# Photovoltaic Hall effect in graphene

Takashi Oka and Hideo Aoki

Department of Physics, University of Tokyo, Hongo, Tokyo 113-0033, Japan (Received 29 July 2008; revised manuscript received 15 January 2009; published 23 February 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 nonlinear effect of a circularly polarized light can open a gap in the Dirac cone, which is predicted to lead to a photoinduced dc Hall current. This is numerically confirmed for a graphene ribbon attached to electrodes with the Keldysh Green's function.

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### I. INTRODUCTION

Nonlinear phenomena in electronic systems are fascinating since they can lead to transport properties qualitatively distinct from those in equilibrium. In this Rapid Communication 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,<sup>1</sup> 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.<sup>2</sup> Extensions along several directions have been done subsequently. Thouless et al. (TKNN) (Ref. 3) have shown that the Kubo formula for the Hall conductivity can be expressed as a topological density. Berry<sup>4</sup> 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. Aharonov and Anandan<sup>5</sup> further extended the notion of geometric phases to nonadiabatic situations, i.e., Aharonov-Anandan (AA) phase.

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, nonadiabatic charge pumping should take place, in which the wave function acquires a nontrivial 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-type formula for the Hall conductivity is obtained, where the Berry curvature is now expressed in terms of Floquet states which depend on the AA phase. We apply this formula to a single Dirac band first, then a graphene. For graphene,<sup>6,7</sup> which is dominated by the chirality, we conclude that a photoinduced 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.

## II. 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  $A_{ac}(t+T)=A_{ac}(t)$  with T the periodicity (while the frequency is  $\Omega=2\pi/T$ ). 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,  $\hbar=1$ . Thus we have a time-dependent Hamiltonian,

$$H(t) = \int \frac{d\mathbf{k}}{(2\pi)^d} \Psi^{\dagger}(\mathbf{k}) h[\mathbf{k} + \mathbf{A}_{\rm ac}(t) + \mathbf{A}(t)] \Psi(\mathbf{k}), \quad (1)$$

where  $h(\mathbf{k})$  is the one-body Hamiltonian and  $\Psi$  a state vector (which is multidimensional for multibands). To represent the states in ac fields, we can employ the Floquet operator (see Refs. 8 and 9)  $\mathcal{H}[\mathbf{k}, \mathbf{A}_{ac}(t), \mathbf{A}(t)] = h[\mathbf{k} + \mathbf{A}_{ac}(t) + \mathbf{A}(t)] - i\partial_{t}$ ,



FIG. 1. (Color online) (a) A trajectory of  $\mathbf{k} + A_{ac}(t)$  around a Dirac point in a circularly polarized light field. (b) The Floquet quasienergy (black curves) plotted against  $k_x$  with  $k_y=0$  for  $F/\Omega = 0.2$ . The color coding represents the weight of the static (m=0) component. (c) Density of states for various field strengths. (d) The photoinduced Berry curvature  $[\nabla_k \times \mathcal{A}_\alpha(k)]_z$  for  $\alpha = (1,m)$  for  $F/\Omega = 0.2$ . The series  $\alpha = (2,m)$  have a similar behavior with an inverted sign. Frequency is  $\Omega/v = 1$  throughout.

