

Unconventional conductivity increase in multilayer black phosphorus

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27 **Abstract**

28 **Multilayers of so-called 2D van der Waals materials have gained considerable**
29 **attention as active components of next generation electronic and**
30 **optoelectronic technologies, with semiconducting black phosphorus (BP)**
31 **regarded as one of the most promising systems.**

32 **The applicability and performance limits of BP in both stand-alone and**
33 **heterostructure-based multilayer devices is determined by individual flake**
34 **charge transport properties, which synergistically depend on the number of**
35 **layers and the strength of interlayer coupling between those.**

36 **In this work, we study the DC electrical transport characteristics of high quality**
37 **BP field-effect devices within a wide range of flake thicknesses at room**
38 **temperature. The experimental data show a non-trivial increase in conductivity**
39 **and hole density with reduced number of layers, while maintaining a constant**
40 **field-effect mobility due to the prevalence of electron-phonon scattering. Based**
41 **on the solution of the 1D Schrödinger-Poisson equation, we find that the**
42 **observed phenomena are a direct consequence of non-negligible interlayer**
43 **coupling, which in turn causes a local redistribution of free charge carriers**
44 **towards the central layers. Our data show that due to the electrostatic**
45 **conditions at the flake surfaces, a naturally protected 2D hole gas can be**
46 **encapsulated in flakes as high as 10 nm, which preserves the bulk-like**
47 **bandgap and effective carrier masses due to the electrostatic environment.**

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50 **Keywords: black phosphorus, 2D van der Waals material, layer dependence,**
51 **interlayer coupling**

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53 **Introduction**

54 Black phosphorus (BP) is a synthetically derived layered semiconductor material [1]
55 representing a class of monoatomic so-called two dimensional (2D) van der Waals
56 materials [2, 3]. The individual layers which form BP – phosphorene – have a
57 puckered hexagonal structure [4] and are bound together by, *both*, van der Waals
58 and ionic forces [5, 6]. As-grown (undoped) BP crystals are hole conductors [7, 8]
59 with distinctive anisotropy of electrical transport properties along the principal axis [8].
60 The *p*-type character of the majority charge carriers has been attributed to the
61 presence of electron trapping sites within the crystal lattice [9, 10].
62 Monolayer phosphorene, in particular, has been identified as one of the most
63 promising 2D electronic materials [11-13]. Its semiconducting properties, including a
64 bandgap comparable to silicon [14] and theoretically predicted hole mobility in the 10^3
65 cm^2/Vs range [15], are expected to significantly boost the electrical conduction and
66 switching properties of associated phosphorene field-effect transistor devices
67 regardless of their actual (effective) carrier concentration or their spatial distribution
68 within the system. On the other hand, few-layer components, which constitute the
69 vast majority of experimental data reported in literature, present a higher degree of
70 structural – and thus electronic – complexity. In particular, the overlap of the
71 electronic wavefunctions between individual phosphorene layers [5,6] is non
72 negligible, therefore black phosphorus is not a pure van der Waals system. This is
73 also reflected by the three to ten times larger layer-to-layer binding energy compared
74 to graphene and MoS_2 , respectively (Supporting Information). Consequently, such
75 non-negligible interaction also facilitates some charge carrier transfer between the
76 neighbouring 2D sheets, while simultaneously preserving the preferential motion of
77 carriers within individual layers under applied in-plane external electric fields.
78 Therefore, the question of how the thickness of a multi-layered BP system affects the

79 spatial distribution of free charge carriers, and, consequently, its overall electrical
80 conduction properties, becomes highly relevant, not only from the fundamental, but
81 also application viewpoint. In particular, the layer number dependent charge transport
82 efficacy will strongly impact the performance of BP-based photodetectors [16, 17],
83 photocatalysts [18], energy conversion and storage devices [19, 20], biomedical
84 devices and sensors [21].

85 In the present work we demonstrate through a comprehensive experimental study
86 that the room temperature electrical conductivity of BP outside of the band-gap
87 opening layer-range reveals an unexpected regime of unconventional increase as the
88 number of layers is reduced. Considering BP in this layer-range in electrical terms to
89 be a simple small-sized bulk semiconductor, such increase is contrary to the
90 expectation - a continuous reduction of size should lead to a decrease of conductivity
91 upon height (layer-number) reduction. The here observed conductivity augmentation
92 is intimately connected to an increase of the effective carrier density in the flakes
93 while charge-carrier mobility remains constant. Using a simple 3D-semiconductor
94 description, we find that the experimentally observed effects can be explained by
95 local charge redistribution towards the central layers due to surface electrostatic
96 conditions. Our results also provide insight to resolve the in literature experimentally
97 observed mobility limit of $\sim 300 \text{ cm}^2/\text{Vs}$ in few-layer BP devices at room temperature
98 [22-25].

