

Anomalous impurity-induced charge modulations in black phosphorus

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We observe anomalous charge modulations induced by ionized indium impurities on the surface of the semiconductor black phosphorus by scanning tunneling microscopy (STM). When the impurities are switched into a negatively charged state by the STM tip, periodic charge modulations emerge around the impurity center, but strictly confined by the nanoscale impurity potential. These modulations form a distorted triangular pattern, whose periodicity remains unchanged in a wide range of positive bias. Furthermore, these local charge orders exhibit an anisotropy opposite to that expected based on the anisotropy of the Fermi surface, challenging a simple band-structure interpretation. Our experiment demonstrates the possibility of creating and manipulating macroscopic charge orders through impurity engineering.

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Introduction. Impurities in low-dimensional electronic materials often act as localized perturbations that reorganize the surrounding charge density or atomic lattice. Such rearrangements can appear as standing-wave-like charge density modulations [1–3], localized impurity states [4], or impurity-induced lattice distortions [5,6], depending on the strength and spatial extent of the impurity potential. Understanding these responses is essential for determining how impurities enable tuning material properties and for revealing the mechanisms behind impurity-driven electronic behavior [7–10]. Despite extensive studies on impurity-induced electronic signatures in various materials, there are still open questions concerning the microscopic mechanism by which an individual impurity reshapes the nearby electronic environment, particularly in systems with strong structural or electronic anisotropy.

Black phosphorus (BP) provides an advantageous platform for investigating these effects. Its puckered structure generates markedly anisotropic effective masses, yielding distinct Fermi velocities [11,12] and screening behaviors depending on crystallographic axes [13,14]. With scanning tunneling microscopy (STM), one can directly tune the ionization state of a surface impurity and thereby control the associated Coulomb (or scattering) potential [15]. This enables real-space visualization of how electrons respond to an impurity whose

charge state and potential profile can be manipulated with an atomic precision. Previous studies have examined impurity charging and scattering in BP [16,17], but it remains uncertain whether an ionized impurity can induce anisotropic charge modulations expected from the intrinsic band anisotropy.

Here, we employ STM to control the charge state of individual indium clusters on BP and uncover anomalous charge modulations. These modulations form a distorted triangular order, remain sharply confined within the tip-induced band-bending region, and exhibit an energy-independent wavevector. Notably, their spatial anisotropy follows the zigzag direction, contrary to expectations based on the effective-mass anisotropy and known screening behavior of BP [14]. These findings show that the tunability of impurity charge states provides a powerful means to elucidate impurity-induced electronic modulations unaccounted for by standard scattering or screening models.

Results and discussion. We begin with a two-dimensional electron system on the surface of black phosphorus. BP is a predominantly p-type semiconductor, and its low-energy electronic band structure can be approximately modeled as simple parabolic dispersions for both conduction and valence bands [11,12]. The (001) surface of BP exhibits a puckered honeycomb lattice structure [inset of Fig. 1(a)], resulting in two distinct directions, designated as zigzag (ZZ) and armchair (AC). One peculiar characteristic of BP's electronic structure is its anisotropy along these two directions, manifested by a substantial difference in the effective mass of the conduction band electrons $m_{AC}^* = 0.07m_e$ for the armchair direction and $m_{ZZ}^* = 1.0m_e$ for the zigzag direction [18], where m_e is the free electron mass. Next, we introduce impurities on BP by depositing indium atoms, which form either clusters on the surface with a diameter of 1~5 nm, or triangular islands with size over 5 nm [19].

