Screening Charged Impurities and Lifting the Orbital Degeneracy in Graphene by Populating Landau Levels

Adina Luican-Mayer,[1] Maxim Kharitonov,[1] Guohong Li,[1] ChihPin Lu,[1] Ivan Skachko,[1]
Alem-Mar B. Goncalves,[1] K. Watanabe,[2] T. Taniguchi,[2] and Eva Y. Andrei[1]

[1]*Department of Physics and Astronomy,*
Rutgers University, Piscataway, NJ 08854, USA
[2]*Advanced Materials Laboratory, National Institute for Materials Science,*
1-1 Namiki, Tsukuba, 305-0044, Japan

(Dated: December 3, 2013)

Abstract

We report the observation of an isolated charged impurity in graphene and present direct evidence of the close connection between the screening properties of a 2D electron system and the influence of the impurity on its electronic environment. Using scanning tunneling microscopy and Landau level spectroscopy we demonstrate that in the presence of a magnetic field the strength of the impurity can be tuned by controlling the occupation of Landau-level states with a gate-voltage. At low occupation the impurity is screened becoming essentially invisible. Screening diminishes as states are filled until, for fully occupied Landau-levels, the unscreened impurity significantly perturbs the spectrum in its vicinity. In this regime we report the first observation of Landau-level splitting into discrete states due to lifting the orbital degeneracy.

Charged-impurities are the primary source of disorder and scattering in two dimensional (2D) electron systems[1]. They produce a spatially localized signature in the density of states (DOS) which, for impurities at the surface, is readily observed with scanning tunneling microscopy and spectroscopy (STM/STS)[2]. In this respect graphene[3,4,5,6], provides a unique playground for elucidating the role of impurities in 2D[6,7,8]. They are particularly important in the presence of a magnetic field when the quantization into highly degenerate Landau levels (LL) gives rise to the quantum Hall effect (QHE). In this regime charged-impurities are expected to lift the orbital-degeneracy causing each LL in their vicinity to split into discrete sub-levels[9]. Thus far however the sub-levels were not experimentally accessible due to the difficulty to attain sufficiently clean samples that would allow isolating a single impurity. Instead, previous experiments[10,11] presented a picture of bent levels which could be interpreted semi-classically in terms of electronic drift trajectories moving along the equipotential lines defined by a dense distribution of charged-impurities.

In this work we employed high quality gated graphene devices to access to the electronic spectrum in the QHE regime in the presence of an isolated charged-impurity. We demonstrate that the strength of the impurity can be controlled by tuning the LL occupation with a back-gate-voltage. For almost empty LLs the impurity is screened and essentially invisible whereas at full LL occupancy screening is very weak and the impurity attains maximum strength. In the unscreened regime we resolve the discrete quantum-mechanical spectrum arising from lifting the orbital-degeneracy.

The low energy spectrum of pristine graphene, gives rise to a linear DOS, which vanishes at the charge neutrality point (CNP). In the presence of a magnetic field B, the spectrum is quantized into a sequence of LLs characteristic of massless Dirac fermions:

$$E_N = \pm\frac{v_F}{l_B}\sqrt{2|N|}, N = 0, \pm1, \pm2, ... \quad (1)$$

where v_F is the Fermi velocity, $l_B=\sqrt{\hbar/eB}$ the magnetic length, e the electron charge, $\hbar$ the reduced Planck constant and $\pm$ refers to electron (hole) states with LL index N>1 (N<1). We employed LL spectroscopy[5] to study the electronic properties of graphene and their modification in the presence of a charged-impurity. The LL spectra were obtained by measuring the bias voltage dependence of the differential tunneling conductance, dI/dV, which is proportional to the $DOS(E, \mathbf{r})$, at the tip position $\mathbf{r}$. Here V=(E-E_F)/e is the bias voltage and E the energy measured relative to the Fermi level, E_F.

