Photovoltaic Hall effect in graphene

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(Dated: March 6, 2009)

Response of electronic systems in intense lights (AC electric fields) to DC source-drain fields is formulated with the Floquet method. We have then applied the formalism to graphene, for which we show that a non-linear effect of a circularly polarized light can open a gap in the Dirac cone, which is predicted to lead to a photo-induced dc Hall current. This is numerically confirmed for a graphene ribbon attached to electrodes with the Keldysh Green’s function.

PACS numbers: 73.43.-f,72.40.+w,78.67.-n,85.60.-q

Introduction — Non-linear phenomena in electronic systems are fascinating since they can lead to transport properties qualitatively distinct from those in equilibrium. In this Letter we seek such a possibility by combining (i) the geometric phase argument extended to electron transports in intense AC fields with (ii) physics of graphene involving chiral states associated with two Dirac cones. Indeed, the geometric phase has become an important ingredient in the modern theory of electric transport[1], which goes back to Thouless’s idea of charge pumping where he showed that an adiabatic deformation of the system may lead to quantized transport[2]. Extensions along several directions have been done subsequently. Thouless et al. (TKNN)[3] have shown that the Kubo formula for the Hall conductivity can be expressed as a topological density. Berry then showed that such phases are present in general quantum systems, and the topological density in the TKNN formula is now called the Berry curvature[4]. Aharonov and Anandan further extended the notion of geometric phases to non-adiabatic situations, i.e., Aharonov-Anandan (AA) phase[5].

With these as a background, the concept of the present study, with graphene in mind, is simple. Let us put a crystal with a Dirac band in a circularly polarized light. As in Fig. 1(a), an intense AC field \(A_{ac}(t)\) will deform the single-body Hamiltonian, and each k-point will follow a circle in the Brillouin zone. If the loop encircles the Dirac point, non-adiabatic charge pumping should take place, in which the wave function acquires a non-trivial AA phase. So the question is: can this be detected with a DC transport measurement? In order to answer this, we have formulated the Kubo formula for the DC response for systems in intense AC fields, which is accomplished with the Floquet-matrix formalism by which we can solve the time-dependent dynamics of k-points within a static approach. We shall show that a TKNN-like formula for the Hall conductivity is obtained, where the Berry curvature is now expressed in terms of Floquet states which depends on the AA phase. We apply this formula to a single Dirac band first, then a graphene.

For graphene[2, 1], which is dominated by the chirality, we conclude that a photo-induced Hall current (despite the absence of uniform magnetic fields) should appear in graphene irradiated by circularly polarized light and attached to two electrodes, where the Hall current can exceed the longitudinal current in magnitude.

Kubo formula in the presence of strong light fields — We first derive the Kubo formula for electric transport for systems in strong AC fields, where we concentrate on the one-body problem for simplicity. The AC electric field is introduced as a time-dependent gauge potential \(A_{ac}(t)\), which satisfies \(A_{ac}(t + T) = A_{ac}(t) + T\) the periodicity (while the frequency is \(\Omega = 2\pi/\Omega\)). On top of this we introduce, as in the linear response, a weak gauge potential \(A(t) = Et\) that changes slowly to represent an infinitesimal DC electric field \(E\), where we have set \(e = 1, \ h = 1\). Thus we have a time-dependent Hamiltonian,

\[
H(t) = \int \frac{d k}{(2\pi)^d} \langle A(k) \rangle (k + A_{ac}(t) + A(t)) \psi(k),
\]

where \(\psi(k)\) is the one-body Hamiltonian and \(\psi\) a state vector (which is multi-dimensional for multibands). To represent the states in AC fields, we can employ the Floquet operator (see 2, 3) \(\overline{\psi}(k, A_{ac}(t), A(t)) = h(k + A_{ac}(t) + A(t)) - i\partial_t\), with which the time-dependent Schrödinger equation reads \(\overline{\psi}(k, A_{ac}(t), A(t))\psi(k; t) = 0\). Since \(A(t)\) is infinitesimal and adiabatically changing, we take it as the adiabatic parameter, while the AC field is intense and rapidly oscillating. So, for each interval of time over which \(A\) may be considered to be constant, we can introduce the Floquet states (a time-analog of Bloch states) satisfying the Floquet equation \(\overline{\psi}(k, A_{ac}(t), A(t))\psi_\alpha(k, A, t) = \varepsilon_\alpha(k, A)\psi_\alpha(k, A, t)\) with a periodicity \(\psi_\alpha(k, A, t + T) = \psi_\alpha(k, A, t)\) \(1\) where \(\varepsilon_\alpha(k, A)\) is called the Floquet quasi-energy which is a sum of the dynamical phase and the AA phase (see eqn. 11 below), and \(\alpha\) labels the eigenstate. The solution of the time-dependent Schrödinger equation for a fixed \(A\) can be expressed as \(\psi_\alpha(t) = e^{-i\varepsilon_\alpha t}\psi_\alpha(0)\). If we define an inner product averaged over a period by \(\langle \alpha|\beta\rangle \equiv \frac{1}{T} \int_0^T dt \langle \alpha(t)|\beta(t)\rangle\), the Floquet states form an orthonormal basis, i.e., \(\psi_\alpha^\dagger(k, A)\psi_\beta(k, A) = \delta_{\alpha\beta}\). With these as a basis the solution to the time-dependent Schrödinger equation for the slow change in \(A\) is

