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To Whom It May Concern,

In the following, I will report and describe the details of my activities regarding my visit to CNR-Nano Pisa from June 12 until June 25 2016, inclusively. In my view, this two week stay was a great success, and most importantly it opens several new doors for important new collaborative works.

-I arrived in Pisa on Sunday June 13 and Dr. Heun graciously came to pick me up the airport. During my stay, I resided at the *Gorki pension house* in the center of Pisa, one block from the *Scuola Normale Superiore* and 10 min walk from CNR-Nano.

-In the beginning of the first week, I have had several important discussions with Dr. Heun, as well as his current Post-doctoral researchers Dr. Francesca Telesio Dr. Yuya Murata and Dr. Shaohua Xiang, and students, Stefano Guiducci and Abhishek Kumar. The purpose of the trip was mostly to write a paper regarding results that were obtained at CNR Nano Pisa in December 2015 with black phosphorus devices made at McGill University.

-I worked intensively (in an intense manner) with Dr. Telesio to write a manuscript treating on “weak localization in black phosphorus field-effect transistors”. Our results at unusual in that the power law that we can extract from extremely high quality data shows an temperature for the dephasing length to be weaker than $\frac{1}{2}$, and instead closer to $\frac{1}{3}$. With Dr. Telesio and Dr. Heun, we agreed to again re-analyse nearly all data to ensure that our results were indeed solid.

-While the results did stand, we did not have an explanation for it. On Friday June 17th Dr. Telesio and I realized that such weaker dephasing had been observed previously in quasi-one-dimensional systems such as carbon nanotubes and semiconductor/metallic nanowires. In fact, this was a great moment as black phosphorus is known to have a strong anisotropic crystal structure and as such this anisotropy was observed last year in both its electronics and optical properties. It is therefore likely that weak localization can effectively be modified by this anisotropy.

-During the second week, Drs Telesio, Heun and I have dedicated much time to explore this possibility, *i.e.* read the literature that exist in other systems. We also generated a manuscript that is now quite close to completion. The manuscript is edited into an NPJ format, and is appended with this report, albeit some sections are still unfinished due one set of analysis that still have to be finalized.

-I have also spent time during both weeks with the whole group of Dr. Heun and two of his visitors. I have had several group meetings where recent data and results from Dr. Heun' group were discussed at length. This gave rise to several intense and extremely interesting brainstorming sessions.

-I also discussed with Dr. Roddaro from CNR Nano. I was very impressed by his recent work on a control strain experiment in graphene and we have discussed possibilities to make a similar measurement but on black phosphorus atomic crystals. In fact, Dr. Roddaro came out with an ingenious scheme which I strongly feel will work. For this to happen, we will gladly prepare at McGill University some samples specially designed for this experiment and Dr. Roddaro and his team will perform the strain experiment in Pisa.

-In closing, our current collaboration is extremely strong at the moment and these two weeks have been tremendously beneficial to both the McGill and CNR Nano' research teams. We foresee in the future to continue such fruitful international collaboration that builds on our respective expertise.

Yours sincerely,

A handwritten signature in black ink, appearing to read 'G. Gervais', with a stylized flourish at the end.

Guillaume Gervais
Professor of Physics
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Quasi-one-dimensional dephasing in a black phosphorus field-effect transistor

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Weak localization was observed and determined in a black phosphorus (bP) field-effect transistor 65 nm thick. The weak localization behaviour was found to be in excellent agreement with the Hikami-Larkin-Nagaoka model for fields up to 1 T, from which characteristic scattering lengths could be inferred. The dephasing length L_ϕ was found to increase linearly with increasing hole density attaining a maximum value of 55 nm at a hole density of approximately 10^{13}cm^{-2} inferred from the Hall effect. The temperature dependence of L_ϕ was also investigated and above 1 K, it was found to decrease weaker than the $L_\phi \propto T^{-\frac{1}{2}}$ dependence characteristic of electron-electron scattering in the presence of elastic scattering in two dimension. Rather, the observed power law was found to be close to that observed previously in other quasi-one-dimensional systems. We attribute our result to the crystal structure of bP which host a ‘puckered’ honeycomb lattice formed by coupled one-dimensional atomic chains.

