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Persistent Current States in Bilayer Graphene

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We argue that at finite carrier density and large displacement fields, bilayer graphene is prone to $\ell = 0$ and $\ell = 1$ Pomeranchuk Fermi surface instabilities. The broken symmetries are driven by non-local exchange interactions which favor momentum space condensation. We find that electron-electron interactions lead first to spontaneous valley polarization, which breaks time-reversal invariance and is associated with spontaneous orbital magnetism, and then under some circumstances to a nematic phase with reduced rotational symmetry. When present, nematic order is signaled by reduced symmetry in the dependence of optical absorption on light polarization.

I. INTRODUCTION

The search for a microscopic theory of superconductivity, ultimately brought to a conclusion by the successful work of Bardeen, Cooper, and Schrieffer¹, led a number of early condensed matter theory researchers²⁻⁴ to speculate on the possibility that interactions could under some circumstances lead to momentum-space order responsible for equilibrium currents. Since orbital magnetism always accompanies spontaneous spin-polarization because of spin-orbit coupling, we now know that $\ell = 0$ Pomeranchuk⁵ instabilities can lead to spontaneous circulating currents. Similarly $\ell = 1$ Fermi surface instabilities can potentially lead to spontaneous longitudinal currents.

An $\ell = 1$ Fermi-surface distortion transfers occupation between quasiparticle states with opposite current components along the continuously variable direction in which overall current flows, as illustrated schematically in Fig. 1. For a translationally-invariant electronic system it is known from both microscopic-quantum-mechanical^{6,7} and Fermi-liquid-theory⁸ points of view that such a distortion corresponds simply to a Galilean boost which raises the center-of-mass kinetic energy and does not change the interaction energy. An $\ell = 1$ charge-channel Pomeranchuk instability is therefore an impossibility in an electron fluid. In this article we show how the simple, but highly unusual electronic structure of graphene bilayers with low carrier densities and large displacement fields can lead to both $\ell = 0$ and $\ell = 1$ Pomeranchuk instabilities, although not to spontaneous longitudinal currents.

In a crystal the Pomeranchuk instability notion refers to Fermi-surface distortions which reduce lattice symmetries. Here $\ell = 1$ charge-channel distortions have also been viewed⁹ as extremely unlikely, although instabilities in p -wave spin channels preserving time reversal symmetry have been proposed under certain conditions.¹⁰ Instabilities with $\ell = 2$, which lead to electron nematic states, do on the other hand appear to occur^{11,12} in a variety of different systems and spin-channel $\ell = 0$ instabilities, which lead to ferromagnetism, are of course common. In this article we point out that a Bernal-stacked graphene bilayer (BLG) in which a gap has been

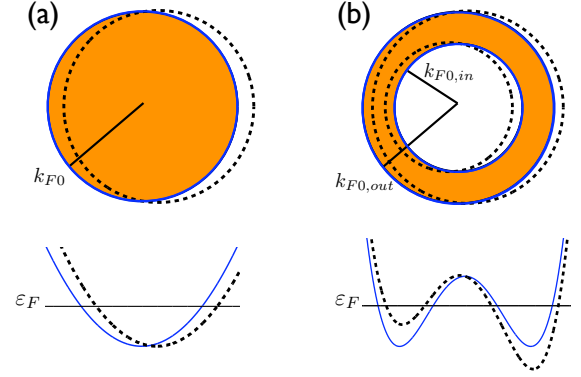


FIG. 1: (color online) $\ell = 1$ Pomeranchuk instability of a conventional Fermi liquid and of density-unbalanced BLG. The lower panels illustrate the quasiparticle band dispersions that correspond to the Fermi surfaces illustrated in the top panels. a) The circular orange Fermi circle with Fermi radius k_{F0} denotes the occupied states of a two-dimensional Fermi sea. A Pomeranchuk instability occurs when energy is reduced by a momentum-space angle-dependent change in Fermi radius that is proportional to $\cos(\ell\theta)$. For $\ell = 1$ the distorted Fermi surface, indicated here by the dashed black circle, is simply shifted in momentum space. b) The disk-shaped orange region with inner and outer Fermi radii $k_{F0,in}$ and $k_{F0,out}$ denotes occupied conduction band states in unbalanced BLG. Shifts in the inner and outer Fermi radii that are proportional to $\cos(\theta)$ and have opposite signs for the inner and outer Fermi lines, thicken and thin the disk in opposite directions as indicated by the black dashed lines. The distorted phase has lower exchange energy because the occupied states are more concentrated in momentum space.

