Photoinduced Valley Hall Effect: Intrinsic or Side-Jump Mechanism?

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Recently, there has started a scientific argument: Which processes underlie the actual, true ground of the valley Hall effect (VHE) in two-dimensional materials? Original VHE emerges in samples with ballistic transport of electrons due to the anomalous velocity terms resulting from the Berry phase effect. However, in disordered samples, there have been suggested alternative mechanisms associated with electron scattering off impurities: (i) the asymmetric electron scattering called the skew-scattering and (ii) the shift of electron wave packet in real space called the side-jump. It has been claimed that the side-jump not only contributes to the VHE but it fully compensates the anomalous terms whatever the drag force is for fundamental reasons, and thus, side-jump (together with skew scattering) becomes the king of the hill. However, this claim is based on equilibrium theories without any external valley-selective optical pumping. It makes the results fundamentally interesting but incomplete and impracticable. We develop the first microscopic theory of the photoinduced VHE using the Keldysh nonequilibrium diagrams technique, and show that the anomalous velocity mechanism is dominating over the side-jump in the vicinity of the interband absorption edge.

Introduction.—The concept of the Hall effect is the emergence of an electric current or other flux of particles in the sample in the direction transverse to both the dragging force and the external magnetic field, which should be finite in order for the effect to take place. If similar phenomena happen in the absence of a magnetic field, they are referred to as the anomalous Hall effects (AHEs) [1]. The eminent examples of the AHE include the Hall effect in magnetic materials (with the built-in sample magnetization), the spin Hall effect, where the Hall effect in magnetic materials (with the built-in sample magnetization), the spin Hall effect, where the Hall effect in magnetic materials (with the built-in sample magnetization), the spin Hall effect, where the Hall effect in magnetic materials (with the built-in sample magnetization), the spin Hall effect, where the Hall effect in magnetic materials (with the built-in sample magnetization), the spin Hall effect, where the Hall effect in magnetic materials (with the built-in sample magnetization), the spin Hall effect, where the Hall effect in magnetic materials (with the built-in sample magnetization), the spin Hall effect, where the Hall effect in magnetic materials (with the built-in sample magnetization), the spin Hall effect, where the Hall effect in magnetic materials (with the built-in sample magnetization), the spin Hall effect, where the Hall effect in magnetic materials (with the built-in sample magnetization). The valley Hall currents that are due to these electrons flow in opposite directions, and being of the same magnitude the currents compensate each other resulting in zero-net VHE current in a sample. To observe a nonzero valley Hall current in actual experiments [3], the sample should be illuminated by an external circularly-polarized electromagnetic field of light. It destroys the time-reversal symmetry and predominantly populates only one of the valleys due to the valley-dependent interband optical selection rules. As the result, the current contributions from nonequivalent valleys do not compensate each other. Hence, it is important to consider nonequilibrium photo-excited electrons since it is them actually contributing to the VHE. This idea has briefly been mentioned in literature [21], however, has not been rigorously studied. In the meantime, in general, the light-induced AHE is now an active field of research [22].

In this Letter, we pose an intriguing question: Do these statements (regarding the partial compensation of the intrinsic contribution) remain valid in the case of optically driven systems based on Dirac materials when circularly polarized light pumps one of the valleys? The answer to this question is of utmost importance not only from the fundamental science viewpoint (since 2D Dirac materials...
are prone to interact with light) but also for the perspective of optoelectronic applications, in particular, in novel van der Waals heterostructures. We consider the intrinsic and side-jump contributions to the valley Hall photocurrent using the nonequilibrium Keldysh diagram technique. Thus we build a microscopic theory of the photoinduced VHE. To investigate the transport properties of nonequilibrium photo-excited electrons and their role in VHE we will assume a small temperature, thus the valence band is fully filled and the conduction band is empty. Then, only the photo-excited electrons contribute to VHE and the possible presence of equilibrium electrons in the valleys does not obscure the phenomena under study.

General theory. — Our 2D system (Fig. 1) is exposed to a circularly-polarized light which results in interband transitions,

\[ \mathbf{A}(t) = \mathbf{A}_c e^{-i\omega t} + \mathbf{A}_v e^{i\omega t}, \]

(1)

thus \( \mathbf{A} = \mathbf{A}(1, i\sigma) \) with \( \sigma = \pm 1 \), and the in-plane alternating drag electric field

\[ \mathbf{A}(t) = \mathbf{A}(e^{-i\Omega t} + e^{i\Omega t}), \]

(2)

where we assumed that the drag field is linearly polarized and thus \( \mathbf{A} \) is real-valued. At the end of the calculations, we will put \( \Omega \to 0 \) to find the static limit, which corresponds to the drag effect. We define coordinates so the drag field is directed along the \( y \) axis, thus our goal is to consider the valley Hall current along \( x \). The full system Hamiltonian reads

\[ H = \frac{\Delta}{2} \vec{s}_z + \mathbf{v} \cdot \mathbf{p} + \gamma \mathbf{v} \cdot \mathbf{A}(t) + \gamma \mathbf{v}_y \mathbf{A}(t), \]