\[
|\psi(k, A(t), t)\rangle = e^{-i\int_0^t dt \varepsilon_\alpha(k; A(t))} |\psi_\alpha(k, A(t), t)\rangle + \sum_{\beta \neq \alpha} |\psi_\beta(k, A(t), t)\rangle \frac{\langle \Phi_\beta(k, A(t))|A(t)\rangle \partial A(t)}{\varepsilon_\beta(k, A(t)) - \varepsilon_\alpha(k, A(t))},
\]
up to first order in time derivatives, with the \( \alpha \)-th Floquet state taken to be the initial state. One can readily derive this result with the two-time method\(^{[11,12]}\). We can immediately notice that the geometrical phase appears in a form \( \langle \{ \Phi_\beta(k; A(t)) \} | \frac{\partial A}{\partial t} | \{ \Phi_\alpha(k; A(t)) \} \rangle = \sum_{\alpha, \beta \neq \alpha} \langle \{ \Phi_\beta(k; A(t)) \} | \frac{\partial A}{\partial t} | \{ \Phi_\alpha(k; A(t)) \} \rangle (\varepsilon_\beta A(t) - \varepsilon_\alpha A(t))^{-1} \). Since the current operator is \( J = \partial h(k + A_{AC}(t) + A)/\partial A \), the above formula for the DC transport in an intense AC background field is rewritten as

\[
\sigma_{ab}(A_{AC}) = i \int \frac{dk}{(2\pi)^d} \sum_{\alpha, \beta \neq \alpha} \frac{f_\alpha(k) - f_\beta(k)}{\varepsilon_\beta(k) - \varepsilon_\alpha(k)} \times \frac{\langle \{ \Phi_\beta(k) | J_0 | \Phi_\alpha(k) \rangle \langle \{ \Phi_\beta(k) | J_0 | \Phi_\alpha(k) \rangle \rangle}{\varepsilon_\beta(k) - \varepsilon_\alpha(k) + i\eta},
\]

(2)

where \( f_\alpha(k) \) is the non-equilibrium distribution (occupation fraction) of the \( \alpha \)-th Floquet state, \( \eta \) a positive infinitesimal, and we have put the perturbation \( A = 0 \) as in the linear-response theory. The essential difference from the conventional Kubo formula in the absence of AC fields is that the energy is replaced with the Floquet quasi-energy, and the inner product with a time averaged one. We note that similar expressions were obtained by Torres and Kunold in their study of microwave-assisted zero-resistance states\(^{[13]}\). The Hall conductivity can be further simplified to a TKNN-like formula,

\[
\sigma_{xy}(A_{AC}) = e^2 \int \frac{dk}{(2\pi)^d} \sum_\alpha f_\alpha(k) [\nabla_k \times A_{\alpha}(k)]_z,
\]

(3)

where \( A_{\alpha}(k) \equiv -i \langle \{ \Phi_\alpha(k) | \nabla_k | \Phi_\alpha(k) \} \rangle \). We note that if we separate the Floquet index into \( \alpha = (i, m) \) where \( i \) labels the original band and \( m \) is the Floquet index, then \( A_{\alpha}(k) \) is independent of \( m \). However, the occupation \( f_\alpha(k) \) depends on both indices. In equilibrium, \( f_{(i,m)}(k) = \delta_{\alpha 0} f_{FD}(E_{i}(k)) \) holds where \( E_{i}(k) \) is the energy of \( i \)-th state and \( f_{FD} \) the Fermi-Dirac distribution. In non-equilibrium, however, the distribution is non-universal, and depends on the detail of the system such as how the electrodes are attached, etc., so that a case-by-case study should be needed to determine the distribution and hence the DC transport of the system.