opened by a transverse electric field is ideally suited to host both $\ell = 0$ and $\ell = 1$ Pomeranchuk instabilities because its electron and hole Fermi seas are disks that are spread widely over momentum space, as illustrated in Fig. 1(b).¹³ Both $\ell = 0$ and $\ell = 1$ Fermi surface distortions can lower energy by compactifying the Fermi sea, thereby realizing the momentum-space condensation envisaged by London⁴ more than seventy years ago.

II. FERMI-SURFACE INSTABILITIES IN BILAYER GRAPHENE

Recent progress¹⁴ has made it possible to prepare and study the electronic properties of two-dimensional (2D) electron systems based on single- and few-layer graphene. This advance has provided researchers with a new family of materials whose electronic structure is at the same time remarkably simple and remarkably variable. Single-layer graphene is described by a massless-Dirac-fermion model with conduction and valence bands that touch at two different points in momentum space and disperse linearly over a wide energy region. The parabolic band dispersion near the Fermi level of a neutral Bernal-stacked bilayer graphene allows electron-interaction-driven instabilities, which have been studied by using mean-field^{15,16} or renormalization-group-based approaches.¹⁷ Clear evidence for strong many-body effects has already been obtained in several recent experiments.^{18–29} In a neutral Bernal-stacked bilayer with a very strong electric field directed perpendicular to the layers³⁰, the Fermi level lies within the conduction band of one layer and the valence band of the other. Inter-layer hybridization with strength γ_1 opens up an avoided crossing gap centered at a finite 2D wavevector magnitude between states localized in opposite layers, yielding an unusual semiconductor with an electrically tunable gap. Our interest here is in the electronic properties of degenerate electrons in the conduction band (or holes in the valence band) of this layer-unbalanced configuration of BLG.

When angular variation of the avoided crossing gap due to trigonal warping effects is neglected,³¹ the conduction band minimum occurs along a circle in momentum space and the non-interacting electron Fermi surface is an annulus as indicated in Fig. 1(b). Because the band density-of-states diverges as energy approaches the conduction band minimum it is clear, as noted previously by others,^{32,33} that interactions may play a central role in determining electronic properties and that instabilities that break symmetries are likely. The possibilities include ferromagnetism³² and density-wave³³ states. In this paper we argue that momentum space condensation of the type imagined by London, Heisenberg and others, which has not previously been observed, is also a possibility.

We illustrate our main point by considering a toy model which ignores trigonal warping and spin and valley degrees-of-freedom, and by using mean-field theory to estimate its Fermi-liquid parameters. The Hartree-Fock energy functional³⁴ is

$$E_{\text{HF}}[\{n_{\mathbf{k}}\}] = \sum_{\mathbf{k}} [\varepsilon_{\text{b}}(k) - \mu] n_{\mathbf{k}} - \frac{1}{2A} \sum_{\mathbf{k}, \mathbf{k}'} n_{\mathbf{k}} V_{\mathbf{k}-\mathbf{k}'} n_{\mathbf{k}'}, \quad (1)$$

where $\varepsilon_{\text{b}}(k)$ are bare-band energies, which are isotropic and thus depend only on $k = |\mathbf{k}|$, and A is the 2D electron system area. In our toy model we assume that the interaction $V_{\mathbf{k}-\mathbf{k}'}$ depends only on $|\mathbf{k} - \mathbf{k}'|$. Expanding

