Charge and Spin Hall Conductivity in Metallic Graphene

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Graphene has an unusual low-energy band structure with four chiral bands and half-quantized and quantized Hall effects that have recently attracted theoretical and experimental attention. We study the Fermi energy and disorder dependence of its spin Hall conductivity $\sigma_{xy}^{\mathrm{SH}}$. In the metallic regime we find that vertex corrections enhance the intrinsic spin Hall conductivity and that skew scattering can lead to $\sigma_{xy}^{\mathrm{SH}}$ values that exceed the quantized ones expected when the chemical potential is inside the spin-orbit induced energy gap. We predict that large spin Hall conductivities will be observable in graphene even when the spin-orbit gap does not survive disorder.

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Introduction.—The low-energy band structure of graphene consists of four chiral bands that realize $(2+1)$-dimensional relativistic field theory models with parity anomalies. The anomalies imply unusual spectra in an external magnetic field and quantized and half-quantized Hall effects [1,2]. Theoretical interest [3] in these unusual electronic systems has increased [4] recently because of experimental progress [5], including measurements of the anticipated *half-quantized* quantum Hall effect. One particularly interesting observation, due to Kane and Mele [6,7], is that because of a gap produced by spin-orbit interactions, the spin Hall conductivity σ_{SH} of undoped graphene is quantized in the absence of a magnetic field. This suggestion is related to recent work on the anomalous Hall effect in ferromagnetic metals [8] and on its paramagnetic cousin, the spin Hall effect [9], in which it was suggested that these transport coefficients can be dominated by an intrinsic momentum-space Berry phase contribution that reduces to quantized values when the Fermi level is in a gap. Here we examine how the quantized spin Hall effect is altered when the Fermi energy in the graphene plane is gated into the metallic regime. We find that the intrinsic spin Hall effect is no longer quantized, that it is enhanced by disorder vertex corrections, and that in the metallic regime skew scattering can potentially lead to parametrically larger spin Hall conductivities. Because the Bloch state disorder broadening in current samples is (according to our estimates) much larger than the clean system spin-orbit gap, these results are necessary for the interpretation of experiment. Spin Hall effects should be observable even when the spin-orbit gap does not survive disorder.

Disordered graphene model.—When spin-orbit interactions are included [6], the low-energy physics of a clean undoped graphene crystal is described by an eight-band envelope function Hamiltonian

$$\hat{H}_0 = v(k_x\tau_z\sigma_x + k_y\sigma_y) + \Delta\sigma_z\tau_z s_z, \tag{1}$$

where $s_z = \pm$ is the up (down) electron spin component perpendicular to the graphene plane, $\tau_z = \pm$ is a valley label that specifies one of the two inequivalent (K and K') points in the crystal Brillouin zone near which low-energy states occur, and the σ_i are Pauli matrices representing a pseudospin degree of freedom corresponding to the two sites per primitive cell of a hexagonal lattice. The parameter Δ is the strength of the spin-orbit coupling, and we take $\hbar = 1$. For $\Delta = 0$ this Hamiltonian defines four spin-degenerate gapless bands in which the pseudospin orientation lies in the $\hat{x}$-$\hat{y}$ plane and winds around the $\hat{z}$ axis, either clockwise or counterclockwise, with a 2π planar wave vector rotation. The operators σ_i, s_z, and τ_z commute with each other. Random defects can in general produce transitions between bands and between spins. Here we assume sufficiently spatially smooth spin-independent disorder so that s_z and τ_z are good quantum numbers, allowing us to consider the cases $\tau_z, s_z = \pm 1$ independently. For this disorder model we evaluate the Kubo-formula Hall conductivity in the self-consistent Born approximation (SCBA) for chemical potentials inside and outside the spin-orbit gap, including both nontrivial pseudospin dependent disorder self-energies and ladder diagram vertex corrections. When the chemical potential lies in the gap, an elementary calculation shows that in the absence of disorder the single-band bulk partial Hall conductivity is given exactly by the half-quantized Berry phase contribution [8,9], $-(s_z e^2/2h)$. Disorder corrections to the intrinsic Hall effect are small near the gap edge but yield substantial enhancement in more strongly gated systems.