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The recent surge of interest in the field of 2D atomic crystals has led to a number of important advances in our understanding of solid-state physics in two dimensions. Most notably is the advent of graphene but recently several new materials have been mechanically exfoliated and transistorized down to a few, or the single atomic layer limit. These include the chalcogenides family (*e.g.* MoS₂), topological insulators such as bismuth selenide, silicene and phosphorene. For the latter, its underlying crystal structure inherited from black phosphorus (bP) can be viewed as a ‘puckered’ graphene-like crystal hosting one-dimensional chains of atoms slightly displaced in the direction perpendicular to the atomic planes. An angle-resolved study involving both electronic transport and optical measurements was recently performed in a thin field-effect bP transistor and demonstrated a substantial anisotropy [1]. Here, we have studied the weak localization in a black phosphorus field-effect transistor and we found that its coherence decays with temperature slower than what is expected for a two-dimensional material. This more robust loss of coherence is reminiscent to what was found previously in quasi-one-dimensional systems such as nanotubes and metallic nanowires.

Black phosphorus is a layered elemental allotrope of phosphorus that can be mechanically exfoliated to atomic layer thickness. Unlike semi-metallic graphene, bP is a direct gap semiconductor [2, 3]. The bandgap is

0.3 eV in bulk and increases by quantum confinement to 1-2 eV in the monolayer limit [4–6], ideal for applications in electronics and optoelectronics [7]. Black phosphorus field effect transistors (FETs) have been demonstrated by various groups [1, 8–12], with hole field effect mobilities reaching up to $1350\text{ cm}^2/\text{Vs}$ and 10^5 current modulation at room temperature [13]. In the presence of a large magnetic field normal to the bP atomic layers, Shubnikov-de Haas oscillations have been observed in bP FETs [13–16].

Magnetotransport in the presence of a weak magnetic field normal to the bP atomic layers is less well studied. In a disordered 2-D system, coherent backscattering of charge carriers gives rise to a peak in the magnetoresistance known as weak localization (WL), see [17] for a review. Previous work on weak localization in 2-D crystals has focused primarily on graphene [18–20] and has also been observed recently in molybdenum disulfide, revealing a phase coherence length L_ϕ of 50 nm at 400 mK that decays with increasing temperature as $T^{-\alpha} = T^{-\frac{1}{2}}$ [21], characteristic of electron-electron interactions in 2D. Importantly, the dimensionality plays an important role in weak localization and previous work in metal and semiconducting mesoscopic structures have shown the dephasing length exponent α to decrease from $\frac{1}{2}$ to $\frac{1}{3}$ as the system geometry was reduced towards the one dimensional limit [22], in agreement with theory of weak localization [23]. This is the central result of this work where we have observed weak localization in a 65 nm thick bP FET whose dephasing length exponent was found to be close to $\alpha = \frac{1}{3}$. We attribute this weak localization behaviour due to the anisotropic nature