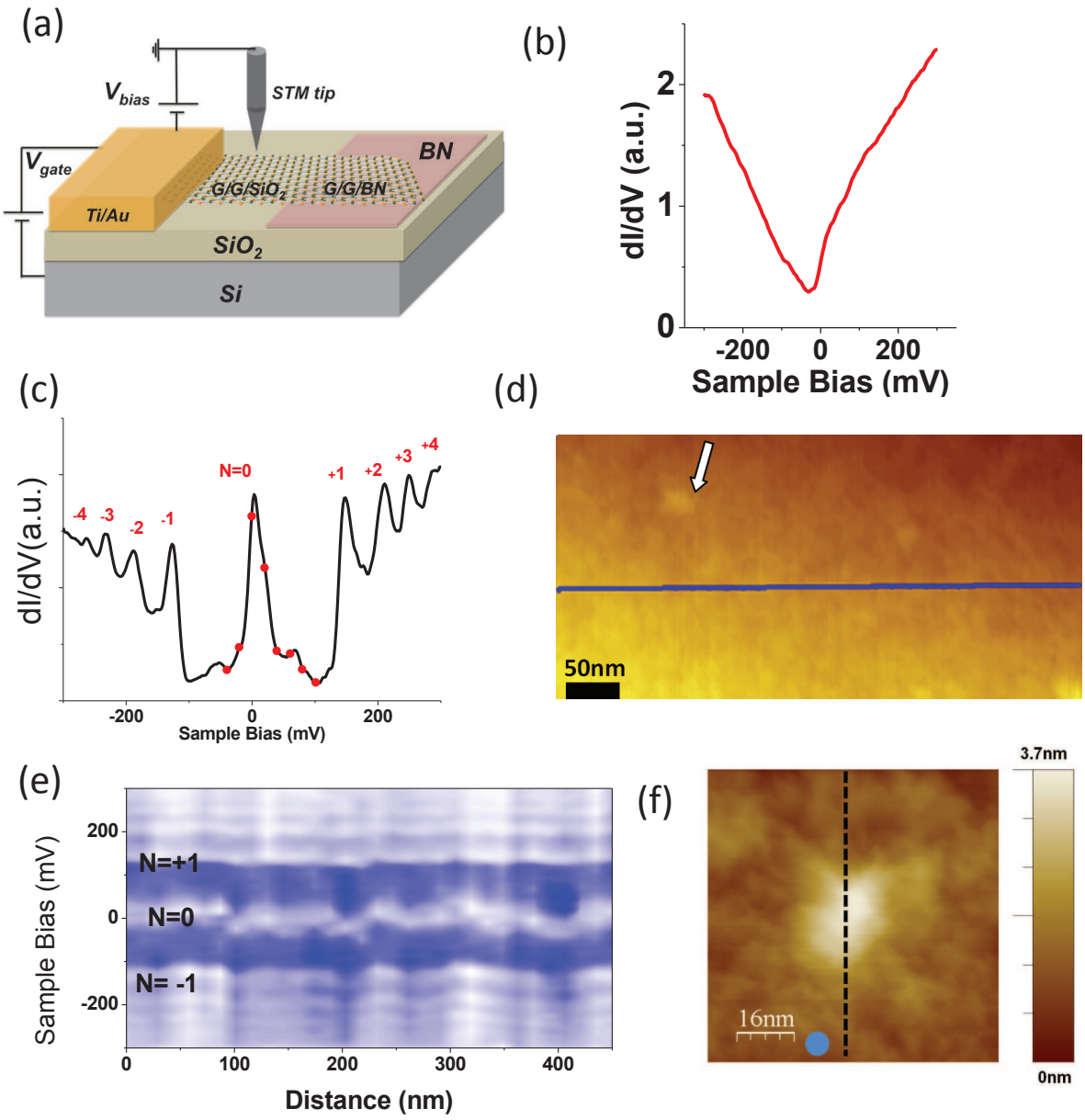

FIG. 1. (a) Schematics of gated graphene device illustrating the two stacked graphene layers deposited on SiO_2 and on h-BN in ($G/G/SiO_2$ and $G/G/BN$). The graphene layers share the same electrode. (b)Zero field STS far from the impurity. (c)STS at B=10T and V_g=0V shows well resolved quantized LLs. (d) Topographic image indicating the line where the spectra in panel e were taken and the position of the isolated impurity. (e)STS line cut along the line shown in panel d; N=0 is clearly resolved. (f) STM topography zoom-into area with isolated impurity (V_B=250mV, I_t=20pA).