2D-Dirac-band Hall effect.—The 2D Dirac Hamiltonian in the spin-$\uparrow$ K valley is

$$\hat{H} = v(k_x\sigma_x + k_y\sigma_y) + \Delta\sigma_z. \tag{2}$$

Spin-orbit coupling opens up a gap which breaks the spectrum into an electron band at positive energies and a hole band at negative energies $\epsilon_{\mathbf{k}}^{\pm} = \pm\sqrt{\Delta^2 + (vk)^2}$, where $k = |\mathbf{k}|$ and $\pm$ refer to electron and hole bands, respectively. (The three other graphene bands differ either in the Dirac band chirality sense, or in the sign of the mass

term, or in both ways.) In what follows, we assume that the Fermi energy is positive; because of the symmetry of the Dirac Hamiltonian, generalization to negative ϵ_F is trivial.

The Kubo formula for the Hall conductivity depends on both band-diagonal and off-diagonal matrix elements of the velocity operator and on the electronic Green function. The disorder-free retarded Green function and velocity operators for this Hamiltonian are $G_0^R(\epsilon) = (\epsilon - \hat{H} + i\eta)^{-1}$, $v_x = v\sigma_x$, and $v_y = v\sigma_y$. We will use the Streda-Smrcka [10] version of the Kubo formula: $\sigma_{xy} = \sigma_{xy}^I + \sigma_{xy}^{II}$, where

$$\sigma_{xy}^I = \frac{-e^2}{4\pi}\int_{-\infty}^{+\infty} d\epsilon \frac{df(\epsilon)}{d\epsilon} \mathrm{Tr}\{v_x[G^R(\epsilon) - G^A(\epsilon)]v_y G^A(\epsilon) - v_x G^R(\epsilon) v_y [G^R(\epsilon) - G^A(\epsilon)]\} \quad (3)$$

and

$$\sigma_{xy}^{II} = \frac{e^2}{4\pi}\int_{-\infty}^{+\infty} d\epsilon f(\epsilon) \mathrm{Tr}\Big[v_x G^R(\epsilon) v_y \frac{G^R(\epsilon)}{d\epsilon} - v_x \frac{G^R(\epsilon)}{d\epsilon} \times v_y G^R(\epsilon) - v_x G^A(\epsilon) v_y \frac{G^A(\epsilon)}{d\epsilon} + v_x \frac{G^A(\epsilon)}{d\epsilon} v_y G^A(\epsilon)\Big]. \quad (4)$$

2D-Dirac-band intrinsic Hall conductivity.—There is a part of Hall conductivity (usually called intrinsic) which is not induced by disorder but rather is due to anomalous trajectories of free electrons under the action of the electric field. It is most simply evaluated by expressing [8] it in terms of matrix elements of the velocity operator between unperturbed Bloch states:

$$\sigma_{xy}^{\mathrm{int}} = \frac{e^2}{\Omega}\sum_{\mathbf{k}} \frac{f_{\mathbf{k}}^+ - f_{\mathbf{k}}^-}{(\epsilon_{\mathbf{k}}^+ - \epsilon_{\mathbf{k}}^-)^2} 2\,\mathrm{Im}[\langle u_{\mathbf{k}}^-|v_y|u_{\mathbf{k}}^+\rangle\langle u_{\mathbf{k}}^+|v_x|u_{\mathbf{k}}^-\rangle], \quad (5)$$

where the $f_{\mathbf{k}}^{\pm}$ are occupation numbers in the electron and hole bands, Ω the area of the system, and $|u_{\mathbf{k}}^{\pm}\rangle$ the $\mathbf{k}$-dependent pseudospinors of the chiral Dirac Hamiltonian, Eq. (2).

$$|u_{\mathbf{k}}^+\rangle = \begin{pmatrix} \cos(\theta/2) \\ \sin(\theta/2)e^{i\phi} \end{pmatrix}, \quad |u_{\mathbf{k}}^-\rangle = \begin{pmatrix} \sin(\theta/2) \\ -\cos(\theta/2)e^{i\phi} \end{pmatrix}, \quad (6)$$

where $\cos(\theta) = \Delta/\sqrt{(vk)^2 + \Delta^2}$ and $\tan(\phi) = k_y/k_x$. For the chemical potential in the upper band with Fermi momentum k_F we find from (5)

$$\sigma_{xy}^{\mathrm{int}} = -\frac{e^2\Delta}{4\pi\sqrt{(vk_F)^2 + \Delta^2}}. \quad (7)$$

When the chemical potential is in the gap we find

$$\sigma_{xy}^{\mathrm{gap}} \equiv -\frac{e^2}{4\pi}. \quad (8)$$

When the Kubo-Streda formula is written in terms of Green functions, as in (3) and (4), the intrinsic contribution (7) and (8) corresponds to the summation of only disorder-free diagrams in σ_{xy}^I and σ_{xy}^{II}. We found that in the metallic regime the disorder-independent part of σ_{xy}^I equals with (7) and $\sigma_{xy}^{II} = 0$.

