Local sublattice symmetry breaking for graphene with a centro-symmetric deformation

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(Dated: January 14, 2015)

In order to investigate measurable strain signatures on graphene, we focus on scattering properties of an infinite graphene sheet with a single centro-symmetric out-of-plane deformation. Open systems offer the advantage that issues concerning to specific boundary conditions are avoided. A perturbative approach is used in the continuum limit for small deformations to obtain the local density of states (LDOS) and results are compared with a Dirac model solved by iterative scattering matrix methods. Real space imaging reveals a characteristic six-fold symmetry pattern with sublattice symmetry breaking within each fold, consistent with experimental and tight-binding observations. We also provide an analytical expression for the contrast between the local density of states of each sub-lattice in each fold for the case of a Gaussian deformation, showing a scaling law as a function of the amplitude and width of the deformation.

PACS numbers: 72.80.Vp, 73.23.-b, 72.10.Fk, 77.80.bn

Introduction.— Graphene under strain has been largely discussed in the literature and explored for different geometries, with particular features providing alternative routes to confine and control charge carrier.4,5,6 Significant development in the theoretical description of strained graphene elucidated how its electronic properties are modified on strained surfaces. At the microscopic level, a general deformation is described by modifications in the atomic positions which reflects in the Hamiltonian as local changes in the hopping parameter.4,5 In the continuum model these changes appear as an effective gauge field, and electrons with momentum around the Dirac valleys move in the deformed region as in the presence of a pseudomagnetic field.6 Strain also produces a deformation potential, i.e., a scalar field similar to a local chemical potential that can affect electron dynamics.4 Very recently, measurements in high-quality graphene samples on particular substrates suggested a strong connection between random fluctuations in strain and transport properties.6

The use of strain effects to engineer graphene electronic properties has also been explored in several experiments in the last years.7,8,9,10,11,12 As one of the most relevant findings, Levy et al. were able to show the presence of pseudo Landau levels generated by giant pseudomagnetic fields induced by homogeneous strain in graphene nanobubble.8 This experimental confirmation of strain can have striking effects on the electronic properties of graphene has been followed by other experiments that explore the effect of different geometries as a path to control graphene electromechanically.10,11 A generic deformation of a graphene sheet can cause inhomogeneous strain, which results in an effective non-uniform pseudomagnetic field and provides an experimental test-bed to explore the interplay between tunable magnetic fields and Dirac fermions. For example, a scanning tunneling microscope (STM) tip has been used not only to probe samples, but also to continuously deform graphene nanomembranes, demonstrating confinement due to non-uniform pseudomagnetic field.10 For a similar experimental setup, Mashoff et al. obtained atomically resolved STM images of stable and lifted regions of graphene.11 Whereas a hexagonal arrangement of the carbon atoms was found at unstrained regions, as expected for monolayer graphene,11 within the strained area a triangular pattern of bright spots was observed, signaling a symmetry breaking between A and B sublattices in some regions. At the time the authors speculated the effect to be caused by an instability in which the different sublattices acquire a zigzag configuration with respect to the substrate. However, a local sublattice re-arrangement requires energies that are prohibitive within the regime of STM imaging, making such scenario rather unlikely. Atomistic tight-binding models12,13 have predicted the development of such asymmetry but a continuum, symmetry-based description remains missing. These results indicate an incomplete understanding of the fundamental electronic properties of graphene samples where local manipulation produce effective inhomogeneous gauge and scalar fields.

In this work, we approach this problem by investigating the electronic properties of a graphene sheet in the presence of a Gaussian out-of-plane deformation. The strain produced by such distortion is represented by a pseudomagnetic field with trigonal symmetry and embodies a good approximation to standard experimental configurations, while still allowing for analytical treatment. We use a scattering formalism based on the continuum description of graphene to address the question of potential confinement of electrons due to this deforma-
tion. In particular we calculate the LDOS and show that a noticeable imbalance in the distribution of charge density between the two graphene nonequivalent sublattices appears even for small deformations, providing a possible explanation for the experimentally observed sublattice asymmetry. We perform exact numerical calculations and show that these results are well described within an analytical perturbative approach for small deformations. We analyze the dependence of the maximum LDOS contrast between sublattices on energy and strain strength, providing a scaling dependence with the parameters of the deformation. Finally, we also show that the effective scalar field introduced by strain minimally modifies the predicted sublattice asymmetry.