**Application to a Dirac band** — Now we study the effect of AC fields on the two-dimensional Dirac band. The Hamiltonian is \( H(t) = t_x \sqrt{\varepsilon + A_{\alpha _x}^\prime(t)} \sigma_x + t_y \sqrt{\varepsilon - A_{\alpha _y}^\prime(t)} \sigma_y \), where \( t_x = \pm 1 \) labels the chirality (\( K \) and \( K' \) points in graphene), \( \varepsilon \) is the velocity (set to \( v \) hereafter), and \( \sigma_1 \) the Pauli matrices. The circularly polarized field is given by \( A_{\alpha _x}^\prime(k) = A_{\alpha _y}^\prime(k) = A(\cos \Omega t, \sin \Omega t) \) where \( A \equiv F/\Omega \) with \( F \) being the field strength. Here we neglect the momentum of light since \( v \ll c \), and consider direct transitions, which is a situation different from the Volkov solution\(^{[14]}\). With Fourier transformed Floquet states \( \Phi(t) = \sum_\alpha e^{-im \Omega t} | v_{\alpha _m}^m \rangle \) the Floquet equation becomes

\[
\sum_n H_{mn}^\alpha | v_{\alpha _m}^m \rangle = (\varepsilon_\alpha + m \Omega) | v_{\alpha _m}^m \rangle,
\]

(4)

where the Floquet Hamiltonian \( H_{mn}^\alpha = \frac{1}{\Omega} \int_0^\tau dt H(t) e^{i(m-n)\Omega t} \) has \( H_{mn}^m = (0_{k_x+i k_y}, 0_{k_x-i k_y}) \) for the diagonal components, whereas the off-diagonal components depend on \( \tau_z \), i.e., \( H_{mn}^{m+1} = (0_{A_0}, \Omega A^{m+1} = (0_{A_0}) \) for \( \tau_z = 1 \) and \( H_{mn}^{-m+1} = (0_{A_0}, \Omega A^{m-1} = (0_{A_0}) \) for \( \tau_z = -1 \). This defines an eigenvalue problem for a block tri-diagonal matrix that can be solved numerically with a truncation at certain \( |m| \).

As an important effect of the AC field, gaps open at \( \omega = \text{integer} \times \Omega / 2 \) in the quasi-energy band structure (Fig.1(b)), reflecting a Dirac-band analog of the AC-Wannier-Stark ladder, as also seen in the density of states \( A(\omega) = -\frac{1}{\pi} \int d\varepsilon \sum_\alpha \text{Im} \langle \{ \Phi_{\alpha _m}^\prime(k) | \Phi_{\alpha _m}^\prime(k) \} \rangle \). The gap at \( \omega = \pm \Omega / 2 \) is the largest, which is related to one-photon assisted transport discussed later. We note that this gap also opens in the linearly polarized case as studied in\(^{[13]}\). More importantly, a new gap opens at \( k = 0, \varepsilon = 0 \) in the circularly polarized case. Indeed, one can show that the solution of the time-dependent Schrödinger equation at \( k = 0 \) is

\[
| \Psi_\alpha(k = 0, t) \rangle \propto e^{-i\varepsilon_\alpha t \frac{1}{\tau}} e^{i\Omega t}
\]

(5)

where the Floquet state is labeled by \( \alpha = (i, m) \) with \( i = 1, 2 \) representing the upper and lower branches of the Dirac band, \( m \) the Floquet index. The quasi-energy is \( \varepsilon_\alpha = \varepsilon_0 + m \Omega \) with \( \varepsilon_1 = \sqrt{4\Delta^2 + \Omega^2} \), \( \varepsilon_2 = -\sqrt{4\Delta^2 + \Omega^2} \). The \( \alpha = (1, -1), (2, 0) \) bands are direct descendants of the original Dirac bands, and the dynamical gap \( 2\kappa \) opening between them is \( \kappa = \frac{\sqrt{4\Delta^2 + \Omega^2} - \Omega}{2} \).
The dynamical gap first grows quadratically with $A$, $2\kappa \sim 2A^2/\Omega$, followed by an asymptote $2\kappa \sim 2A - \Omega$. An important property of the quasi-energy is that it is a sum of the dynamical phase and the AA phase, i.e.,

$$\varepsilon_\alpha = \langle \Phi_\alpha | H(t) | \Phi_\alpha \rangle + \gamma_{\alpha A}/T,$$

