Valley hydrodynamics in graphene

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Recent experiments have elucidated that novel nonequilibrium states consistent with the hydrodynamic description of electrons are realized in graphene, which hosts the valley degrees of freedom (DOF) at the corners of the Brillouin zone. Here, we formulate a theory of electron hydrodynamics with the valley DOF for noncentrosymmetric graphene and find that the effective theory has a close analogy with micropolar fluids. Our theory reveals nonlinear valley dynamics including a longitudinal valley current and a circular dichroic valley polarization induced by off-resonant light.

Introduction.— The investigation of internal quantum degrees of freedom (DOF) of electrons in solids has played the central role in condensed matter physics. The most-studied example is that of the electron spin, which gives rise to the vast field of spintronics with an eye on their potential for future electronics [1, 2]. The advent of novel 2D materials which support massive Dirac fermions, exemplified by gapped graphene and transition metal dichalcogenides, has triggered research on alternative future electronics [3–9]. In these systems, two inequivalent valleys $K$ and $-K$ reside at the corners of the hexagonal Brillouin zone. Similar to the spin, the valley labeling constitutes a discrete DOF for low energy carriers. From this perspective, the valley DOF has a potential use for information carriers, giving rise to an active research field called valleytronics as a promising concept for the next-generation electronics [10–17]. Especially, the valley polarization, a nonequilibrium charge carrier imbalance between valleys, is the key to create valleytronic devices [18–31]. Therefore, a necessary requirement for valleytronic applications is the ability to generate and control the valley polarization.

Hydrodynamics composed of the evolution of conserved quantities well describes the nonequilibrium behavior of interacting systems close to equilibrium. Recent experiments on ultrapure graphene have provided clear evidence for a hydrodynamic behavior of charge carriers [32–45]. Graphene hosts a high-quality electron system with weak electron-phonon coupling such that electron-electron scatterings can become the dominant scattering process at elevated temperatures. In addition, the electronic structure of graphene inhibits Umklapp processes, which ensures that electron-electron scatterings are momentum conserving. These features lead to novel nonequilibrium states inherent in the hydrodynamic regime, with the momentum taking on the role of a conserved quantity that governs local equilibrium.

The phenomenon of angular momentum conversion between internal DOF of quantum particles and mechanical rotation have attracted great interest in various fields, ranging from nuclear physics to condensed matter physics. Specifically, the past decades have seen a profound growth of interest in the field of hydrodynamics which deals with the microstructures of the fluid elements so-called micropolar fluids [46–49] because of its many applications in liquid crystals [50, 51], ferrofluids [52–54], spintronics [55–58], quark-gluon plasma [59–61], and active matter [62–65]. The micropolar fluids have received special attention due to angular momentum conservation, which gives rise to an emergent equation for the microrotation: the internal angular momentum stems from the valley polarization $P_v$ of fluid elements. The microrotation relaxes towards the vorticity due to the rotational viscosity.

![FIG. 1. Schematics of the microrotation in valley hydrodynamics. The non-uniform velocity $u(r)$ gives rise to the vorticity $\nabla \times u$. On the other hand, the microrotation $\omega$ is the internal angular momentum stems from the valley polarization $P_v$ of fluid elements. The microrotation relaxes towards the vorticity due to the rotational viscosity.](image-url)
presence of the microrotation and the rotational viscosity affect the hydrodynamics. However, the role of the internal DOF in electron hydrodynamics has not been well investigated yet. This naturally motivates us to study electron hydrodynamics with the valley DOF, which can be regarded as an internal angular momentum in noncentrosymmetric systems [66]. Our study gives a new strategy for controlling the valley polarization in gapped graphene and may open up various avenues for research on valley hydrodynamics.

In this Letter, we derive the hydrodynamic equations for 2D honeycomb lattice systems with broken spatial inversion symmetry, which are correct up to the first order in the drift velocity and the microrotation. We uncover that our theory acquires an emergent conservation law for the microrotation, therefore, it has a close analogy to micropolar fluids owing to the valley-microrotation coupling. From a symmetry viewpoint, we find that this interaction can appear only in the systems without inversion symmetry. A key ingredient for valleytronics is a controllable way of population imbalance between the two valleys, thereby producing a valley polarization. Previous works showed that a valley polarization can be conserved in the drift velocity and the microrotation. We also predict nonlinear valley dynamics including a valley polarization induced by off-resonant light [Fig.3] and a circular dichroic longitudinal valley current [Fig.2].