Model.— The electronic properties of undeformed graphene are, for low energies and large system sizes, governed by two copies of a two-dimensional (2D) Dirac Hamiltonian $H_0 = v_F \mathbf{p} \cdot \mathbf{\sigma}$ where $v_F \approx 10^6 m/s$ is the velocity of graphene electrons, $\mathbf{p}$ the electronic momentum around the K (K') point, and $\mathbf{\sigma} = (\sigma_x, \sigma_y)$ are Pauli matrices reflecting the pseudospin degree of freedom associated with the sublattice structure of the honeycomb lattice. The strain is produced by a mechanical deformation modeled with a height-profile $h(\mathbf{r})$ that is centro-symmetric and is written generically as $h(\mathbf{r}) = A h_0(r/b)$, where $h_0$ contains the radial profile, and the parameters $A$ and $b$ describe amplitude and effective radius of the deformation. In the following, to illustrate our results, we consider the case of a Gaussian bump with height profile $h_0(x) = e^{-x^2}$. Note however that our results below are qualitatively valid for a generic profile $h_0$ with axial symmetry.

The effect of such deformation on the electronic properties in the continuum limit are described within the theory of elasticity. For an out-of-plane deformation the strain tensor of elasticity is derived from the height profile according to $\epsilon_{ij} = \frac{1}{2} \partial_i h \partial_j h$, which in polar coordinates $(r, \theta)$ reads

$$
\epsilon = \alpha f(r/b) \begin{pmatrix}
\cos^2 \theta & \sin \theta \cos \theta \\
\sin \theta \cos \theta & \sin^2 \theta
\end{pmatrix},
$$

where $\alpha = A^2/b^2$ sets the strength of the strain, while its spatial distribution is contained in the function $f(x) = \frac{1}{2} [h_0'(x)]^2$. For the Gaussian profile, one has $f(x) = \frac{2 x^2 e^{-2x^2}}{2 x^2 e^{-2x^2}}$.

In the presence of the deformation, electrons experience the strain as a gauge field

$$
\mathbf{A}(\mathbf{r}) = -\frac{g_v}{v_F} \alpha f(r/b) \begin{pmatrix}
\cos 2\theta \\
-\sin 2\theta
\end{pmatrix},
$$

where we chose the zigzag direction to lie along the $x$-axis. The coupling constant is $g_v = \beta h v_F / 2 a \approx 7 eV$. The coupling constant is $\beta = |\partial \log t / \partial \log a| \approx 3$, and $t$ and $a$ the hopping parameter and the lattice constant of graphene.

For the radial symmetric deformation, the associated pseudomagnetic field $\mathbf{B} = \nabla \times \mathbf{A}$ shares the trigonal symmetry of the graphene lattice,

$$
B_z(\mathbf{r}) = \frac{h}{e} \left( -\frac{\beta}{2ab} \right) 0 b_0(r/b) \sin 3\theta
$$

where the spatial profile is given by the function $b_0(x) = \frac{2}{x^2} - f'(x)$. For the Gaussian-shaped deformation, one has $b_0(x) = 8 x^3 e^{-2x^2}$.

In addition to the gauge field, the electrons are also exposed to a scalar potential proportional to the trace of the strain tensor, in our model given by $V(\mathbf{r}) = -g_s \alpha f(r/b)$ with a value of $g_s = 3 eV$. The low-energy electronic properties in the presence of the deformation are hence described by

$$
H = v_F [\mathbf{p} + e \mathbf{A}(\mathbf{r})] \cdot \mathbf{\sigma} + V(\mathbf{r})
$$

In this article we consider a bump that is smooth on the scale of the lattice constant, such that a coupling between the valleys can be neglected. Moreover, we consider an infinite graphene sheet containing a single deformation, hence our results are independent of finite size effects and boundary conditions.

Perturbation theory.— In this section we present analytic results for the change in the LDOS produced by the scattering of electrons off the Gaussian deformation obtained with a perturbative approach in real space. We consider therefore small deformations that allow for an expansion in the parameter $\alpha$.