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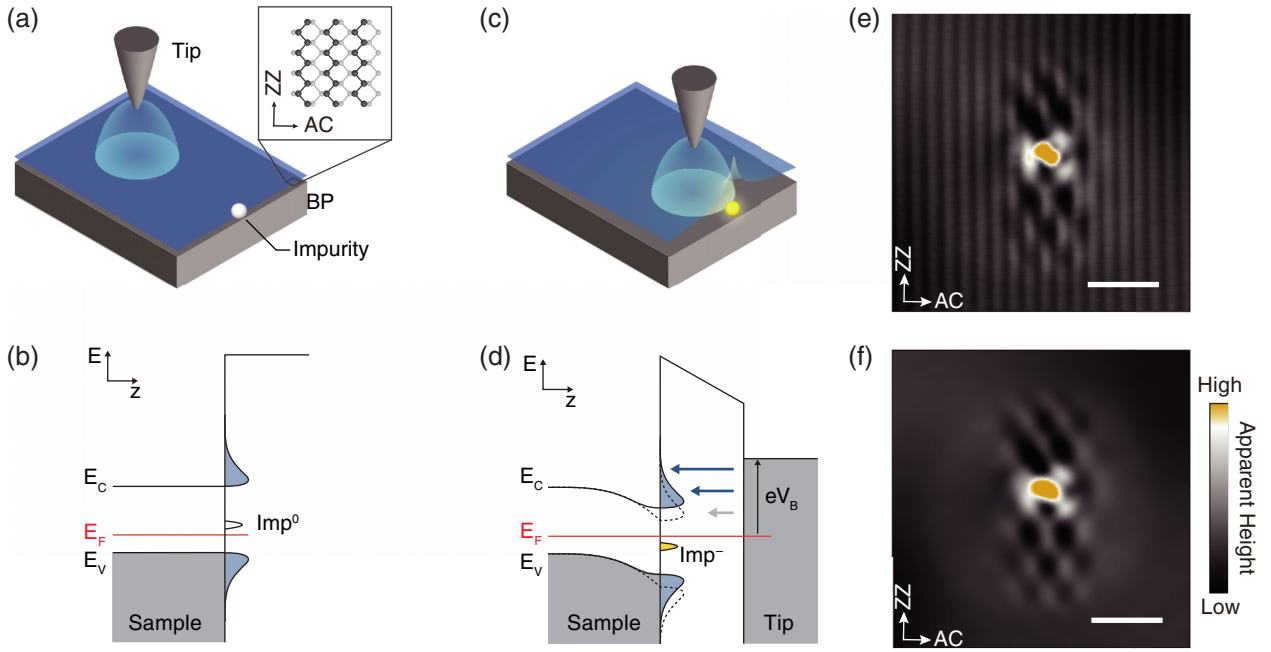


FIG. 1. Charge modulations near ionized adatom impurities. (a) Schematic illustration of the experimental setup. The STM tip is brought close to the sample (gray), and the electric field between them (cyan) locally penetrates the sample. A neutral impurity (white ball) is placed on the surface. When the tip is away from the impurity, the surface electrons (blue) remain spatially uniform. The inset shows the crystal structure of the (001) surface of the black phosphorus (BP) samples used. The darker and brighter balls represent surface and subsurface phosphorus atoms, respectively. (b) Local energy diagram near the impurity for the case in (a). E_C and E_V denote band edges of the conduction (white) and valence (gray) bands, respectively. The red line indicates the Fermi level, E_F . The blue shades illustrate the density of states on the surface. The impurity level (white) above E_F indicates the impurity's charge-neutral (Imp^0) state. (c) Ionization (yellow ball) occurs when the local electric field reaches the impurity, and the impurity's Coulomb potential disturbs locally the energy of surface electrons (blue). (d) Local energy diagram near the impurity for the case in (c). V_B is the applied bias voltage. Ionization (Imp^-) is represented by the impurity level (yellow) moving below the E_F . While the dashed curves illustrate the tip-induced band bending, the Coulomb potential of ionized impurity further lifts the bands (solid curves). The blue arrows represent the tunneling channels, with their lengths indicating the tunneling probabilities. (e) STM image taken near indium adatoms showing charge modulations. Setup conditions: $V_B = 0.7$ V, $I_{\text{set}} = 0.1$ nA. (f) Fourier-filtered image of (e), highlighting the charge modulations. Scale bar: 2 nm.

