly-defined phonon gap, as phonon-mediated tunneling no longer dominates in the presence of additional tunneling channels, similar to previous observations in STM [26].

At finite magnetic fields, the DOS of graphene evolves into sharp peaks at energies of LLs, given by

$$E_N = E_D + v_F \cdot \text{sgn}(N) \sqrt{2e\hbar|N|B},$$  \hspace{1cm} (4)

where $N$ is the index for the $N$th LL (LL$_N$), and $E_N$ is the energy of LL$_N$. For clarity, we remove the global background that is independent of magnetic field by subtracting the differential conductance $G$ measured at $B = 12.5$ T from its counterpart $G_0$ measured at zero field, resulting in the $G - G_0$ maps shown in Figs. 4c and d for D1 and D2, respectively. Both spectra show distinct LLs, with each LL indicated by unique color in the maps. In Fig. 4c, we identify the locations of $E_N$...
In summary, by studying the spectra of NbSe$_2$ and MLG at low temperature and high magnetic fields with various tunnel junction areas and hBN barrier thicknesses, we demonstrate that the via-based planar tunneling method has multiple advantages over conventional planar tunneling geometries in vdW heterostructures and over STM. The via platform avoids exposing any interface to air or lithographic polymers and permits tunneling areas that are limited, in principle, only by lithographic techniques. We found that for ~1 µm$^2$-area tunnel barriers, 2L hBN optimizes the combination of low interface transparency $Z$ and low interface disorder $\Gamma$ for tunneling into 2D superconductors; 2L hBN has also been found to be the ideal thickness as a tunnel barrier for spin injection in graphene-based spintronic devices [6, 7]. In the limit of small tunneling area (down to (44 ± 14)$^2$ nm$^2$) and 0L hBN, we found that point-contact spectra could be realized. This suggests that via-based tunneling devices could be a powerful probe of the spin structure of triplet or mixed singlet-triplet superconductors by studying the tunneling into the same sample from multiple via point contacts having different degrees of spin polarization [38]. Such high quality tunneling structures could also be applied to other low-dimensional systems, such as superconducting contacts to semiconductors [39], superconducting-ferromagnetic-superconducting (SFS) junctions, or high-$T_c$ superconductors, or combined with other techniques such as the capacitive detection of vertical planar tunneling currents [24] to reduce the series transport resistance in semiconducting/insulating devices.
SUPPORTING INFORMATION

Identification and contrast calibration of ultra-thin hBN; device fabrication details; measurement setup; BTK calculation of NbSe2 tunneling spectra; simulation of graphene tunneling spectra; extended transport and tunneling data.

AUTHOR CONTRIBUTIONS

Q.C. and E.J.T. contributed equally to this work. Q.C. fabricated, measured, and analyzed data from the graphene-based tunneling devices. E.J.T., A.B., and I.K. fabricated, measured, and analyzed data from NbSe2-based tunneling devices. E.J.T. performed the scanning electron microscopy. A.Z. performed the transmission electron microscopy. K.W. and T.T. provided hBN and carbon-defected hBN crystals. C.R.D. and B.M.H. supervised the research. All authors contributed to writing the manuscript.

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SUPPLEMENTARY INFORMATION
TUNNELING SPECTROSCOPY OF TWO-DIMENSIONAL MATERIALS BASED ON VIA CONTACTS

Identification and Contrast Calibration of Ultra-thin hBN

To quickly and reliably identify the thickness of ultra-thin hBN flakes (<10 layers), we developed a contrast calibration curve for flakes exfoliated on 90 nm SiO$_2$/Si$^{++}$ substrates. Substrates with 90 nm SiO$_2$/Si$^{++}$ were chosen over 285 nm SiO$_2$/Si$^{++}$ substrates due to the increased contrast for few-layer hBN [40]. First, a series of images of hBN flakes with varying thicknesses was collected using a Nikon Eclipse LV150N microscope fitted with a Nikon DS-Fi3 camera (Figure S11 a). The images were then shading corrected and the flake contrast was extracted using Gwyddion (Figure S11 b,c). We found that red color contrast was the most significant. The contrast was then correlated to flake thickness (Figure S11 e), which was measured through atomic force microscopy and tunneling resistance [41].

