Two-dimensional multi-valley electronic systems in which the dispersion of individual pockets has low symmetry give rise to quantum Hall ferroelectric and nematic states in the presence of strong quantising magnetic fields. We investigate local signatures of these states arising near impurities that can be probed via Scanning Tunnelling Microscopy (STM) spectroscopy. For quantum Hall ferroelectrics, we demonstrate a direct relation between the dipole moment measured at impurity bound states and the ideal bulk dipole moment obtained from the modern theory of polarisation. We also study the many-body problem with a single impurity via exact diagonalization and find that near strong impurities non-trivial excitonic state can form with specific features that can be easily identified via STM spectroscopy.

Introduction. In recent years we have witnessed an explosion of high-quality two-dimensional electronic systems with strongly anisotropic dispersions that can be driven into the quantum Hall regime in the presence of strong quantizing perpendicular fields [1, 2], such as (111) surface of Bismuth [3–5], AlAs heterostructures [6, 7], PbTe(111) quantum wells [8] and (001) surface of the topological crystalline insulators like Sn_{1−x}Pb_{x}(Te,Se) [9]. In these systems, at integer fillings of the Landau levels, the Coulomb interaction tends to spontaneously break symmetry by driving the formation of valley-polarized states [1, 10–12]. States resulting from this valley-polarization can be generally divided into nematic or ferroelectric states according to whether or not the Fermi surface of individual valleys preserves inversion symmetry [1]. Additionally, recent advances in STM have made it possible to directly image the shape of Landau level orbitals near impurities [3–5, 13], providing a new exciting window into these correlated states. Evidence of the quantum Hall ferroelectric state has recently been reported in Bismuth (111) [4]. The surface of SnPb(Te,Se) based topological crystalline insulator (TCI) is also a promising platform to realise these states [14–18].

In this letter we investigate the behaviour of quantum Hall ferroelectrics and nematics near short range impurities. One of our goals is to elaborate on how to measure an “order parameter” for the quantum Hall ferroelectric state. In trivial ferroelectric insulating states in which the bulk and the boundary are simultaneously gapped a natural order parameter is the ferroelectric dipole moment, which can be computed from the Berry phase based approach in the modern theory of polarization [19, 20]. In quantum Hall ferroelectrics, although such polarization is well defined in an ideal setting in which the system is placed in periodic boundary conditions, it is not clear how to directly measure it because the metallic states at the boundaries invariably screen the charge that would otherwise accompany the presence of a bulk polarisation. As we will show, states bound near impurities can be used to measure the degree of inversion symmetry breaking in the quantum Hall ferroelectric states, by measuring the dipole moment relative to the impurity center. Given the knowledge of the electronic structure of the host material, one can mathematically relate the ideal dipole moment defined by the modern theory of polarization to that of impurity bound states, as we will demonstrate explicitly for the case of tilted Dirac cones that are relevant for the surface of SnPb(Te,Se) based TCI’s.
In this section we consider $\varphi_n$ where the lowest-lying excited states corresponds to a non-trivial inter-valley excitonic state, in which an electron is added to another valley. We will discuss how these new many-body states have clear signatures that can be identified in the STM spectra.

Impurity states for Dirac cones. In this section we consider a model that is relevant to the (001) surface of SnPb(Se,Te) based TCI’s. In these materials, at temperatures below a ferroelectric transition their surface states comprise four Dirac cones, two of which are massive and two massless. Each of the massive/massless pair is degenerate \( [13] \) and the dispersions also have a small tilt in momentum \( [16, 21] \). Therefore, their low energy effective Hamiltonian reads as:

\[
H = v_x \sigma_x p_x + v_y \sigma_y p_y + \Delta \sigma_z + \delta v_x p_x,
\]

where $\sigma_i$ are Pauli matrices, $\Delta$ controls the mass (gap) and $\delta v_x$ represents the tilt of the Dirac cone. The sign of $\delta v_x$, $\Delta$, $v_y$ depends on the valley. In the presence of external magnetic fields Landau levels will form. We consider a partial filling $\nu = 1$ for any of the resulting 2-fold degenerate valley doublets. In the ferroelectric state the electrons will further spontaneously polarize into a single one of these valleys \( [11] \). Figure 1(a) and (b) provide simplistic illustrations of this model. Inspired by recent STM experiments \( [3, 4] \), we study states near short-range impurities modelled as delta-function potentials \( [22] \):

\[
H_{\text{imp}} = V_0 \delta_B (\mathbf{r}).
\]

Assuming that the impurity potential \( (V_0) \) is smaller than the Landau level spacing, we project the Hamiltonian to the Landau level of interest. Only states with a finite probability at the origin will be affected by the impurity potential. For a parabolic dispersion, there would be a single state per Landau level with non-zero probability at the origin, as demonstrated in the Bismuth experiments \( [3] \). However, the situation is richer for Dirac Landau levels. The wavefunctions of the $n^{th}$ Dirac Landau level in the massless and un-tilted limit (for the general case see the Supplement \( [23] \)) are:

\[
\psi_{n,m} = \frac{1}{Z_n} \left( \phi_{|n|,m} s_n \phi_{|n|-1,m} \right),
\]

where $n \in \mathbb{Z}$, $s_n = \text{sign}(n)$ (with $s_0 = 0$), $Z_n = 2^{\frac{1}{2}|n|}$, and $\phi_{|n|,m}$ is the wavefunction for a parabolic Landau level in the symmetric gauge with angular momentum $m - |n|$. For the $n = 0$ LL only the $m = 0$ state would have probability at the origin, however, for $n \neq 0$, two states with $m = |n|$ and $m = |n| - 1$ would have probability at the origin and opposite pseudospin polarization \( [24, 25] \). These two states are exactly degenerate for a massless and un-tilted cone, but either of these perturbations produces an energy splitting of these impurity states as illustrated in Fig. 1. Thus the impurity states are generically resolvable in STM measurements. In the Supplement \( [23] \) we demonstrate that these perturbations do not produce extra impurity states, and therefore, only these two states are split from the bulk Landau level and bound to the impurity. Let us introduce dimensionless parameters to characterise the tilt $\tau \equiv \delta v_x/(2v_x)$ and the mass $\lambda \equiv \Delta l_B/(\sqrt{2v_x}v_y)$. In Sn$_{1-x}$Pb$_x$(Te,Se) these are approximately $\tau = 0.1$, $\lambda = 0.5$ and $v_x/v_y = 1.6$ \( [23] \). It is therefore justified to use perturbation theory in $\tau$. The splitting of the two impurity states from the bulk Landau level, to leading order in $\tau$, are then estimated to be: $E_1 \approx 0.12V_0$, $E_2 \approx 0.04V_0$. Figure 2 displays the spatial profile of these two states.