Samples were prepared by exfoliating graphene from analyzer-grade HOPG and deposited on a doped Si back-gate capped with 300nm of chlorinated SiO_2[12]. To achieve high quality we used two superposed graphene layers twisted away from Bernal stacking by a large angle. This ensures that the spectrum of single layer graphene is preserved[13][14] while reducing the random potential fluctuations due to substrate imperfections.Hexagonal boron-nitride (h-BN) flakes which significantly reduce the corrugation of graphene[15] were also employed (Figure 1a), but the data reported is restricted to the SiO_2 substrate.

Using the STM tip as a capacitive antenna[16] we located the samples at low temperature and performed STS to identify areas of interest. A typical zero-field spectrum taken far from an impurity in Figure 1b reveals the V-shaped DOS characteristic of single layer graphene (SLG). In finite-field the spectrum develops pronounced peaks (Figure 1c) at energies corresponding to the LLs[17] that are well resolved up to N=4 in both electron and hole sectors, attesting to good sample quality. Fitting the field and level index dependence to Equation 1 confirms the massless-Dirac-fermion nature of the quasiparticles with v_F=1.2 $\times 10^6 m/s$, consistent with measurements on SLG. Charged-impurities were located using LL-spectroscopy to measure the separation between E_F and the CNP which coincides with the N=0 level. LL-spectroscopy is more sensitive to the position of the CNP than the broad zero-field spectrum. The search for impurities starts with a topography image (Figure 1d) followed by STS. An intensity map of the LLs as a function of position (Figure 1e) shows the fluctuations of the N=0 level in response to charged-impurities. Zooming into this area the impurity appears as an isolated bright region in the center of Figure 1f. To visualize its effect on the spatial distribution of the electronic-wavefunction we measured constant energy DOS maps (Figure 2). The maps are roughly radially symmetric, consistent with a charged-impurity at the center. We note that for energies within a gap between LLs the electronic DOS (bright region) is tightly localized on the impurity. In contrast, for energies in the center of the LL the electronic DOS extends across the entire field of view while avoiding the impurity. This fully supports the picture which attributes the QHE plateaus to the existence of localized impurity states in the gaps between LLs.

We next studied the effect of LL occupancy (filling) by tuning the gate-voltage, V_g , to progressively fill the LLs. The LL filling factor is ν=n/n_0(B) where n$\approx 7\times 10^{10}$ V_g (V) cm^{-2} is the carrier density and $n_0(B)=g_l g_v g_s \frac{Be}{2\pi\hbar}$ is the degeneracy/area of the LL. Here g_l=g_v=g_s=2 represent the layer, valley and spin degeneracy respectively. Placing the STM

