

Materials design from non-equilibrium steady states: driven graphene as a tunable semiconductor with topological properties

Thomas Iadecola,¹ David Campbell,¹ Claudio Chamon,¹ Chang-Yu Hou,^{2,3}
Roman Jackiw,⁴ So-Young Pi,¹ and Silvia Viola Kusminskiy⁵

¹*Physics Department, Boston University, Boston, Massachusetts 02215, USA*

²*Department of Physics and Astronomy, University of California at Riverside, Riverside, California 92521, USA*

³*Department of Physics, California Institute of Technology, Pasadena, California 91125, USA*

⁴*Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA*

⁵*Dahlem Center for Complex Quantum Systems and Fachbereich Physik,
Freie Universität Berlin, 14195 Berlin, Germany*

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Controlling the properties of materials by driving them out of equilibrium is an exciting prospect that has only recently begun to be explored. In this paper we give a striking theoretical example of such materials design: a tunable gap in monolayer graphene is generated by exciting a particular optical phonon. We show that the system reaches a steady state whose transport properties are the same as if the system had a static electronic gap, controllable by the driving amplitude. Moreover, the steady state displays topological phenomena: there are chiral edge currents, which circulate a fractional charge $e/2$ per rotation cycle, with frequency set by the optical phonon frequency.

Non-equilibrium quantum systems constitute a natural frontier in physics that is only beginning to be probed by theory and experiment. Non-equilibrium methods can be used to study the properties of condensed matter systems—for instance, pump-probe spectroscopy is emerging as a powerful way of studying strongly-correlated materials [1]. Such methods can also be used to control the properties of materials, as recent work on metallization of dielectrics by optical driving has shown [2, 3]. Particularly exciting is the possibility that such driving, whether optical, mechanical, or otherwise, could be used to control the properties of novel materials such as graphene, paving the way for applications to devices. Graphene’s gaplessness poses a critical challenge to such applications, as the development of graphene-based semiconductors is predicated on the ability to induce a gap.

In this Letter we present a theoretical study in graphene of this non-equilibrium approach to materials design. We demonstrate the possibility of inducing a gap in monolayer graphene by the excitation of optical phonon modes in the lattice. The gap is controlled by a time-dependent Kekulé-pattern bond density wave, which appears in the effective field theory as a complex-valued order parameter Δ that rotates with the frequency Ω of the driven phonon mode. The time-dependence in this order parameter falls into a particularly simple class in that it is completely removable by an axial (valley) gauge transformation. This gauge transformation, which can be viewed as a kind of “boost” to a co-moving “reference frame,” has no effect on the coupling of the system to a heat bath, thereby guaranteeing thermal equilibration in the new frame. Furthermore, the fermion currents are invariant under the transformation, which implies that the electric response of the system is equivalent to that of one with a static gap. In this way, all non-equilibrium aspects of the problem are removed and the system can

be studied as if it were at equilibrium.

The topological consequences of the presence of a Kekulé gap have been studied in the static case, revealing that fractionally charged states can emerge that are bound to vortices in the order parameter Δ [4]. In the driven case, we show that further nontrivial topological phenomena arise: the system supports chiral edge currents of magnitude $J_{\text{edge}} = e\Omega/4\pi$, corresponding to the pumping of a charge $e/2$ per rotation cycle, while the current in the bulk vanishes. Similar currents appear near the center of vortices in the order parameter Δ . These results suggest the possibility that driven graphene could be used as a tunable semiconductor with nontrivial topological properties.

Let us consider spinless electrons [16] hopping on a honeycomb lattice Λ according to the *time-dependent* tight-binding Hamiltonian

$$H = - \sum_{\mathbf{r} \in \Lambda_A} \sum_{j=1}^3 [t + \delta t_{\mathbf{r},j}(\tau)] a_{\mathbf{r}}^\dagger b_{\mathbf{r}+\mathbf{s}_j} + \text{h.c.}, \quad (1)$$