Ferroelectric dipole moments. In the modern theory of electric polarization \( [19, 20] \), the dipole moment of an insulator is computed by ignoring its boundary and placing it under periodic boundary conditions. The dipole is computed from the change of the electronic position induced while varying the Hamiltonian along an adiabatic path in which the bulk gap remains open and that starts from an inversion symmetric reference state and ends at the state of interest. Following this principle a dipole moment for the ferroelectric quantum Hall
state was introduced in Ref. [1]. In our case of tilted Dirac cones, this dipole moment per particle to leading order in the tilt is found to be:

$$D_n = \tilde{s}_n \sqrt{2} \tau e B \left( \frac{2\lambda^2 + 3|n|}{\sqrt{\lambda^2 + |n|}} \right) \sqrt{\nu_{ex}},$$

(4)

here $\tilde{s}_n = \text{sign}(n)$ (with $\tilde{s}_0 = 1$). Notice that the dipole along the tilt of Dirac cone (x-axis) vanishes [1]. The limitation of this definition is that one would like to assume the charge that flows through the bulk will appear intact at its surface providing a net electric polarization. However, in an insulating topological phase with a metallic boundary, the latter assumption is not justified since the charge that would otherwise accumulate at the surface can flow, leading to no net macroscopic polarization in the sample. Therefore, this notion is hard to access by conventional measurements of the macroscopic electric dipole moment, and, it is therefore important to devise alternative diagnostics of the degree of inversion symmetry breaking in topological states with metallic boundaries such as the quantum Hall ferroelectric state.

As we will describe, states bound to impurities, which can be directly imaged via STM, offer a valuable window into the inversion asymmetry of the ferroelectric state. For any given impurity state one can define a dipole moment as the expectation value of the position measured relative to the center of the impurity potential. If the impurity potential is inversion symmetric, then, this dipole moment serves to characterise the inversion asymmetry of the host state. Figure 2(a) and (b) display the average position of the impurity states in tilted Dirac cones as a function of their mass and tilt. Interestingly, the average position of the impurity states is a non-analytic function of tilt and mass near $\tau = 0$, $\lambda = 0$, as evidenced by the fact that the limits of $\tau \to 0$, $\lambda \to 0$ do not commute in Fig. 2. This is a consequence of the fact that in this limit both impurity states are degenerate and hence the expectation values on individual states become ambiguous. However, the sum of the average positions in both impurity states is free from ambiguities and vanishes as $\tau \to 0$, $\lambda \to 0$. We therefore introduce the notion of the impurity dipole moment, $D^{\text{imp}}$, as the sum of the expectation value of position of the impurity states, $\psi_i$, that are split from the bulk of the Landau level [26]:

$$D^{\text{imp}} = e \sum_i \langle \psi_i | \mathbf{r} | \psi_i \rangle.$$ (5)

By explicitly computing these expectation values to leading order in the Dirac cone tilt ($\tau$), we obtained the following explicit relation between the adiabatic bulk dipole moment, in Eq. (4), and the impurity dipole moment:

$$D^{\text{imp}} = \frac{2|n|}{3|n| + 2\lambda^2} D_n,$$ (6)

for the $n^{\text{th}}$ Dirac Landau level in a Dirac cone of mass $\lambda$. This formula succinctly summarises one of the key messages of our study: that the measurement of the impurity dipole moment, $D^{\text{imp}}$, combined with the knowledge of the electronic structure, can be used to directly estimate the bulk adiabatic dipole moment that follows from the modern theory of polarisation, $D_n$, in a quantum Hall ferroelectric state.

In the limit of massless cones, i.e. $\lambda \ll \sqrt{|n|}$, the two dipole moments have a simple proportionality relation, $D^{\text{imp}} = (2/3)D_n$. However, a notable difference between these two notions appears in the large mass limit, i.e. $\lambda \gg \sqrt{|n|}$, for which the adiabatic dipole grows linearly with the mass, $|D_n| \propto \lambda$, whereas $|D^{\text{imp}}_n| \propto 1/\lambda$. This markedly different behavior is a consequence of the approach to the parabolic mass limit as we detail in the Supplement [23].

Many-body physics near impurities. So far we have largely ignored the role of electron-electron interactions by imagining that a large self-consistent exchange field sets in that selects a single valley. In this section, we will study the many-body problem in the presence of the impurity potential from Eq. (3) by means of exact numerical diagonalization of the interacting Hamiltonian on a torus. We concentrate here on the ferroelectric states where two valleys are described by the tilted massless Dirac cone with the same axis orientation and velocity ratio but opposite tilt. We expect the states at Landau level $n = +3$ to essentially carry over to the case of Bismuth Surfaces [3-5]. In the Supplement [23], we also present a nematic model of two valleys with anisotropic masses whose principal axes are rotated by $\pi/2$, as in AlAs quantum wells [6, 7], which gives a simpler picture of what we find.

In the absence of impurity ($V_0 = 0$) at the $n = +3$ Dirac LL and partial filling $\nu = 1$, the ground state of the system spontaneously polarizes into a single valley and an exchange splitting, $\Delta_X$, between the two valleys develops [1, 11, 12]. This is schematically depicted in Fig. 1(a) and (b). In the forthcoming discussion we choose the chemical potential to lie exactly in the middle of the charge gap, namely, we add a single particle term to the Hamiltonian so that far away from the impurity the energy to add one electron equals the energy to add one hole. In STM spectra this is satisfied when the two peaks corresponding to the occupied and empty valleys in the Landau level are located symmetrically away from zero bias with no impurity, as illustrated in Fig. 3. We assume a sufficiently strong tilt so that the lowest energy charged excitations are not skyrmions [1].