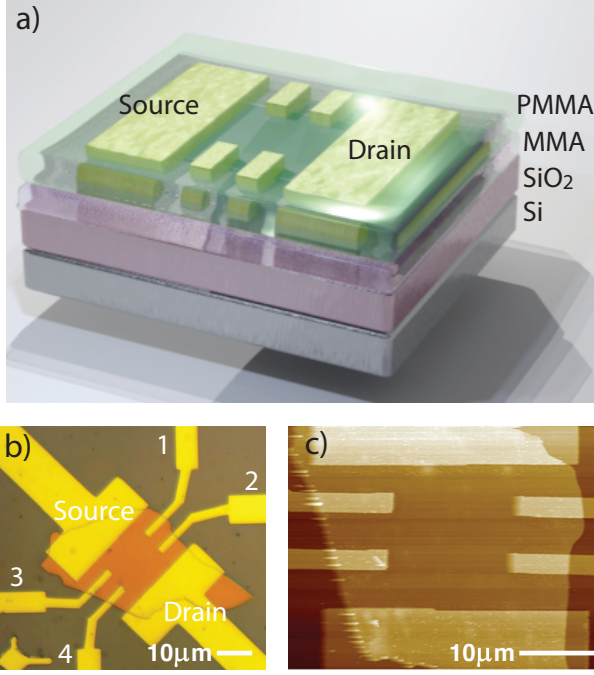


FIG. 1. **Black phosphorus field-effect transistor.** a) Schematic of the back-gated bP FET in a Hall bar geometry. b) Optical reflection image of the bP FET. The longitudinal resistance R_{xx} was measured with voltage probes 1 and 2, and the Hall resistance R_{xy} was measured with voltage probes 1 and 3. c) AFM image of the bP FET. The bP thickness was measured to be 65 ± 2 nm.

of the puckered bP atomic crystal structure forming effective one-dimensional chains in the $x - y$ plane.

The design, as well as photograph and atomic force microscope images of the bP FET is shown in Fig.1. The longitudinal resistance R_{xx} versus back gate voltage V_g is plotted in Fig. 2(a), with sample temperature T as a parameter. The sample is highly resistive for $V_g > -30$ V, and exhibits clear p-type conduction for $V_g < -30$ V. The Hall resistance R_{xy} versus magnetic field B is plotted in Fig. 2(b) at $T = 0.26$ K and at gate voltage $V_g = -40$ V and -80 V, with the component symmetric in B removed. Hole carrier density p inferred from the Hall resistance is found to depend linearly on gate voltage V_g over the range of p-type conduction $V_g < -30$ V. The field effect hole mobility, $\mu_{FE} = (L/W) \cdot \partial G_{xx} / \partial (C_g V_g)$ was found to reach a peak value of $300 \text{ cm}^2/\text{Vs}$ at a gate voltage $V_g = -70$ V, with a negligible dependence upon temperature over the measured range $0.26 \text{ K} < T < 20 \text{ K}$.

The measured weak localization peak in longitudinal resistance is observed in the plot of $\Delta R_{xx}/R_{xx}(0) =$