where τ is time and $a_{\mathbf{r}}^\dagger$ and $b_{\mathbf{r}+\mathbf{s}_j}^\dagger$ are fermionic creation operators at sites $\mathbf{r} \in \Lambda_A$ and $\mathbf{r} + \mathbf{s}_j \in \Lambda_B$, with Λ_A and Λ_B the two triangular sublattices forming the hexagonal lattice Λ . The three vectors \mathbf{s}_j , with $j = 1, 2, 3$, connect a site at $\mathbf{r} \in \Lambda_A$ to its three nearest neighbors at $\mathbf{r} + \mathbf{s}_j \in \Lambda_B$ located a distance $|\mathbf{s}_j| = d$ away. The uniform hopping amplitudes t are modulated by time- and site-dependent perturbations $\delta t_{\mathbf{r},j}(\tau)$. In the absence of such perturbations, *i.e.* $\delta t_{\mathbf{r},j}(\tau) = 0$, the Hamiltonian (1) can be diagonalized in momentum space, and the single particle spectrum has two Dirac points at $\mathbf{k} = \mathbf{K}_\pm = \pm \frac{4\pi}{3\sqrt{3}d} (1, 0)$.

We shall now consider the time- and site-dependent perturbations $\delta t_{\mathbf{r},j}(\tau)$ that result from the excitation of the highest-energy optical phonon modes at wavevectors

\mathbf{K}_\pm , with frequency denoted by Ω . The atomic displacements from the lattice sites $\mathbf{r}_{A,B} \in \Lambda_{A,B}$ are

$$\begin{aligned} \mathbf{u}_{\mathbf{K}_\pm}^A(\mathbf{r}_A, \tau) &= c_\pm e^{i\mathbf{r}_A \cdot \mathbf{K}_\pm} e^{-i\Omega\tau} \mathbf{u}_\pm^A + \text{c.c.} \\ \mathbf{u}_{\mathbf{K}_\pm}^B(\mathbf{r}_B, \tau) &= c_\pm e^{i\mathbf{r}_B \cdot \mathbf{K}_\pm} e^{-i\Omega\tau} \mathbf{u}_\pm^B + \text{c.c.} \end{aligned} \quad (2)$$

The coefficients c_\pm are the amplitudes of the excited waves. The normal mode vectors $\mathbf{u}_\pm^{A,B}$ for the highest-energy optical modes with frequency Ω at wavevectors \mathbf{K}_\pm can be determined from a classical analysis of the lattice displacements [5, 6] and are given by

$$\mathbf{u}_\pm^A = \frac{1}{2} \begin{pmatrix} 1 \\ \mp i \end{pmatrix} \quad \text{and} \quad \mathbf{u}_\pm^B = \frac{1}{2} \begin{pmatrix} 1 \\ \pm i \end{pmatrix}. \quad (3)$$

To determine the form of the hopping modulations $\delta t_{\mathbf{r},j}(\tau)$ brought about by the phonon modes, we consider the changes in bond lengths due to the atomic displacements (2) when either the mode at \mathbf{K}_+ or the one at \mathbf{K}_- is excited. For small displacements, the change in the length $d_{\mathbf{r},j}(\tau)$ of the bond connecting site \mathbf{r} and $\mathbf{r} + \mathbf{s}_j$ is [7]

$$\begin{aligned} \frac{\delta d_{\mathbf{r},j}^\pm(\tau)}{d} &= \left| \frac{\mathbf{s}_j}{d} - \left[\frac{\mathbf{u}_{\mathbf{K}_\pm}^A(\mathbf{r}, \tau)}{d} - \frac{\mathbf{u}_{\mathbf{K}_\pm}^B(\mathbf{r} + \mathbf{s}_j, \tau)}{d} \right] \right| - 1 \\ &\approx -\frac{\mathbf{s}_j}{d} \cdot \left[\frac{\mathbf{u}_{\mathbf{K}_\pm}^A(\mathbf{r}, \tau)}{d} - \frac{\mathbf{u}_{\mathbf{K}_\pm}^B(\mathbf{r} + \mathbf{s}_j, \tau)}{d} \right] \end{aligned} \quad (4)$$