Atomic force microscopy was performed in a Bruker Dimension Icon® using OTESPA-R3 tips in tapping mode. Flake thicknesses were extracted using Gwyddion to measure histograms of the height difference between the substrate and the desired hBN flake.

Fabrication of Au/hBN/NbSe$_2$ Tunneling Devices

Tunneling devices were fabricated from NbSe$_2$ flakes using the via contact technique [42] in which hBN with embedded Au electrodes was used to pick up ultra-thin flakes of hBN (ranging in thickness from 1-3 layers) and subsequently placed onto the desired NbSe$_2$ flake using the dry-polymer-transfer technique [15]. The via contacts and hBN tunnel barrier were prepared under ambient conditions, whereas the NbSe$_2$ flakes were prepared under inert conditions in an N$_2$ glovebox with <5 ppm O$_2$ and <0.5 ppm H$_2$O. All exfoliated flakes were prepared using mechanical exfoliation with Scotch® Magic™ tape [43, 44]. First, the via contacts and hBN tunnel barrier were picked up in air then transferred into a glovebox where the heterostructure was placed onto the desired NbSe$_2$ flake under inert conditions. Bonding pads (Cr + Au: 2nm + 80nm) were then designed and deposited using conventional electron-beam lithography and deposition techniques. All devices were diced by hand and bonded to a 16-pin DIP socket for measurement in cryogenic systems. Between fabrication steps, all devices were stored in the N$_2$ glovebox to avoid sample degradation.

Tunneling Measurements on Au/hBN/NbSe$_2$ Tunnel Junctions

Tunnel junction resistance was measured in a 2-terminal differential configuration whereby a small AC voltage bias was superimposed on top of a DC voltage bias. The corresponding AC current was measured as a function of applied DC voltage, temperature, and magnetic field. For all data presented in the manuscript, the tunnel conductance is defined as $G = \frac{dI}{dV}(V_{DC}) = \frac{I_{AC}}{V_{AC}}(V_{DC})$, where $V_{AC}$ and $I_{AC}$ are the AC voltage bias and current and $V_{DC}$ is the applied DC voltage bias. The normal state resistance is defined as the junction resistance when the DC voltage bias is greater than the superconducting gap, $R_N = \frac{1}{dI/dV}(eV_{DC} > \Delta_{NbSe_{2}})$. For all data presented in the manuscript, $V_{AC}$ was set to <50 $\mu$V. The AC voltage bias was applied using an SRS830 lock-in amplifier with a reference frequency of 17.777Hz and the AC current was measured using the same lock-in amplifier. The DC voltage bias was applied using a Keithley 2400. The AC and DC voltage sources were connected in parallel to a voltage divider, the output of which was directly connected to the sample (Figure S12). To ensure the DC voltage was dropped predominantly across the relevant Au/hBN/NbSe$_2$ interface, the DC+AC voltage source was injected at the tunnel barrier and drained from low-resistance Au/NbSe$_2$ contacts (Figure S12). For measurements of low-resistance tunnel junctions (with <2 layers of hBN) multiple Au/NbSe$_2$ contacts were used as the drain to reduce the total drain contact resistance. Measurements were performed either in a $^3$He cryostat with temperatures ranging from 300 mK up to 10 K or a pumped $^4$He cryostat with temperatures ranging from 1.6 K up to 10 K.

Synthesis of Carbon-defected hBN

Carbon-defected hBN crystals were obtained by post treatment of ultra-clean hBN crystals by annealing with graphite powder using the procedure outlined in reference [34]. As carbon diffusion takes place, the color of the hBN crystals changes from white to yellow (Figure S13).
Scanning Electron Microscopy

Scanning electron micrographs were collected on a Zeiss Sigma VP scanning electron microscope (SEM) using a beam energy of 5 kV.

Transmission Electron Microscopy

Thermo Scientific Helios NanoLab 660 (equipped with focused ion-beam) was used to prepare thin foils for transmission electron microscopy (TEM). In order to protect the surface against the ion-milling process, amorphous platinum (2 µm thick) was sputtered on the top surface by the electron and ion-beam, respectively. High-resolution TEM images were acquired by Thermo Scientific Talos F200X (S)TEM at an accelerating voltage of 200 kV using a 100 µm objective aperture.