where the AA phase is given by

$$\gamma_{\alpha A} = T \langle \langle \Phi_\alpha | i\partial_t | \Phi_\alpha \rangle \rangle = \pm \pi \left\{ 4(A/\Omega)^2 + 1 \right\}^{-1/2} - 1,$$

where $\pm$ refers to $\alpha = (1, -1), (2, 0)$. In the adiabatic limit ($\Omega \rightarrow 0$ with a fixed $A$) it approaches $\pm \pi$. We note that only the $k$-points with $|k| < A$ acquires the AA phase, since otherwise the Dirac cone is not encircled. In the Berry curvature for the $\alpha = (2, 0)$ Floquet state (Fig. 4d)) there is a conspicuous peak around $k = 0$,

$$[\nabla_k \times \mathbf{A}_\alpha(k)]_z \sim \pm \tau_2 \frac{1}{2} \kappa ([k|^2 + \kappa^2]^{-3/2})^{-1},$$

where $\pm$ corresponds to $\alpha = (1, m), (2, m)$. In this expression, two geometric quantities appear, while the Berry curvature comes from the perturbative treatment of the weak DC electric field whereas the AA phase emerges because of the time-periodic dynamics of $k$-points in intense AC fields. Due to the factor $\tau_2$, the contribution from $K$ and $K'$ points in graphene will cancel with each other if the distribution is identical between them. However, we shall see that, if we apply a static DC bias across the system, chirality (the valley symmetry) is degraded, which will lead to a non-trivial curvature, hence to a Hall current.

**Keldysh approach to photovoltaic transport in graphene**

— So we move on to a Keldysh Green’s function analysis of transport properties in a graphene irradiated by a circularly polarized light and attached to two electrodes. The system is described by an action, $S = \int dt (\mathcal{L}_{\text{graphene}} + \mathcal{L}_{\text{mix}} + \mathcal{L}_{\text{electrodes}})$, where $\mathcal{L}_{\text{graphene}} = \sum_{ijA} \frac{\hbar^2}{2} c_i^{\dagger}(i\partial_t - t_{ij} e^{iA^{\text{ac}}_{ij}(t)})c_j$ is the tight-binding model for graphene with a hopping $t_{ij}$, $\mathcal{L}_{\text{mix}} = \sum_{k} \mathcal{V}^{\text{mix}}_{\text{ac}}(\alpha_k) c_k + \text{h.c.}$ represents the coupling between the electrodes and graphene, with the spin degrees of freedom ignored. The AC field is introduced by $A^{\text{ac}}_{ij}(t) = (r_i - r_j) \cdot A^{\text{ac}}(t)$ with $A^{\text{ac}}(t) = (F/\Omega)(\cos \Omega t, \sin \Omega t)$, where $F = e\alpha E$ is the normalized field strength ($\alpha$: lattice const.). We assume that the electrodes are described by a fermion operator $a^\dagger (r \in \{L, R\}$ labeling the left and right electrodes), for which a Fermi-Dirac distribution $\langle a^\dagger a^\dagger \rangle = f_r = [e^{\beta (\omega - \mu_r)} + 1]^{-1}$ is assumed with electrode-dependent chemical potential $\mu_r$. They are related to the DC bias $V$ across the electrodes by $\mu_L = V/2$, $\mu_R = -V/2$. Imposing a periodic boundary condition in the direction $(y)$ of the graphene ribbon, the Keldysh Green’s functions for each momentum $k_y$ become a matrix labeled by the site in the $x$-direction ($i = 1, \ldots, N$) and by the Floquet index. The Green’s functions satisfy

$$\left( \begin{array}{cc} G^R_{kj}(\omega) & G^K_{kj}(\omega) \\ 0 & G^A_{kj}(\omega) \end{array} \right)^{-1} = \left( \begin{array}{cc} (\omega + n\Omega + i\eta)\delta_{mn} \delta_{ij} - (\hat{H})_{ij}^{mn}(k_y) & 0 \\ 0 & (\omega + n\Omega - i\eta)\delta_{mn} \delta_{ij} - (\hat{H})_{ij}^{mn}(k_y) \end{array} \right),$$

where $G^{K, R, A}$ are the Keldysh, retarded, and advanced Green’s function, respectively, $\Gamma \propto |V^{\text{mix}}|^2$ is the imaginary part of the self-energy due to the sample-electrode coupling. The effective Floquet Hamiltonian is defined by $(H)^{mn}(k_y) = \frac{\hbar}{\Omega} \int_0^\Omega dt e^{i(m-n)\Omega t} H(k_y; A^{\text{ac}}(t))$, where the indices $i, j$ are suppressed. The current between sites $i, j$ is determined from the lesser component $G^\ast$ by