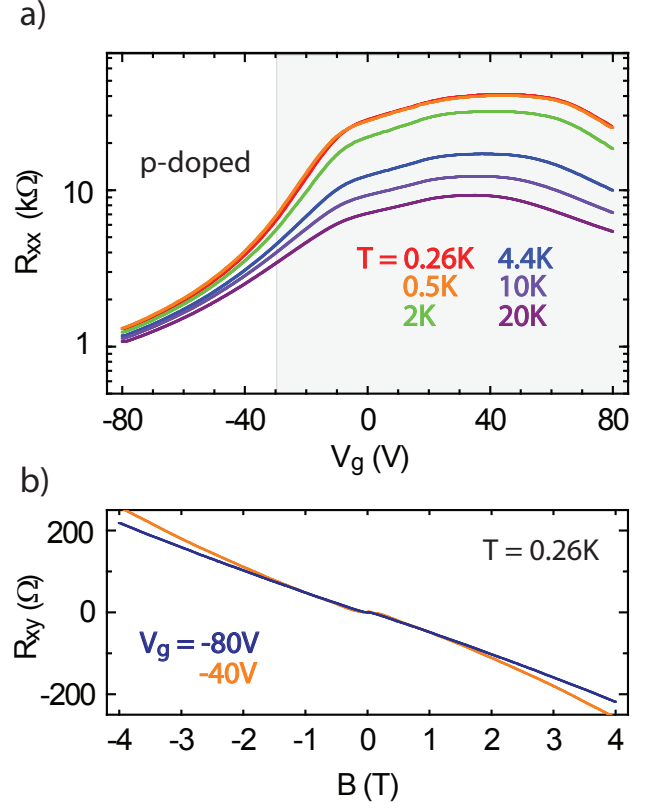


FIG. 2. **Transport measurements versus backgate voltage and temperature.** a) The measured longitudinal resistance R_{xx} versus gate voltage V_g , at temperatures $T = 0.26$ K to 20 K. The region of strong p-doping is identified. b) The Hall resistance R_{xy} versus magnetic field B at $T = 0.26$ K, with the component symmetric in B removed, as measured at $V_g = -80$ V and -40 V.

$(R_{xx}(B) - R_{xx}(0))/R_{xx}(0)$ versus magnetic field B and gate voltage V_g in Fig. 3(a). The amplitude of the WL peak increases with increasing hole density attaining a maximum value of 55 nm at approximately 10^{13} cm^{-2} hole density. The temperature dependence of the WL peak at $V_g = -80$ V and $V_g = -40$ V is plotted in Fig. 3(b) and Fig. 3(c), respectively. The WL correction to the resistance decreases with increasing temperature eventually disappearing at temperatures above 20 K, as expected.

The Hikami-Larkin-Nagaoka theory [17, 24, 25] gives a quantitative prediction for the WL correction to the sheet conductance,

$$\Delta\sigma = -\frac{e^2}{2\pi^2\hbar} \left(\Psi\left(\frac{1}{2} + \frac{B_1}{B}\right) - \Psi\left(\frac{1}{2} + \frac{B_2}{B}\right) + \frac{1}{2}\Psi\left(\frac{1}{2} + \frac{B_3}{B}\right) - \frac{1}{2}\Psi\left(\frac{1}{2} + \frac{B_2}{B}\right) \right), \quad (1)$$

where Ψ is the digamma function. The field parameters

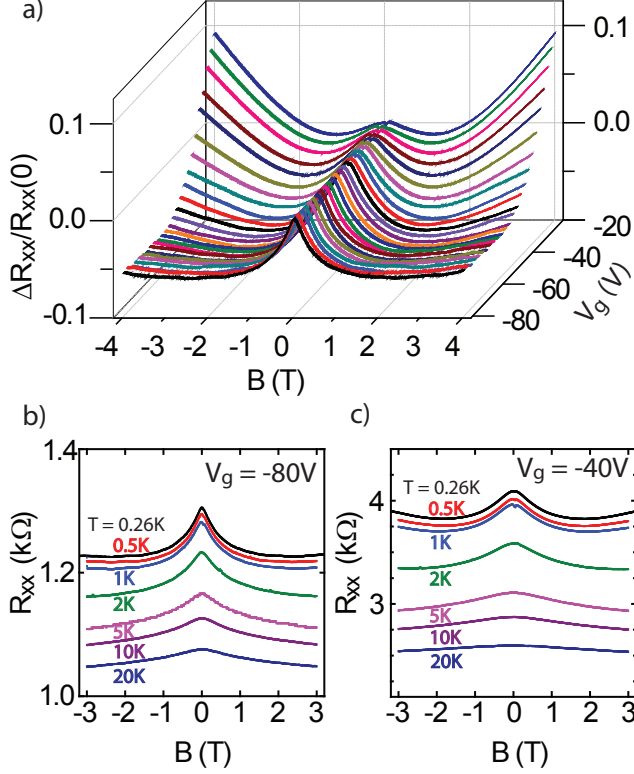


FIG. 3. **Weak localization measurements.** a) A weak localization peak is observed in a plot of the normalized longitudinal resistance $(R_{xx}(B) - R_{xx}(0))/R_{xx}(0)$ versus magnetic field B and gate voltage V_g at $T = 0.26$ K. The temperature dependence of the longitudinal resistance R_{xx} versus magnetic field B at b) $V_g = -80$ V and at c) $V_g = -40$ V.

in the above expression are given by:

$$B_1 = B_0 + B_{so} + B_s \quad (2)$$

$$B_2 = \frac{4}{3}B_{so} + \frac{2}{3}B_s + B_\phi \quad (3)$$

$$B_3 = 2B_s + B_\phi \quad (4)$$

where B_0 , B_{so} , B_s , and B_ϕ are the characteristic fields associated with elastic scattering, spin-orbit scattering, magnetic scattering, and inelastic scattering (dephasing), respectively.