Fabrication of Au/hBN/graphene Tunneling Devices

Tunneling devices were fabricated from graphene using the via contact technique where hBN was embedded with Au or Pd electrodes that were arranged in two rows. First, the via contact hBN flakes were picked up using the dry-polymer-transfer technique [15]. Thin hBN tunnel barriers were subsequently picked up such that the few-layer hBN was located under only one of the rows of vias, after which we picked up the graphene flake as well as the remaining hBN capping layer (20-50 nm thick) and the bottom graphite gate (>30 nm thick). The entire heterostructure was then transferred onto a SiO$_2$/Si$^{++}$ substrate. One row of the via contacts served as tunneling electrodes with precisely defined and located tunneling areas, and the other row of via contacts made direct contact to the graphene and served as drain electrodes. All exfoliated flakes were prepared using mechanical exfoliation with Scotch® Magic™ tape [43, 44]. As a final step, bonding pads (Cr + Pd + Au: 2nm + 40nm + 50nm) were designed and deposited using standard electron-beam lithography and deposition. All fabrication steps were performed under ambient conditions.

Tunneling Measurements on Au/hBN/graphene Tunnel Junctions

Tunnel junction resistance was measured in a 2-terminal differential configuration whereby a small AC voltage bias was superimposed on top of a DC voltage bias. The corresponding AC current was measured as a function of applied DC voltage, temperature, and magnetic field. For all data presented in the manuscript, the tunnel conductance is defined as $G = \frac{dI}{dV}(V_{DC}) = \frac{I_{AC}}{V_{AC}}(V_{DC})$, where $V_{AC}$ and $I_{AC}$ are the AC voltage bias and current and $V_{DC}$ is the applied DC voltage bias. For all data presented in the manuscript, $V_{AC}$ was set between 5-10 mV. The AC voltage bias was applied using an SRS860 lock-in amplifier with a reference frequency of 13Hz and the AC current was measured using the same lock-in amplifier. An SRS570 preamplifier was placed in series before the current-measuring lockin to reduce the noise (lowpass filter, 12db, cutoff 1kHz). The DC voltage bias was applied using a Keithley 2400. The AC and DC voltage sources were connected in parallel to a voltage divider, the output of which was directly connected to the sample (Figure S12). To ensure the DC voltage was dropped predominantly across the relevant Au/hBN/graphene interface, the DC+AC voltage source was injected at the tunnel barrier and drained from low-resistance Au/graphene contacts (Figure S12). The electrostatic gate voltage was applied using a Yokogawa GS200 DC voltage source. All measurements were performed in a dilution refrigerator with temperature ranging from 35 mK to 4.5 K. Figure S14 shows the microscope images and transport characterization of a representative MLG device.

Blonder-Tinkham-Klapwijk (BTK) Calculations of NbSe$_2$ Tunneling Spectra

Overview of BTK Theory Calculations

Using BTK theory [45][46], we can directly model the tunneling conductance versus $V_{DC}$ with various barrier strengths $Z$ and inhomogeneity parameters $\Gamma$. The tunneling rate $I_{NS}$ of the normal-insulator-superconductor (NIS) junction as a function of $V_{DC}$ and temperature $T$ can be written as

$$I_{NS}(V_{DC}) \propto \int_{-\infty}^{\infty} (f(E - eV_{DC}; T) - f(E, T)) [1 + AR(E) - R(E)] dE$$  \hspace{1cm} (S5)
where $f$ is the Fermi function and $AR(E)$ and $R(E)$ are the probabilities for Andreev reflection and normal reflection, respectively. For comparison to experiments, we normalize the NIS current by the magnitude of the current in the normal state ($I_{NS}$ in the limit $AR(E) \rightarrow 0$ and $\Delta \rightarrow 0$, $I_{NN}(V) \propto \frac{V}{1+Z^2}$. Our final expression for interface current is the following

$$I_{NS}(V_{DC}) = \frac{1 + Z^2}{en_D} \int_{-\infty}^{\infty} (f(E - eV_{DC}, T) - f(E, T)) [1 + AR(E) - R(E)] dE$$