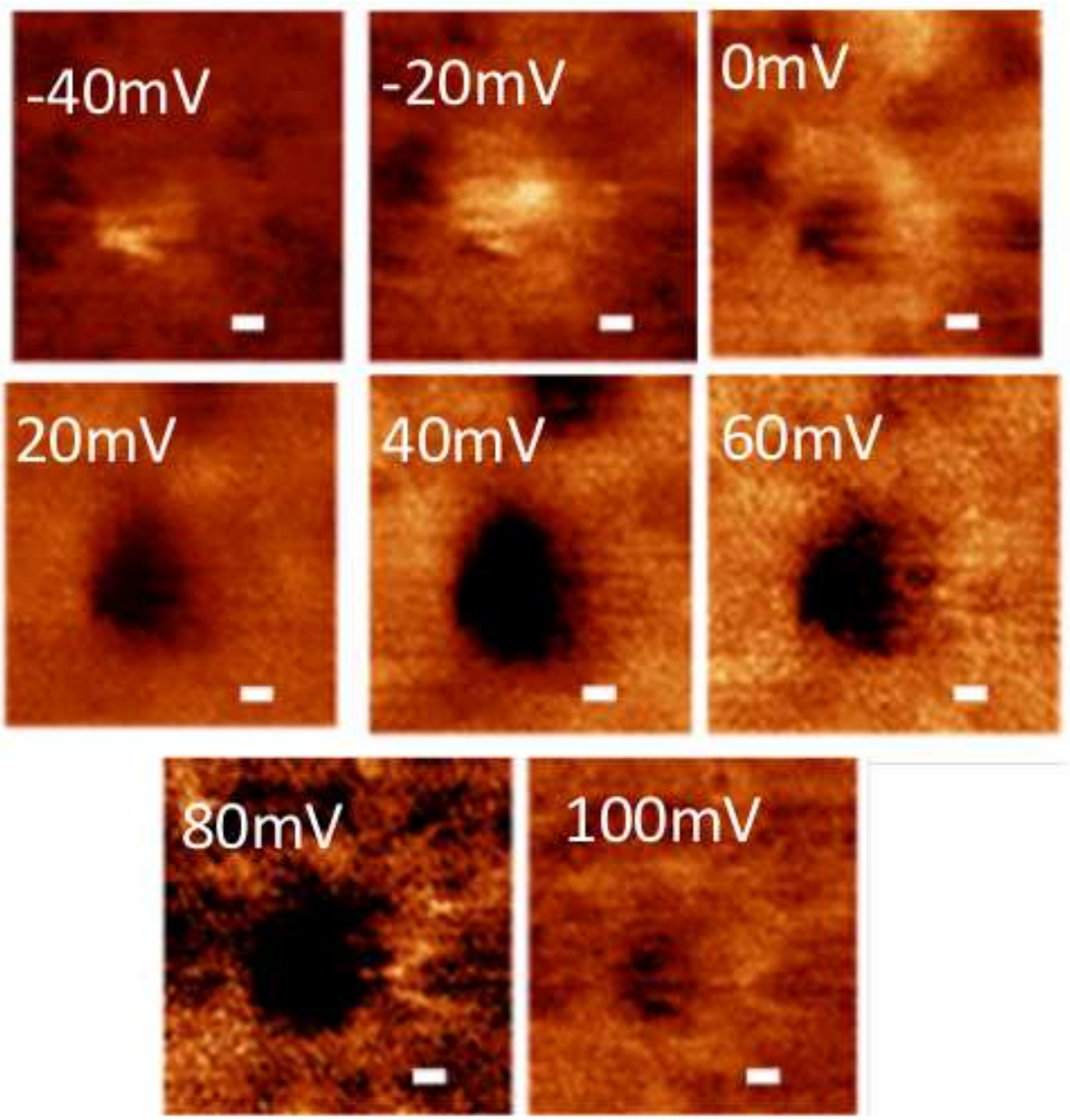

FIG. 2. Spatial dI/dV maps at B=10T near the impurity taken at indicated bias voltages. Scale bar for all maps:8.2nm=l_B.

tip far from the impurity we find that as V_g is swept the LL peaks produce a distinctive step-like pattern seen as bright traces in the intensity map of Figure 3a[12,18]. Each step consists of a nearly horizontal plateau separated from its neighbors by steep slopes. The separation between the centers of steep segments, $\Delta V_g \approx 28$V, gives the LL degeneracy $\approx 2\times10^{12}$ cm^{-2} for B=10T, as expected for this double-layer sample[19]. The plateau indicates that the Fermi-energy remains pinned within a narrow energy band around the center of the

LL until the plateau states are filled. A further increase in V_g populates the sparse states in the gap producing the steep slopes[20].

To explore the influence of the impurity on the LLs we follow the spatial evolution of spectra along a trajectory traversing it (Figure 1f) for a series of gate-voltages. As shown in Figure 3b, for certain gate-voltages the spectra become significantly distorted close to the impurity, with the N=0 level shifting downwards toward negative energies. The downshift indicates an attractive potential produced by a positively charged-impurity. Its strength, as measured by the distortion of the N=0 LL, reveals a surprisingly strong dependence on LL filling. In the range of gate-voltages corresponding to filling the N=0 LL (-15V< V_g <9V) the distortion grows monotonically with filling. At small fillingit is almost absent indicating that the impurity is effectively screened and it reaches its maximum value close to full occupancy. At full occupancy the N=0 level shifts by as much as ≈0.1eV indicating that the effect would survive at room temperature. We note that this distortion is only present in the immediate vicinity of the impurity. Farther away no distortion is observed for all the carrier densities studied here.