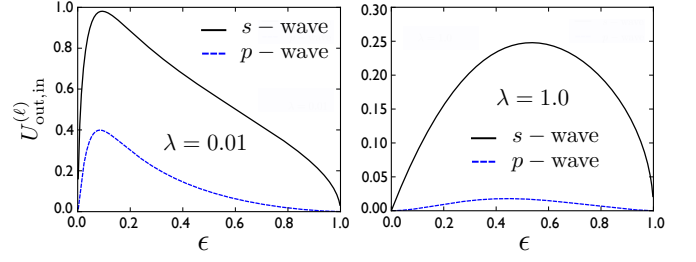


FIG. 2: (color online) The dimensionless “outer-inner” pseudopotential $U_{\text{out},\text{in}}^{(\ell)}$ vs. $\epsilon = b/(2\bar{k})$ for $\ell = 0$ (*s*-wave) and $\ell = 1$ (*p*-wave). This plot is for $\delta = 2/(a_{\text{eff}}\bar{k}) = 1$ where a_{eff} and \bar{k} are defined in the main text. The parameter b measures the thickness of the ring and $b \rightarrow 0$ in the limit in which the Fermi energy ε_{F} equals the band-edge energy ε_{min} . The left panel is for a very small screening parameter, $\lambda = 0.01$, while the right panel is for fully screened Thomas-Fermi interactions, $\lambda = 1.0$.

this energy functional in powers of the deviation $\delta n_{\mathbf{k}}$ from the occupation numbers $n_{\mathbf{k}}^{(0)}$ corresponding to the undistorted Fermi surface yields an energy expression of the Fermi-liquid-theory form:

$$E_{\text{HF}}[\{n_{\mathbf{k}}\}] = E_0 + \sum_{\mathbf{k}} \varepsilon_{\text{b}}^{\text{HF}}(k) \delta n_{\mathbf{k}} - \frac{1}{2A} \sum_{\mathbf{k}, \mathbf{k}'} \delta n_{\mathbf{k}} V_{\mathbf{k}-\mathbf{k}'} \delta n_{\mathbf{k}'}, \quad (2)$$

where the Hartree-Fock band energy $\varepsilon_{\text{b}}^{\text{HF}}(k)$ is defined by

$$\varepsilon_{\text{b}}^{\text{HF}}(k) = \varepsilon_{\text{b}}(k) - \int \frac{d^2 \mathbf{k}'}{(2\pi)^2} n_{\mathbf{k}'}^{(0)} V_{\mathbf{k}-\mathbf{k}'}. \quad (3)$$

Because the total energy is prone to cancellations between quasiparticle-velocity renormalizations and quasiparticle-interaction effects, we must treat the two contributions on an equal footing. Below we consider the quasiparticle-velocity renormalization first and use this analysis to define our notation.

We begin by calculating the Hartree-Fock velocities, *i.e.* the momentum-space radial derivatives of the quasiparticle energy at the outer ($n = \text{out}$) and inner ($n = \text{in}$) Fermi circles. The velocities are most conveniently evaluated by replacing the derivative with respect to \mathbf{k} in $V_{\mathbf{k}-\mathbf{k}'}$ by a derivative with respect to \mathbf{k}' . When the interaction correction to the velocity is integrated by parts, this derivative then acts on $n_{\mathbf{k}'}^{(0)}$ picking out states at the Fermi energy. We find that

$$v_n^{\text{HF}}(k) = \text{sgn}(n) |v_n(k)| + \frac{1}{2\pi\hbar} \int_0^{2\pi} \frac{d\varphi}{2\pi} \cos(\varphi) \times \{ [k' V_{\mathbf{k}-\mathbf{k}'}]_{k'=k_{\text{F0,out}}} - [k' V_{\mathbf{k}-\mathbf{k}'}]_{k'=k_{\text{F0,in}}} \}, \quad (4)$$

where ϕ is the difference between the angular coordinates of \mathbf{k} and \mathbf{k}' , and we have noted that $V_{\mathbf{k}-\mathbf{k}'}$ depends only on ϕ once k and k' are fixed³⁵. This leads to

$$\frac{v_{\text{out}}^*}{v_{\text{out}}} = 1 + \alpha_{\text{out}} \left(U_{\text{out,out}}^{(1)} - \sqrt{\frac{k_{\text{F0,in}}}{k_{\text{F0,out}}}} U_{\text{out,in}}^{(1)} \right), \quad (5)$$