Substituting (2) and (3) into (4) and using $e^{i\mathbf{K}_\pm \cdot \mathbf{s}_j} = e^{\pm i \frac{2\pi}{3}(j-1)}$, one obtains

$$\frac{\delta d_{\mathbf{r},j}^\pm(\tau)}{d} = \pm i \frac{c_\pm^*}{d} e^{i\mathbf{K}_\pm \cdot \mathbf{s}_j} e^{\pm i\mathbf{G} \cdot \mathbf{r}} e^{\pm i\Omega\tau} + \text{c.c.}, \quad (5)$$

where the vector $\mathbf{G} = \mathbf{K}_+ - \mathbf{K}_- = 2\mathbf{K}_+$ connects the two Dirac points. The modulation in the hopping matrix element is related to the change in bond length through $\delta t_{\mathbf{r},j}(\tau)/t = \alpha \delta d_{\mathbf{r},j}^\pm(\tau)/d$, where $\alpha \approx 3.7$ is the dimensionless electron-phonon coupling. The resulting $\delta t_{\mathbf{r},j}(\tau)$ can be written as

$$\delta t_{\mathbf{r},j}(\tau) = \frac{1}{3} \Delta(\tau) e^{i\mathbf{K}_+ \cdot \mathbf{s}_j} e^{i\mathbf{G} \cdot \mathbf{r}} + \text{c.c.}, \quad (6)$$

where

$$\Delta(\tau) = i3\alpha t \frac{c_+^*}{d} e^{+i\Omega\tau} \quad \text{for the } \mathbf{K}_+ \text{ mode} \quad (7a)$$

$$\Delta(\tau) = i3\alpha t \frac{c_-}{d} e^{-i\Omega\tau} \quad \text{for the } \mathbf{K}_- \text{ mode}. \quad (7b)$$

The form of the hopping modulations (6) is precisely that of a Kekulé distortion in terms of an order parameter $\Delta(\tau)$ [4] that is time-dependent. Therefore, exciting either the \mathbf{K}_+ or the \mathbf{K}_- mode independently yields a Kekulé order parameter that rotates in time with a frequency Ω in opposite directions for these two phonon modes.

Without loss of generality, we henceforth consider the case where the \mathbf{K}_+ mode is excited, and write $\Delta(\tau) = |\Delta| e^{i\phi(\tau)}$, where $\phi(\tau) = \Omega\tau + \varphi$. All the results for when the \mathbf{K}_- mode is excited instead are obtained from those below by taking $\Omega \rightarrow -\Omega$.

We wish to study the consequences of this rotating order parameter in the context of the effective Dirac field theory of the system, which is valid in the limit where the fermions have relativistic (hyperbolic) dispersion. In order to ensure the validity of this approximation it is necessary to stipulate that $|\Delta|/t \ll 1$ and $\Omega/t \ll 1$, where the uniform hopping amplitude t sets the kinetic energy scale of the problem. In this regime the Hamiltonian (1) corresponds, to first order in a gradient expansion, to the Dirac Lagrangian density [4, 8]

$$\mathcal{L} = \bar{\Psi} \left[\gamma^\mu (i\partial_\mu + \gamma_5 A_{5\mu}) - |\Delta| e^{-i\gamma_5 \phi(\tau)} \right] \Psi, \quad (8)$$

with $\mu = 0, 1, 2$, $\bar{\Psi} = \Psi^\dagger \gamma^0$ and 4×4 Dirac matrices

$$\begin{aligned} \gamma^0 &\equiv \begin{pmatrix} 0 & \mathbb{1} \\ \mathbb{1} & 0 \end{pmatrix}, \quad \gamma^i \equiv \begin{pmatrix} 0 & -\sigma_i \\ \sigma_i & 0 \end{pmatrix}, \\ \gamma_5 &\equiv i\gamma^0 \gamma^1 \gamma^2 \gamma^3 = \begin{pmatrix} \mathbb{1} & 0 \\ 0 & -\mathbb{1} \end{pmatrix}, \end{aligned}$$

where $\mathbb{1}$ is the 2×2 unit matrix and σ_i are the three Pauli matrices. The Dirac spinor $\Psi_{\mathbf{p}}^\dagger = (b_{\mathbf{p},+}^\dagger, a_{\mathbf{p},+}^\dagger, a_{\mathbf{p},-}^\dagger, b_{\mathbf{p},-}^\dagger)$ collects the creation operators $a_{\mathbf{p},\pm}^\dagger$ and $b_{\mathbf{p},\pm}^\dagger$ for the \pm species on sublattices A and B, respectively. The axial gauge field $A_{5\mu}$, examined in a different context in Ref. 8, will play an important role in the discussion of the asymptotic steady state of the driven system. The spatial components A_{5i} correspond physically to acoustic phonons and strain in the graphene lattice. If the lattice is strained uniaxially, the hopping amplitudes change, and the Dirac points shift away from \mathbf{K}_\pm . In this case, the A_{5i} acquire a non-zero average value. In addition, acoustic phonons, either in-plane or out-of-plane, dynamically stretch the bonds and lead to fluctuations of A_{5i} around the average. These acoustic phonons provide a thermal bath and their coupling to the electronic degrees of freedom provides a system-bath interaction, which enables the system to reach an out-of-equilibrium steady state.