$$\langle J^R_{ij}(t) \rangle = -\frac{e}{\hbar} \frac{1}{N_{k_y}} \sum_{k_m} \sum_{m_n} \int_0^\Omega \frac{d\omega}{2\pi} e^{-i(m-n)\Omega t} \left( \begin{array}{c} 0 \\ \Gamma R/2 \end{array} \right),$$

where $J_{ij} = \delta \hat{H} / \delta A_{ij}$ is the current operator. In practice, we do not calculate the Keldysh component but use the Keldysh equation (c.f. 10) to relate $G^{\ast}$ with $G^A, G^R$ obtained by diagonalizing the Floquet Hamiltonian.

In the obtained current distribution (Fig. 2a)) the polarized light induces locally circulating currents in the absence of the bias $V$ across the electrodes. There is no net current in the $y$-direction as it should. This current resembles the orbital magnetism which was predicted to arise when a perturbation induce a gap in the Dirac cone 17, 18. Indeed, the circularly polarized light in the present case opens a gap around the band crossing (arrow in Fig. 2b)), which has led to a similar DC effect.

A striking finding here is that, when we switch on the bias voltage $V$, we have a photo-induced DC Hall current as well as a longitudinal ($|| x $) current (Fig. 2b)). The Hall current is naturally inverted when the right circularly polarized field $A^{\text{ac}} \propto (\cos \Omega t, \sin \Omega t)$ is changed into the left polarization $\propto (\cos \Omega t, -\sin \Omega t)$, or the bias voltage is inverted. The $I - V$-characteristics is shown in Fig. 2c) for $J_x$ and $J_y$, the averaged cur-
FIG. 2: DC current distribution in an armchair graphene ribbon attached to electrodes subject to a circularly polarized light for a finite AC field $F = 0.025w$, $\Omega = 0.3w$ with no DC bias (a) and with a finite bias $V = 0.005w$ (b). (c) $I-V$ characteristics of the longitudinal $J_x$ (black) and the Hall current $J_y$ (blue) for various values of $F$. (d) DC conductance $G = J/V$ plotted against field strength $F$ for a fixed bias $V = 0.005w$. System size in the $x$-direction $N = 34$ throughout.

rent in $x$ and $y$ directions, respectively. The photo-induced net Hall current grows linearly with the bias $V$, but saturates and then decrease when $V$ becomes large. Figure 2(d) depicts the dependence of the conductance, $G_{xx} = J_x/V$, $G_{xy} = J_y/V$ on the intensity $F$ of the circularly polarized light for a fixed bias $V$. The conductance, unlike the conductivity discussed in the first part of the paper, is less universal and depends on the contact $\Gamma_r$ (here we set $\Gamma_L = \Gamma_R = 0.2w$) etc, but we expect that the effect will be qualitatively robust. As we increase $F$, the longitudinal $G_{xx}$ first decreases, and increase again ($F > 0.03w$). The decrease can be explained by the gap opening at the Dirac points, while the increase is due to photo-assisted transport. We note that similar features have been experimentally observed in microwave irradiated carbon nanotubes [10]. The Hall conductance $G_{xy}$, on the other hand, initially grows quadratically with $F$ and then increases linearly, which is a dependence similar to the gap $\kappa$ in the Dirac cone (eqn. (3)), which may indicate $J_y \sim \kappa V$. We note that a similar expression was obtained in the case where the chirality is broken in a static manner [20].

To summarize, we have found that a combined effect of an intense AC field of a circularly polarized light and a (weak) DC bias can produce a photo-voltaic dc Hall current in graphene, despite the absence of a uniform magnetic field. The typical intensity of laser conceived here, $F \sim 0.001w$, corresponds to $E \sim 10^7V/m$ for photon energy $E \sim 1eV$, $w = 2.7eV$, $a = 2.6\AA$, which should be within the experimental feasibility. Inclusion of dissipation etc will be an interesting future problem.

We wish to thank Andre Geim for fruitful discussion in the initial stage of this work. TO thanks Jun Okubo and Naoto Tsuji for eliminating discussions on the Floquet method. HA was supported by a Grant-in-Aid for Scientific Research on Priority Area “Anomalous quantum materials” from the Japanese Ministry of Education, TO by Grant-in-Aid for young Scientists (B).

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