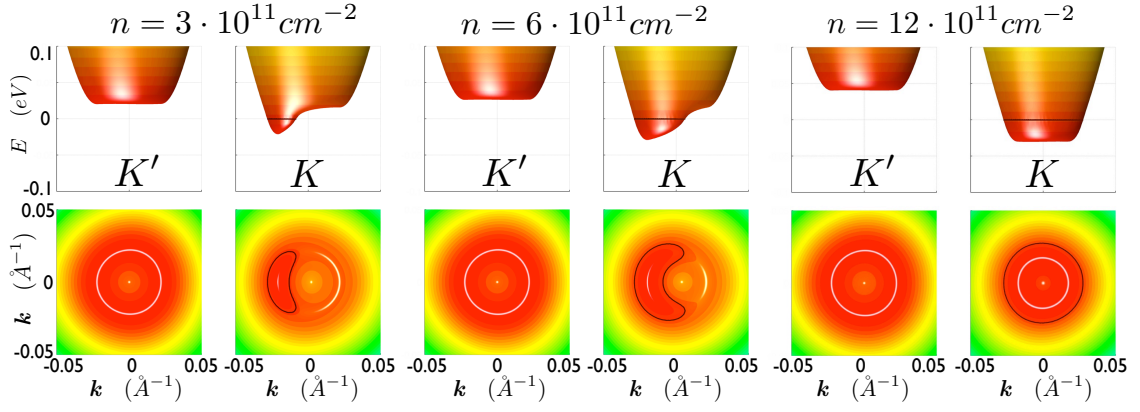


FIG. 3: (color online) Valley polarization and deformation of quasiparticle bands due to $\ell = 0$ and $\ell = 1$ Pomeranchuk instabilities in unbalanced BLG. These results were obtained with an external potential difference corresponding to a perpendicular electric field of 1 V/nm between layers. Valley polarized quasiparticle bands are plotted for three different total carrier densities. The upper and lower panels illustrate conduction band energy dispersions and constant energy contours respectively. The left and right columns are for momenta near the K' and K valleys. The $\ell = 1$ instability is signaled by broken rotational symmetry within an occupied valley. The solid black line marks the position of the Fermi level, chosen as the zero of energy: $\varepsilon_F = 0$. For this calculation we used a k -point sampling density near the Dirac cones equivalent to a density of 4608×4608 points in the whole Brillouin zone.

where $v_{\text{out}}^* \equiv v_{\text{out}}^{\text{HF}}(k_{\text{F0,out}})$, $\alpha_{\text{out}} \equiv e^2/(\hbar v_{\text{out}})$, $U_{n,n'}^{(m)} \equiv V_m(k_{\text{F0},n}, k_{\text{F0},n'}) (k_{\text{F0},n} k_{\text{F0},n'})^{1/2} / (2\pi e^2)$ is a dimensionless interaction parameter that is symmetric in the n, n' inner/outer indices, and the V_m 's are Fourier components of the interaction's ϕ -dependence. The corresponding expression for v_{in}^* can be obtained by interchanging the in and out labels.

To look for Pomeranchuk instabilities we parameterize the inner ($n = \text{in}$) and outer ($n = \text{out}$) Fermi surfaces in terms of dimensionless distortion functions:

$$k_{\text{F},n} = k_{\text{F0},n} [1 + a_n(\theta)] \equiv k_{\text{F0},n} + \delta k_{\text{F},n}(\theta), \quad (6)$$

and expand these in terms of their angular momentum components $a_n(\theta) = \sum_{\ell=-\infty}^{+\infty} a_{n\ell} e^{i\ell\theta}$. The distorted state has a $\delta n_{\mathbf{k}}$ which is non-zero only in the vicinity of the inner and outer Fermi lines. It is easy to verify that the first-order correction to the energy vanishes. To obtain the second-order correction we linearize the Hartree-Fock energies around $k_{\text{F0,in}}$ and $k_{\text{F0,out}}$:

$$\varepsilon_{\text{b}}^{\text{HF}}(k) \simeq \begin{cases} -\hbar v_{\text{in}}^*(k - k_{\text{F0,in}}) \\ \hbar v_{\text{out}}^*(k - k_{\text{F0,out}}) \end{cases}$$

and add the quasiparticle interaction contribution to obtain the energy change for small distortions:

$$\begin{aligned} \frac{E^{(2)}}{A} &= \frac{\hbar}{4\pi} \sum_{n,n'} \sum_{\ell=-\infty}^{+\infty} \left\{ v_n^* \delta_{n,n'} \right. \\ &\quad \left. - \frac{e^2}{\hbar} \text{sgn}(nn') U_{n,n'}^{(\ell)} \right\} k_{\text{F0},n}^{3/2} k_{\text{F0},n'}^{3/2} a_{n\ell} a_{n'\ell}^* . \end{aligned} \quad (7)$$

This is our principal result. Note that for a Galilean-invariant system which has only an outer Fermi radius

the two interaction contributions to the $\ell = 1$ distortion energy cancel, recovering the *no go* theorem^{6–8} mentioned previously. An $\ell = 1$ Pomeranchuk instability occurs when the determinant of the matrix

$$\mathbf{F} = \begin{pmatrix} v_{\text{out}}^* - (e^2/\hbar) U_{\text{out,out}}^{(1)} & (e^2/\hbar) U_{\text{out,in}}^{(1)} \\ (e^2/\hbar) U_{\text{out,in}}^{(1)} & v_{\text{in}}^* - (e^2/\hbar) U_{\text{in,in}}^{(1)} \end{pmatrix} \quad (8)$$

is zero. Using Eq. (5) (and the corresponding equation for v_{in}^*) we finally find the following criterion:

$$1 - \left(\alpha_{\text{out}} \sqrt{\frac{k_{\text{F0,in}}}{k_{\text{F0,out}}}} + \alpha_{\text{in}} \sqrt{\frac{k_{\text{F0,out}}}{k_{\text{F0,in}}}} \right) U_{\text{out,in}}^{(1)} = 0 . \quad (9)$$

Notice that this instability criterion depends only on the “outer-inner” interaction.

We estimate the disk Fermi-liquid interaction parameters using

$$V_{\mathbf{k}-\mathbf{k}'} = \frac{2\pi e^2}{|\mathbf{k} - \mathbf{k}'| + \lambda q_{\text{TF}}} , \quad (10)$$

where q_{TF} is the Thomas-Fermi screening wave vector, and $\lambda \in [0, 1]$ is a dimensionless control parameter that allows us to interpolate between bare-Coulomb ($\lambda = 0$) and Thomas-Fermi ($\lambda = 1$) limits. The Thomas-Fermi screening wave vector is proportional to the density-of-states at the Fermi energy, $N(0) = 2m_{\text{eff}}\bar{k}/(\pi\hbar^2 b)$ where $b = k_{\text{F0,out}} - k_{\text{F0,in}}$, $\bar{k} = (k_{\text{F0,out}} + k_{\text{F0,in}})/2$, and m_{eff} parameterize the band-energy dispersion at its minimum. It follows that $q_{\text{TF}} = 2\pi e^2 N(0) = 4\bar{k}/(a_{\text{eff}} b)$ where $a_{\text{eff}} = \hbar^2/(m_{\text{eff}} e^2)$ is an effective Bohr radius. The inner-outer interaction parameters $U_{\text{out,in}}^{(\ell)}$ obtained using this approximation depend on two dimensionless quantities, $\epsilon = b/(2\bar{k})$ and $\delta = 2/(a_{\text{eff}}\bar{k})$, and are plotted for $\ell = 0, 1$