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with which the time-dependent Schrödinger equation reads  $\mathcal{H}[\mathbf{k}, \mathbf{A}_{ac}(t), \mathbf{A}(t)] | \Psi(\mathbf{k}; t) \rangle = 0$ . Since  $\mathbf{A}(t)$  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  $\mathcal{H}[k, A_{ac}(t), A] | \Phi_{\alpha}(k; A, t) \rangle = \varepsilon_{\alpha}(k; A) | \Phi_{\alpha}(k; A, t) \rangle$  with a periodicity  $|\Phi_{\alpha}(\mathbf{k}; \mathbf{A}, t+T)\rangle = |\Phi_{\alpha}(\mathbf{k}; \mathbf{A}, t)\rangle$  (Ref. 10), where  $\varepsilon_{\alpha}(k;A)$  is called the Floquet quasienergy which is a sum of the dynamical phase and the AA phase [see Eq. (6) below], and  $\alpha$  labels the eigenstate. The solution of the timedependent Schrödinger equation for a fixed A can be expressed as  $|\Psi_{\alpha}(t)\rangle = e^{-i\varepsilon_{\alpha}t} |\Phi_{\alpha}(t)\rangle$ . If we define an inner product averaged over a period by  $\langle \langle \alpha | \beta \rangle \rangle \equiv \frac{1}{T} \int_{0}^{T} dt \langle \alpha(t) | \beta(t) \rangle$ , the Floquet states form an orthonormal basis, i.e.,  $\langle \langle \Phi_{\alpha}(\boldsymbol{k}; \boldsymbol{A}) | \Phi_{\beta}(\boldsymbol{k}; \boldsymbol{A}) \rangle \rangle = \delta_{\alpha\beta}$ . With these as a basis the solution to the time-dependent Schrödinger equation for the slow change in A is

$$\begin{split} \Psi[\boldsymbol{k}; \boldsymbol{A}(t), t] \rangle &= e^{-i \int_{0}^{t} dt' \varepsilon_{\alpha}[\boldsymbol{k}; \boldsymbol{A}(t')]} \\ &\times \Bigg[ |\Phi_{\alpha}[\boldsymbol{k}; \boldsymbol{A}(t), t] \rangle + \sum_{\beta \neq \alpha} |\Phi_{\beta}[\boldsymbol{k}; \vec{A}(t), t] \rangle \\ &\times \frac{\langle \langle \Phi_{\beta}[\boldsymbol{k}; \boldsymbol{A}(t)] | \frac{\partial \boldsymbol{A}}{\partial t} \cdot \frac{\partial}{\partial \boldsymbol{A}} |\Phi_{\alpha}[\boldsymbol{k}; \boldsymbol{A}(t)] \rangle \rangle}{\varepsilon_{\beta}[\boldsymbol{k}; \boldsymbol{A}(t)] - \varepsilon_{\alpha}[\boldsymbol{k}; \boldsymbol{A}(t)]} \Bigg], \end{split}$$

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.<sup>11,12</sup> We can immediately notice that the geometrical phase appears in a form

$$\left\langle \left\langle \Phi_{\beta}[\boldsymbol{k};\boldsymbol{A}(t)] \middle| \frac{\partial \boldsymbol{A}}{\partial t} \cdot \frac{\partial}{\partial \boldsymbol{A}} \middle| \Phi_{\alpha}[\boldsymbol{k};\boldsymbol{A}(t)] \right\rangle \right\rangle$$
$$= \sum_{\beta \neq \alpha} \left\langle \left\langle \Phi_{\beta}[\boldsymbol{k};\boldsymbol{A}(t)] \middle| \frac{\partial \boldsymbol{A}}{\partial t} \cdot \frac{\partial \mathcal{H}}{\partial \boldsymbol{A}} \middle| \Phi_{\alpha}[\boldsymbol{k};\boldsymbol{A}(t)] \right\rangle \right\rangle$$
$$\times \{ \varepsilon_{\beta}[\boldsymbol{A}(t)] - \varepsilon_{\alpha}[\boldsymbol{A}(t)] \}^{-1}.$$

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

$$\sigma_{ab}(A_{ac}) = i \int \frac{d\mathbf{k}}{(2\pi)^d} \sum_{\alpha,\beta\neq\alpha} \frac{[f_{\beta}(\mathbf{k}) - f_{\alpha}(\mathbf{k})]}{\varepsilon_{\beta}(\mathbf{k}) - \varepsilon_{\alpha}(\mathbf{k})} \times \frac{\langle\langle \Phi_{\alpha}(\mathbf{k}) | J_b | \Phi_{\beta}(\mathbf{k}) \rangle \rangle \langle\langle \Phi_{\beta}(\mathbf{k}) | J_a | \Phi_{\alpha}(\mathbf{k}) \rangle \rangle}{\varepsilon_{\beta}(\mathbf{k}) - \varepsilon_{\alpha}(\mathbf{k}) + i\eta}, \quad (2)$$