We now observe that the time-dependent mass term in the Lagrangian (8) can be brought to a constant one by the axial (valley) gauge transformation

$$\tilde{\Psi} = e^{-i\gamma_5 \frac{\Omega}{2}\tau} \Psi, \quad \tilde{A}_{50} = A_{50} + \frac{\Omega}{2}, \quad \tilde{A}_{5i} = A_{5i}, \quad (9)$$

where $i = 1, 2$. The transformed Lagrangian is found to be

$$\tilde{\mathcal{L}} = \tilde{\Psi} \left[\gamma^\mu (i\partial_\mu + \gamma_5 \tilde{A}_{5\mu}) - |\Delta| e^{-i\gamma_5 \varphi} \right] \tilde{\Psi}, \quad (10)$$

where we used $\{\gamma_5, \gamma^\mu\} = 0$. One can think of this gauge transformation as mapping the problem to a new frame

of reference which is co-moving with the Kekulé mass and in which the Lagrangian is therefore no longer explicitly dependent on time.

The vector current operator $j^\mu = \bar{\Psi}\gamma^\mu\Psi$, which is associated with the electric response of the system, and the axial current operator $j_5^\mu = \bar{\Psi}\gamma^\mu\gamma_5\Psi$ are invariant under (9), as can be seen by rewriting the currents in terms of the transformed fields and applying the identity $[\gamma_5, \gamma^0\gamma^\mu] = 0$. Furthermore, the spatial components A_{5i} of the axial gauge field are invariant under the gauge transformation (9). Since we have taken the fluctuations in A_{5i} to act as a heat bath, we conclude that this transformation leaves the bath invariant. Moreover, it also leaves the system-bath coupling $A_{5i}j_5^i$ invariant. Therefore *all* time-dependences—those of the system, the bath, and the system-bath interactions—are removed by the gauge transformation. The remarkable consequence is that the non-equilibrium steady state of the time-dependent system corresponds to a thermal equilibrium state in the co-moving frame!

Consequently, the gauge-transformed Lagrangian (10) can be analyzed quantum mechanically in the time-independent Schrödinger picture at thermal equilibrium. The Hamiltonian \mathcal{H} corresponding to the Lagrangian (10) takes a particularly simple form in the absence of strain, in which case the externally applied $A_{5\mu} = 0$ for $\mu = 0, 1, 2$, so that $\tilde{A}_{50} = \Omega/2$ and $\tilde{A}_{5i} = 0$:

$$\mathcal{H} = \begin{pmatrix} \boldsymbol{\sigma} \cdot \mathbf{p} - \frac{\Omega}{2} \mathbb{1} & |\Delta| e^{i\varphi} \mathbb{1} \\ |\Delta| e^{-i\varphi} \mathbb{1} & -\boldsymbol{\sigma} \cdot \mathbf{p} + \frac{\Omega}{2} \mathbb{1} \end{pmatrix}, \quad (11)$$

where $\boldsymbol{\sigma}$ is the 2D vector of Pauli matrices and $\mathbf{p} = -i\nabla$. The eigenvalue problem $\mathcal{H}\psi = E\psi$ has been solved in [9] in the context of the superconducting proximity effect in topological insulators [10]; the four energy eigenvalues of the Hamiltonian (11) are given by

$$E_{\pm, \mp} = \pm \sqrt{(p \mp \Omega/2)^2 + |\Delta|^2}. \quad (12)$$

Evidently the gauge transformation (9) maps the time-dependent quantum mechanical problem described by the Lagrangian (8) to a time-independent problem with an energy gap $2|\Delta|$.

It is important to observe that, because the vector current operator j^μ is invariant under (9), *all* observables associated with this current can be calculated from the static Lagrangian (10) without the need for dealing with the original time-dependent mass. In particular, the conductivity tensor σ_{ij} that follows from the Kubo formula written in terms of the current operator j^μ can be computed from the static Lagrangian (10). Consequently, the driven graphene system effectively behaves as a semiconductor with a gap $2|\Delta|$ tunable by the amplitude of the optical phonon modes.