From now on we set $\hbar = v_F = 1$, and work around the K valley. The effect of the K' valley is discussed at the end of this section. We split the Hamiltonian in the kinetic part $H_0$ and the perturbation $V$,

$$
V(\mathbf{r}) = e \mathbf{A}(\mathbf{r}) \cdot \mathbf{\sigma} = \begin{pmatrix}
0 & A_-(\mathbf{r}) \\
A_+ (\mathbf{r}) & 0
\end{pmatrix},
$$

FIG. 1: (Color online) Schematic view of the graphene lattice with a magnified out-of-plane deformation. (b) Pseudomagnetic field created by a deformation with a Gaussian height profile as in (a). (c) Spatial profile of the LDOS for sublattice A in the presence of a bump, see Eq. (13). Bright (dark) spots indicate an increase (decrease) of LDOS compared to the undeformed graphene sheet. For sublattice B, the effect is exactly opposite.
which contains the Green’s function of graphene, determined by the Lippmann-Schwinger equation

$$\langle \Phi_m^{(0)}(r) | H | \Phi_n^{(0)}(r') \rangle = \frac{\varepsilon}{4\pi} e^{i m g} \left( e^{-i\theta/2} J_{m-1/2}(\varepsilon r) - i sgn(m) e^{i\theta/2} J_{m+1/2}(\varepsilon r) \right),$$

where $\varepsilon$ denotes the energy of the Dirac fermions (which we assume to be positive, for simplicity), and $m$ is a half-integer index labeling the states according to their angular momentum. $J_n(x)$ denotes the Bessel function of $n$-th order. We chose a normalization such that

$$\int dr' \langle \Phi_m^{(0)}(r') | \Phi_n^{(0)}(r') \rangle = \delta_{nm},$$

where $\delta$ is the Kronecker delta. This equation is orthogonal to the unperturbed state. Since we are interested in the different sublattice occupations, we further introduce the sublattice-resolved LDOS

$$\nu_{A/B}(\varepsilon, r) = \sum_m \langle \Psi_m(r) | P_A/B | \Psi_m(r) \rangle$$

where $P_A/B$ are projectors on the respective sublattice $A/B$. For undeformed graphene, evaluating Eq. (11) with the free states $|\Phi_m^{(0)}(r)\rangle$ produces the well-known value of

$$\nu_{A,B}(\varepsilon, r) = \frac{\varepsilon}{4\pi} \sum_m \left[ J_{m-1/2}(\varepsilon r) \right]^2 = \frac{\varepsilon}{4\pi},$$

for the LDOS per sublattice.

We now want to discuss the effect of the deformation on the LDOS. Specifically we address the limit $\varepsilon b\ll 1$, which is the relevant case for experiments with a radius of a few lattice constants. In this case, one may simplify the results by using the asymptotic expressions of the Bessel and Hankel functions for small arguments $r \lesssim \langle \varepsilon b \rangle$. Upon retaining only the leading contribution for small energies, one finds for the corrections $\delta \nu_{A,B} = \nu_{A,B} - \nu_{A,B}^{(0)}$ to leading order in $\alpha$

$$\frac{\delta \nu_A}{\nu_A} = -\frac{\delta \nu_B}{\nu_B} = -\frac{\beta A^2 b}{\alpha} \sin 3\theta g(r/b)$$

with the function

$$g(x) = \frac{1}{x^3} \int_0^x dy y^3 f(y)$$

To leading order in $\alpha$, one can replace $\nu_{A,B}^{(0)}$ by $\nu_{A,B}$ in the denominator of Eq. (13). Notice that the relative LDOS correction has no dependence on energy. Thus, the deformation changes the local occupation in the different sublattices in opposite directions, and their spatial distribution shares the symmetry of the underlying pseudomagnetic field with a radial distribution governed by the function $g(x)$. Specifically for a Gaussian height profile, one finds

$$g(x) = \frac{1}{x^3} \left[ 1 - e^{-2x^2} \left( 1 + 2x^2 + 2x^4 \right) \right].$$

The spatial distribution of the change in LDOS for sublattice $A$ according to Eq. (13) is shown in Fig. 1(c) for a Gaussian deformation of typical dimensions.

A quantity of experimental relevance is the LDOS contrast between sublattices, defined as

$$C = 2\frac{\nu_A - \nu_B}{\nu_A + \nu_B}$$

which is plotted as a function of the radial distance in Fig. 2(a). Note that, for a fixed width $b$ of the deformation, Eq (13) indicates that the contrast $C$ scales quadratically with the amplitude of a centro-symmetric deformation. This scaling is shown for a Gaussian deformation in Fig. 3 and compared with the exact numerical results presented in the next section.