When the chemical potential is in the gap we found $\sigma_{xy}^I = 0$ but $\sigma_{xy}^{II} = -\frac{e^2}{4\pi}$, which is the same as in (8). This is the 2D-Dirac model's half-quantized (in units $e^2/2\pi\hbar$) Hall conductivity, which after summing over bands is responsible for the quantum spin Hall effect discussed in Refs. [6,7,11]. It may be surprising that the Hall conductivity in Eq. (8) is a half-integer in units $e^2/(2\pi\hbar)$ given general arguments that it must be an integer for a filled band of noninteracting electrons. The resolution of this paradox is that Dirac bands come in pairs. The sum of the K and K' valley bulk conductivities is quantized; correspondingly only one band of edge states is induced by the truncation of both K and K' bulk bands.

Influence of disorder on σ_{xy}.—We assume a δ-correlated spin-independent random potential with Gaussian correlations $\langle V(\mathbf{r}_1)V(\mathbf{r}_2)\rangle_{\mathrm{dis}} = nV_0^2\delta(\mathbf{r}_1 - \mathbf{r}_2)$.

The SCBA that we employ includes only contributions from Feynman diagrams without crossed disorder correlation lines. This common approximation is self-consistent but incomplete. We assume that crossed-disorder-line contributions give rise to parametrically distinguishable effects and do not affect our qualitative conclusions about Hall effects in metallic graphene. Figure 1 illustrates the SCBA self-energy diagram which can be evaluated to obtain $\Sigma^R = -\frac{i}{4\tau^q}(1 + \sigma_z\cos(\theta))$ where τ^q is a quantum life time at the Fermi surface:

$$1/\tau^q = nV_0^2\int kdk\delta(\epsilon_F - \epsilon_{\mathbf{k}}^+) = \frac{nV_0^2 k_F}{v_F}. \quad (9)$$

Following the notation of Dugaev *et al.* [12] and Inoue *et al.* [13], the SCBA retarded Green function is

$$G^R = \frac{1}{1/G_0^R - \Sigma^R} = \frac{\epsilon_F + i\Gamma_0 + v(k_x\sigma_x + k_y\sigma_y) + (\Delta - i\Gamma_1)\sigma_z}{(\epsilon_F - \epsilon^+ + i\gamma^+)(\epsilon_F - \epsilon^- + i\gamma^-)}, \quad (10)$$

where $\Gamma_0 = 1/(4\tau^q)$, $\Gamma_1 = \Gamma_0\cos(\theta)$, $\gamma^+ = \Gamma_0 + \Gamma_1\cos(\theta)$, and $\gamma^- = \Gamma_0 - \Gamma_1\cos(\theta)$. For these chiral bands disorder not only gives the quasiparticle states a finite lifetime but also changes the quasiparticle eigenspinors. The SCBA includes in addition ladder diagram vertex correction illustrated in Fig. 2. For large $vk_F\tau$ the terms in σ_{xy}^I which are products of retarded and advanced Green functions dominate so that the 2D matrix vertex function for which we must solve satisfies

$$\Upsilon_y = \sigma_y + nV_0^2\int \frac{d^2\mathbf{k}}{(2\pi)^2} G^R\Upsilon_y G^A. \quad (11)$$

This equation is most easily solved by assuming that $\Upsilon_y = a\sigma_0 + b\sigma_x + c\sigma_y + d\sigma_z$ and deriving equations for a, b, c, and d. We find that $c = \frac{2[(vk)^2 + 2\Delta^2]}{4\Delta^2 + (vk)^2}$, $b = -\frac{8\Gamma_0\Delta[(vk)^2 + 2\Delta^2]}{[4\Delta^2 + (vk)^2]^2}$,

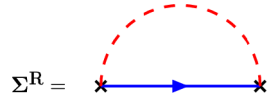

FIG. 1 (color online). Self-energy Feynman diagram.

and $a = d = 0$. The vertex correction is not trivial even when $\Delta = 0$ because then $c = 1/2$. This means that the approximation of disorder effects only by a level broadening is usually invalid even in the weak disorder limit. The SCBA σ^I_{xy} is obtained by substituting the disorder-dressed Green function [Eq. (10)] for the bare Green function and $v\Upsilon_y$ for v_y in the Kubo formula Eq. (3). We find that

$$\sigma_{xy} = \frac{-e^2\Delta}{4\pi\sqrt{(vk_F)^2+\Delta^2}}\left[1 + \frac{4(vk_F)^2}{4\Delta^2+(vk_F)^2} + \frac{3(vk_F)^4}{[4\Delta^2+(vk_F)^2]^2}\right]. \quad (12)$$