We denote the valley polarization of states by a vector $(N_1, N_2)$, where $N_i$ is the number of electrons in valley $i$ ($i = A, B$). The ground state at $\nu = 1$ in the absence of the impurity therefore has polarization $(N_1, 0)$. The number of orbits in a single valley is taken to be $N_{\phi} = 40$. STM is customarily viewed as a probe of the density of states of the single particle charged excitations, because it requires the removal or injection of electrons from the sample. As we will see, however, near strong impurities, it is possible to use STM to probe excitonic states. For a weak impurity, $V_0 \ll \Delta_X$, as the STM tip is brought near the impurity one expects sim-
We have studied how to locally probe quantum Hall ferroelectric and nematic states near short range impurities. Impurities provide a valuable window to experimentally determine the degree of inversion symmetry.

FIG. 3: (a) Spectra with increasing impurity potential: \((N_1, N_2)\) labels the state with \(N_1\) electrons in valley A and \(N_2\) electrons in valley B. Only the lowest states of \((N_0 - 1, 0)\) and \((N_0 - 1, 1)\) are drawn, while the lowest states of \((N_0, 1)\) sector are nearly degenerate and indistinguishable by tunneling spectroscopy, and are shown in dashed lines. All energies are measured relative to that of \((N_0, 0)\). \(A, B, C, D, C', D', C'', D''\) correspond to the tunneling peaks in the lower panel. (b) Illustration of tunneling spectroscopy peaks measured by the STM. The peaks \(A, B\) denote tunneling from \((N_0, 0)\) to \((N_0 - 1, 0)\) and \((N_0, 1)\) at weak impurity; the symmetric dashed peaks denote the same tunneling with no impurity; \(C', C''\) denote tunneling from \((N_0 - 1, 0)\) to \((N_0, 0); D', D''\) denote tunneling from \((N_0 - 1, 0)\) to \((N_0 - 1, 1)\).

(a) differential conductance at \(A, C, C''\)

(b) differential conductance at \(B\)

(c) differential conductance at \(D', D''\)

FIG. 4: The local density of states at energy levels \(A, B, C', D', C'', D''\), which is proportional to the differential conductance obtained by direct STM measurements. The unit of length is set to be \(l_B\). The tilt \(\tau = 0.1\) and velocity ratio \(v_x/v_y = 5\) are used.

given by the local density of state (LDOS) at energy \(\varepsilon\):

\[
G(\mathbf{r}) \propto \sum_m \sum_j \left( c_{A,j}^\dagger \phi^*_{A,j}(\mathbf{r}) + c_{B,j}^\dagger \phi_{B,j}(\mathbf{r}) \right) |\phi_0\rangle^2,
\]

where \(|\phi_0\rangle\) is the lowest energy state. For a weak impurity below the threshold, \(|\phi_0\rangle = |N_0, 0\rangle\). Above the threshold, \(|\phi_0\rangle = |N_0 - 1, 0\rangle\), which is the hole state created by the impurity. \(c_{i,j}^\dagger\) and \(\phi_{i,j}\) are the creation operator and single electron wavefunction for an orbit \(j\) on valley \(i\). \(|\phi_m\rangle\) is the state with energy \(\varepsilon\), the sum over \(m\) is taken for all degeneracy. The case of removing an electron follows from Eq. (7) by replacing \(c_{i,j}^\dagger\) and \(\phi^*_{i,j}\) with \(c_{i,j}\) and \(\phi_{i,j}\) respectively.

Figure 4 depicts the expected shape of the differential conductance in STM at the energy and impurity indicated in Fig. 3. The \(B\) peak in the spectroscopy includes multiple nearly degenerate states, here in Fig. 4 we treat them as degenerate at energy \(\varepsilon\) and average over them. The first two panels of Fig. 4 depict tunneling between a single-hole or electron state and the fully polarized state, which only involves single-body physics; while the last panel is the tunneling between the hole state and the excitonic state, though only reflects the LDOS of valley B with one electron, its shape is modified via the interaction with the hole in valley A. The significant difference between Fig. 4(a) and (c) allows for distinguishing this non-trivial excitonic state in STM.

Summary and discussion. We have studied how to locally probe quantum Hall ferroelectric and nematic states near short range impurities. Impurities provide a valuable window to experimentally determine the degree of inversion symmetry.
breaking in these states. We have shown that the dipole moment of states bound to impurities, which can be measured via STM, can be used to estimate the degree of inversion symmetry breaking and the bulk adiabatic electric dipole of the system that follows from the modern theory of polarisation. We have also studied the many-body problem near short range impurities and found that strong impurities can be used to image non-trivial excitonic states. These states, which are typically invisible in STM in the clean regions of the sample or near weak impurities, become accessible near strong impurities which can change the ground state by locally removing/adding an electron to the sample.

We are grateful to Benjamin Feldman, Mallika Randeria and Ali Yazdani for illuminating discussions. TL would like to thank Zheng Zhu for helping with the numerical study. This work is supported by DOE Oce of Basic Energy Sciences under Award de-sc0018945. PMT was in part supported by the Croucher Scholarship for Doctoral Study from the Croucher Foundation.