(S6)

where $E$ is the electron energy, $V_{DC}$ is the applied DC voltage, $e$ is the electron charge, $T$ is the sample temperature, $AR(E)$ and $R(E)$ are the Andreev reflection and normal electron reflection probabilities, respectively, and $f$ is the Fermi function. Ordinary reflection of electrons reduces the overall tunneling current, but Andreev reflections enhance it by transmitting a Cooper pair for each reflected hole. $AR(E)$ and $R(E)$ can be determined by matching wave-function and wave-function-derivative boundary conditions on either side of the junction, assuming the gap $\Delta$ is zero on the metal side and constant on the superconductor side. The interface potential is modeled as a delta function $V(z) = H\delta(z)$ with a dimensionless barrier strength parameter $Z = \frac{k_F}{\epsilon_F}H$, where $k_F$ and $\epsilon_F$ are the Fermi wave vector and energy, respectively. The total electron transmission probability can be written as

$$1 + AR(E) - R(E) = \tau \frac{1 + |\gamma(E)|^2 + (\tau - 1)|\gamma(E)|^2}{|1 + (\tau - 1)|\gamma(E)|^2|^2}$$

(S7)

where $\tau = \frac{1}{1+Z^2}$ is the transparency of the junction and

$$\gamma(E) = \frac{N_q(E) - 1}{N_p(E)}, N_q(E) = \frac{E}{\sqrt{E^2 - \Delta^2}}, N_p(E) = \frac{\Delta}{\sqrt{E^2 - \Delta^2}}$$

(S8)

In our experiments, when we extract quantitative information about the tunnel barrier, the sample temperature is much lower than the superconducting gap of NbSe$_2$, so we can take the limit of $T \rightarrow 0$, simplifying the expression for barrier conductance

$$\frac{G_{NS}}{G_{NN}} = \frac{d}{\tau d(eV_{DC})} \int_{0}^{eV_{DC}} [1 + AR(E) - R(E)] dE$$

(S9)

To account for the effects of energy broadening and inhomogeneity, the model can be modified by replacing $E \rightarrow E + i\Gamma$ where $\frac{1}{\tau}$ is interpreted as a finite quasiparticle life time.

Extracting $Z$ and $\Gamma$ from Tunneling Spectra

In determining the transparency of our measured junctions, it is important to accurately capture the role of $\Gamma$ on the tunnel spectra. A common procedure for extracting $Z$ is to extract the excess current $I_{EXCESS}$ defined as the intersection of the normal-state $I - V$ curves with the $I$ axis (Figure S15a). The excess current depends strongly on $Z$ in the low $Z$ regime (Figure S15b). However, the $Z$ dependence of the excess current is greatly suppressed with increasing $\Gamma$ (Figure S15b). As such, we use a different method to extract $Z$ from our data. The main qualitative role of $\Gamma$ is to suppress sharp features in the spectra (quasiparticle peaks) and push any deviations from the normal-state conductance to the normal-state value (Figure 2a). We can accurately extract $Z$ and $\Gamma$ values from our junctions by finding intersecting contours of the zero-bias conductance versus $Z$ and $\Gamma$ (Figure S7a) and the maximum junction conductance versus $Z$ and $\Gamma$ (Figure S7b).