where  $f_{\alpha}(\mathbf{k})$  is the nonequilibrium distribution (occupation fraction) of the  $\alpha$ th Floquet state,  $\eta$  is a positive infinitesimal, and we have put the perturbation  $\mathbf{A}=0$  as in the linearresponse theory. The essential difference from the conventional Kubo formula in the absence of ac fields is that the energy is replaced with the Floquet quasienergy, and the inner product with a time averaged one. We note that similar expressions were obtained by Torres and Kunold<sup>13</sup> in their

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study of microwave-assisted zero-resistance states. The Hall conductivity can be further simplified to a TKNN-type formula,

$$\sigma_{xy}(\boldsymbol{A}_{\rm ac}) = e^2 \int \frac{d\boldsymbol{k}}{(2\pi)^d} \sum_{\alpha} f_{\alpha}(\boldsymbol{k}) [\nabla_{\boldsymbol{k}} \times \boldsymbol{\mathcal{A}}_{\alpha}(\boldsymbol{k})]_{z}, \qquad (3)$$

where  $\mathcal{A}_{\alpha}(\mathbf{k}) \equiv -i\langle\langle \Phi_{\alpha}(\mathbf{k}) | \nabla_{\mathbf{k}} | \Phi_{\alpha}(\mathbf{k}) \rangle\rangle$ . 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  $\mathcal{A}_{\alpha}(\mathbf{k})$  is independent of *m*. However, the occupation  $f_{\alpha}(\mathbf{k})$  depends on both indices. In equilibrium,  $f_{(i,m)}(\mathbf{k}) = \delta_{m0} f_{\text{FD}}[E_i(\mathbf{k})]$  holds where  $E_i(\mathbf{k})$  is the energy of *i*th state and  $f_{\text{FD}}$  the Fermi-Dirac distribution. In nonequilibrium, however, the distribution is *nonuniversal* 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.

#### **III. APPLICATION TO A DIRAC BAND**

Now we study the effect of ac fields on the twodimensional Dirac band. The Hamiltonian is  $H(t) = \tau_z v$  $[k^x + A_{ac}^x(t)]\sigma_x + v[k^y + A_{ac}^y(t)]\sigma_y$ , where  $\tau_z = \pm 1$  labels the chirality (K and K' points in graphene), v is the velocity (set to v = 1 hereafter), and  $\sigma_i$  are the Pauli matrices. The circularly polarized field is given as  $(A_{ac}^x, A_{ac}^y) = A(\cos \Omega t, \sin \Omega t)$ 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.<sup>14</sup> With Fourier transformed Floquet states  $|\Phi(t)\rangle = \sum_m e^{-im\Omega t} |u_{\alpha}^m\rangle$  the Floquet equation becomes

$$\sum_{n} H^{mn} |u_{\alpha}^{n}\rangle = (\varepsilon_{\alpha} + m\Omega) |u_{\alpha}^{m}\rangle, \tag{4}$$

where the Floquet Hamiltonian  $H^{mn} = \frac{1}{T} \int_0^T dt H(t) e^{i(m-n)\Omega t}$  has  $H^{mm} = \begin{pmatrix} 0 & k_x - ik_y \\ k_x + ik_y & 0 \end{pmatrix}$  for the diagonal components, whereas the off-diagonal components depend on  $\tau_z$ , i.e.,  $H^{mm+1} = \begin{pmatrix} 0 & A \\ 0 & 0 \end{pmatrix}$ ,  $H^{mm-1} = \begin{pmatrix} 0 & 0 \\ A & 0 \end{pmatrix}$  for  $\tau_z = 1$  and  $H^{mm+1} = \begin{pmatrix} 0 & 0 \\ -A & 0 \end{pmatrix}$ ,  $H^{mm-1} = \begin{pmatrix} 0 & 0 \\ -A & 0 \end{pmatrix}$ ,  $H^{mm-1} = \begin{pmatrix} 0 & 0 \\ -A & 0 \end{pmatrix}$  for  $\tau_z = -1$ . This defines an eigenvalue problem for a block tridiagonal matrix that can be solved numerically with a truncation at certain |m|.