We then proceed to switch the ionization state of the surface impurities. A surface impurity creates a strong Coulomb potential, but only when the impurity is charged. A neutral impurity, otherwise, would only weakly perturb the electron density around it. Experimentally, it is well established that the ionization state of an impurity in a semiconductor can be controlled by STM [15,20,21], as illustrated in Fig. 1. When an STM tip approaches the sample surface, the strong electric field (typically on the order of 1 V/nm) exerted by the tip extends toward the sample. If screening is insufficient, this field penetrates the local surface area under the tip [Fig. 1(a)], modifying the local potential inside the sample. This often results in a rigid shift of the conduction and valence band edges [dashed lines in Fig. 1(d)], known as tip-induced band bending (TIBB) [22]. When the impurity is far from the tip, it stays neutralized with the surrounding background [Fig. 1(b)]. However, when the spatial extension of the tip's field reaches the impurity [Fig. 1(c)] and shifts down the lowest unoccupied impurity level across E_F , the impurity becomes negatively ionized. This ionization subsequently enhances the impurity's Coulomb potential, which in turn lifts the bands at the surface. The combination of tip- and impurity-induced band bending effects have been understood in previous reports [23,24], as shown by comparing the solid and dashed curves in Fig. 1(d).

In this case, as the tunneling channels with higher transmission probability (blue arrows) align closer to a peak in the surface density of states (blue shades) [25], the tunneling current is instantaneously increased compared to the case before ionization [19]. In constant-current mode, the feedback control of STM immediately retracts the tip. It is then anticipated that this change in current occurs roughly at the same distance from the impurity in all directions, manifesting as a disklike feature with enhanced contrast in the STM image [15,20,21]. As illustrated in Fig. 1(e), this is indeed what we observed around a dimer impurity. Behind the vertical stripes originating from the zigzag ridges and valleys, we observe a faint disklike feature around the impurity. The area inside the disk exhibits a larger apparent height at positive bias voltages, which is better discernible after filtering out the surface corrugation in Fig. 1(f) [19].

Charge modulations near ionized impurities. The primary finding of this study is the discovery of additional modulations in the vicinity of the impurity, which exist within the commonly observed disklike feature. These modulations can be clearly visualized as oscillations in both filtered and unfiltered images in Figs. 1(e) and 1(f), as well as in differential conductance images taken simultaneously [19]. They appear to form an approximately triangular lattice that is distorted

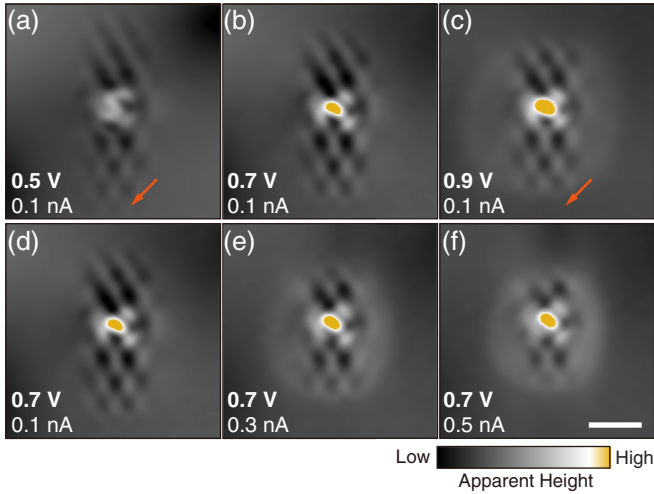


FIG. 2. Charge modulations controlled by the electric field between tip and sample. (a)–(c) Fourier-filtered STM images taken at a fixed current setpoint but with varying bias voltages. As the bias voltage increases, the size of the disk decreases. The orange arrows indicate that a charge modulation peak shown in (a) disappears at the same location in (c). (d)–(f) Fourier-filtered STM images taken at a fixed bias but with varying current setpoints. Scale bar: 2 nm.

because of the underlying orthorhombic surface Bravais lattice, leading to a centered rectangular charge modulation; additional distortions are visible at the disk edge and near the central impurity. One important observation is that these modulations are strictly confined to the disk. To demonstrate this, we change the disk radius by varying the bias voltage or the tip-sample distance, as TIBB depends on the electric field between the tip and sample.

As shown in Fig. 2, the disk shrinks in size with an increasing bias or a decreasing tip-sample distance (increasing current setpoint). This band-bending behavior confirms that the impurity is negatively charged when the tip is positioned inside the disk [19]. As the disk size decreases, the area that contains the modulations also shrinks accordingly. This confinement is clearly visible when comparing the same field of view, for example, at a fixed current setpoint but increasing bias in Figs. 2(a)–2(c). In Fig. 2(a), the modulations extend to over 4 nm away from the impurity along the zigzag direction; a density peak appears at the location indicated by the arrow. In contrast, in Fig. 2(c) the modulations disappear at the same location as the disk shrinks. We observe the same phenomenon when comparing images taken at a fixed bias of 0.7 V but with increasing current setpoint [Figs. 2(d)–2(f)]. These observations rule out the possibility of structural deformation and suggest an electronic origin of the modulations.