99

100 **RESULTS AND DISCUSSION**

101 ***Sample contacting, Raman and AFM characterisation***

102 BP flakes were exfoliated from bulk crystals on substrate and immediately capped
103 with PMMA, both, under inert atmosphere (cf. Methods section). Optical images of
104 PMMA capped exemplary flakes are shown in **Fig. 1a**. After identification of flakes,

Ti/Au electrodes were electron-beam-lithographically defined on top (cf. Methods section) resulting in devices as shown in **Fig. 1b**. Immediately after electrode definition the devices were again covered by a protective PMMA layer. For further characterization Raman spectroscopy was carried out (cf. Methods section) on fully processed devices revealing a spectrum as shown in **Fig. 1c**. The spectrum shows only the well-known intrinsic first (A_g^1 , B_{2g} and A_g^2) and second order (ca. 790-920 nm^{-1} range) Raman signatures [26,27], while there is no trace of lines associated with oxidation [28,29] or any other uncommon lines discernible. Combined, the presence of the two intrinsic Raman responses and the absence of any other signs indicative of oxidation or degradation shows that the BP processed into devices is free of any unintentional influences.

Fig. 1. Mechanical exfoliation and electrical contacting of multilayer BP.

Exemplary optical micrographs of **a** BP flakes transferred onto the SiO_2 -coated silicon substrate. **Inset:** single BP flake (scale bar 10 μm). **b** A flake after electrical contacting with Ti/Au electrodes in 4-point configuration. Large electrodes are current-carrying leads, side-electrodes are 4-point voltage probes (cf. Methods). **c** Raman spectrum of two flakes of different thickness after device processing. Only the

common BP signature lines A_g^1 , B_{2g} and A_g^2 and the intrinsic BP second order Raman response (ca. 790-920 nm^{-1} range) are observed (Si line from substrate as expected at 520 cm^{-1}). No trace of other lines associated with oxidation or degradation is present. The expected positions of theoretically predicted prominent oxidation lines are indicated by the arrows [28,29]. **d** Flake height profile extracted from an atomic force micrograph (inset) after electrical characterisation. The extracted flake height is (15.1 ± 1.8) nm corresponding to (29 ± 3) phosphorene layers.

This is further supported by AFM characterisation (cf. Methods) of our processed devices from which we also extracted the height of the samples (**Fig. 1d**). Regarding the latter, the height range of the electrically characterised flakes ranged from ca. 6 nm up to 160 nm. That is, the thinnest flakes characterised were still beyond the bandgap broadening limit [14]. Also, our AFM imaging directly after electrical measurements (after PMMA removal) did not show any discernible blistering associated with BP flake degradation (**Fig. 1d** inset) in agreement with the Raman findings. Specifically, the data analysis revealed root mean square (RMS) values consistent with the absence of any relevant surface oxidation or degradation in general (Supporting Information) in agreement with previous AFM studies [30].

Layer-dependent charge transport properties

Electrical measurements were carried out in four-point configuration at room temperature and under DC conditions. The highly doped silicon-substrate (300 nm SiO_2 gate dielectric on top) served as a global back-gate for transfer characteristic measurements. In-plane-orientation selection of the flakes was discarded as experimental evidences showed that the conductivity and mobility of BP flakes in

armchair and in zigzag direction differ at room temperature by less than a factor of 1.6 [11]. All devices showed linear two-point current-voltage (I-V) characteristics demonstrating low contact resistance and ohmicity of the electrode/BP interface (cf. Supporting Information).

In all devices studied, unipolar *p*-type field switching behaviour was observed, with weak on/off switching for gate bias exceeding +/- 60 V (**Fig. 2a**). The hole mobilities, μ , extracted from the linear part of the subthreshold slope were found to be largely independent of the flake height (as corroborated by Hall measurements, see Supporting Information), and to lie around 200 cm²/Vs (**Fig. 2b**), which is in agreement with the majority of earlier experimental studies [22-25].