[1] I. Sodemann, Z. Zhu, and L. Fu, Phys. Rev. X 7, 041068 (2017).
[2] S. A. Parameswaran and B. E. Feldman, J. Phys.: Condens. Matter 31, 273001 (2019).
[3] B. E. Feldman, M. T. Randeria, A. Gyenis, F. Wu, H. Ji, R. J. Cava, A. H. MacDonald, and A. Yazdani, Science 354, 316 (2016).
[4] M. T. Randeria, B. E. Feldman, F. Wu, H. Ding, A. Gyenis, H. Ji, R. J. Cava, A. H. MacDonald, and A. Yazdani, Nat. Phys. 14, 796 (2018).
[5] M. T. Randeria and K. Agarwal and B. E. Feldman and H. Ding and H. Ji and R. J. Cava and S. L. Sondhi and S. A. Parameswaran and A. Yazdani, Nature 566, 363 (2019).
[6] M. Shayegan, E. P. De Poortere, O. Gunawan, Y. P. Shkolnikov, E. Tutuc, K. Vakili, Phys. Stat. Sol. b 243, 3629 (2006).
[7] T. Gokmen, M. Padmanabhan, and M. Shayegan, Nat. Phys. 6, 621 (2010).
[8] V. A. Chitta, W. Desrat, D. K. Maude, B. A. Piot, N. F. Oliveira Jr., P. H. O. Rapp, A. Y. Ueta, E. Abramof, Physica E: Low-dimensional Systems and Nanostructures 34, 124 (2006).
[9] Okada, Y., M. Serbyn, H. Lin, D. Walkup, W. Zhou, C. Dhital, M. Neupane, S. Xu, Y. J. Wang, R. Sankar, F. Chou, A. Bansil, M. Z. Hasan, S. D. Wilson, L. Fu, and V. Madhavan, Science 341, 1496 (2013).
[10] X. Li, F. Zhang, A. H. MacDonald, Phys. Rev. Lett. 116, 026803 (2016).
[11] D. A. Abanin, S. A. Parameswaran, S. A. Kivelson, and S. L. Sondhi, Phys. Rev. B 82, 035428 (2010).
[12] A. Kumar, S. A. Parameswaran, and S. L. Sondhi Phys. Rev. B 88, 045133 (2013).
[13] Zlatko Papić, Roger S. K. Mong, Ali Yazdani, and Michael P. Zaletel Phys. Rev. X 8, 011037 (2018).
[14] P. Dziawa, B. J. Kowalski, K. Dybko, R. Buczko, A. Szczersbakow, M. Szot, E. Lusakowska, T. Balasubramanian, B. M. Wojek, M. H. Berntsen, O. Tjernberg and T. Story, Nature Materials 11, 1023 (2012).
[15] T. H. Hsieh, H. Lin, J. Liu, W. Duan, A. Bansil, and L. Fu, Nat. Commun. 3:982 (2012).
[16] Y. Tanaka, Z. Ren, T. Sato, K. Nakayama, S. Souma, T. Takahashi, K. Segawa, and Y. Ando, Nat. Phys. 8, 800 (2012).
[17] J. Liu, W. Duan, and L. Fu, Phys. Rev. B 88, 241303(R) (2013).
[18] M. Serbyn and L. Fu, Phys. Rev. B 90, 035402 (2014).
[19] D. J. Thouless, Phys. Rev. B 27, 6083 (1983).
[20] R. D. King-Smith and D. Vanderbilt, Phys. Rev. B 47, 1651(R) (1993).
[21] I. Sodemann and L. Fu, Phys. Rev. Lett. 115, 216806 (2015).
[22] For simplicity we neglect inter-valley scattering although it has recently been shown that certain detailed aspects of the states near the impurity require understanding the intervalley scattering properties [11].
[23] Supplementary Materials.
[24] A. H. MacDonald, arXiv:cond-mat/9410047v1 (1994).
[25] G. Giuliani and G. Vignale, Quantum Theory of the Electron Liquid (Cambridge University Press, 2008) p. 559.
[26] This average coincides with the minus of dipole moment weighted by the charge distribution of the hole that is left in Landau level which can also be directly accessed by STM measurements.
[27] Here we describe the behavior for repulsive impurities $V_0 > 0$, but equivalent statements hold for attractive impurities after performing a particle-hole conjugation $(N_1, N_2) \rightarrow (N_0 - N_2, N_0 - N_1)$. Particularly, the quasihole state in the repulsive case is replaced by a quasi-particle state in the attractive case.
Supplementary Materials for “Local Probes for Quantum Hall Ferroelectrics and Nematics”

In this supplementary, we provide more details about the setup of our theoretical and numerical studies. In Sec. A-D, we explain how to study integer quantum Hall states on the surface of topological crystalline insulator, which has Dirac dispersion that is both tilted and massive. Particularly, we argue in Sec. B that in the presence of delta-potential impurity, there are exactly two states per Landau level that are perturbed away in energy. In Sec. C we explain the values of parameters adopted in our model, while in Sec. D we distinguish two notions of electric dipole moments and identify the one that can reveal ferroelectricity in our system. In Sec. E we present the setup for carrying out exact diagonalization which leads to the prediction of non-trivial excitonic states near strong impurities. While the experimental signatures of these many-body states in systems with Dirac dispersion have been discussed in the main text, a simpler situation with anisotropic parabolic dispersion (such as in AlAs quantum well) is analyzed in Sec. F.

A. Massive and tilted Dirac Landau levels

Under an out-of-plane magnetic field $-B\hat{z}$, in the un-tilted limit, the massive Dirac Hamiltonian in Eq. (1) can be written as

$$H_0 = \frac{\sqrt{2}}{l_B} v \begin{pmatrix} \lambda & a^\dagger \\ a & -\lambda \end{pmatrix}$$

(A.1)

where $v = \sqrt{v_x v_y}$, magnetic length $l_B = \sqrt{\hbar c/eB}$ and the mass parameter $\lambda = \Delta l_B/(\sqrt{2}v)$. Here, $a^\dagger, a$ are parabolic Landau level raising and lowering operators respectively, and are related to the momentum operators by:

$$p_x = \frac{1}{l_B} \sqrt{\frac{v_y}{2v_x}} (a^\dagger + a) , \quad p_y = \frac{i}{l_B} \sqrt{\frac{v_x}{2v_y}} (a - a^\dagger)$$

$$[a, a^\dagger] = 1$$

(A.2)

The wavefunctions of the massive Dirac Landau levels and their corresponding energy can then be solved exactly. For the $n$-th Landau level with $n \neq 0$:

$$\psi_{n,m} = \frac{1}{\sqrt{1 + \gamma_n^2}} \begin{pmatrix} \phi_{|n|,m} \\ \gamma_n \phi_{|n|-1,m} \end{pmatrix} , \quad E_n = s_n \frac{\sqrt{2}}{l_B} \sqrt{\lambda^2 + |n|}$$