The measured WL peak (normalized) resistances were inverted into conductivity by the usual tensor relation $\sigma_{xx} = \rho_{xx}/(\rho_{xx}^2 + \rho_{xy}^2)$, with negligible contributions from the (transverse) Hall resistivity (see 2(b)). To account for the background resistivity, the relation $\Delta\sigma = -(L/W) \cdot (R_{xx}(B) - R_\eta)/R_{xx}(B)^2$ was used, where R_η is the classical Drude resistance in the absence of a WL correction. The determination of the constant R_η is not trivial, and was thus left as a fit parameter, however it was verified that its trend in temperature followed

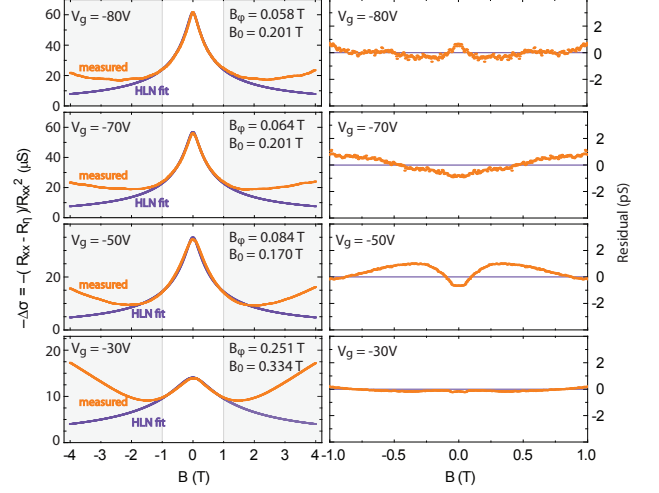


FIG. 4. **HLN weak localization fits.** The measured longitudinal conductance correction $-\Delta\sigma$ versus magnetic field B and a best fit to a Hikami-Larkin-Nagaoka theory over the field range $-1 \text{ T} < B < 1 \text{ T}$, where weak localization dominates, is shown for various gate voltages V_g and hole density p at $T = 0.26$ K. The extracted characteristic field B_0 for elastic and B_ϕ for inelastic (dephasing) scattering are indicated.

that of the measured transport mobility of the bP. This WL correction to conductivity was numerically fitted to the HLN model under the approximation of negligible spin-orbit coupling, $B_{so} = 0$, and negligible magnetic impurity scattering, $B_s = 0$. Both approximations are appropriate for our bP crystals [26]. Measurements and numerical fits of the WL contribution to conductivity $\Delta\sigma$ are plotted in Fig. 4 versus backgate voltage at temperature $T = 0.26$ K. The fit was performed over the magnetic field range $-1 \text{ T} < B < +1 \text{ T}$, where the WL feature dominates the magnetoresistance. The fit quality was evaluated over the same B -range, with an R^2 coefficient of determination of at minimum 0.99. The residuals are also shown in the right panels of Fig. 4 in parts per million.