As an important effect of the ac field, gaps open at  $\omega$ =integer  $\times \Omega/2$  in the quasienergy band structure [Fig. 1(b)], reflecting a Dirac-band analog of the ac-Wannier-Stark ladder, as also seen in the density of states  $A(\omega)$ = $-\frac{1}{\pi}\int dk \Sigma_{\alpha} Im \frac{\langle u_{\alpha}^{0} | u_{\alpha}^{0} \rangle}{\omega - \varepsilon_{\alpha} + i \eta}$  in Fig. 1(c). 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 Ref. 15. 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 timedependent Schrödinger equation at k=0 is

$$\left|\Psi_{\alpha}(\boldsymbol{k}=0,t)\right\rangle \propto e^{-i\varepsilon_{\alpha}t} \begin{pmatrix} 1\\ \frac{\widetilde{\varepsilon}_{i}}{A}e^{i\Omega t} \end{pmatrix},$$
(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

and *m* the Floquet index. The quasienergy is  $\varepsilon_{\alpha} = \tilde{\varepsilon}_i + m\Omega$ with  $\tilde{\varepsilon}_1 = \frac{\sqrt{4A^2 + \Omega^2 + \Omega}}{2}$ ,  $\tilde{\varepsilon}_2 = \frac{-\sqrt{4A^2 + \Omega^2 + \Omega}}{2}$ . 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  $\kappa = \varepsilon_{(1, -1)}$  $= \frac{\sqrt{4A^2 + \Omega^2 - \Omega}}{2}$ . 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 quasienergy is that it is a sum of the dynamical phase and the AA phase, i.e.,

$$\varepsilon_{\alpha} = \langle \langle \Phi_{\alpha} | H(t) | \Phi_{\alpha} \rangle \rangle + \gamma_{\alpha}^{AA} / T, \tag{6}$$

where the AA phase is given by

$$\gamma_{\alpha}^{\text{AA}} \equiv T \langle \langle \Phi_{\alpha} | i \partial_t | \Phi_{\alpha} \rangle \rangle = \pm \pi \{ [4(A/\Omega)^2 + 1]^{-1/2} - 1 \},$$
(7)

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

$$[\nabla_k \times \boldsymbol{\mathcal{A}}_{\alpha}(\boldsymbol{k})]_z \sim \pm \tau_z \frac{1}{2} \kappa (|\boldsymbol{k}|^2 + \kappa^2)^{-3/2}, \qquad (8)$$

where  $\pm$  corresponds to  $\alpha = (1,m), (2,m)$ . In this expression, two geometric quantities appear, where 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  $\tau_z$ , 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 nontrivial curvature, hence to a Hall current.

### IV. 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,  $S = \int_C dt (\mathcal{L}_{graphene} + \mathcal{L}_{mix} + \mathcal{L}_{electrodes}),$ where  $\mathcal{L}_{\text{graphene}} = \sum_{i \neq j} c_i^{\dagger} (i \partial_t - t_{ij} e^{i A_{ij}^{ac}(t)}) c_j$  is the tight-binding model for graphene with a hopping  $t_{ij} = -w$  for nearest neighbors, while  $\mathcal{L}_{\text{mix}} = \sum_{k,r} V_{\text{mix}}^r [(a_k^r)^{\dagger} c_r + \text{H.c.}]$  represents the coupling between the electrodes and graphene, with the spin degrees of freedom ignored. The ac field is introduced by  $A_{ij}^{\mathrm{ac}}(t) = (\mathbf{r}_i - \mathbf{r}_j) \cdot A^{\mathrm{ac}}(t)$  with  $A^{\mathrm{ac}}(t) = (F/\Omega)(\cos \Omega t, \sin \Omega t)$ , where F = eaE is the normalized field strength (a: lattice const.). We assume that the electrodes are described by a fermion operator  $a^r$  ( $r \in \{L, R\}$  labeling the left and right electrodes), for which a Fermi-Dirac distribution  $\langle a^{r'}a^{r}\rangle = f_r$ = $[e^{\beta(\omega-\mu_r)}+1]^{-1}$  is assumed with electrode-dependent chemical potential  $\mu_r$ . They are related to the dc bias V across the electrodes by  $\mu_I = V/2$ ,  $\mu_R = -V/2$ . With 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