Possible origin of the charge modulations. To discuss possible electronic mechanisms, we first consider Friedel oscillations, often referred to as quasiparticle interference (QPI) and widely utilized in determining electronic band structures by STM [1–3]. Specifically, the oscillations should have a scattering wave vector $\mathbf{q}(E) = \mathbf{k}_f(E) - \mathbf{k}_i(E)$, connecting the wave vectors $\mathbf{k}_i(E)$ and $\mathbf{k}_f(E)$ of the initial and final states at a certain energy E in the energy bands, respectively. A Fourier transform of the real-space oscillation pattern at each energy reveals the dispersion relation $\mathbf{q}(E)$, which can be used

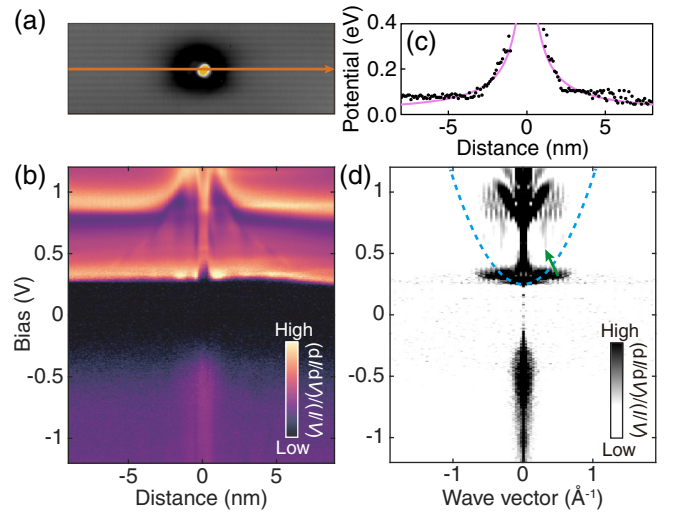


FIG. 3. Energy dependence of charge-modulation periodicity. (a) STM image taken near a two-atom impurity. Setup conditions: $V_B = 1.2$ V, $I_{\text{set}} = 0.1$ nA. (b) The normalized differential conductance $(dI/dV)/(I/V)$ spectra along the line marked by the arrow in (a). Setup conditions: $V_B = 1.2$ V, $I_{\text{set}} = 0.2$ nA. (c) Band edge (black dots) appearing near $V_B = 0.8$ V in (b), extracted by finding the bias that corresponds to a maximum in the first derivative of each spectrum above 0.7 V. The impurity potential (purple) is estimated by a screened Coulomb potential [19]. (d) Linewise Fourier transform of (b). The periodic feature of the charge order is marked with the green arrow. The blue dashed curve corresponds to the expected dispersion of quasiparticle interference along the zigzag direction in the conduction band.

to reconstruct the energy bands. In our case, an anisotropic parabolic dispersion mode is expected from the first-principle calculations and photoemission experiments [11,18].

To obtain the wave vector $\mathbf{q}(E)$ associated with our observed modulations, we measure the differential conductance along a line in the zigzag directions where modulations appear [Fig. 3(b)] and then perform a linewise Fourier transform [Fig. 3(d)]. We first note the conduction band that starts at 0.3 V, and slightly bends upwards right on top of the impurity; this results in a flat dark feature in the Fourier transform in Fig. 3(d). We then focus on the bias range of 0.5 V–1.2 V where the modulations appear. The spatial extent of the modulations diminishes due to the reduced disk radius with increasing bias [Fig. 3(b)], as also indicated by the V-shaped dark feature with multiple sidebands in Fig. 3(d) due to quantum confinement effects. Most importantly, we note an almost constant wave vector $|\mathbf{q}| \sim 0.32 \text{ \AA}^{-1}$ that corresponds to the observed modulations. This energy-independent mode clearly deviates from the expected parabolic dispersion [blue dashed line in Figs. 3(d)], thus indicating that trivial Friedel oscillations would not suffice to explain the observed modulations. Interestingly, it is known that the QPI signal can be significantly influenced by quantum geometric form factors [26]. In this case, we do find the possibility that an apparent energy-independent mode appears in our simulated QPI. Specifically, nonlocal impurity potentials with selective orbital couplings suppress scatterings along the zigzag direction [19].