We attribute the variation of the impurity strength with filling to the screening properties of the electron system. For a positively (negatively) charged-impurity and almost empty (full) LLs, unoccupied states necessary for virtual electron transitions are readily available in the vicinity of the impurity, resulting in substantial screening. By contrast for almost filled (empty) LLs, unoccupied states are scarce, which renders local screening inefficient[21]

Remarkably, when screening is minimal (V_g=7V) the N = 0 LL does not shift smoothly, but rather splits into a series of well resolved discrete spectral lines in the immediate vicinity of the impurity. As shown in Figure 4a,b the evolution of the spectra radially outwards from the center of the impurity exhibits a progression of peaks within the N=0 LL. Starting with a single peak at the center of the impurity, it evolves into a well resolved double peak and then a triplet at distances ≈13 nm,20 nm from the center respectively. This behavior can be understood by considering the quantum-mechanical electron motion in the presence of a magnetic-field and a charged-impurity. In one valley and for each spin projection, the two-component wave-function $\psi=(\psi_A,\psi_B)^T$ satisfies an effective Dirac Hamiltonian:$\hat{H}\psi=(\hbar v_F\boldsymbol{\sigma}(\mathbf{p}-\mathbf{eA})+\mathbf{U}(\mathbf{r}))\psi=E\psi$ where, $\boldsymbol{\sigma}=(\sigma_x,\sigma_y)$ are the Pauli matrices in the sublattice space, $\mathbf{p}=-i\hbar\nabla$, $\mathbf{B}=\nabla\times\mathbf{A}$, and $\mathbf{A}=\frac{1}{2}[\mathbf{B}\times\mathbf{r}]$ is the vector potential. We assume a radially symmetric impurity-potential $U(r)$, and neglect spin. In the symmetric-

gauge the eigenstates are characterized by the orbital quantum-number m. For the unperturbed spectrum in Equation 1, the eigenfunctions are $\psi^0_{NmA}(r), \psi^0_{NmB}(r)$ (Figure 4c) where m≥-| N | . Since E_N are independent of m, the LLs have infinite orbital-degeneracy. The impurity lifts this orbital-degeneracy and the eigenenergies split into series of sublevels, E_{Nm}.

To illustrate impurity-induced orbital-splitting we numerically solved the problem for a Coulomb potential U(r)=$\frac{Z}{\kappa}\frac{e^2}{4\pi\epsilon_0}\frac{1}{\sqrt{r^2+a^2}}$ corresponding to a charge Z located a distance a below the graphene plane with κ the effective dielectric constant and ϵ_0 the permitivity of free-space. The resulting simulated-spectrum in Figure 4d, shows that the orbital-degeneracy is lifted resulting in an m dependent energy downshift. The downshift is largest for E_{00}, and diminishes with increasing m and/or N where the unperturbed LLs are approached. For comparison with the STS data we calculated the local tunneling DOS assuming a finite linewidth γ[22]. If $\gamma<\Delta E_{Nm}$ (ΔE_{Nm} spacing between adjacent levels) the discreteness of the spectrum is resolved, but for $\gamma\geq\Delta E_{Nm}$ peaks of adjacent states overlap and merge into a continuous band.

Thus, even if the spectrum is discrete, but the resolution insufficient or if impurities too close together, the measured $D(E,\mathbf{r})$ will still display "bent" LLs, whose energies seemingly adjust to the local potential. The resulting $D(E,\mathbf{r})$, shown in Figure 4d, captures the main features of the data. Upon approaching the impurity the N=0 LL splits into discrete peaks, attributed to specific orbital states. In both experiment and simulation the states $\psi_{0m}(\mathbf{r})$ with m=0,1,2 are well resolved close to the impurity but higher order states merge into a continuous line. Similarly, the discreteness of the spectrum is not resolved for N≠0, consistent with the weaker impurity effect at larger distances. We note that for partial filling (V_g=-5V,0V) as screening becomes more efficient and orbital-splitting is no longer observed the unresolved sublevels merge into continuous lines of "bent" Landau levels (Figure 3b). Thus the capability to tune the strength of the impurity allows us to trace the evolution between the discrete and the previously observed quasi-continuous regimes[11].