$$\begin{pmatrix} G_{k_y}^R(\omega) & G_{k_y}^K(\omega) \\ 0 & G_{k_y}^A(\omega) \end{pmatrix}_{ij;mn}^{-1} = \begin{pmatrix} (\omega + n\Omega + i\eta) \delta_{mn} \delta_{ij} - (\hat{H})_{ij}^{mn}(k_y) & 0 \\ 0 & (\omega + n\Omega - i\eta) \delta_{mn} \delta_{ij} - (\hat{H})_{ij}^{mn}(k_y) \end{pmatrix} \\ + \delta_{i1} \delta_{mn} \begin{pmatrix} i\Gamma_L/2 & -i\Gamma_L[1 - 2f_L(\omega + m\Omega)] \\ 0 & -i\Gamma_L/2 \end{pmatrix} + \delta_{iN} \delta_{mn} \begin{pmatrix} i\Gamma_R/2 & -i\Gamma_R[1 - 2f_R(\omega + m\Omega)] \\ 0 & -i\Gamma_R/2 \end{pmatrix},$$

where  $G^{K,R,A}$  are the Keldysh, retarded, and advanced Green's functions, respectively, and  $\Gamma_r \propto |V_{\text{mix}}^r|^2$  is the imaginary part of the self-energy due to the sample-electrode coupling. The effective Floquet Hamiltonian is defined by  $(\hat{H})^{mn}(k_y) = \frac{1}{T} \int_0^T dt e^{i(m-n)\Omega t} \hat{H}[k_y; A^{\text{ac}}(t)]$ , where the indices i, j are suppressed. The current between sites i, j is determined from the lesser component  $G^{<}$  by

$$\langle J_{ij}^{a}(t) \rangle = -i \frac{e}{\hbar} \frac{1}{N_{k_{y}}} \sum_{k_{y}} \sum_{mn} \int_{0}^{\Omega} \frac{d\omega}{2\pi} e^{-i(m-n)\Omega t}$$

$$\times \left[ J_{ij}(t) (G_{k_{y}}^{<})_{mn;ji}(\omega) + J_{ji}(t) (G_{k_{y}}^{<})_{mn;ij}(\omega) \right], \quad (9)$$

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 (cf. Ref. 16) to relate  $G^{<}$  with  $G^{A}, G^{R}$  obtained by diagonalizing the Floquet Hamiltonian.

In the obtained current distribution [Fig. 2(a)] 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 there should be. This current resembles the orbital magnetism which was predicted to arise when a perturbation induces a gap in the Dirac cone.<sup>17,18</sup> Indeed, the circularly polarized light in the present case opens a gap around the band crossing [arrow in Fig. 3(b)], 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 *photoinduced dc Hall current* as well as a longitudinal ( $||x\rangle$  current [Fig. 2(b)]. The Hall current is naturally inverted when the right circularly polarized field  $A_{ac} \propto (\cos \Omega t, \sin \Omega t)$  is changed into the left polarization

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FIG. 2. (Color online) 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.