We shall next demonstrate that the rotating Kekulé mass in the Lagrangian (8) gives rise to topological phenomena beyond those that have been found in the static

case. To do this, we follow [11] in studying a variant of (8):

$$\mathcal{L} = \bar{\Psi} [\gamma^\mu (i\partial_\mu + \gamma_5 A_{5\mu}) - |\Delta| e^{-i\gamma_5\phi} - \gamma^3 \mu] \Psi, \quad (13)$$

where the scalar field $\mu = \mu(\mathbf{x})$ corresponds to a staggered chemical potential that establishes an energy imbalance between the sites of Λ_A and Λ_B . The Kekulé field $\Delta = |\Delta(\mathbf{x})| e^{i\phi(\mathbf{x}, \tau)}$, where $\phi(\mathbf{x}, \tau) = \Omega\tau + \varphi(\mathbf{x})$, now carries an explicit spatial dependence. The fields μ and Δ play the role of coexistent masses in the Lagrangian (13) and add in quadrature, *i.e.* the total effective mass of the charge carriers is $\sqrt{\mu^2 + |\Delta|^2}$. In Ref. 11 it is shown that the vector current density in the presence of (space- and time-dependent) order parameters μ and Δ is given by

$$\langle j^\mu \rangle = e \frac{i}{2\pi} \epsilon^{\mu\alpha\beta} \{ \partial_\alpha \chi^* \partial_\beta \chi - i \partial_\alpha [(1 - 2|\chi|^2) A_{5\beta}] \}, \quad (14)$$

where e is the electron charge, $\epsilon^{\mu\alpha\beta}$ is the Levi-Civita symbol, and the complex-valued auxiliary field $\chi \equiv \sin(\theta/2) e^{i\phi}$, where

$$\cos \theta = \frac{\mu}{\sqrt{\mu^2 + |\Delta|^2}}, \quad \sin \theta e^{i\phi} = \frac{\Delta}{\sqrt{\mu^2 + |\Delta|^2}}, \quad (15)$$

with $0 \leq \theta < \pi$ and $0 \leq \phi < 2\pi$. Equations (14) and (15) will form the basis of our discussion of the topological currents that arise from the time-dependence of the mass term in (13) generated by the excitation of the optical phonon modes. We will use μ to examine finite systems, setting $\mu \rightarrow 0$ in the bulk and using the limit $|\mu| \rightarrow \infty$ to define an insulating region outside the sample [17].

The current density in (14) is gauge invariant, and therefore one has the choice of computing it in the frame of reference where ϕ has a time dependence or in the co-moving frame where the time dependence of ϕ (and χ) is removed. It follows that the averaged charge and current densities are

$$\langle \rho \rangle = e \frac{i}{2\pi} \epsilon^{0ij} \partial_i \chi^* \partial_j \chi = \langle \rho \rangle_{\text{static}} \quad (16a)$$

$$\langle \mathbf{j} \rangle = e \frac{\Omega}{2\pi} \hat{\mathbf{z}} \times \nabla |\chi(\mathbf{x})|^2, \quad (16b)$$

where $\hat{\mathbf{z}}$ is the unit vector perpendicular to the plane of the sample.

Several observations are in order. First, notice that the charge density in the case of the time-dependent Kekulé mass is identical to that in the static case. Second, the current density in the case of the time-dependent Kekulé mass is non-vanishing and proportional to the rotation frequency Ω . Notice that the rotating mass breaks time-reversal symmetry, and therefore it is possible to have a non-vanishing current. Third, if the magnitude of the order parameter $|\Delta|$ does not vary spatially, the current vanishes; this is the case in the bulk (interior) of a

uniform graphene sample, where we take $|\Delta|$ to be constant. Fourth, there are necessarily edge currents, which we shall now discuss in detail.