We now turn to the effective charge of the impurity and how its screening is affected by LL occupancy. Although we do not have in-situ chemical characterization of the impurity we can assume based on chemical-analysis of similar samples that the most likely candidate consistent with our observations is an Na^+ ion adsorbed on the surface of the SiO_2 substrate[23]. It is well known that Na^+ ions are ubiquitous in cleanrooms and laboratory environments

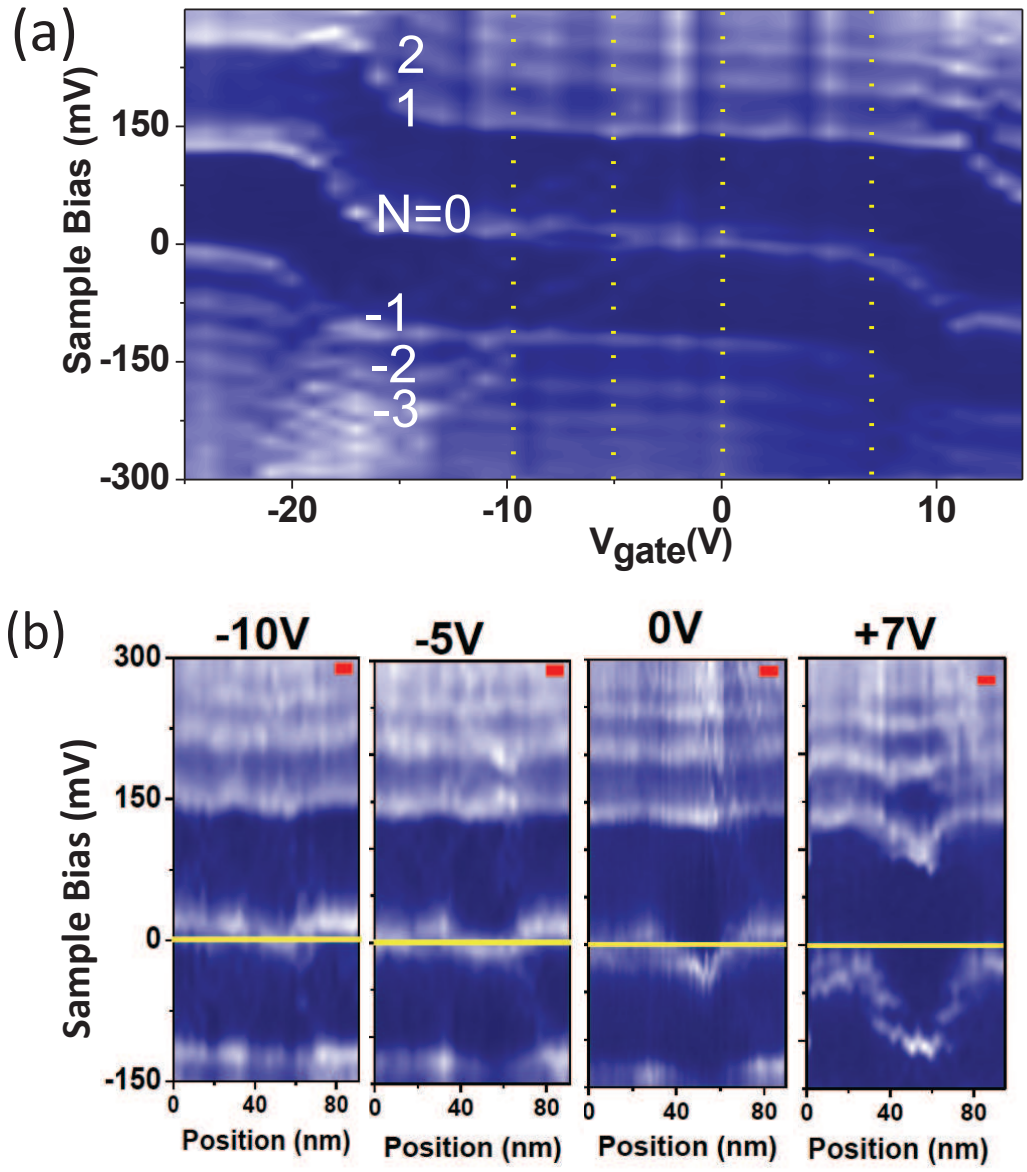

FIG. 3. Impurity screening by populating Landau levels. (a)DOS map at B=10T showing evolution of LLs as a function of gate-voltage taken far from the impurity at the position indicated by the blue dot in Figure 1f. Dashed lines indicate gate-voltages at of the spectra in b. (b)LL maps across the impurity for indicated gate-voltages. The distortion of the LL sequence by the impurity is strongest for filled levels (V_g=+7V), diminishing as filling is reduced and becoming negligible at V_g=-10V.