Furthermore, our results show a spatial anisotropy opposite to what one would expect from the Fermi surface. Due to the anisotropy of the effective masses along the zigzag and armchair directions, the Fermi surface (of doped BP) has an elliptical shape elongated along the zigzag directions [11,27]. This means that the screening of the extra charges by band electrons should have a longer decay length along the armchair direction [14], and that any charge modulation, e.g., originating from Fermi surface nesting, should have a longer length scale along the armchair direction. Surprisingly, upon close inspection of the charge modulations in Fig. 1(f), we find that the individual density peaks have an elliptical shape elongated along the zigzag direction instead of the armchair direction. In addition, the modulations extend to the boundary of the disk along the zigzag direction but are more confined spatially along the armchair direction. Inside the disk, we also observe a smooth transition from modulations to a relatively uniform background when moving toward the disk edge along the armchair direction. Both the spatial extent of the modulations and the shape of the individual density peaks are intrinsic to the charge orders and irrespective of the appearance of the impurity, as confirmed by imaging charge modulations around different impurities and nanoislands [19]. While local impurities can mix bands in a way consistent with a shorter wavelength along the armchair direction [12,28], there is no indication of such a strong redistribution across the band gap, which stays open with a nearly constant size when approaching the impurity [Fig. 3(b)].

We also consider whether enhanced interaction effects give rise to the charge modulations. The presence of a strong, localized impurity potential can trap electrons and one might hypothesize that it simultaneously reduces the screening of Coulomb interactions between them, potentially stabilizing a charge-ordered phase within the confined region. This interpretation is supported by our observation of the modulations being strictly confined by the potential of the ionized impurity (Fig. 2). From the differential conductance plot [Fig. 3(b)], we can extract the impurity potential, which follows the expected screened Coulomb form at the dielectric-vacuum interface [15,29]. Assuming a tip-sample distance of 0.5 nm, and that each atom in the indium dimer acquires one electron charge after ionization (see the Supplemental Material [19] and also Refs. [16,30–38] therein), we obtain a potential profile shown as the curve in Fig. 3(c). The expected impurity potential aligns well with a band edge appearing at a bias of ~ 0.8 V, indicating the effect of the impurity potential acting on this band.

Finally, while the charge modulations exhibit features reminiscent of a Wigner crystal [39]—including a distorted triangular lattice incommensurate with the host lattice—quantitative analysis reveals fundamental discrepancies: if each peak in Fig. 1(f) in the disk corresponded to a “frozen” electron, we would have an electron density of $5 \times 10^{13} \text{ cm}^{-2}$; this corresponds to a Wigner-Seitz radius $r_s \approx 0.5$, which is at least an order of magnitude lower than what is required for crystallization [40–42]. The unique appearance of these charge modulations near indium impurities—absent for phosphorus vacancies [13], ionized potassium [14], or cobalt adatoms [43]—suggests that the underlying mechanism likely involves an intricate interplay between (at least a subset of)

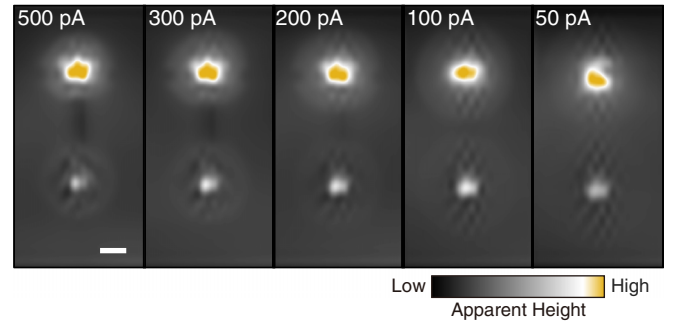


FIG. 4. Fourier-filtered STM images taken around two nearby cluster impurities. The bias is fixed at 0.6 V, while the current setpoint decreases from 500 pA to 50 pA. The charge modulations around both impurities are initially separated at 500 pA, and gradually expand their extensions and start to merge at 50 pA. Scale bar: 2 nm.

the nonlocal impurity potential, quantum geometric effects, and enhanced local electronic correlations.