The extracted characteristic fields B_i are related to the scattering lengths L_i by considering the phase shift of diffusing charge carriers under the influence of a magnetic field, $B_i L_i^2 = \hbar/4e$. From these fields, we can deduce an elastic as well as inelastic (dephasing) characteristic lengths for the electrons (in our case holes), L_0 and L_ϕ respectively. These lengths are plotted versus gate voltage V_g in Fig. 5(a) at $T = 0.26$ K. In particular, the dephasing length L_ϕ clearly increases with increasing hole density reaching a maximum of 55 nm , whereas L_0 remains nearly independent over a broad range of carrier density. We have also verified that L_0 scale linearly with the the transport mobility, as expected for an elastic scattering process occurring in the presence of both a

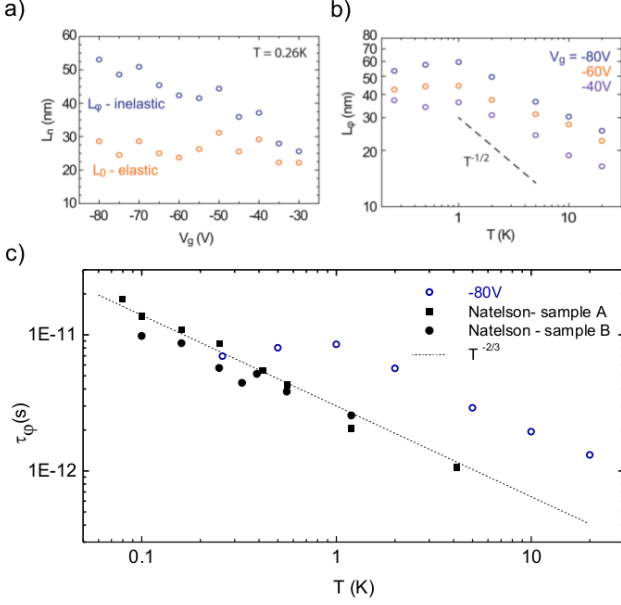


FIG. 5. **Scattering lengths and power laws.** a) The inelastic scattering length L_i and elastic scattering length L_o versus gate voltage V_g , and carrier density p , at temperature $T = 0.26$ K. b) The inelastic scattering length L_i versus temperature T at various gate voltages V_g . The $T^{-1/2}$ temperature dependence associated with electron-electron scattering in the diffuse limit is shown for comparison. c) Comparison of our data at gate voltage $V_g = -80$ V converted dephasing time τ_ϕ with those of Natelson *et al.*[22] measured in metallic nanowires in the quasi-one-dimensional regime. The dotted lines shows a dephasing time decaying as $T^{-2/3}$, *i.e.* corresponding to a power law exponent $\alpha = \frac{1}{3}$ for L_ϕ .

phonon bath and disorder.

The most intriguing aspect of the data is certainly the temperature dependence of L_ϕ shown in Fig. 5(b) in a log-log plot at fixed gate voltages $V_g = -40, -60, -80$ V. The error bars are an interval of confidence from a visual inspection of the fit when varying one parameter and keeping all other constant. The saturation of L_ϕ at temperatures below 1 K is most likely due to impurities, as is been previously observed in a variety of metallic and semiconducting 2D systems. However, at temperature above 1 K the temperature dependence of L_ϕ does not follow the $T^{-1/2}$ (dashed line) behaviour expected *a priori* from electron-electron scattering in the presence of elastic scattering. A weighted fit of data rather show a dephasing length exponent α corresponding to $0.27 \pm 0.01, 0.22 \pm 0.01, 0.29 \pm 0.01$ (and R^2 coefficient all greater than 0.99) at gate voltages $V_g = -40, -60, -80$ V, respectively.

For weak localization occurring in typical disordered

two-dimensional systems, the dephasing length is related to the inelastic scattering time via $L_\phi^2 = D\tau_\phi$, where D is the elastic diffusion coefficient. Electron-electron scattering in the absence of elastic scattering is expected to give a scattering rate $1/\tau_i \propto T^2$, and hence $L_\phi \propto T^{-1}$ [27]. In the presence of strong elastic scattering, appropriate here since $L_o < L_i$, the electron-electron scattering rate is expected to follow $1/\tau_i \propto T$ [23, 28, 29], and hence $L_i \propto T^{-1/2}$. This characteristic inelastic scattering rate has been observed in graphene [18–20, 27] and MoS₂ [21], however our weak localization measurements in a black phosphorus thin films are clearly not following this trend.