(A.3)

where

$$\gamma_n = \frac{-\lambda + s_n \sqrt{\lambda^2 + |n|}}{\sqrt{|n|}}$$

(A.4)

Here $s_n = \text{sign}(n)$, and $\phi_{|n|,m}$ are the wavefunctions for a parabolic Landau level in the symmetric gauge with angular momentum $m - |n|$. For the 0-th Dirac Landau level, we have:

$$\psi_{0,m} = \begin{pmatrix} \phi_{0,m} \\ 0 \end{pmatrix} , \quad E_0 = \frac{\sqrt{2}}{l_B} \lambda$$

(A.5)

When the tilt of Dirac cone $\delta v_y$ is turned on, we can do first order perturbation theory to obtain the approximate eigenstates. To leading order in $\tau = \delta v_y/(2v_x)$, for the $n \neq 0$ massive and tilted Dirac LL, we obtain (up to normalization):

$$\psi_{n,m} = \begin{pmatrix} \phi_{|n|,m} \pm \tau [\alpha_{-1} \phi_{|n|-1,m} + \alpha_1 \phi_{|n|+1,m}] \\ \gamma_n [\phi_{|n|-1,m} \mp \tau (\alpha_0 \phi_{|n|,m} + \alpha_{-2} \phi_{|n|-2,m})] \end{pmatrix}$$

(A.6)

where

$$\alpha_{-1} = \frac{(2|n| - 1) \sqrt{\lambda^2 + |n|} \pm \lambda}{\sqrt{|n|}} , \quad \alpha_1 = -2\sqrt{|n| + 1} \sqrt{\lambda^2 + |n|}$$

$$\alpha_0 = \frac{(2|n| + 1) \sqrt{\lambda^2 + |n|} \pm \lambda}{\sqrt{|n|}} , \quad \alpha_{-2} = -2\sqrt{|n| - 1} \sqrt{\lambda^2 + |n|}$$

(A.7)
As for the massive and tilted 0-th Dirac LL:

$$\psi_{0,m} = \left( \frac{\phi_{0,m} - 2\tau \lambda \phi_{1,m}}{-\tau \phi_{0,m}} \right)$$  \hspace{1cm} (A.8)

These expressions allow us to calculate dipole moments, and energy shifts under the influence of impurity, straightforwardly.

### B. Number of impurity states for massive and tilted Dirac cones

Here we demonstrate that there are only two states that have probability amplitudes at the impurity site, and which therefore are split from the Landau level, even in the presence of perturbations in mass and tilt of the Dirac cone.

We consider a delta-function impurity $H_{imp} = V_0 l_B^2 \delta(\vec{x})$. Upon projection to a specific Landau level, the impurity Hamiltonian has matrix elements:

$$\langle n, m | H_{imp} | n', m' \rangle = V_0 l_B^2 \Psi_{n,m}^\dagger \Psi_{n,m'}$$  \hspace{1cm} (B.1)

where we have defined $\Psi_{n,m} \equiv \psi_{n,m}(\vec{0})$, i.e. the amplitude of the Landau level orbital at the impurity site. To the first order in tilt $\tau$, the Dirac Landau level is found in Eq. (A.6). To prove our claim in full generality, let us assume we have carried out a $k$-th order perturbation theory in $\tau$, so that the $n$-th tilted Dirac Landau level $\psi_{n,m}$ is expressed in terms of $\phi_{p,m}$ with $p = |n| - k - 1, ..., |n| + k$. The only states that are relevant to our impurity problem are those that have non-vanishing probability amplitudes at the impurity site, which correspond to those $\psi_{n,m}$ with $m = |n| - k - 1, ..., |n| + k$. We thus study the degenerate perturbation theory within this subspace, and consider linear combinations of $\Psi_{n,m}$:

$$\Phi = r_1 \psi_{n,|n| - k - 1} + r_2 \psi_{n,|n| - k} + ... + r_{2k+2} \psi_{n,|n| + k}$$  \hspace{1cm} (B.2)

If there is a choice of $(r_1, r_2, ..., r_{2k+2})$ such that $\Phi = (0, 0)^T$, the corresponding linear combination of intra-Landau level orbitals are guaranteed to diagonalize the impurity Hamiltonian and thus remain at the same energy as the Landau level in the absence of impurity. Below, we argue that there are $2k$ such solutions.

Denote $\Psi_{n,m} = (\psi_{n,m}^1, \psi_{n,m}^2)^T$. Only the intra-Landau level index $m$ is made explicit here. Notice that $\psi_{m}^1$ and $\psi_{m}^2$ are both real or both imaginary. This is because each of them is proportional to the wavefunction of parabolic Landau level $\phi_{m,r}$ evaluated at the origin, which is real when $m$ is even and is imaginary when $m$ is odd. Redefining $i\Psi_{n,m} \mapsto \Psi_{n,m}$ for odd $m$, Eq. (B.2) with $\Phi = (0, 0)^T$ becomes a set of simultaneous equations for real unknowns $r_i$. Setting $r_{2k+2} = 1$ without loss of generality, we reach the following set of equations for $r_i \in \mathbb{R}$:

$$\begin{align*}
 r_1 \psi_{n-k-1}^1 + r_2 \psi_{n-k}^1 + ... + r_{2k+1} \psi_{n+k}^1 &= -\psi_{n+k}^1 \\
 r_1 \psi_{n-k-1}^2 + r_2 \psi_{n-k}^2 + ... + r_{2k+1} \psi_{n+k}^2 &= -\psi_{n+k}^2
\end{align*}$$  \hspace{1cm} (B.3)

With $2k+1$ unknowns and only two linear equations, there are in general $2k$ linearly independent solutions, leading to $2k$ states that have vanishing amplitudes at the impurity site. Since we start with a $(2k + 2)$-dimensional subspace, only $2k + 2 - 2k = 2$ states are allowed to have non-vanishing amplitudes at the origin. These are the two impurity bound states whose energy is split from the bulk Landau level, and are the ones employed in our construction of impurity dipole moment in the main text.

The above argument also works for the 0-th Landau level. However, only one impurity state is significantly shifted away from the bulk Landau level, while the shift of the second impurity state is minuscule (controlled by the size of the tilt), so practically, in the quantum Hall ferroelectric system that we consider, only one impurity state can be probed in this special case.