To conclude this section let’s discuss the role of the two valleys $K$ and $K'$ in these results. As mentioned above, the deformation is smooth enough that does not couple the valleys and their contributions add directly. To see that these are identical, note that the Dirac Hamiltonian
Numerics.— In this section we discuss briefly our exact numerical approach for the continuum model, which takes the same form in both valleys when the spinors are written in the valley symmetric representation (\(\psi_A, \psi_B\)) around valley K, and (\(\psi_B, -\psi_A\)) around valley K'. Note that the components referring to A and B sublattices are interchanged between different valleys. On the other hand, the pseudomagnetic field enters the Dirac equation with opposite sign for each valley (in contrast to a real magnetic field that has the same sign in both valleys). These two effects ensure that their contributions to the sublattice occupancy contrast are identical.

Acknowledgments.— We gratefully acknowledge support by CNPq, and DAAD (D.F.), DFG SPP 1459 and...
Appendix A: Greens functions

In this appendix, we show a derivation of the Green function for graphene given in Eq. 9 in the main text. The defining equation reads

$$ (\varepsilon + i\sigma \cdot \nabla)G(r, r') = \delta(r - r'). $$

(A1)

For the derivation, it is customary to introduce the auxiliary Green function $G_s(r, r')$ via

$$ G(r, r') = (\varepsilon - i\sigma \cdot \nabla)G_s(r, r') $$

(A2)

The advantage of the Green function $G_s$ is that it satisfies a scalar equation (i.e. the pseudospin degree of freedom has been eliminated).

$$ (\varepsilon^2 + \nabla^2)G_s(r, r') = \delta(r - r') $$

(A3)

Moreover, $G_s$ is the solution to the Helmholtz equation in two dimensions. We would like to express the Green function in polar coordinates, thus we express the Laplacian as

$$ \Delta = \frac{\partial^2}{\partial r^2} + \frac{1}{r} \frac{\partial}{\partial r} + \frac{1}{r^2} \frac{\partial^2}{\partial \theta^2}, $$

(A4)

and we write for the $\delta$-function

$$ \delta(r - r') = \frac{1}{r} \delta(r - r') \sum_{\mu} \frac{1}{2\pi} e^{i\mu(\theta - \theta')}, $$

(A5)

where the summation is running over integer $\mu$. We then expand $G_s$ as

$$ G_s(r, \theta; r', \theta') = \sum_{\mu} \frac{1}{2\pi} e^{i\mu(\theta - \theta')} g_{\mu}(r, r') $$

(A6)

where $g_{\mu}(r, r')$ is determined by

$$ \left(\varepsilon^2 + \frac{1}{r} \frac{\partial}{\partial r} + \frac{\mu^2}{r^2}\right) g_{\mu}(r, r') = \frac{1}{r} \delta(r - r'). $$

(A7)

For $r \neq r'$, $g_{\mu}$ is thus satisfying a Bessel differential equation. We therefore write

$$ g_{\mu}(r, r') = a_{\mu} J_{|\mu|}(\varepsilon r <) H^{(+)}_{|\mu|}(\varepsilon r >) $$

(A8)

with $r_> = \max(r, r')$, $r_< = \min(r, r')$. In this way, we respect the conditions, that

- $g_{\mu}$ is regular at $r \to 0$,
- $g_{\mu}$ is behaving as an outgoing wave ($\propto \frac{e^{\varepsilon r}}{\sqrt{r}}$) for $r \to \infty$ (this is the proper boundary condition for the retarded Green function $\varepsilon \to \varepsilon + i0$),
- $g_{\mu}$ is continuous at $r = r'$.

The remaining coefficient $a_{\mu}$ is determined by the jump condition for the derivative of $g_{\mu}$,

$$ [\partial_r g_{\mu}(r, r')]_{r=r'+0} - [\partial_r g_{\mu}(r, r')]_{r=r'-0} = \frac{1}{r'}, $$

(A9)

which yields $a_{\mu} = \frac{\pi}{2\mu}$. Thus, the result for the scalar Green function reads

$$ G_s(r, \theta; r', \theta') = \frac{1}{4\pi} \sum_{\mu} e^{i\mu(\theta - \theta')} J_{|\mu|}(\varepsilon r <) H^{(+)}_{|\mu|}(\varepsilon r >). $$

(A10)

The Green function for graphene is then found in a straightforward albeit lengthy calculation from Eq. (A2).
Appendix B: Scattering states

The scattering states in the Born approximation are obtained by inserting Eq. 5, Eq. 6 and Eq. 9 of the main text into the following equation

$$|\Psi_m(r)\rangle = |\Phi_m^{(0)}(r)\rangle + \int dr' G(r, r') \nu(r') |\Phi_m^{(0)}(r')\rangle.$$