Simulations of Graphene Tunneling Spectra

Tunneling under Zero Magnetic Field

We modeled the measured conductance of monolayer graphene based on four contributions: direct tunneling through the via contacts (1 contribution for each contact), tunneling in the presence of the phonon gap in graphene, and in-plane transport. At zero magnetic field, graphene’s band structure is a Dirac cone where the location of the charge neutrality point shifts by phonon energy $E_T \sim 63$ meV for phonon-assisted tunneling, and remains unchanged for in-plane transport. We also included several defect states in the graphene band structure to model the bright features in devices with 3L of hBN. Figure S16 shows simulations of conductance for devices with various tunnel barrier thickness and area in comparison with the measured conductance through via contacts. We can see that the phonon-assisted tunneling dominates in the device with 7L of hBN, and the bright features in the spectra of 3L devices can be explained by defect states in graphene. Moreover, all devices exhibit features associated with in-plane transport (dip in conductance near $n_0 = 0$).
We performed a self-consistent calculation \cite{47} to model the density of states in graphene with varying doping density and tunneling energy. Figure S17 shows a representative simulation at $B = 12.5\text{T}$ with a similar parameter range as in the experiments.
Fig. S6. Via-based tunneling into NbSe$_2$ with 3L $h$BN. Measurements of differential conductance $G = dI/dV_T$ versus junction bias normalized to the normal-state resistance $R_N$ for $T < T_C$ (solid black dots and line) and $T > T_C$ (solid red dots and line) for a 3L $h$BN Au/$h$BN/NbSe$_2$ tunnel junction. The cross-sectional area of the via tunnel contact is $\sim 0.6 \ \mu m^2$. 
Fig. S7. Select tunneling spectra parameters versus $Z$ and $\Gamma$. a) Zero-bias conductance ($G_{eV=0}$) normalized by the normal-state conductance ($G_N$) versus $\Gamma$ and $Z$. Contours of $G_{eV=0}/G_N = 1.5$, $G_{eV=0}/G_N = 1.0$, $G_{eV=0}/G_N = 0.5$ are denoted by solid black lines with the corresponding $G_{eV=0}/G_N$ values given. b) Maximum spectra conductance ($G_{max}$) normalized by the normal-state conductance ($G_N$) versus $\Gamma$ and $Z$. Contours for various $G_{max}/G_N$ are given by solid white lines with the corresponding $G_{max}/G_N$ values given. All calculations were performed at $T = 0$. 
Fig. S8. Characterization of small-area via contacts. a) Schematic of the lithography design used to dose test the small-area via contacts on SiO₂. The design contains circles ranging in diameter from 100 nm down to 30 nm in increments of 5 nm. Large 1 µm holes are also written for reference. b-e) Scanning electron microscopy (SEM) images of etched holes in SiO₂ with intended diameters of 1 µm (b), 100 nm (c), 65 nm (d), and 30 nm (e). All scale bars are 100 µm. f) Measured hole diameter versus electron dose for 30 nm (solid green dots), 65 nm (solid blue dots), and 100 nm (solid red dots) hole designs. The error bars represent the standard deviation.
**Fig. S9. Effect of junction size on interface inhomogeneity $\Gamma$ for 1L hBN junctions.** Au/hBN/NbSe$_2$ tunnel conductance versus junction bias normalized to the normal-state conductance for various contact cross-sectional areas for 1L hBN tunnel barriers. Corresponding junction sizes, normal-state resistances, and extracted $\Gamma$ values are given in the inset.

**Fig. S10. Number of observed defects in hBN tunnel barrier estimated from monolayer graphene tunnel spectra.** a-d Measured dI/dV spectra in $E_T$-$n_0$ space with 3L of hBN tunnel barriers and 0.07, 0.20, 0.38 and 0.95 $\mu m^2$ tunnel area, respectively. Diameters of tunnel contacts are given in the insets. e Number of observed defects in (a)-(d) versus tunnel area, adapted from Fig. 5b.
**Fig. S11. Identification and characterization of few-layer hBN.** a,b) Unprocessed (a) and processed (b) optical images of mono, bi, and trilayer hBN flakes on a 90 nm SiO$_2$/Si$^{++}$ substrate. In (b), the mono, bi, and trilayers of hBN are labeled by their corresponding layer number. c) Red optical contrast versus hBN thickness. A linear fit to the data is given by the solid red line. The fit parameters are given in the inset. Lower right inset: atomic force microscope topography of the flake in (a) and (b).

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**Fig. S12. Schematic of the tunneling measurement setup.** For the Au/hBN/NbSe$_2$ experiments, $R_1 = 100$ kΩ, $R_2 = 1$ MΩ, $R_S = 100$ Ω. The RC filters are homemade low-pass filters with 3 dB attenuation at $\sim$5 kHz [48, 49]. For the Au/hBN/graphene experiment, $R_1 = R_2 = 10$ kΩ, $R_S = 100$ Ω. The RC filters are also low-pass filters with 3 dB attenuation at $\sim$5 kHz. The dashed square represents the fabricated tunnel junction devices.
Fig. S13. Optical images of bulk hBN crystals. Optical images of carbon-defected (a) and ultra-pure (b) bulk hBN crystals. Synthesis and characterization of pristine and carbon-defected hBN are outlined in references [34, 50].
Fig. S14. Characterization of a representative monolayer graphene device. a, b Microscopic image of a MLG device with 100x magnification under bright field (a) and dark field (b). The black dashed line in (a) outlines the hBN tunnel barrier. Both scale bars are 5 µm. c Four-terminal resistance measurement (without hBN tunnel barrier) as a function of back gate voltage and magnetic field.