Figure 2. Height-dependent electronic properties of BP flakes at room temperature. **a** Representative four-point field-effect characteristic of a BP flake showing unipolar *p*-type behaviour and weak on/off switching. The hole mobility was extracted from the linear part of the subthreshold slope (denoted in the graph by dashed grey line). **b** Charge carrier mobility values estimated based on the field-effect characterisation. The mobility values do not show a height-dependent trend and show some few variations around 200 cm²/Vs, which fall within the range of the weakly developed in-plane anisotropy in charge transport at room temperature [11]. **c** Electrical conductivity and **d** charge carrier density show a steep increase for flakes with height below ca. 20 nm (dashed lines as guide to eye), whereas in thicker flakes the values remain approximately constant. **Inset d:** Linear dependence between conductivity and carrier density implies a height-independent average charge carrier mobility of ca. 215 cm²/Vs. Error bars result from data acquisition and error propagation in data evaluation.

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183 **Fig. 2c** shows the electrical conductivity, σ , as function of flake height h . In the case
184 of thicker flakes, the electrical conductivity is relatively constant and of the order of
185 few tens of S/cm. Notably, a marked increase in σ is observed for flakes with
186 thicknesses below *ca.* 20 nm, reaching values in the range of 10^2 S/cm. This result is
187 non-trivial, as in the conventional nanoscaled, but still bulk, semiconductor case, one
188 would expect a decrease in conductivity towards smaller layer numbers due to the
189 mere reduction of the cross-sectional area.

190 Taking the experimentally measured μ values into account, we estimated the free
191 mobile charge carrier (hole) density n_C (**Fig. 2d**). It needs to be emphasised here,
192 that the hole densities of about $1 \times 10^{18} \text{ cm}^{-3}$ for our thicker flakes is consistent with
193 recent experimental work on the density of *p*-type vacancies (about 60 to 63 ppm) in
194 high-quality black phosphorus crystals [10]. For a BP monolayer with atomic density
195 of $2.53 \times 10^{15} \text{ cm}^{-2}$ it can be therefore estimated that there is only one acceptor-
196 inducing electron trap per several tens of thousands of P atoms in the crystal lattice.
197 This demonstrates the high quality of the BP material used in our study.

198 Remarkably, we found that n_C – being the effective density of mobile charge carriers
199 contributing to the electrical conduction – also changes with the number of layers
200 (height). Specifically, n_C first remains rather constant for thick(er) flakes, while from
201 about 20 nm height on it displays a continuous increase, reaching values in the range
202 from 5 to $8 \times 10^{18} \text{ cm}^{-3}$ for heights in the 10 nm range. Intriguingly, such volume
203 carrier densities in this height range are commonly observed. For instance, a recent
204 publication by Yan *et al.* [28] reports on experimental sheet free carrier densities of 5
205 $\times 10^{12} \text{ cm}^{-2}$ in a 10 nm high flake; in a similar height range of 8 to 15 nm, Xia *et al.*
206 [11] report on an average sheet carrier density of about $6.7 \times 10^{12} \text{ cm}^{-2}$. Strikingly,
207 these values result in a volume free carrier density of $5 \times 10^{18} \text{ cm}^{-3}$ and $4.5\text{-}8.4 \times 10^{18}$

cm⁻³, respectively, which falls quantitatively into our experimental values in the same height range. While our experimentally determined hole densities are consistent with other experimental studies, the continuous increase with reduction of height (equivalently layer number) is non-trivial.

The experimental data also reveal that σ is directly proportional to n_C (**inset Fig. 2d**), which allows to estimate the average charge carrier mobility of (215 ± 49) cm²/Vs in all devices. This indicates that a dominant scattering mechanism limiting the motion of the charge carriers exists, which is independent of the layer number and charge carrier density. In fact, recent theoretical [31] and experimental work [32] has shown that the hole mobility in BP flakes is limited exclusively by electron-phonon scattering at room temperature. Specifically, the theoretically expected mobility in monolayer phosphorene at room temperature is about 250 cm²/Vs regardless of the carrier density. It should be noted, that any ionized charged impurity scattering becomes only dominant at temperatures below 100 K and is at room temperature negligible [32]. Comparison of this value with our experimental (average) mobility suggests therefore that indeed the main scattering contribution of the charge carriers is in-plane electron-phonon scattering, however, also that additional scattering channels exist. These are naturally occurring defects, electronic trapping sites as well as layer-to-layer hopping of free charge carriers.