 $\propto$ (cos  $\Omega t$ ,  $-\sin \Omega t$ ), or the bias voltage is inverted. The *I*-*V* characteristics are shown in Fig. 2(c) for  $J_x$  and  $J_y$ , the averaged current in the *x* and *y* directions, respectively. The photoinduced net Hall current grows linearly with the bias *V*, but saturates and then decreases 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 this Rapid Communication, 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 increases again (*F* > 0.03w). The decrease can be explained by the gap opening at the Dirac points, while the increase is due to photoassisted





FIG. 3. (Color online) Floquet quasienergy diagram for an armchair (Ref. 21) ribbon in an ac field for (a) F=0.005w, (b) 0.02wwith frequency  $\Omega=0.3w$ . The color coding represents the static weight  $|\langle u_{\alpha}^{0}|u_{\alpha}^{0}\rangle|^{2}$  as in Fig. 1(b).

transport. We note that similar features have been experimentally observed in microwave irradiated carbon nanotubes.<sup>19</sup> 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 [Eq. (8)], and indicates  $J_y \sim \kappa V$ . We note that a similar expression was obtained in the case where the chirality is broken in a static manner.<sup>20</sup>

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 photovoltaic dc Hall current in graphene, despite the absence of a uniform magnetic field. The typical intensity of the laser conceived here,  $F \sim 0.001w$ , corresponds to  $E \sim 10^7$  V/m for photon energy  $\Omega \sim 1$  eV, w=2.7 eV, a=2.6 Å, which should be within the experimental feasibility. Inclusion of dissipation, etc. will be an interesting future problem.

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- <sup>1</sup>Geometric phase in nonequilibrium is evoked in, e.g., the dielectric breakdown of a Mott insulator in T. Oka and H. Aoki, Phys. Rev. Lett. **95**, 137601 (2005).
- <sup>2</sup>D. J. Thouless, Phys. Rev. B 27, 6083 (1983).
- <sup>3</sup>D. J. Thouless *et al.*, Phys. Rev. Lett. **49**, 405 (1982).
- <sup>4</sup>M. V. Berry, Proc. R. Soc. London, Ser. A **392**, 45 (1984).
- <sup>5</sup>Y. Aharonov and J. Anandan, Phys. Rev. Lett. 58, 1593 (1987).
- <sup>6</sup>K. S. Novoselov *et al.*, Proc. Natl. Acad. Sci. U.S.A. **102**, 10451 (2005).
- <sup>7</sup>K. S. Novoselov *et al.*, Science **306**, 666 (2004).
- <sup>8</sup>P. Hänggi, *Quantum Transport and Dissipation* (Wiley, New York, 1988), Chap. 5.
- <sup>9</sup>S. Kohler et al., Phys. Rep. 406, 379 (2005).
- <sup>10</sup>H. Sambe, Phys. Rev. A 7, 2203 (1973).
- <sup>11</sup>U. Peskin and N. Moiseyev, J. Chem. Phys. 99, 4590 (1993).

- <sup>12</sup>H. P. Breuer and M. Holthaus, Phys. Lett. A **140**, 507 (1989).
- <sup>13</sup>M. Torres and A. Kunold, Phys. Rev. B **71**, 115313 (2005).
- <sup>14</sup>D. M. Wolkov, Z. Phys. **94**, 250 (1935).
- <sup>15</sup>S. V. Syzranov, M. V. Fistul, and K. B. Efetov, Phys. Rev. B 78, 045407 (2008).
- <sup>16</sup>A. P. Jauho, N. S. Wingreen, and Y. Meir, Phys. Rev. B **50**, 5528 (1994).
- <sup>17</sup>D. Xiao, J. Shi, and Q. Niu, Phys. Rev. Lett. **95**, 137204 (2005).
- <sup>18</sup>T. Thonhauser et al., Phys. Rev. Lett. **95**, 137205 (2005).
- <sup>19</sup>J. Kim et al., Phys. Rev. B 70, 153402 (2004).
- <sup>20</sup>D. Xiao, W. Yao, and Q. Niu, Phys. Rev. Lett. **99**, 236809 (2007).
- <sup>21</sup>We have also studied zigzag nanoribbons to find that the photovoltaic Hall current behaves similarly, and that the zero-energy zigzag edge modes do not alter the physics.