Manipulation of charge modulations. To complete the story, we demonstrate how the impurity potential provides a microscopic means of manipulating the charge orders. To do so, we monitor the evolution of charge modulations around two nearby clusters (Fig. 4) while controlling the spatial profile of the impurity potentials.

As previously demonstrated in Fig. 2, we can expand the spatial extent of the impurity potential by reducing the size of the TIBB disk. With a decreasing current setpoint, we find that the charge modulations around the two clusters extend toward each other. For example, at a setpoint of 50 pA, the modulations eventually merge. This microscopic enlargement of the charge modulations implies that, through impurity engineering such as nanopatterning [44], a macroscopic phase of such charge order can form with an appearance analogous to a charge density wave.

Conclusions. In conclusion, we present a direct observation of anomalous charge modulations near indium impurities in black phosphorus. The charge modulations appear as a distorted triangular lattice, with an apparent anisotropy opposite to that expected based on the Fermi surface’s anisotropy. This unexpected spatial anisotropy and the energy-independent behavior of these modulations cannot be explained by standard scattering between the quasiparticle energy bands. Instead, it might be the result of nontrivial wave-function effects, the interplay of localization and strong-coupling physics, or a combination of both. Our work establishes a versatile experimental platform for exploring how impurities can stabilize intriguing charge orders in a system with strong intrinsic anisotropy.

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Data availability. The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