Spatial distribution of charge carriers

To elucidate the origin of the observed conductivity and carrier density increase, we first note that due to the presence of a finite charge density between the individual layers [5,6], the electrical current carrying charges can move both within a single layer and in between neighbouring ones. In a simplified picture, such electronic

system can therefore be treated as a 3D semiconductor with some degree of in-plane preference.

As schematically shown in **Fig. 3a**, the BP devices in our experiment are bound on each side by insulating SiO₂ and PMMA, which act both as (rigid) insulating barriers for charge transfer. In nanoscaled systems, such barriers are well known to induce valence and conduction band bending and produce sub-surface space-charge (carrier depletion and accumulation) regions in semiconductor materials [33].

Consequently, in the case of BP multilayers, the few layers close to the surfaces become depleted of charge carriers, and thus do not participate in the overall charge transport [34]. Since typical space-charge regions extend into standard semiconductor body by up to a few nm, the impact of surface electrostatics is non-negligible in the case of multilayer BP and therefore must be taken into account.

In view of the above complexity, we carried out the self-consistent evaluation of the one-dimensional Schrödinger-Poisson equation [35] to estimate the extent of band bending at the interfaces of BP with SiO₂ and PMMA perpendicular to the layers (Supplementary Information).

Within our approach, we excluded the contributions from the contact interfaces and flake edges. The one-dimensional description used here was suitable as the lateral dimensions of our flakes are by orders of magnitude larger (μm) than our experimental height range (nm). The simulations were carried out for layer numbers ranging from relatively thick, bulk-like >100 layer stacks down to flakes consisting of only 10 layers (ca. 6 nm height – thinnest flakes measured in our study). We also note, that we are here outside of the band-gap opening regime for BP. For hole density we took the experimentally observed average carrier density in thick flakes of about $2 \times 10^{18} \text{ cm}^{-3}$ in the constant conductivity regime (details on parameters for model in the Supporting Information). To visualize the impact of space charge

formation on the free carrier landscape, we also estimated the spatial distribution of holes across individual flakes. Although the theoretical approach used here is simplistic, it provides not only a qualitative, but also a good quantitative description of the multilayer BP system as shown below.

Figure 3. Space charge region formation and local redistribution of majority charge carriers in multilayer BP close to the flake surfaces. a Schematic diagram depicting an electrically contacted BP multilayer on SiO₂ substrate capped with a PMMA protective layer. **b** Valence band energies within BP with layer number varied from 30 to 10. The dashed grey line denotes the VB position expected in bulk BP as reference. For clarity the flake heights are normalised as indicated by the double arrow. **c** Layer number dependence of the VB bottom energies (orange) and associated with it the augmentation of local carrier density within the central layers of BP flakes. **d** Examples of local distribution of carriers within BP flakes of different layer numbers (flake heights normalised as in b) and **e** the corresponding colour maps depicting local carrier densities within BP flakes as function of height/layer number. As the layer number is reduced, the majority of charge carriers become confined to the innermost layers. Already for flakes with heights of 10 nm the free holes are electrically confined into the innermost 1-2 layers with the carrier depleted layers not contributing to the charge transport. In all calculations the average carrier density of $2 \times 10^{18} \text{ cm}^{-3}$ was used.

Fig. 3b shows the energetic positions of the valence band (VB) with respect to the Fermi level ($E_F = 0$) and the extent of band bending at the SiO₂/BP and BP/PMMA