We outline how to derive the hydrodynamic equations for noncentrosymmetric graphene with a staggered sublattice potential. We start from the Boltzmann equation which governs the evolution of the electron distribution function \( f_{\alpha \tau} \) for band \( \alpha \) and valley \( \tau \),

\[
\frac{\partial f_{\alpha \tau}}{\partial t} + \mathbf{v}_{\alpha \tau} \cdot \frac{\partial f_{\alpha \tau}}{\partial \mathbf{r}} + \mathbf{\dot{k}}_{\alpha \tau} \cdot \frac{\partial f_{\alpha \tau}}{\partial \mathbf{k}} = -\frac{f_{\alpha \tau} - f_{\alpha \tau}^N}{\tau_N} - \frac{f_{\alpha \tau} - f_0}{\tau_R} - \frac{f_{\alpha \tau} - f_{\alpha - \tau}}{\tau_{\text{vf}}},
\]

where \( f_0 \) is the Fermi-Dirac (global equilibrium) distribution function. \( \tau_N, \tau_R \) and \( \tau_{\text{vf}} \) are the relaxation times for normal (N), resistive (R) and valley flipping processes. Here, N process conserves the linear momentum, while R process does not. If we construct an electron wave packet near the valley center, the semiclassical equations of motion read [67]

\[
\mathbf{\dot{v}}_{\alpha \tau} = \frac{1}{\hbar} \frac{\partial \epsilon_{\alpha \tau}}{\partial \mathbf{k}} - \mathbf{\dot{k}}_{\alpha \tau} \times \mathbf{\Omega}_{\alpha \tau}, \quad \mathbf{\dot{k}}_{\alpha \tau} = -\frac{e}{\hbar} \mathbf{E},
\]

where electric fields \( \mathbf{E} \) can be time-dependent. \( \epsilon_{\alpha \tau}(\mathbf{k}) \) and \( \mathbf{\Omega}_{\alpha \tau}(\mathbf{k}) \) are the band energy and the Berry curvature of the Bloch electrons respectively. Due to the lack of inversion symmetry, \( \mathbf{\Omega}_{\alpha \tau} \) is allowed to have nonzero values for any \( \mathbf{k} \).

Following the standard approach [68–73], the continuity equations for the carrier density and the linear momentum are obtained as follows:

\[
\frac{\partial n}{\partial t} + \nabla \cdot \mathbf{j} = 0,
\]

\[
\frac{\partial P_i}{\partial t} + \frac{\partial \Pi_{ij}}{\partial x_j} = -enE_i - \frac{P_i}{\tau_R},
\]

where \( n \) and \( P \) are the carrier and the linear momentum densities respectively, \( -en \mathbf{E} \) is the driving force due to external electric fields. In Eqs. (3) and (4), \( \mathbf{j} \) and \( \Pi_{ij} \) are the corresponding fluxes of each density.

A monolayer graphene with a staggered sublattice potential breaking the inversion symmetry is a concrete example for considering valley hydrodynamics. Staggered sublattice potential is generally expected in epitaxial graphene on SiC substrates [74–80]. The effective Hamiltonian describing electron states in the vicinity of the \( K \) and \( -K \) points is given by [3]

\[
H_{\tau} = at(\sigma_x \epsilon_{\tau \sigma} + k_y \sigma_y) + \frac{\Delta}{2} \sigma_z,
\]

where \( a \) and \( t \) are the lattice constant and the nearest-neighbor hopping parameter, \( \mathbf{k} = (k_x, k_y) \) are the two components of the wave vector measured from the valley center, \( \sigma's \) are the Pauli matrices representing a pseudospin from the sublattice DOF, and \( \tau = \pm 1 \) is the valley index labeling the two inequivalent valleys. Note that its band structure \( \epsilon_{\alpha \tau}(\mathbf{k}) \) has no dependence on the valley, while the Berry curvature \( \mathbf{\Omega}_{\alpha \tau}(\mathbf{k}) \) has a valley-contrasting property. Because of large separation of two valleys in the momentum space, intervalley scatterings are strongly suppressed [81–83], implying the potential for regarding the valley polarization as a conserved quantity. Therefore, the effective hydrodynamic theory acquires an emergent continuity equation for the valley polarization:

\[
\frac{\partial P_i}{\partial t} + \nabla \cdot \mathbf{j}_v = -\frac{P_i}{\tau_R} - \frac{2P_v}{\tau_{\text{vf}}},
\]

where \( P_v \) and \( \mathbf{j}_v \) are the valley polarization and the valley current. Here, we have defined the valley polarization \( P_v \equiv n_K - n_{-K} \) as a population imbalance between the two valleys in analogy to the spin polarization. We should note that not only valley flipping processes but also R process contribute to the relaxation of the valley polarization. This indicates that the valley DOF combines a linear momentum and an angular momentum.