It follows from (16b) that the currents are perpendicular in direction to the gradient of $|\Delta|$. At the boundary of the sample $|\Delta|$ must go from constant to zero. Therefore an edge current should flow parallel to the boundary, within the region of width where $|\Delta|$ varies in space, as depicted in Fig. 1. The edge current is given by

$$\begin{aligned} J_{\text{edge}} &= \int_{\text{in}}^{\text{out}} (\hat{z} \times d\ell) \cdot \langle \mathbf{j} \rangle \\ &= e \frac{\Omega}{2\pi} \int_{\text{in}}^{\text{out}} d\ell \cdot \nabla |\chi|^2 \\ &= e \frac{\Omega}{2\pi} (|\chi_{\text{out}}|^2 - |\chi_{\text{in}}|^2), \end{aligned} \quad (17)$$

where ℓ is a path that traverses the boundary. In the interior of the sample $|\Delta|$ is non-vanishing, so we can set $\mu \rightarrow 0$, and using Eq. (15) we obtain that $|\chi_{\text{in}}|^2 \rightarrow 1/2$. In the exterior of the sample $|\Delta| \rightarrow 0$ and $|\mu| \rightarrow \infty$. Depending on whether μ is positive or negative we obtain $|\chi_{\text{out}}|^2 \rightarrow 0$ or 1, respectively. Therefore, we arrive at the edge current

$$J_{\text{edge}} = -\frac{e}{2} \frac{\Omega}{2\pi} \text{sgn } \mu. \quad (18)$$

The linear relation between J_{edge} and Ω has a quantized coefficient. Note that because $\Omega = 2\pi/T$, where T is the rotation period, the current J_{edge} carries a fractional charge $\pm e/2$ per rotation cycle [18]. This chiral current at the boundary of the steady state bulk insulator is a topological property of the out-of-equilibrium system; the currents are quantized and protected against details at the edge, including disorder.

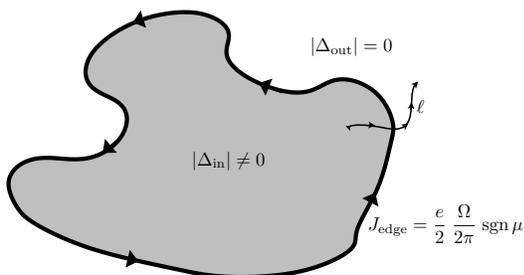


FIG. 1: Chiral edge current resulting from the out-of-equilibrium topological steady-state arising from the excitation of optical phonons at a wavevector \mathbf{K}_+ . The direction of the current is inverted for \mathbf{K}_- phonons, for which the time-dependent order parameter rotates in the opposite direction in time.

The chirality of the edge currents depends on which optical phonon mode is excited, either that with wavevector

\mathbf{K}_+ or \mathbf{K}_- . (Recall that the results for the \mathbf{K}_- excitations are obtained from those for the \mathbf{K}_+ ones by taking $\Omega \rightarrow -\Omega$, and therefore the currents are inverted.) However, the overall sign of the current also depends on $\text{sgn } \mu$.

We now offer a physical explanation of this fact. The mass μ was included in the Lagrangian (13) as a means of terminating the sample with an insulating region. In a physical graphene flake, our findings therefore indicate that the sign of the edge current is determined by the specific shape of the sample. Notice that the direction of the current obtained from the field theory cannot change unless μ changes sign outside the sample. But if this is the case, there will be domain walls separating these regions which support gapless modes. Indeed these walls serve as quantum wires [12] that connect to the sample, as shown in Fig. 2. The direction of the edge currents reverses at the contacts, as shown in the figure. Conservation of current requires that currents of magnitude $J_{\text{wire}} = e\Omega/2\pi$ flow on the wires and split equally at the contacts, traveling as edge states around the sample. The graphene flake in this scenario becomes a charge pump [13] that transports a charge e each time the mass Δ completes a rotation cycle.

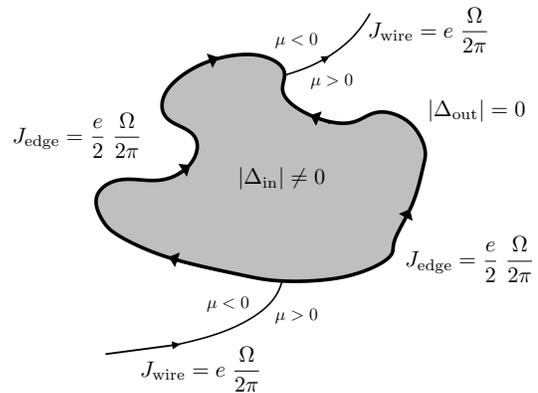


FIG. 2: Currents in the presence of wires described by a domain wall between regions with $\mu > 0$ and $\mu < 0$. The chiralities of the edge currents are opposite on the two sides of the wires, and the current pumped per cycle is an integer multiple of e , while a fraction $e/2$ goes around each side during the cycle.