(B1)

After insertion of the corresponding expressions, one finds

$$|\Psi_m(r)\rangle = |\Phi_m^{(0)}(r)\rangle - i\pi \sum_n |\Phi_n^{(+)}(r)\rangle \int_{r'<r} r' dr' \int d\theta (\Phi_n^{(0)}(r') |\nu(r')\rangle |\Phi_m^{(0)}(r')\rangle)$$

$$- i\pi \sum_n |\Phi_n^{(0)}(r)\rangle \int_{r'>r} r' dr' \int d\theta (\Phi_n^{(-)}(r') |\nu(r')\rangle |\Phi_m^{(0)}(r')\rangle).$$

(B2)

The angular integration is nonzero for $n = m + 3$ and $n = m - 3$. Calculating the matrix elements, one obtains the scattering states given by

$$|\Psi_m(r)\rangle = |\phi_m^{(0)}(r)\rangle + \pi \frac{\beta}{4a} \varepsilon \alpha \left( a_m(r) |\phi_{m+3}^{(+)}(r)\rangle + b_m(r) |\phi_{m+3}^{(+)}(r)\rangle + c_m(r) |\phi_{m-3}^{(0)}(r)\rangle + d_m(r) |\phi_{m-3}^{(0)}(r)\rangle \right)$$

(B3)

with

$$a_m(r) = - \text{sgn}(m) \int_0^r dr' r' f(r'/b) J_{|m+5/2|}(\varepsilon r') J_{|m+1/2|}(\varepsilon r')$$

$$b_m(r) = \text{sgn}(m-3) \int_0^r dr' r' f(r'/b) J_{|m-5/2|}(\varepsilon r') J_{|m-1/2|}(\varepsilon r')$$

$$c_m(r) = - \text{sgn}(m) \int_r^\infty dr' r' f(r'/b) H_{|m+5/2|}^{(+)}(\varepsilon r') J_{|m+1/2|}(\varepsilon r')$$

$$d_m(r) = \text{sgn}(m-3) \int_r^\infty dr' r' f(r'/b) H_{|m-5/2|}^{(+)}(\varepsilon r') J_{|m-1/2|}(\varepsilon r').$$

(B4)

The leading order correction to the local density of states per sublattice up to first order on $\alpha$, when the deformation is present, is given by

$$\frac{\delta \nu_A}{\nu_A}(\varepsilon, r) = - \pi \frac{\beta}{4a} \varepsilon \alpha e^{3\theta} \sum_m J_{|m-1/2|}(\varepsilon r) H_{|m+5/2|}^{(+)}(\varepsilon r) a_m(r)$$

$$+ \pi \frac{\beta}{4a} \varepsilon \alpha e^{-3\theta} \sum_m J_{|m-1/2|}(\varepsilon r) H_{|m+7/2|}^{(+)}(\varepsilon r) b_m(r)$$

$$- \pi \frac{\beta}{4a} \varepsilon \alpha e^{3\theta} \sum_m J_{|m-1/2|}(\varepsilon r) J_{|m+5/2|}(\varepsilon r) c_m(r)$$

$$+ \pi \frac{\beta}{4a} \varepsilon \alpha e^{-3\theta} \sum_m J_{|m-1/2|}(\varepsilon r) J_{|m+7/2|}(\varepsilon r) b_m(r) + c.c.$$

(B5)

with $\nu_A = \frac{\nu}{4\pi}$ for clean graphene. Using the asymptotic forms of the Bessel functions for small arguments

$$J_n(x) \approx \frac{1}{2^n n!} x^n,$$

(B6)

$$Y_n(x) \approx \begin{cases} 
\frac{2}{\pi} \ln \left( \frac{e}{2x} \right), & n = 0 \\
-\frac{2^n (n-1)!}{\pi x^n}, & n \geq 1
\end{cases}$$

(B7)
one finds the dominant contribution to $\delta \nu_A$ from the second line, for $m=1/2$ (+c.c.)

$$\frac{\delta \nu_A}{\nu_A} = -\frac{\beta b}{a} \alpha \sin 3 \theta \ g(r/b),$$

which corresponds to Eq. (13) in the main text. The LDOS correction for sublattice B is $\delta \nu_B/\nu_B = -\delta \nu_A/\nu_A$. 

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