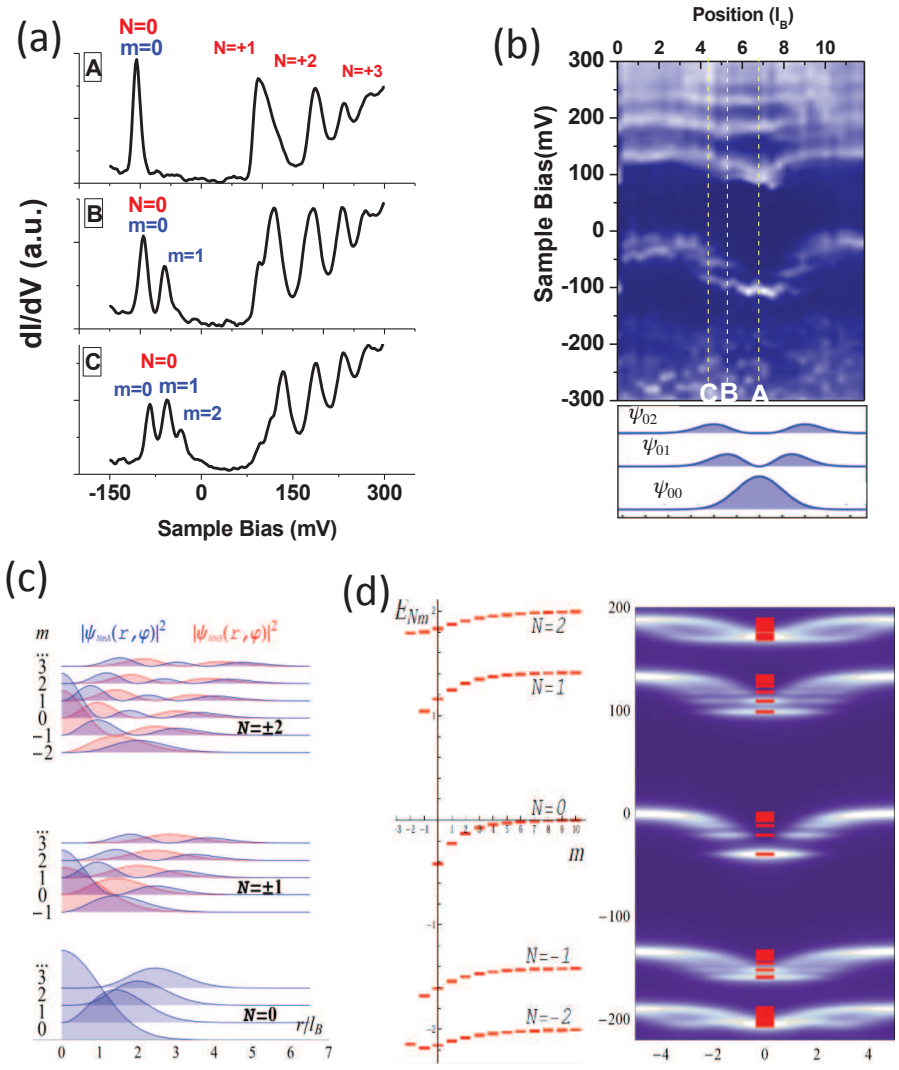

FIG. 4. Lifting the orbital-degeneracy. (a)dI/dV spectra for B=10T and V_g=7V at the positions indicated inl b reveal peaks corresponding to N=0 and m=0, m=0,1 and m=0,1,2. (b)Top: LL map across the impurity for V_g=7V. Dashed lines at 0nm, 13nm and 20nm from the center of the impurity indicate the position of the spectra in a. Bottom: calculated probability densities for ψ_{0m} with m=0,1,2 are consistent with the spatial distribution of the spectral lines in the top panel. (c)Calculated probability densities on the two graphene sublattice A (blue) and B (red). (d) Left: simulated spectrum near an impurity illustrating lifting the orbital-degeneracy in different LLs. Right: simulated DOS near an impurity. Linewidth γ=0.05 v_F/l_B . Red lines represent calculated energies, E_{Nm} shown in the left panel.