- [1] Y. Hasegawa and P. Avouris, Direct observation of standing wave formation at surface steps using scanning tunneling spectroscopy, *Phys. Rev. Lett.* **71**, 1071 (1993).
- [2] M. Crommie, C. Lutz, and D. Eigler, Imaging standing waves in a two-dimensional electron gas, *Nature (London)* **363**, 524 (1993).
- [3] J. E. Hoffman, K. McElroy, D.-H. Lee, K. M. Lang, H. Eisaki, S. Uchida, and J. C. Davis, Imaging quasiparticle interference in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$, *Science* **297**, 1148 (2002).
- [4] D. Kitchen, A. Richardella, J.-M. Tang, M. E. Flatté, and A. Yazdani, Atom-by-atom substitution of Mn in GaAs and visualization of their hole-mediated interactions, *Nature (London)* **442**, 436 (2006).
- [5] M. Reticcioli, M. Setvin, X. Hao, P. Flaeger, G. Kresse, M. Schmid, U. Diebold, and C. Franchini, Polaron-driven surface reconstructions, *Phys. Rev. X* **7**, 031053 (2017).
- [6] Y. Zhang, Q.-Q. Guo, S.-Y. Li, and L. He, Nanoscale probing of broken-symmetry states in graphene induced by individual atomic impurities, *Phys. Rev. B* **101**, 155424 (2020).
- [7] A. V. Balatsky, I. Vekhter, and J.-X. Zhu, Impurity-induced states in conventional and unconventional superconductors, *Rev. Mod. Phys.* **78**, 373 (2006).
- [8] F. Evers and A. D. Mirlin, Anderson transitions, *Rev. Mod. Phys.* **80**, 1355 (2008).
- [9] O. V. Yazyev and L. Helm, Defect-induced magnetism in graphene, *Phys. Rev. B* **75**, 125408 (2007).
- [10] S. Park, M. Huh, C. Jozwiak, E. Rotenberg, A. Bostwick, and K. S. Kim, Electronic rotons and wigner crystallites in a two-dimensional dipole liquid, *Nature (London)* **634**, 813 (2024).
- [11] J. Kim, S. S. Baik, S. H. Ryu, Y. Sohn, S. Park, B. G. Park, J. Denlinger, Y. Yi, H. J. Choi, and K. S. Kim, Observation of tunable band gap and anisotropic Dirac semimetal state in black phosphorus, *Science* **349**, 723 (2015).
- [12] S. S. Baik, K. S. Kim, Y. Yi, and H. J. Choi, Emergence of two-dimensional massless Dirac fermions, chiral pseudospins, and Berry's phase in potassium doped few-layer black phosphorus, *Nano Lett.* **15**, 7788 (2015).
- [13] B. Kiraly, N. Hauptmann, A. N. Rudenko, M. I. Katsnelson, and A. A. Khajetoorians, Probing single vacancies in black phosphorus at the atomic level, *Nano Lett.* **17**, 3607 (2017).
- [14] B. Kiraly, E. J. Knol, K. Volckaert, D. Biswas, A. N. Rudenko, D. A. Prishchenko, V. G. Mazurenko, M. I. Katsnelson, P. Hofmann, D. Wegner, and A. A. Khajetoorians, Anisotropic two-dimensional screening at the surface of black phosphorus, *Phys. Rev. Lett.* **123**, 216403 (2019).
- [15] K. Teichmann, M. Wenderoth, S. Loth, R. G. Ulbrich, J. K. Garleff, A. P. Wijnheijmer, and P. M. Koenraad, Controlled charge switching on a single donor with a scanning tunneling microscope, *Phys. Rev. Lett.* **101**, 076103 (2008).
- [16] H. Fang, A. Gallardo, D. Dulal, Z. Qiu, J. Su, M. Telychko, H. Mahalingam, P. Lyu, Y. Han, Y. Zheng, Y. Cai, A. Rodin, P. Jelnek, and J. Lu, Electronic self-passivation of single vacancy in black phosphorus via ionization, *Phys. Rev. Lett.* **128**, 176801 (2022).
- [17] Y.-L. Zou, J. Song, C. Bai, and K. Chang, Impurity scattering and friedel oscillations in monolayer black phosphorus, *Phys. Rev. B* **94**, 035431 (2016).
- [18] Z. Chen, J. Dong, E. Papalazarou, M. Marsi, C. Giorgetti, Z. Zhang, B. Tian, J.-P. Rueff, A. Taleb-Ibrahimi, and L. Perfetti, Band gap renormalization, carrier multiplication, and stark broadening in photoexcited black phosphorus, *Nano Lett.* **19**, 488 (2019).
- [19] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/39jp-9dkp> for further analysis and more detailed description of methods.
- [20] J. Repp, G. Meyer, F. E. Olsson, and M. Persson, Controlling the charge state of individual gold adatoms, *Science* **305**, 493 (2004).
- [21] F. Marczinowski, J. Wiebe, F. Meier, K. Hashimoto, and R. Wiesendanger, Effect of charge manipulation on scanning tunneling spectra of single Mn acceptors in InAs, *Phys. Rev. B* **77**, 115318 (2008).
- [22] R. Feenstra and J. Stroscio, Tunneling spectroscopy of the GaAs(110) surface, *J. Vac. Sci. Technol. B* **5**, 923 (1987).
- [23] A. P. Wijnheijmer, J. K. Garleff, K. Teichmann, M. Wenderoth, S. Loth, R. G. Ulbrich, P. A. Maksym, M. Roy, and P. M. Koenraad, Enhanced donor binding energy close to a semiconductor surface, *Phys. Rev. Lett.* **102**, 166101 (2009).
- [24] C.-L. Song, Y.-P. Jiang, Y.-L. Wang, Z. Li, L. Wang, K. He, X. Chen, X.-C. Ma, and Q.-K. Xue, Gating the charge state of single Fe dopants in the topological insulator Bi_2Se_3 with a scanning tunneling microscope, *Phys. Rev. B* **86**, 045441 (2012).
- [25] E. Golias, M. Krivenkov, and J. Sánchez-Barriga, Disentangling bulk from surface contributions in the electronic structure of black phosphorus, *Phys. Rev. B* **93**, 075207 (2016).
- [26] D.-B. Zhang, Q. Han, and Z. D. Wang, Local and global patterns in quasiparticle interference: A reduced response function approach, *Phys. Rev. B* **100**, 205112 (2019).
- [27] N. Ehlen, A. Sanna, B. V. Senkovskiy, L. Petaccia, A. V. Fedorov, G. Profeta, and A. Grüneis, Direct observation of a surface resonance state and surface band inversion control in black phosphorus, *Phys. Rev. B* **97**, 045143 (2018).
- [28] J. Jang, S. Ahn, and H. Min, Optical conductivity of black phosphorus with a tunable electronic structure, *2D Mater.* **6**, 025029 (2019).
- [29] P. Ebert, X. Chen, M. Heinrich, M. Simon, K. Urban, and M. G. Lagally, Direct determination of the interaction between vacancies on InP(110) surfaces, *Phys. Rev. Lett.* **76**, 2089 (1996).
- [30] C. Hellenthal, R. Heimbuch, K. Sotthewes, E. S. Kooij, and H. J. W. Zandvliet, Determining the local density of states