### C. Choice of Parameters

In the main text, we use the following parameters to study the quantum Hall ferroelectrics in topological crystalline insulator Sn$_{1-x}$Pb$_x$(Te,Se):

$$\tau = 0.1, \quad \lambda = 0.5, \quad v_x/v_y = 1.6$$  \hspace{1cm} (C.4)

Here we explain why these values match with the low-energy physics of the system obtained either from experiments or *ab initio* calculations.
The tilting effect of Dirac cones (at $\bar{\Lambda}$) has been observed in the ARPES measurements by Tanaka et al. [1]. The left-hand and right-hand branches were measured to have different Dirac velocities, which were 4.5 eV Å and 3.0 eV Å respectively. According to the effective Hamiltonian in Eq. (1), the Dirac velocity for the left-hand branch is $v_x + \delta v_x$, while that for the right-hand branch is $v_x - \delta v_x$. Thus we can estimate the tilting parameter to be $\tau = \delta v_x/(2v_x) = 0.1$. The acquisition of mass in topological crystalline insulators was observed by Okada et al. [2]. By applying symmetry-breaking distortions, two of the four surface Dirac cones were measured to obtain mass of about $\Delta = 10$ meV. Assuming the proposed measurement of Dirac Landau orbitals to be performed at 10 T, we shall then approximate $\lambda = \Delta l_B/(\sqrt{2}v) \approx 0.5$. The values of $v_x$ and $v_y$ have been obtained by Liu et al. by fitting with ab initio calculations [3]. For the effective Dirac Hamiltonian (around $\bar{\Lambda}$) that we are considering, $v_x = 1.3$ eV Å and $v_y = 0.83$ eV Å. Thus, the anisotropy $v_x/v_y = 1.6$.

We studied the energy-shift and inversion asymmetry of the impurity states as functions of tilt and mass in Figure 1 and 2 of the main text respectively, allowing us to understand the behavior away from these specific values of parameters.

D. Two different notions of the electric dipole moment

To further clarify the difference between the adiabatic bulk dipole moment (following the modern theory of polarization) and the impurity dipole moment $D^{\text{imp}}$ introduced in this letter, we consider a toy model with a parabolic dispersion:

$$H = \frac{(p_x - a_x)^2}{2m_x} + \frac{p_y^2}{2m_y} \tag{D.1}$$

The parameter $a_x$ plays a similar role as the tilt $\delta v_x$ in the Dirac Hamiltonian.

Now, apply a magnetic field $B\hat{z}$ on the system. Denote a Landau orbital as $|\psi\rangle$ for $a_x=0$, and the corresponding Landau orbital when $a_x$ is tuned from zero to some finite value as $|\tilde{\psi}\rangle$. According to the polarization theory based on Berry phase, the difference of polarization between these two Landau orbitals is:

$$\Delta D_y = -\left|e\right|^2\left[\langle \tilde{\psi} | p_x | \tilde{\psi}\rangle - \langle \psi | p_x | \psi\rangle\right] = -\left|e\right|^2\left[\langle \tilde{\psi} | \tilde{p}_x + a_x | \tilde{\psi}\rangle - \langle \psi | p_x | \psi\rangle\right] = -\left|e\right|^2\left[\langle \tilde{\psi} | \tilde{p}_x | \tilde{\psi}\rangle - \langle \psi | p_x | \psi\rangle + a_x\right] = -\frac{a_x}{|B|} \tag{D.2}$$

The last equality is obtained because $\tilde{p}_x = p_x - a_x$ is just a gauge transformation, while the expectation value $\langle \psi | p_x | \psi\rangle$ should be gauge-invariant.

However, this dipole moment does not reflect the inversion asymmetry of the Landau orbital. In this example, there is simply no inversion asymmetry to begin with, and this can be verified if one examine $D^{\text{imp}} = e\langle \phi|\tau|\phi\rangle$, for the Landau orbital bound to a delta-potential impurity. By a proper gauge transformation, one can move the center of unperturbed Landau orbitals to the impurity site, irrespective of what $a_x$ is. After all, the presence of $a_x$ can be viewed as a gauge-transformation. In the presence of a delta-function impurity, only one state in each Landau level is bound to the impurity. That is the state $\phi_{n,n}$, which has a non-zero amplitude at the origin where the impurity sits. As this state is inversion symmetric, and the perturbation (i.e. the delta potential) preserves this symmetry, the bound state should also be inversion symmetric. Thus $D^{\text{imp}} = 0$.

In this extreme example, which can be considered as the parabolic limit ($\lambda \rightarrow \infty$) of the Dirac Hamiltonian, $D$ measures solely the effect of Landau orbital displacement, which cannot be detected in a quantum Hall system due to edge screening. On the other hand, $D^{\text{imp}}$ measures only the inversion asymmetry of Landau orbitals, and therefore gives a local experimental signature for quantum Hall ferroelectrics.

E. Numerical Setup of Exact Diagonalization

Anisotropic parabolic dispersions

To exact-diagonalize the Hamiltonian with Coulomb interaction, one has to project the Coulomb term onto the Landau orbitals. In the main text, we deal with Landau levels arising from the tilted Dirac cones dispersion, while in this appendix we will also consider the case with parabolic dispersion. The parabolic case is the cornerstone for case with tilted Dirac cones dispersion.
since the Dirac Landau orbitals are spinors consisting of parabolic Landau orbitals. The parabolic dispersion Hamiltonian is:

$$H = \frac{1}{2m^*} p_a g_{ab} p_b = \frac{1}{l_B^2 m^*} (a^\dagger a + 1/2)$$  \hspace{1cm} (E.1)

where $p = \nabla / -eA$, $g = Q^T S^2 Q$ is a $2 \times 2$ tensor, $Q \in SO(2)$ describes the rotation around principal axes in real space, the valleys we are interested in are vertical oriented, so we can set the real space axes along the principal axes of rotation, thus $Q = I$, and simply $g = S^2$. $S = \text{diag} \{(m_x/m_y)^{1/4} (m_y/m_x)^{1/4}\}$ is the mass tensor for the valley, effective mass $m^* = (m_x m_y)^{1/2}$.