The next observation concerns zero modes in graphene, which are supported in the presence of vortices in the Kekulé order parameter Δ [4]. An external chiral gauge potential \mathbf{A}_5 was added to render finite the vortex energies, thereby deconfining them [8]. Such an unpinned vortex background can also be present in our time-dependent scenario. In the co-moving frame this involves adding $A_{5,0} = \Omega/2$ to the static problem. We find that zero-energy modes persist both with and without \mathbf{A}_5 , consistent with the findings of [14, 15].

Our final observation concerns the sizes of the gaps

that can be achieved by excitation of the optical phonon modes at \mathbf{K}_{\pm} . From Eqs. (7a) and (7b) we obtain that $|\Delta| = 3\alpha t |c_{\pm}|/d$, where $|c_{\pm}|/d$ is a measure of the relative displacement of the atoms from their equilibrium position due to the phonons, and is controlled by the intensity of the excitations. Using $\alpha \approx 3.7$ and $t \approx 2.8 \text{ eV}$ for graphene, one obtains that for a relative displacement $|c_{\pm}|/d \approx 4 \times 10^{-4}$, or a displacement that is 0.04% of the atomic distance d , the gap should be at the room temperature scale, *i.e.* $2|\Delta| \approx 0.025 \text{ eV}$.

In summary, we have illustrated in this Letter a mechanism for opening a tunable Kekulé gap in graphene by exciting phonon modes in the lattice. This gap corresponds to a complex-valued order parameter Δ in the effective Dirac field theory whose phase rotates in time with a frequency Ω . The time dependence of Δ is exceptionally simple in that it is completely removable by a gauge transformation which has no effect on bath degrees of freedom and leaves the fermion currents unaffected. The electric response of the system is therefore equivalent to that of one with a static gap. Furthermore, the system is found to support chiral quantized currents that are localized in regions where $|\Delta|$ varies in space. In particular, the system supports edge currents whose chirality depends on the shape of the sample.

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- [1] D. Basov, R. Averitt, D. van der Marel, M. Dressel, and K. Haule, *Rev. Mod. Phys.* **83**, 471 (2011).
 - [2] M. Schultze et al., *Nature* **493**, 75 (2013).
 - [3] M. Durach, A. Rusina, M. Kling, and M. Stockman, *Phys. Rev. Lett.* **107**, 086602 (2011).
 - [4] C.-Y. Hou, C. Chamon, and C. Mudry, *Phys. Rev. Lett.* **98**, 186809 (2007).
 - [5] L. Woods and G. Mahan, *Phys. Rev. B* **61**, 10651 (2000).
 - [6] H. Suzuura and T. Ando, *J. Phys. Soc. Jpn.* **77**, 044703 (2008).
 - [7] C. Chamon, *Phys. Rev. B* **62**, 2806 (2000).
 - [8] R. Jackiw and S.-Y. Pi, *Phys. Rev. Lett.* **98**, 266402 (2007).
 - [9] C. Chamon, R. Jackiw, Y. Nishida, S.-Y. Pi, and L. Santos, *Phys. Rev. B* **81**, 224515 (2010).
 - [10] L. Fu and C. Kane, *Phys. Rev. Lett.* **100**, 096407 (2008).
 - [11] C. Chamon, C.-Y. Hou, R. Jackiw, C. Mudry, S.-Y. Pi, and G. Semenoff, *Phys. Rev. B* **77**, 235431 (2008).
 - [12] G. W. Semenoff, V. Semenoff, and F. Zhou, *Phys. Rev. Lett.* **101**, 087204 (2008).
 - [13] D. J. Thouless, *Phys. Rev. B* **27**, 6083 (1983).
 - [14] B. Seradjeh, H. Weber, and M. Franz, *Phys. Rev. Lett.* **101**, 246404 (2008).
 - [15] R. Jackiw and S.-Y. Pi, *Phys. Rev. B* **78**, 132104 (2008).
 - [16] This is not a restrictive assumption. The inclusion of spin just doubles the size of all matrices and vectors used here.
 - [17] In the limit $|\mu| \rightarrow \infty$, propagation into one sublattice costs infinite energy, while propagation into the other is blocked by the Pauli principle.
 - [18] When spin is included, (18) acquires a factor of 2, so that a charge e is pumped per cycle.