In hydrodynamic regime, \( \tau_N \ll \tau_R, \tau_{\text{vf}} \), the system reaches local equilibrium via N electron-electron scatterings, which conserve both the linear momentum and the valley polarization of the electron system. For this reason, we assume that the distribution functions are described as

\[
f_{\alpha \tau}^N = \left[ \exp \left( \frac{\epsilon_{\alpha \tau} - \hbar \mathbf{k} \cdot \mathbf{u} - \tau \hbar \omega_z - \mu}{k_B T} \right) + 1 \right]^{-1},
\]
which is referred to as the local equilibrium distribution function. Here, the drift velocity \( \mathbf{u} \) and the microrotation \( \omega_z \) are corresponding parameters for conserved quantities \( \mathbf{P} \) and \( \mathbf{P}_\nu \). From a symmetry viewpoint, the absence of inversion symmetry allows for the interplay between the valley DOF and an angular momentum; examples include the spin-valley coupling \([5, 84]\) and the valley-vorticity coupling \([85]\). Here, the microrotation is the internal angular momentum of the fluid elements and we referred to \( \tau \omega_z \) as the valley-microrotation coupling.

Since the relevant conduction and valence bands are well described by Eq.\((5)\) for low doping level, we use a quadratic dispersion \( \epsilon_{\alpha \tau} = \alpha [\Delta / 2 + \hbar^2 k^2 / 2m^*] \) with an effective mass \( m^* \equiv \hbar^2 \Delta / 2a^2 t^2 \) in the vicinity of the \( K \) and \(-K\) points in the following analysis. Under this assumption, we obtain the valley polarization and the valley current in terms of hydrodynamic variables:

\[
P_v = \hbar \omega_z \sum_{\alpha, \tau} \int [\mathbf{k}] \left( -\frac{\partial f_0}{\partial \epsilon} \right) \approx \hbar \omega_z D(\mu),
\]

\[
j_v = P_v \mathbf{u} + \frac{e}{\hbar} \mathbf{E} \times \sum_{\alpha, \tau} \int [\mathbf{k}] \Omega_{\alpha \tau} f_0,
\]

with the density of states \( D(\epsilon) \) and \( \int [\mathbf{k}] \equiv \int d\mathbf{k} / (2\pi)^2 \). These results indicate that the microrotation leads to a valley polarization. The second term in Eq. \((9)\) is the well-known valley Hall effect \([3]\), on the other hand, the valley polarization. The second term in Eq. \((9)\) is the well-known valley Hall effect \([3]\). In gapped graphene, the orbital magnetization consists of the orbital moment of carriers plus a correction from the Berry curvature \([86]\):

\[
M_{\text{orb}}^z = \sum_{\alpha, \tau} \int [\mathbf{k}] m_{\alpha \tau}^z f_0 + \frac{e}{\beta} \sum_{\alpha, \tau} \int [\mathbf{k}] \Omega_{\alpha \tau} \log [1 + e^{-(\epsilon_{\alpha \tau} - \hbar \mathbf{k} \cdot \mathbf{u} - \hbar \omega_z \tau \mathbf{r} - \mu)]].
\]

After straightforward calculation, we obtain the orbital magnetization,

\[
M_{\text{orb}}^z = \hbar \omega_z \sum_{\alpha, \tau} \int [\mathbf{k}] \left[ m_{\alpha \tau}^z \left( -\frac{\partial f_0}{\partial \epsilon} \right) + \frac{e}{\hbar} \Omega_{\alpha \tau}^z f_0 \right].
\]

This result also supports that the microrotation has a meaning of an angular momentum. The spatial profile of the orbital magnetization can be detected with magneto-optical Kerr rotation microscopy \([29, 87-89]\). By using this experimental setup, a population difference in the two valleys can be also detected as a signal of the orbital magnetization [see Fig. 2b, as a demonstration].