The second and third terms in square brackets in Eq. (12) represent disorder corrections to the intrinsic Hall conductivity of the 2D-Dirac model. We note that all terms are independent of the disorder potential strength and of the concentration of scatterers and in this sense are parametrically similar. They do, however, have different dependences on the position of the Fermi level. Note that when the chemical potential approaches the gap the contribution remains finite and disorder corrections vanish, recovering the model's half-quantized Hall effect [6]. In our previous publications [14] we discussed the semiclassical interpretations of such disorder effects in terms of the coordinate shift (side jump [15]) and asymmetry of the collision term corenel. Our result (12) is in perfect agreement with the semiclassical theory of the anomalous Hall effect presented in [14]; however, we postpone the detailed discussion to our future publications.

Non-Gaussian disorder.—We have so far made the usual approximation of assuming Gaussian disorder correlations. Although normally small, nonzero third moments of the disorder potential distribution can [16,17] alter σ_{xy} qualitatively since they can favor scattering with a particular chirality (skew scattering) and consequently lead to a σ_{xy} contribution that diverges in the limit of weak disorder scattering. The size of this contribution to σ_{xy} is particularly difficult to estimate since it depends very strongly on the details of the scattering potential. To illustrate its potential role we consider for concreteness a model of uncorrelated δ-function scatterers: $V(\mathbf{r}) = \sum_i V_i \delta(\mathbf{r} - \mathbf{R}_i)$, R_i random, $\langle V_i\rangle = 0$, $\langle (V_i)^2\rangle = V_0^2 \neq 0$, and $\langle (V_i)^3\rangle = V_1^3 \neq 0$.

Asymmetric scattering can be described directly using either Boltzmann transport theory or the Kubo formula, including the nonstandard Feynman diagrams implied by non-Gaussian disorder models. We apply results, which have been derived previously, to the graphene case. Let $\psi^+_{\mathbf{k}} = (1/\sqrt{\Omega})e^{i\mathbf{kr}}|u^+_{\mathbf{k}}\rangle$ be a Bloch state in the electron band with positive energy and $V_{\mathbf{k},\mathbf{k}'} = \langle\psi^+_{\mathbf{k}}|\hat{V}|\psi^+_{\mathbf{k}'}\rangle$ be a

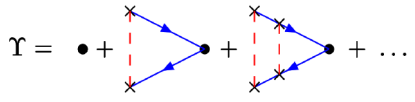

FIG. 2 (color online). Vertex correction Feynman diagram. Black dots represent the Pauli operator.

disorder potential matrix elements within the band. Following Eqs. (32)–(36) in Ref. [18] for zero temperature and a single band, we find

$$\frac{\sigma^{\rm sk}_{xy}}{(e\tau^{\rm tr})^2} = -\int\frac{d^2\mathbf{k}}{(2\pi)^2}\left(\frac{-\partial f_0}{\partial\epsilon}\right)\frac{v_x^2(\mathbf{k})}{\tau^\perp} = -\frac{v_F k_F}{4\pi\tau^\perp} \quad (13)$$

where $v_x(\mathbf{k}) = \partial\epsilon^+_{\mathbf{k}}/\partial k_x$, v_F is the Fermi velocity, and

$$\begin{aligned} 1/\tau^{\rm tr} &= \int\frac{d^2\mathbf{k}'}{(2\pi)^2}\omega_{\mathbf{k},\mathbf{k}'}[1-\cos(\phi-\phi')], \\ 1/\tau^\perp &= \int\frac{d^2\mathbf{k}'}{(2\pi)^2}\omega_{\mathbf{k},\mathbf{k}'}\sin(\phi-\phi'). \end{aligned} \quad (14)$$

Since the scattering rate $\omega_{\mathbf{k},\mathbf{k}'}$ is usually only weakly chiral ($\tau^{\rm tr} \ll \tau^\perp$), $\omega_{\mathbf{k},\mathbf{k}'}$ can be estimated from time-dependent perturbation theory [16,19]. The lowest order symmetric scattering rate is given by the golden rule expression, while the lowest order antisymmetric contribution appears at third order [see, for example, Eqs. (2.7) and (3.11) in Ref. [16]].