- in the constant current STM mode, *Phys. Rev. B* **88**, 035425 (2013).
- [31] R. J. Hamers and D. F. Padowitz, in *Scanning Probe Microscopy and Spectroscopy*, 2nd ed., edited by D. Bonnell (John Wiley & Sons, Nashville, TN, 2000), Chap. 4, pp. 59–110.
- [32] C. Q. Han, M. Y. Yao, X. X. Bai, L. Miao, F. Zhu, D. D. Guan, S. Wang, C. L. Gao, C. Liu, D. Qian, Y. Liu, and J.-F. Jia, Electronic structure of black phosphorus studied by angle-resolved photoemission spectroscopy, *Phys. Rev. B* **90**, 085101 (2014).
- [33] I. Battisti, V. Fedoseev, K. M. Bastiaans, A. de la Torre, R. S. Perry, F. Baumberger, and M. P. Allan, Poor electronic screening in lightly doped Mott insulators observed with scanning tunneling microscopy, *Phys. Rev. B* **95**, 235141 (2017).
- [34] G. D. Nguyen, L. Liang, Q. Zou, M. Fu, A. D. Oyedele, B. G. Sumpter, Z. Liu, Z. Gai, K. Xiao, and A.-P. Li, 3D imaging and manipulation of subsurface selenium vacancies in PdSe₂, *Phys. Rev. Lett.* **121**, 086101 (2018).
- [35] M. T. Woodside and P. L. McEuen, Scanned probe imaging of single-electron charge states in nanotube quantum dots, *Science* **296**, 1098 (2002).
- [36] C. Brun, K. H. Müller, I.-P. Hong, F. Patthey, C. Flindt, and W. D. Schneider, Dynamical coulomb blockade observed in nanosized electrical contacts, *Phys. Rev. Lett.* **108**, 126802 (2012).
- [37] Z. Tian, Y. Gan, T. Zhang, B. Wang, H. Ji, Y. Feng, and J. Xue, Isotropic charge screening of anisotropic black phosphorus revealed by potassium adatoms, *Phys. Rev. B* **100**, 085440 (2019).
- [38] T. Low, R. Roldán, H. Wang, F. Xia, P. Avouris, L. M. Moreno, and F. Guinea, Plasmons and screening in monolayer and multilayer black phosphorus, *Phys. Rev. Lett.* **113**, 106802 (2014).
- [39] Y. C. Tsui, M. He, Y. Hu, E. Lake, T. Wang, K. Watanabe, T. Taniguchi, M. P. Zaletel, and A. Yazdani, Direct observation of a magnetic-field-induced Wigner crystal, *Nature (London)* **628**, 287 (2024).
- [40] B. Tanatar and D. M. Ceperley, Ground state of the two-dimensional electron gas, *Phys. Rev. B* **39**, 5005 (1989).
- [41] S. T. Chui and B. Tanatar, Impurity effect on the two-dimensional-electron fluid-solid transition in zero field, *Phys. Rev. Lett.* **74**, 458 (1995).
- [42] R. Egger, W. Häusler, C. H. Mak, and H. Grabert, Crossover from Fermi liquid to Wigner molecule behavior in quantum dots, *Phys. Rev. Lett.* **82**, 3320 (1999).
- [43] B. Kiraly, A. N. Rudenko, W. M. J. van Weerdenburg, D. Wegner, M. I. Katsnelson, and A. A. Khajetoorians, An orbitally derived single-atom magnetic memory, *Nat. Commun.* **9**, 3904 (2018).
- [44] Z. Han, A. Allain, H. Arjmandi-Tash, K. Tikhonov, M. Feigel'man, B. Sacépé, and V. Bouchiat, Collapse of superconductivity in a hybrid tin-graphene Josephson junction array, *Nat. Phys.* **10**, 380 (2014).