Fig. S15. Excess current versus $Z$ and $\Gamma$. a) Calculated $I - V$ curves for various barrier strengths $Z$. The excess current is found by extrapolating the normal-state $I - V$ behavior to zero energy. b) Excess current versus barrier strength $Z$ for various $\Gamma$ values (given with respect to the superconducting gap $\Delta$). All calculations were performed at $T = 0$. 
Fig. S16. Measured tunnel spectra for monolayer graphene and simulations under zero magnetic field. a, b, c Measured dI/dV spectra in $E_F$-$n_0$ space with 7L, 3L and 3L of hBN tunnel barriers and 3.08, 0.07 and 0.38 µm$^2$ tunnel area, respectively. d, e, f Corresponding simulations of dI/dV with contributions from tunneling, in-plane transport, and phonon gap.

Fig. S17. Self-consistent calculation of DOS in monolayer graphene. Simulation at $B = 12.5T$ within the experimentally reachable parameter ranges.
Fig. S18. Normal-state NIS junction resistance versus hBN thickness. a, c. Histograms of normal-state resistances for various Au/hBN/NbSe$_2$ junctions fabricated with ultra-clean hBN flakes (a) and intentionally carbon-defected hBN flakes (c). Resistances were measured at $T = 10$ K. Dashed black lines separate the histograms into discrete hBN thicknesses. b, d) Plot of normal-state resistance versus hBN thickness for Au/hBN/NbSe$_2$ junctions fabricated with ultra-clean (b) and carbon-defected (d) hBN. Thickness of hBN was determined from optical contrast. Solid dark green (b) and solid blue (d) lines are fits to the data. Extracted fit parameters are given in the insets. Error bars and grey boxes represent the standard deviation and data range for each hBN thickness, respectively. In (d), the dashed dark green line is the fit from (b) for comparison. For all devices, the cross-sectional area of the via tunnel contacts is $\sim 0.6 \ \mu$m$^2$. 
Fig. S19. NIS junction barrier strength $Z$ versus $h$BN thickness. 

\( a, c \), Histograms of barrier strength $Z$ for various Au/$h$BN/NbSe$_2$ junctions fabricated with ultra-clean $h$BN flakes (\( a \)) and intentionally carbon-defected $h$BN flakes (\( c \)). $Z$ values were measured at $T = 300$ mK. Dashed black lines separate the histograms into discrete $h$BN thicknesses. 

\( b, d \), Plot of barrier strength $Z$ versus $h$BN thickness for junctions fabricated from ultra-clean (\( b \)) and carbon-defected (\( d \)) $h$BN, respectively. $h$BN thicknesses were determined by optical contrast. Solid red (\( b \)) and pink (\( d \)) lines are fits to the data. Extracted fit parameters are given in the insets. Error bars and grey boxes represent the standard deviation and data range for each $h$BN thickness, respectively. In (\( d \)), the dashed red line is the fit to the data in (\( b \)) for comparison.

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Fig. S20. Magnetic-field dependence of Au/hBN/NbSe₂ tunneling spectra. Contour maps of tunnel conductance versus junction bias and external magnetic field (B) for tunnel junctions with 0 (a), 1 (b), and 2 (c) layers of hBN as the barrier. Each trace is normalized to the normal-state conductance. All data was acquired at $T \sim 300$ mK. For all devices, the cross-sectional area of the via tunnel contacts is $\sim 0.6 \ \mu m^2$.

Fig. S21. Temperature dependence of Au/hBN/NbSe₂ tunneling spectra. Contour maps of tunnel conductance versus junction bias and temperature (T) for tunnel junctions with 0 (a), 1 (b), and 2 (c) layers of hBN as the barrier. Each trace is normalized to the normal-state conductance. All data was acquired at $B = 0$ T. For all devices, the cross-sectional area of the via tunnel contacts is $\sim 0.6 \ \mu m^2$.