$$\begin{aligned} \omega^{(3a)}_{\mathbf{k},\mathbf{k}'} &= -(2\pi)^2\delta(\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}'}) \\ &\times \int\frac{d^2\mathbf{k}''}{(2\pi)^2}\,{\rm Im}\langle V_{\mathbf{k},\mathbf{k}'}V_{\mathbf{k}',\mathbf{k}''}V_{\mathbf{k}'',\mathbf{k}}\rangle_{\rm dis}\delta(\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}''}). \end{aligned} \quad (15)$$

This yields

$$\frac{1}{\tau^{\rm tr}} = \frac{(vk_F)^2 + 4\Delta^2}{4\tau^q[(vk_F)^2 + \Delta^2]}, \quad (16)$$

$$\frac{1}{\tau^\perp} = \frac{V_1^3}{(\tau^q)^2 n V_0^4}\frac{\Delta(vk_F)^2}{8[(vk_F)^2 + \Delta^2]^{3/2}}, \quad (17)$$

so that the skew scattering Hall conductivity contribution due to non-Gaussian disorder correlations is

$$\sigma^{\rm sk}_{xy} = -\frac{e^2V_1^3}{2\pi n V_0^4}\frac{\Delta(vk_F)^4}{[4\Delta^2+(vk_F)^2]^2}. \quad (18)$$

The Hall conductivity contribution (18) is inversely proportional to the impurity concentration n, and therefore can in principle dominate in relatively clean samples. Since the size of third disorder correlation moment in a particular sample is unlikely to be reliably known and can be exceedingly small, we expect that the relative importance of skew scattering will always have to be assessed experimentally.

Application to graphene.—A finite charge Hall conductance requires broken time reversal symmetry. In graphene the vanishing conductance results from cancellation between bands of opposite spin. The Hall conductance we evaluate here could be measured in graphene if the Fermi levels in the two spin-$\uparrow$ and the two spin-$\downarrow$ bands differed. It may be possible to generate spin polarization in graphene by optical orientation [20], by tunneling through ferromagnetic contacts, or by hyperfine coupling to polarized nuclei. We note that the $\hat{z}$ component of spin is expected to relax

particularly slowly in graphene because of the planar character of the crystal and the π character of the orbitals near the Fermi energy. The alternative of studying the physics we address here, by applying an external magnetic field, is not favorable since it leads to an ordinary Hall effect in addition to the anomalous Hall effect. When the chemical potentials of spin-up and spin-down electrons are different our Hall effect calculation for each band remains valid. The total Hall current is therefore

$$\sigma_{xy}^{\mathrm{AHE}} = 2[\sigma_{xy}(\mu_\uparrow) - \sigma_{xy}(\mu_\downarrow)], \tag{19}$$

where the coefficient 2 reflects equal contributions from the K and K' valleys.

The Hall conductivity we evaluate appears in the spin Hall response even in the absence of external magnetic fields. To find the magnitude of the spin Hall effect one should remember that instead of charge e we are interested in spin $\pm 1/2$ carried by electrons: $\sigma_{xy}^{\mathrm{SH}} = 4\sigma_{xy}/2e$. Here the coefficient 4 is due to the 4 Dirac bands which contribute equally to the spin Hall effect. The spin Hall effect could be measured by using ferromagnetic leads, in the extreme case measuring transport only in one spin subsystem. For that case the charge Hall conductivity becomes $2\sigma_{xy}$. Alternately the spin Hall conductivity could be measured optically using either the Faraday effect [21] or polarization selective electroluminescence [22]. Both approaches should also be successful in graphene. We expect that the results we derive here are valid for $\epsilon_F \gtrsim \tau^{-1}$, whereas the quantized spin Hall conductivity will be observable only if $\Delta \gtrsim \tau^{-1}$. The value of τ^{-1} in current samples can be estimated roughly from measured mobilities [5] which are roughly constant except for Fermi energies below $\sim$50 meV. Associating the change in mobility at low carrier densities with disorder mixing between electron and hole bands implies a τ^{-1} value of the same order. The value of Δ is difficult to estimate accurately. Based on the relevant potential energy and length scales Kane and Mele have estimated that $\Delta \sim 0.2$ meV. This is likely to be an overestimate since the splitting represents an average of spin-orbit interactions that vary in sign over the system. We [23] have separately estimated on the basis of a tight-binding model with atomic spin-orbit interactions and *ab initio* electronic structure calculations that $\Delta \sim 0.001$ meV. In any event, it appears clear that sample quality will need to improve substantially in order to realize the quantum spin Hall effect. As our calculation shows, however, the surprisingly large spin Hall currents that flow from the chiral graphene bands should still be measurable in the metallic regime.

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