285 interfaces for flakes with varying layer number (for completeness, bulk-VB is also
 286 plotted as a limit of very thick flakes as grey dashed line).
 287 The VB bends upwards at both interfaces, indicating the presence of charge
 288 accumulation close to the surfaces. As the flake height is reduced, the VB bottom
 289 (corresponding to the central parts of the flakes) is shifted towards the Fermi level,
 290 and no flat-band (potential) landscape is observed within the flake centre for flakes
 291 with *ca.* 35 to 45 layers, which falls close to our experimentally observed onset of
 292 conductivity increase. With further reduction of layer number (below *ca.* 25 layers),
 293 also the VB edges at both surfaces move closer to the Fermi level, and for flakes with
 294 10 to 11 layers (corresponding to the thinnest flakes measured in our study), the VB
 295 appears to cross the E_F close to the flake surfaces.
 296 In **Fig. 3c** the distance of the VB bottom (orange line) from E_F is plotted as function of
 297 layer number. The energetic separation of the VB from the Fermi level is well-known
 298 to be intimately connected with the charge carrier density [33]. Based on that, we
 299 estimated the effective augmentation of the charge carrier density, $n_{\text{centre}}/n_{\text{bulk}}$, in the
 300 central layers of the flakes as compared to the corresponding bulk value. As shown in
 301 **Fig. 3c**, we would expect up to a 3-fold increase in the charge carrier density within
 302 the central layers for the thinnest flakes. Remarkably, this is of the same order as
 303 observed in our experiments, which further supports the validity of our approach.
 304 For completeness, we had a closer look at the spatial distribution of charge carriers
 305 among the layers in individual flakes. **Fig. 3d** shows the (local) spatial distribution,
 306 n_{loc} , of free charge carriers (holes) in flakes with varied layer numbers. In all cases,
 307 there is a distinct carrier depletion region beneath both surfaces of the flakes. For
 308 thicker flakes (>75 layers) two narrow charge carrier accumulation layers are formed
 309 below the depleted regions, as seen in the ‘rabbit ear’ curve shapes, while the hole
 310 density in the central layers remains constant. With height reduction, however, the

free charge carriers are squeezed towards the central layers due to the electrostatic potential at the surface, increasing the hole density in the centre. Eventually, for layer numbers below 18, a delta-like dopant profile is observed (**Fig. 3e**).

Since within the depletion region the electrical current flow is by large negligible [34], the findings imply that the charge carriers in the thin BP flakes are spatially limited to the innermost layers only. This effect becomes more prominent for flake heights smaller than about 20 nm, remarkably consistent with our experimental findings that σ and n suddenly increase (**Fig. 2c** and **d**, respectively). In fact, the calculations suggest that for flake heights as large as 10 nm already only one to two central phosphorene layers are able to carry the electrical current since all mobile charges are confined to these.

This finding is crucial, as it reveals that an electrostatically protected, quasi-2D hole gas confined to only one or two monolayers of phosphorene in the middle of a multilayer flake can be achieved in flakes with about 10 nm height. In particular, it is then this 2D hole gas which carries (predominantly) the electrical current in the entire system. It is worthwhile noting that the confinement of the mobile charge carriers to only one or two layers within the flake does not necessarily imply that these layers experience a bandgap opening such as isolated monolayer phosphorene [14]. The layers here are still embedded within a larger structure, that is, are subject to the inherent interlayer interactions present in BP. Therefore, the bandgap of these layers to which the 2D hole gas is confined is expected to have still its bulk-value [36].

Remarkably, this (rather simplistic) theoretical divagation reflects the experimentally observed phenomena. Indeed, the development of space-charge regions close to the flake surface can only be possible due to the quasi-3D nature of the BP system. In other words, the combined experimental and model data unequivocally prove the non-pure van der Waals character of interlayer bonding in BP.

Summarizing, our findings demonstrate that due to non-negligible interlayer coupling in multilayer BP, an unprecedented increase in the electrical conductivity with decreasing number of layers is found, where the flow of charge carriers is not entirely limited to the constituting monolayers. Consequently, the presence of an insulating environment induces the formation of charge depletion regions extending beneath the flake surfaces. The observed experimentally drastic increase of electrical conductivity and effective charge carrier density with reduction of layer number is a natural consequence of electrostatic confinement of free holes towards the central BP layers. Most notably, already in BP flakes as thick as 10 nm, a naturally encapsulated quasi-2D hole gas can be expected, as the free charge carriers are limited to one or two central layers. This gas should be by large protected from external influences and preserve the bulk electronic properties of BP such as a narrow bandgap. This is further corroborated by the limited, layer-number and charge-carrier density independent hole mobility at room temperature. Its limitation in this temperature range is associated with electron-phonon scattering [28,29] and, in particular, its magnitude coincides well with according theoretical predictions [29]. Remarkably, we demonstrate that this phenomenon holds at room temperature, which opens the perspective to re-evaluate the use of multilayered BP in particular and comparable multi-layered semiconductor materials for on-chip integrated applications in general.