We introduce the mass ratio: $\alpha = m_x/m_y$ that specifies aspect ratio of the valley. The rescaled momenta along the principal axes of the tensor $\pi_a = S_{ab} p_b$ satisfy:

$$[\pi_a, \pi_b] = i l_B^2 \epsilon_{ab}$$  \hspace{1cm} (E.2)

and the LL lowering operator is:

$$a = \frac{l_B}{\sqrt{2}} (\pi_x + i \pi_y), \quad [a, a^\dagger] = 1$$  \hspace{1cm} (E.3)

Numerically, the electrons are on the 2D surface of torus, $L_x (L_y)$ represents the circumference of the torus along $x(y)$ direction and they satisfy relation $L_x L_y = 2\pi N_0$, $N_0$ represents the number of orbitals for each valley.

Choosing the Landau gauge, $\vec{A} = (0, x)B$, the wavefunction of LL orbital is expressed as:

$$\phi_{n,j}^\alpha(r) = \left( \frac{2\pi}{L_y l_B} \right)^{1/2} \sum_{k=-\infty}^{\infty} \overline{H}_n \left[ x - k L_x - X_j \over \alpha l_B \right] \times \exp \left[ i X_j + k L_x y/l_B - (X_j + k L_x - x)^2/(2\alpha l_B^2) \right]$$  \hspace{1cm} (E.4)

where $X_j = \frac{2\pi j}{L_y}$ to fulfill the periodic boundary condition, $\overline{H}_n$ is the physicist’s Hermite polynomial that has been normalized so that:

$$\int_{-\infty}^{+\infty} \left( \overline{H}_n(x) \right)^2 e^{-x^2} dx = 1$$  \hspace{1cm} (E.5)

With $\phi_{n,j}^\alpha(r)$ normalized as $\int_0^{L_x} dy \int_0^{L_y} dx |\phi_{n,j}^\alpha(r)|^2 = 2\pi$, when a Landau level is completely filled and thus the electron density is uniformly distributed, $\int_0^{L_x} dy \int_0^{L_y} dx \sum_j |\phi_{n,j}^\alpha(r)|^2 = 2\pi N_0 = L_x L_y$ would then imply $\sum_j |\phi_{n,j}^\alpha(r)|^2 = 1$.

Next, we define $f_{nm}$ as the form factor for the parabolic Landau levels calculated in the Landau gauge:

$$f_{nm}(q^\alpha) = \langle n, \alpha | e^{i q^\alpha \cdot \vec{r}} | m, \alpha \rangle = e^{-l_B^2 (q_y^2)^{1/4}} \int_{-\infty}^{+\infty} \overline{H}_m(x - \frac{q_y^2}{2}) \overline{H}_n(x + \frac{q_y^2}{2}) e^{-x^2} e^{i q^\alpha \cdot x} dx$$  \hspace{1cm} (E.6)

where the wavevector $q^\alpha$ is not the natural wavevector $q$ but rotated as

$$q^\alpha = -S^{-1} \epsilon q$$  \hspace{1cm} (E.7)

where $\epsilon$ is the rank-2 levi-civita symbol, $Q$ and $S$ are the matrices associated with the mass ratio $\alpha$. This definition will become clear later when we project the electron interaction on the LLs.

**Tilted Dirac cone dispersion**

The massless Dirac Hamiltonian is just Eq. (A.1) with $\lambda = 0$. Similar to the case with a parabolic Hamiltonian in Eq. (E.1), here we would define $S = \text{diag} \{(v_x/v_y)^{1/2}, (v_y/v_x)^{1/2}\}$ and rescale the momentum by $\pi_a = S_{ab} p_b$, which explains Eq. (A.2). One can relate the mass ratio $\alpha$ in the anisotropic parabolic dispersion and velocity ratio $r = v_x/v_y$ in the Dirac dispersion as:

$$\alpha = r^2$$
Tilting of the Dirac cone along the \(x\)-direction is described by the following perturbation:

\[
H_1 = \delta v_x p_x = \delta v_x \frac{v_y (a + a^\dagger)}{v_x \sqrt{2}\ell_B} = \tau \frac{\sqrt{2}v}{\ell_B} (a + a^\dagger)
\]

(E.8)

where \(\tau = \delta v_x/(2v_x)\). Using the general expression for the tilted Dirac LL in Eq. (A.6), we have the following expression for the \(n = +3\) Dirac Landau level:

\[
| +3, \tau \rangle = \frac{1}{\sqrt{2}} \left( |3\rangle + \tau (-4\sqrt{3}|4\rangle + 5|2\rangle) \right)
\]

(E.9)

Here, for simplicity, we have suppressed the intra-Landau level indices and the mass ratio \(\alpha\) that would label the parabolic Landau orbitals. The form factor for the Dirac Landau level is then obtained as follows:

\[
F^3(\mathbf{q}^\alpha, \tau) = \langle +3, \tau | e^{i\mathbf{q}^\alpha \cdot \mathbf{r}} | +3, \tau \rangle
\]

(E.10)

\[
= \frac{1}{2} [f_{33} + f_{22} - 2\tau (f_{32} + f_{23}) - 4\sqrt{3}\tau(f_{34} + f_{43}) + 2\sqrt{6}\tau(f_{12} + f_{21})]
\]

where \(f_{nm}\) is the form factor for the parabolic Landau levels (Eq. E.6).