and they are readily adsorbed on SiO_2[24]. For a Na^+ ion adsorbed on the substrate underneath the first graphene layer a≈0.6nm≪l_B. A rough estimate of its effect on the spectrum can be obtained by equating the measured energy shift ΔE_{00}≈0.1eV, obtained at V_g=+7V to the calculated value in first order perturbation theory[25] $\Delta E_{00} \approx \frac{Z}{\kappa}\frac{e^2}{4\pi\epsilon_0 l_B}(\pi/2)^{1/2}$ we obtain Z/κ=2.5. Factoring out the contribution of the SiO_2 substrate, κ_{SiO_2}=4, from the expression for the dielectric constant[26] $\kappa=\kappa_{gr}(\kappa_{SiO_2}+1)/2$, gives $Z/\kappa_{gr} \cong 1$ where κ_{gr} is the static dielectric constant of graphene. This indicates that graphene provides no screening for fully occupied LLs and that bare charge of the impurity is Z=+1. In the limit of almost empty LLs (V_g=-10V) the estimated $\kappa_{gr} \approx 5$ indicates that graphene strongly screens the impurity potential[27]. The absence of screening for filled states implies strong Coulomb interactions when E_F lies in a gap consistent with the observation of a fractional QHE in suspended graphene[28] .

This work demonstrates that screening in graphene is controlled by LL occupancy and that it is possible to tune charge-impurities and their effect on the environment by applying a gate voltage or by varying the magnetic field. Due to the large enhancement of the effective fine-structure constant in graphene a charged-impurity with Z≥1 is expected to become supercritical[38] but tuning the effective charge to observe supercritically is extremely difficult[29]. The ability demonstrated here to tune the strength of the impurity in-situ opens the door to exploring Coulomb criticality and to investigate a hitherto inaccessible regime of criticality in the presence of a magnetic field[9].

ACKNOWLEDGMENTS

Funding provided by DOE-FG02-99ER45742 (E.Y.A and G.L), Lucent (A.L-M), NSF DMR 1207108 (I.S.,C.P.L), IAMDN, DOE DE-FG02-99ER45790 (M. K.), Brazilian agency Capes BEX 5115-09-4 (A.M.B.G). We thank M. Devoret for useful discussions and M. Aronson for the HOPG crystal.

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(Van-Hove singularities) which flank the charge neutrality point and are separated from each other by an energy which increases with twist-angle[13]. For twist-angles exceeding 10 degrees the low energy spectrum ($< 1eV$) is indistinguishable from that of single layer graphene. The absence of Van-Hove singularities and the single layer LL spectrum in the data reported here provide direct evidence of layer decoupling. Although there is no topographic signature of the associated Moire pattern, which would require a very sharp tip, the above signatures are taken as evidence for a large twist angle.

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[20] Although STM explores only a small area of the sample, the gate-voltage dependence of the data in Figure 3a reflects the available states in the entire sample including those that are outside the field of view of the STM. This is because the gate covers the entire sample and can populate all available states.

[21] These properties are readily understood by examining the local DOS, $D_s(E,r) = \int d^2r D(E,r)/S$, averaged over a finite-size region, S, around the impurity. Unlike the DOS averaged over the whole sample, $D_s(E,r)$, is manifestly particle-hole asymmetric within a given LL which translates to the particle-hole asymmetry of the local screening.

[22] $D(E,\mathbf{r}) = 4\sum_{Nmi}\delta_\gamma(E-E_{Nm})\psi^\dagger_{Nm}(\mathbf{r})\psi_{Nm}(\mathbf{r})$. Here i is the sublattice index and $\delta_\gamma(E-E_{Nm}) = \gamma/[\pi((E-E_{Nm})^2+\gamma^2)]$ represents the broadened LL. The peak intensity is determined by the probability-density $\psi^\dagger_{Nm}(\mathbf{r})\psi_{Nm}(\mathbf{r})$ and is position dependent.

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[27] This value is comparable to the zero field RPA estimate[26] for double layer graphene $\kappa_{gr} \approx 2.4$,$\kappa_{gr} = 1 + g_l g_s g_v \pi r_s/8 \approx 3.75$ suggesting that when the LLs are almost empty screening of positive charges in graphene is not very different from the zero field case. Here $r_s = 4\pi e^2/hv_F(\kappa_{SiO_2} + 1)$ is the dimensionless Wigner-Seitz radius which measures the relative strength of the potential and kinetic energies in an interacting quantum Coulomb system with linear dispersion. We note that for single layer graphene, $g_l = 1$, screening would be significantly weaker, $\kappa_g r \approx 2.4$.

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