METHODS

Sample preparation

BP flakes were mechanically cleaved from high purity single crystals (2D Semiconductors), transferred onto SiO₂(300 nm)/Si(n++) substrates and then capped with ca. 300 nm PMMA. This entire process was carried out under Ar atmosphere

within a glove-box. Individual flakes covered under PMMA were identified *via* optical means and electrically contacted by standard electron-beam lithography. As electrode material electron-beam-evaporated Ti/Au (5 nm/50 nm thick) was used. Immediately after fabrication the devices were capped again by a PMMA layer (ca. 300 nm) and baked-out.

Electrical measurements

Electrical measurements were carried out with a device-protecting PMMA layer protected devices on a standard probe-station. Driving and gate voltage were applied by K2450 source-units. Voltage drops and current were measured with the aid of K2000 digital multimeters and SR530 current-voltage amplifiers. Both, conductivity and mobility were measured by 4-point probing independently. Conductivities were measured with zero gate bias in an open-circuit configuration. All devices were electrically contacted in standard Hall-bar configuration, 2 oppositely oriented source/drain electrodes for the driving voltage and 4 voltage probes inbetween (cf. Figure 1b and inset Figure 1d). The voltage probes were used to measure the 4-point voltage drop V_{4p} along the sample when current I (measured simultaneously) was flowing through the device. The intrinsic conductivity then reads $\sigma = G_{4p} \frac{L}{wh}$ where $G_{4p} = \frac{I}{V_{4p}}$, L is the channel length (distance between probing electrodes), w the channel width (distance between probing electrodes) and h is the flake height as measured by AFM. The intrinsic 4-point mobility was extracted from the linear (subthreshold) slope of the G_{4p} vs. backgate voltage V_g curve using the expression

$$\mu = \left| \frac{\partial G_{4p}}{\partial V_g} \right| \frac{L}{w} \frac{t_{ox}}{\epsilon_r \epsilon_0} \text{ where } t_{ox} = 300 \text{ nm is the thickness of the SiO}_2, \epsilon_r = 3.9 \text{ the dielectric}$$

constant of SiO_2 and ϵ_0 the vacuum permittivity. The carrier density n was obtained via $\sigma = en\mu$ where e is the electron charge. By intrinsic we refer here to the electronic transport properties as inherent to the real material under study under the given experimental conditions.

AFM measurements

The height of each flake was determined by a Park Systems NX10 AFM directly after the electrical measurements after stripping the protective PMMA layer.

Raman microscopy

Raman spectroscopic characterisation was carried out on a Horiba Jobin Yvon LabRAM Aramis confocal Raman microscope (excitation wavelength: 532 nm) with a laser spot size of $<1 \mu\text{m}$ at room temperature. To not compromise the Raman measurements the spectra were taken without the removal of the device-protecting PMMA layer. No relative position adjustment of flake and laser beam to account for the in-plane anisotropy of BP was undertaken.

Schrödinger-Poisson calculations

We used a FreeWare program available online (<https://www3.nd.edu/~gsnider/>) which solves the one-dimensional Poisson and Schrödinger equations self-consistently. We assumed the thickness of both SiO_2 and PMMA to be 300 nm and varied the thickness of the BP sandwiched between them incrementally layer-by-layer. The physical parameters of the system are listed in Table 1 in the Supplementary information.

DATA AVAILABILITY

The data supporting the findings of this study are available within the paper and its Supplementary Information file. Data are also available from the corresponding author upon reasonable request.

CODE AVAILABILITY

The FreeWare program we used is available fully online under <https://www3.nd.edu/~gsnider/>.

INCLUSION & ETHICS

This research study has included local researchers throughout the research process with well defined responsibilities. The research presented is locally relevant and is due to a profound collaboration of the local partners.

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COMPETING INTERESTS

The Authors declare no Competing Financial or Non-Financial Interests.

AUTHOR CONTRIBUTIONS

V.K. designed and supervised the study. M.K.G., L.M., T.U. and M.S. carried out device preparation, electrical characterization and AFM measurements. M.S. performed the Raman measurements and G.E. the numerical simulations. T.H., A.H., J.S., M.N., M.Sz. and M.K.G. did AFM measurements and analysis. G.A., S.W., V.L., U.M., M.R., F.H. and A.Hi. carried out the flake preparation. M.K.G. and V.K. analysed the data. All authors contributed equally to the manuscript write-up.

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