**Impurity potential**

The impurity potential is \(U(\mathbf{r}) = V_0 \ell_B^2 \delta(\mathbf{r})\). The matrix elements of impurity potential projected to the \(n\)-th and \(m\)-th parabolic Landau levels are:

\[
U^\alpha_{j_1, j_2, n, m} = V_0 \frac{2\pi \ell_B}{L_y} \sum_{l=0}^{\infty} \sum_{k=0}^{\infty} X_{j_1} + iL_x \left[ \frac{X_{j_2} + kL_x}{\ell_B \alpha^{1/4}} \right] \times \left[ \frac{X_{j_2} + kL_x}{\ell_B \alpha^{1/4}} \right] e^{(X_{j_1} + iL_x)^2 + (X_{j_2} + kL_x)^2} \sqrt{2\pi} \sqrt{\hat{\alpha}}
\]

(E.11)

In the parabolic dispersion case, one only need to consider the case \(n = m\), and in the main text we focus on the lowest Landau level, so \(n = m = 0\); on the other hand, in the tilted Dirac case there exist non-trivial terms with \(n \neq m\), the impurity matrix elements \(\langle +3, \tau | \hat{U}(\mathbf{r}) | +3, \tau \rangle_{j_1, j_2}\) are linear combinations of \(U_{j_1, j_2, n, m}\) with \(n, m = 1, 2, 3, 4\), which is similar to the form factor in Eq. (E.10).

**Coulomb interaction**

The Coulomb interaction in a finite system has the form

\[
V(\mathbf{r}) = \frac{1}{L_x L_y} \sum_{\mathbf{q}} V(\mathbf{q}) e^{i\mathbf{q} \cdot \mathbf{r}}
\]

(E.12)

where \(V(\mathbf{q}) = \frac{2\pi e^2}{\ell_B}\) for finite size torus with the circumference \(L_x\) and \(L_y\). Here \(\mathbf{q} = \frac{2\pi s}{L_x}, \frac{2\pi t}{L_y}\) takes discrete values to ensure the periodicity.

The projected Coulomb interaction between two electrons in the valleys \(i\) and \(j\) (\(i, j\) can either be the same valley or two different valleys) into the \(n\)-th Landau level has the form:

\[
P_n V(\mathbf{r}_i - \mathbf{r}_j)P_n = \frac{1}{L_x L_y} \sum_{\mathbf{q}} V(\mathbf{q}) F^n_i(\mathbf{q}_i) F^n_j(\mathbf{q}_j)^* e^{i\mathbf{q} \cdot (\mathbf{R}_i - \mathbf{R}_j)}
\]

(E.13)

Here we have introduced the guiding center operator \(\mathbf{R}_i\) for valley \(i\), which is related to the position operator as follows:

\[
\mathbf{r}_i = \mathbf{R}_i - \ell_B^2 e \mathbf{p}_i = \mathbf{R}_i - \ell_B^2 e S_i^{-1} \mathbf{p}_i
\]

where \(e\) is the rank-2 levi-civita symbol and \(S_i\) is the \(S\) tensor associated to valley \(i\), which has been defined earlier for both parabolic and Dirac dispersions. Accordingly, wavevector \(\mathbf{q}_i\) is defined as:

\[
\mathbf{q}_i = -S_i^{-1} e \mathbf{q}
\]

(E.15)
For the numerical results presented in the main text, the valleys have the same velocity ratio \( r \) (or mass ratio \( \alpha = r^2 \)) and opposite \( \tau \). Thus we have \( \mathbf{q}_i = \mathbf{q}_j = \mathbf{q}^\alpha, F^n_{ij}(\mathbf{q}^\alpha) = F^n(\mathbf{q}^\alpha, \pm \tau) \). While for the numerics to be presented in the next section for anisotropic parabolic dispersion at \( n = 0 \) LL, different valleys have different mass ratio \( \alpha \) and \( \beta \), where \( \beta = \frac{1}{\alpha} \) for the two orthogonal-orientated valleys of interest. There we have \( \mathbf{q}_{i/j} = \mathbf{q}^{\alpha/\beta} \), and \( F^n_{ij} = f_{00} \).

F. Quantum Hall Nematics with Anisotropic Parabolic Dispensions

After considering electron-electron interaction in ferroelectric states in the main text, here we illustrate a simpler scenario where the anisotropic parabolic dispersion is used so that the impurity only hosts a single bound state. The two valleys A and B are parabolic dispersive with the same aspect ratio, but vertical elliptical axes, meaning that if we choose the principal axes along the same direction for two valleys, there mass ratio will satisfy \( \alpha = \frac{1}{\beta} \). A smaller system size with \( N_0 = 20 \) single-valley orbitals is enough to demonstrate this case. The corresponding energy spectra with disorder are shown in Fig. 5 and some representative tunneling density profiles are shown in Fig. 6 with various mass ratios. Again, just like what happens in the ferroelectric state around an impurity, when the impurity potential is larger than a certain threshold a quasihole state becomes the new ground state. Adding an electron to this state would lead to an exciton state, and the resulting density profile can be captured by STM measurements.

![Energy spectra with disorder](image)

FIG. 5: The energy spectra with increasing impurity potentials, as indicated in the legend, blue lines represent \( \{N_0, 0\} \), red lines \( \{N_0, 1\} \), orange lines \( \{N_0 - 1, 0\} \), purple lines \( \{N_0 - 1, 1\} \) and green lines \( \{N_0 - 2, 0\} \). The mass ratios(\( \alpha = m_x/m_y \)) in panels (a), (b), (c), (d) are 1, 2, 4, 8, respectively. The orbital number \( N_0 = 20 \).
FIG. 6: The tunneling matrix elements from the ground state \( \{ N_0 - 1, 0 \} \) to the excitonic state \( \{ N_0 - 1, 1 \} \) for different mass ratio: \( \alpha = 2 \) in (a, b) and \( \alpha = 8 \) in (c,d), which are proportional to the differential conductance obtained by direct STM measurements. The strength of impurity potential is set to be \( 0.6 \frac{e^2}{2l B N_0} \) and the length scale is in the unit of \( l_B \).

[1] Y. Tanaka, Z. Ren, T. Sato, K. Nakayama, S. Souma, T. Takahashi, K. Segawa, and Y. Ando, Nat. Phys. 8, 800 (2012).
[2] Okada, Y., M. Serbyn, H. Lin, D. Walkup, W. Zhou, C. Dhital, M. Neupane, S. Xu, Y. J. Wang, R. Sankar, F. Chou, A. Bansil, M. Z. Hasan, S. D. Wilson, L. Fu, and V. Madhavan (2013), Science 341 (6153), 1496.
[3] J. Liu, W. Duan, and L. Fu, Phys. Rev. B 88, 241303(R) (2013).