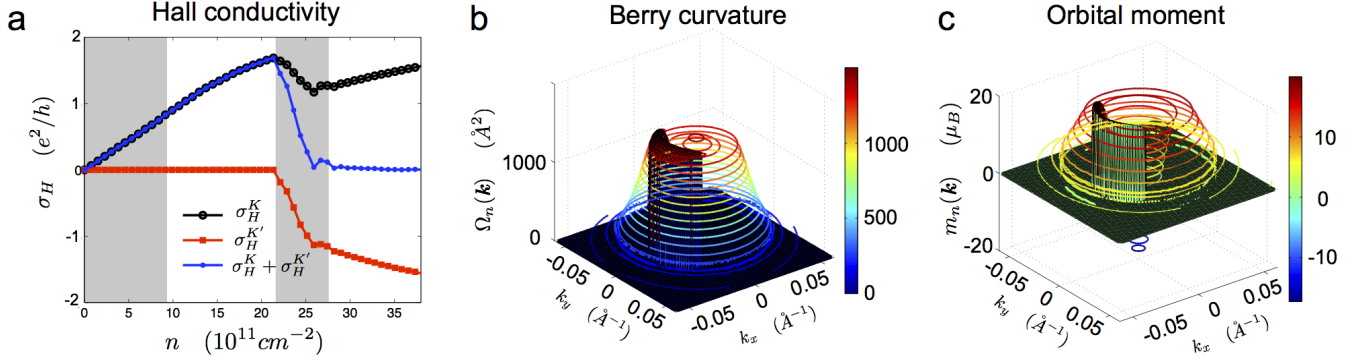


FIG. 4: (color online) **a.** Spontaneous Hall effect *vs.* carrier density. The Hall effect [Eq. (11)] reflects spontaneous valley polarization. We find that the Hall conductivity increases monotonically with carrier density until the minority (K') valley with opposite Hall conductivity starts to be occupied. The shaded regions indicate intervals of carrier density over which nematic phases due to $\ell = 1$ and higher order channel Pomeranchuk instabilities appear. The lower density instability occurs within the majority valley and the higher density instability within the minority valley. **b-c.** Berry curvature and Bloch state orbital magnetic moment in units of Bohr magnetons near the K valley in the limit of low carrier density.

in Fig. 2. The rapid variation at small ϵ is due to strong Thomas-Fermi screening in this limit and are certainly an artifact of using such a simple screening approximation to describe the $\epsilon \rightarrow 0$ limit in which Fermi-level density-of-states diverges.

We can make two conclusions based on these estimates: i) Interactions in the $\ell = 1$ channel are strong enough to produce an $\ell = 1$ Pomeranchuk instability and ii) Interactions in the $\ell = 0$ channel are almost certainly stronger than those in the $\ell = 1$ channel. A single valley, $\ell = 0$ charge channel instability, would imply phase separation into low and high density regions, is forbidden by the long-range Coulomb interaction. However, the presence of valley and spin degrees of freedom allows $\ell = 0$ instabilities that lead to spin³² or valley polarized states. $\ell = 1$ Pomeranchuk instabilities are likely only in states which are already spin or valley polarized. Although momentum space is occupied asymmetrically when the $\ell = 1$ instability occurs, we find that there is no longitudinal current because of a cancellation between inner and outer Fermi surface contributions.

III. TIGHT-BINDING CALCULATIONS AND HALL CONDUCTIVITY SIGNATURES OF THE INSTABILITIES

We have corroborated the conclusions made on the basis of our toy-model calculations by performing self-consistent π -band lattice Hartree-Fock calculations similar to those described in Refs. 16,36. For these calculations we added interactions to a band model with nearest-neighbor intra-layer hopping $\gamma_0 = 3.12$ eV and inter-layer hopping $\gamma_1 = 0.377$ eV, ignoring other hopping parameters for the sake of simplicity.³⁷ The resulting bands near the K and K' points are shown in Fig. 3. We have suppressed spin-polarization instabilities in these calculations so that $\ell = 0$ instabilities are manifested by spon-

taneous valley and not spin polarization. Because external electric fields in unbalanced BLG lead to large Berry curvatures $\Omega_{n,\mathbf{k}}$ of opposite sign in the vicinity of K and K' valley points³⁸ the anomalous Hall conductivity can be used as an observable for spontaneous valley polarization. The Hall conductivity (per spin) is calculated by integrating $\Omega_{n,\mathbf{k}}$ over occupied quasiparticle states near the Dirac points,

$$\sigma_H = \frac{e^2}{h} \int \frac{d^2\mathbf{k}}{(2\pi)^2} \sum_n f_{n,\mathbf{k}} \Omega_{n,\mathbf{k}}, \quad (11)$$

where $f_{n,\mathbf{k}}$ is the Fermi-Dirac distribution and a sum is carried over the band index n .