**Valley hydrodynamic generation.** — The most significant consequences of our theory is that the interplay between the valley-microrotation coupling and the viscous effects gives rise to an unprecedented longitudinal nonlinear valley current in finite size systems, which has not been addressed so far. We consider the Poiseuille flow in gapped graphene with finite width \( w \) in the \( y \)-direction, which most clearly characterizes the hydrodynamic transport. When we apply DC electric fields in the \( x \)-direction and take no-slip boundary conditions \( u_x(\pm w/2) = 0 \), the electron fluids form the Poiseuille flow with the velocity profile given by

\[
u_x(y) = -\frac{en\tau_R}{\rho} \left[ 1 - \cosh(y/\ell) \right] E.
\]

The microrotation is also calculated as

\[
\omega_z(y) = -\frac{\tau_{\text{eff}}}{\tau_r} \frac{1}{2} \frac{\partial u_x}{\partial y} = -\frac{en\tau_R}{2\rho\ell} \tau_{\text{eff}} \sinh(y/\ell) E.
\]
FIG. 2. (a) Schematics of the valley hydrodynamic generation. In hydrodynamic regime, the valley polarization $P_v$ is induced by the microrotation via the valley-microrotation coupling with non-uniform electron velocity $u(x)$, and the longitudinal valley current is generated as $j_v = P_v u$. Plot of (b) the valley polarization and (c) the valley current in the $y$-range $[-w/2, w/2]$ under DC electric fields for several rotational viscosities. We use the parameters: $w = 10 \, \mu m$, $E = 1 \times 10^6 \, \text{V/m}$, $\nu = 0.1 \, \text{m}^2/\text{s}$, $I = 10^{-12} \, \text{m}^2$, $\tau_r = \tau_{ct} = 1 \times 10^{-12} \, \text{s}$, $a = 2.46 \, \text{Å}$, $t = 2.82 \, \text{eV}$, $\Delta = 0.28 \, \text{eV}$, $\mu = 0.15 \, \text{eV}$, $n = 1.4 \times 10^{15} \, \text{m}^{-2}$, $\rho = 2.8 \times 10^{-17} \, \text{kg/m}^2$, and the valley Hall current $j_{vR} = 2.76 \times 10^{21} \, \text{m}^{-1}\text{s}^{-1}$.

where

$$\ell \equiv \sqrt{(\nu + \nu_r) \frac{\tau_r}{\tau_r + \tau_{\text{inter}}}} \, \tau_R,$$

is a characteristic length that determines the scale of viscous effects. Here, $\tau_r^{-1} \equiv 4\nu_r / I$ and $\tau_{\text{eff}} \equiv (1/\tau_{\text{inter}} + 1/\tau_r)^{-1}$ are the rotational and effective relaxation times. From Eq.(9), we obtain the longitudinal valley current profile as [Fig.2c]

$$j_{v,||}^{\text{DC}}(y) = \left(\frac{e\nu_r}{\rho} \frac{E}{\ell} \right)^2 \frac{\hbar D(\mu)}{2\ell} \frac{\tau_r}{\tau_{\text{eff}}} \times \frac{\sinh(y/\ell)}{\cosh(y/\ell)^2} \left[1 - \frac{\cosh(y/\ell)}{\cosh(\ell/2)} \right].$$

We should note that the rotational viscosity $\nu_r$ is necessary for realizing a longitudinal nonlinear valley current under DC electric fields. Similar to DC electric fields, an AC electric field along the $x$-direction $E_x(t) = R[\dot{E} e^{-\alpha t}]$ also induces the Poiseuille flow and leads to the same solutions:

$$\tilde{u}_x(y, \Omega) = \frac{u_x(y)}{1 - i\Omega \tau_R}, \quad \tilde{\omega}_z(y, \Omega) = \frac{\omega_z(y)}{(1 - i\Omega \tau_{\text{eff}})(1 - i\Omega \tau_R)},$$

except for the replacement of $\ell$ by

$$\tilde{\ell}(\Omega) = \sqrt{(\nu + \nu_r) \frac{\tau_r}{\tau_r + \tau_{\text{inter}} - i\Omega \tau_{\text{inter}}} \frac{1}{1 - i\Omega \tau_R}}.$$

Because of the intrinsic nonlinearity of the longitudinal valley current, $j_{v,||}$ is composed of the valley counterparts of the rectification and the second harmonic generation:

$$j_{v,||}^{\text{AC}}(y, t) = j_{v,||}^0(y) + j_{v,||}^{2\text{D}}(y, t).$$

Circular photovalley generation. The rotational viscosity-induced valley transport discussed above can be interpreted as a phenomenon of angular momentum conversion between the fluid vorticity and the valley DOF. From this viewpoint, we consider a different scenario for generating a valley polarization by circularly polarized light (CPL). CPL with the electric component $E(t) = E_0(\cos \Omega t, \sin \Omega t)$ induces a circular motion of electrons, which in turn generates a DC orbital magnetization:

$$M_{\text{orb}} = -\frac{ne^3}{4m^2\gamma^3} \xi E_0^2.\quad (24)$$

This phenomenon is known as the inverse Faraday effect [90–93] owing to the fact that CPL has a spin angular momentum proportional to $\xi E_0^2$ [94–98]. Here, the different chirality indices $\xi = \pm 1$ correspond to the clockwise/counterclockwise circular polarizations. In our hydrodynamic formulation, the orbital magnetization is described as Eq. (16), therefore, the inverse Faraday effect can be regarded as a direct transfer mechanism of angular momentum from CPL to the microrotation.