Our WL data reveal the presence of an inelastic scattering mechanism that is characterized by a weaker temperature dependence, leading to a coherence loss slower than $T^{-1/2}$. This contrast with previous measurements in a few-layer bP films [30] where this power law was deduced, albeit with saturation of L_ϕ at higher temperatures than ours (5 K as opposed to 1 K in our case) and by making use in their analysis of a fitting parameter describing valley degeneracy in bP. This parameter was found to differ from unity (no valley degeneracy) and to vary from ~ 1.2 to 0.3 over the range of temperatures where the power law exponent for L_ϕ was extracted. To the best of our knowledge, the band structure of bP has never shown any hint of valley degeneracies and as such one can question whether an intrinsic $T^{-1/2}$ behaviour was observed in Ref.[30].

A weaker loss of the coherence versus temperature, as compared to the 2D case, has previously observed in the presence of a strong 1D confinement potential, *e.g.* in carbon nanotubes [31], and L_ϕ shown to be well ascribed per the theoretical prediction of a $T^{-1/3}$ power law. Perhaps more remarkably is the systematic study undertaken by Natelson *et al.*[22] whereby the dephasing time in metallic nanowires was measured as a function of the wire width down to 5 nm. In this regime of width and low temperatures, several heuristic lengthscales can be estimated placing the wires well into a quasi-one-dimensional regime. In this study a lower saturation temperature was observed however the dephasing time τ_ϕ was determined to follow closely a $T^{-2/3}$ power law, corresponding to a dephasing length exponent $\alpha = \frac{1}{3}$. These data, together with our own taken at -80 V backgate voltage and converted into dephasing times are shown for comparison in Fig. 5(c). The expected $T^{-2/3}$ power law predicted from weak localization in quasi-1D is also shown as a dotted line.

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Methods

Our bP FETs were prepared by mechanical exfoliation of bulk bP crystals onto a degenerately doped Si wafer with a 300 nm SiO₂ layer prepared by dry thermal oxidation. The bP crystals were prepared by heating commercially available red phosphorus in a muffle oven, together with a tin-gold alloy and a catalytic amount of SnI₄, following a published procedure [32]. The solids were charged in a quartz tube, which was then evacuated, put in the oven and heated up to 650°C. The sample stayed for three days at this temperature in the oven. Afterwards, a slow cooling rate was chosen (0.1°C/min) to afford the formation of good shape crystals of BP (typical size: 2 mm × 3 mm). Exfoliation was performed in a nitrogen glove box to suppress photo-oxidation, and the SiO₂ surface was treated with a hexamethyldisilazane (HMDS) layer to suppress charge transfer doping. Standard electron beam lithography was used to define 5 nm Ti/ 80 nm Au metal electrodes in a Hall bar geometry. The samples were made environmentally stable by encapsulation with 300 nm of methyl methacrylate (MMA) and 200 nm of polymethyl methacrylate (PMMA). A schematic of the device structure is shown in Fig. 1(a), with an optical micrograph shown in Fig. 1(b) and an atomic force microscope (AFM) image in Fig. 1(c). The thickness of the bP was determined to be 65 ± 2 nm by AFM. Importantly, the hole accumulation layer induced in a bP FET at cryogenic temperatures is estimated to be < 3 nm thick, corresponding to a hole gas occupying 5-6 bP layers [33]. Charge transport measurements were performed using standard ac lock-in techniques in a ³He cryostat with sample temperature ranging from $T = 0.26$ K to $T = 20$ K. A 100 nA ac current at frequency $f = 17$ Hz was applied through the source and drain terminals of the bP FET and both the longitudinal and transverse (Hall) voltages were measured. A dc gate voltage V_g was applied to tune the carrier density through the back gate capacitance $C_g = 11.5$ nF/cm².

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Supplementary information