Fig. 4 plots the Hall conductivities evaluated for this model and demonstrate that spontaneous valley polarization occurs for carrier densities smaller than $\sim 25 \times 10^{11} \text{ cm}^{-2}$, and that nematic order occurs for carrier densities smaller than $\sim 9 \times 10^{11} \text{ cm}^{-2}$ and again near the onset of minority valley occupation. The associated Berry curvatures and the orbital moments of the gapped chiral band edges are represented together with the Hall conductivity to represent the \mathbf{k} space resolved contribution.

IV. OPTICAL CONDUCTIVITY SIGNATURES

The presence of either valley polarization or nematic order should be observable via interband optical conductivity measurements. Circular dichroism measurements can detect valley polarization because the associated optical transition matrix elements³⁸ are valley dependent. Valley-dependent population of states near the band edges then leads to valley-dependent Pauli blocking and contrast between the absorption of left and right circularly polarized light.

Broken rotational symmetry in a nematic phase also has observable signatures in optical conductivity mea-

measurements comparing absorption of light that is linearly polarized along different directions. For example, the real part of the conductivity per valley and spin for light polarized along the x axis is given by

$$\sigma_{xx}(\omega) = \frac{\pi e^2}{\hbar \omega A} \sum_{\mathbf{k}} |W_{\mathbf{k}\pm}|^2 (f_{+, \mathbf{k}} - f_{-, \mathbf{k}}) \delta(\hbar \omega - \Delta E_{\mathbf{k}}) \quad (12)$$

where $W_{\mathbf{k}\pm} = \langle +\mathbf{k} | j_x | -\mathbf{k} \rangle$, $|+\mathbf{k}\rangle$ and $|-\mathbf{k}\rangle$ are conduction and valence band quasiparticle states, $\Delta E_{\mathbf{k}}$ is the band splitting and $j_x = \partial H_{\mathbf{k}} / \partial k_x$ is the current operator in the x -direction. At the band extrema, the quasiparticle states are symmetric and antisymmetric combinations of the conduction bands of the low-potential graphene sheet and the valence bands of the high-potential graphene sheet. It follows that for states near the band edges $W_{\mathbf{k}\pm} \simeq \cos \theta_{\mathbf{k}}$. To illustrate how the momentum-direction dependence of the optical conductivity is sensitive to the spontaneous anisotropy of the nematic ground state, we evaluate the real part of the conductivity for incident light polarized along direction φ and a nematic state with conduction bands occupied between orientation angles θ_i and θ_f . We find that because absorption is reduced by Pauli-blocking the conductivity at the absorption edge is proportional to

$$\sigma_{\varphi} \propto \left(\theta_d + \sin \theta_d \cos \left(\frac{\theta_s}{2} + \varphi \right) \right)^2 \quad (13)$$

where $\theta_d = \theta_f - \theta_i$ and $\theta_s = \theta_f + \theta_i$. Note that in the absence of nematicity ($\theta_d = 2\pi$) the optical absorption is independent of φ .

V. DISCUSSION

Because the Fermi surface instabilities discussed in this paper appear only at low carrier densities, comparable to or smaller than typical disorder-induced density-fluctuation scales for bilayer samples on silicon oxide substrates, and because large electric fields are favorable for their occurrence, we anticipate that momentum space condensation is at present a realistic possibility only for dual-gated bilayer graphene samples on h-BN substrates.³⁹ We expect that trigonal warping of the unbalanced bilayer conduction bands will favor momentum space condensation over competing³³ density-wave instabilities. Because the $\ell = 1$ Pomeranchuk instability is likely only within states in which spin or valley polarization, or both, has already occurred, its appearance should be signaled most clearly by observables which detect reduced orientational symmetry, for example polarization direction dependence in interband optical absorption.

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