We are now ready to discuss the generation of a valley polarization by CPL dubbed circular photovalley generation [Fig.3]. In contrast to the above discussion, we consider bulk systems with normally-incident CPL. We start from the hydrodynamic equations:

$$\frac{D\mathbf{u}}{Dt} = (\nu + \nu_r) \Delta \mathbf{u} + 2\nu_r \nabla \times \mathbf{u} - \frac{en \mathbf{E}}{\rho} \frac{\mathbf{u}}{\tau_R},$$

$$\frac{D\mathbf{\omega}_z}{Dt} = \frac{1}{\tau_r} \left[(\nabla \times \mathbf{u})_z - \omega_z\right] - \frac{\omega_z}{\tau_{\text{inter}}} + g \xi E_0^2,$$

where an external angular momentum pumping term stems from the inverse Faraday effect is introduced. By solving Eqs. (25) and (26), the DC component of the microrotation in the second order in electric fields is obtained as

$$\omega_z^0 = \tau_{\text{eff}} g \xi E_0^2.$$

(27)
FIG. 3. Schematics of circular photovalley generation. The microrotation is directly induced by irradiating CPL via the inverse Faraday effect. As a result, a DC valley polarization is generated.

giving rise to a nonlinear DC valley polarization:

\[ P_\nu^0 = \hbar D(\mu) \tau_{\text{eff}} g \xi E_0^2. \]  

(28)

Here, the coefficient of \( g \) is estimated as

\[ \tau_{\text{eff}} g = \text{sgn}(\mu) \frac{\Delta^2}{4}\hbar (\Omega \mu)^3 \frac{a^2 t^2}{\Delta^2/4}, \]  

(29)

in consistent with Eqs.(16) and (24). In stark contrast to the previous works [18–23], we do not rely on inter-band transition processes, therefore, on-resonant light is not required. This suggests that our hydrodynamic approach broadens the frequency range of CPL and also allows ultrafast manipulation of the valley polarization. This can be achieved by combining the valley DOF with the concept of micropolar fluids. Furthermore, the sign of the valley polarization can be tuned by the chirality \( \xi \) of CPL, and hence circular dichroism appears in the valley polarization.

Discussion.—We propose an experimental setup how to determine \( \nu_r \) below. In order to estimate the rotational viscosity experimentally, valley injection provides a reasonable measure. When we inject valley current from the proximity valley Hall materials into valley hydrodynamic materials, the induced valley polarization leads to the non-uniform microrotation profile \( \omega(\mathbf{r}) \) due to the valley-microrotation coupling. Then, the fluid velocity \( \mathbf{u} \) is generated by the term \( \nu_r \nabla \times \omega \) in Eq.(12). Therefore, we can estimate the rotational viscosity from the observed velocity profile.

Conclusion.—In summary, we have developed a basic framework of valley hydrodynamics in noncentrosymmetric graphene with a staggered sublattice potential, which is composed of the Euler equation Eq.(10) and the balance equation for the microrotation Eq.(11). In addition, we have investigated the interplay between the valley DOF and the microrotation, and elucidated that the valley polarization can be controlled by the microrotation. Our hydrodynamic theory also reveals nonlinear valley dynamics. For example, the rotational viscosity \( \nu_r \) provides a longitudinal nonlinear valley current, which gives rise to the valley counterparts of the rectification and the second harmonic generation. As discussed, \( \nu_r \) can be measured by valley-induced hydrodynamic flow generation. Furthermore, the concept of micropolar fluids sheds light on a rich physics of angular momentum conversion, exemplified by a circular dichroic valley polarization induced by off-resonant light. These results are summarized in Table I.

The conventional strategy for designing electronic devices in spintronics or valleytronics has been creating confined nanostructure in order to achieve functionality. On the other hand, in hydrodynamic regime, the flow of electrons can become spatially non-uniform due to the viscosities even when the material structure is homogeneous. This suggests a new design guideline for innovative device functionality without nanostructure. Therefore, we believe that the present results provide a building block for future electronics and will pave the way to valleytronic applications of electron hydrodynamics.

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