(3)

where \( \Delta \) is the monolayer material bandgap, \( \mathbf{p} = \mathbf{p}(\cos \phi, \sin \phi) \) is the electron momentum, \( \mathbf{v} = v_0 (\eta \hat{s}_x, \hat{s}_y) \) is the velocity, \( \eta = \pm 1 \) is the valley index, and \( \hat{s}_x \), the Pauli matrices with \( \alpha = x, y, z \). The Hamiltonian (3) is written in sub-lattices basis since the honeycomb lattice of a TMDC monolayer can be looked at as two triangle sub-lattices inserted into each other. However, it is instructive and physically transparent to work in the conduction and valence band basis (the \( cv \)-basis in what follows). In our case, the external fields in (3) are uniform in space thus conserving the electron momentum (which, hence, can be considered as a \( c \)-number). To transform into the \( cv \)-basis, we use a unitary operator which depends only on the electron momentum [23],

\[ U = \begin{pmatrix} \cos(\theta/2) & \sin(\theta/2) e^{i\eta\phi} \\ \sin(\theta/2) e^{-i\eta\phi} & -\cos(\theta/2) \end{pmatrix}, \]

(4)

where \( \cos \theta = \Delta/2\epsilon_p, \sin \theta = \eta \nu_0 p/\epsilon_p, \) and \( \epsilon_p = \sqrt{(\Delta/2)^2 + v_0^2 p^2} \approx \Delta/2 + p^2/2m, \) where the electron effective mass is \( m = \Delta/(2v_0^2) \) at small electron momenta, \( \nu_0 p \ll \Delta \). Using \( \mathcal{H} = U^* \mathcal{H} U \), we find

\[ \mathcal{H} = \mathcal{H}_0 + e\mathbf{v} \cdot \mathbf{A}(t) + ev_y \mathbf{A}(t), \]

(5)

where

\[ \mathcal{H}_0 = \begin{pmatrix} \epsilon_c(p) & 0 \\ 0 & -\epsilon_v(p) \end{pmatrix}, \quad \mathbf{v} = \begin{pmatrix} \mathbf{v}_c & \mathbf{v}_v \\ \mathbf{v}_v & -\mathbf{v}_c \end{pmatrix} \]

(6)

are the bare Hamiltonian and the velocity operator in \( cv \)-basis; \( \epsilon_c(p) \equiv \epsilon_p, \epsilon_v(p) = -\epsilon_p \) (we will write just \( \epsilon_c \) and \( \epsilon_v \) in what follows, keeping in mind that they both depend on the absolute value of the momentum; we will also omit \( \hbar \) in the expressions below but restore it in final formulas).

The valley Hall current being the linear response to external drag field \( \mathbf{A} \) reads [24],

\[ j_x(t) = \int \frac{d\omega}{c} Q_{xy}(t,t') \mathbf{A}(t'), \]

(7)

\[ Q_{xy}(t,t') = -i e^2 \text{Tr} [v_x \mathcal{G}(t,t') v_y \mathcal{G}(t,t')], \]

(8)

where \( C \) stands for the Keldysh contour, \( \text{Tr} \) is the trace operator which should be taken over the bands, and

\[ [i\partial_t - \mathcal{H}_0 - e\mathbf{v} \cdot \mathbf{A}(t)] \mathcal{G}(t,t') = \delta(t-t') \]

(9)

defines the matrix Green’s function in the \( cv \)-basis. It should be stressed that this (matrix) Green’s function accounts exactly for the external pumping field. We can also write (7) as

\[ j_x(t) = j_x^{(1)}(\Omega) e^{-i\Omega t} + j_x^{(1)}(-\Omega) e^{i\Omega t}. \]

(10)

The in-plane electric field is \( E_y(t) = -\partial_t \mathbf{A}(t) \), and the current is \( j_x(\Omega) = \mathbf{A}[Q_{xy}(\Omega,\omega) + Q_{xy}(-\Omega,\omega)] \). Also we define \( j_x = \sigma H E_y \). Then, it is possible to express the static (with respect to in-plane electric field \( E_y \)) valley Hall photocurrentivity by the standard formula [24],

\[ \sigma_H(\omega) = \lim_{\Omega \to 0} \frac{Q_{xy}(\Omega,\omega) - Q_{xy}(-\Omega,\omega)}{2i\Omega}. \]

(11)
Since we are interested in resonant processes when the frequency of the external light is close to the bandgap \( \omega \approx \Delta \), we can keep only the interband matrix elements of the velocity and disregard the intraband ones, yielding
\[
\begin{pmatrix}
  i\partial_t - \varepsilon_e & -ev_{cv} \cdot A(t) \\
  -ev_{vc} \cdot A(t) & i\partial_t - \varepsilon_v
\end{pmatrix} G(t, t') = \delta(t - t').
\] (12)

We will also assume that the conduction band is “most of the time” empty, and only the photoinduced electrons take part in the conductivity. This regime is the most interesting for us since electrons find themselves in the c-band due to the optical absorption or scattering on impurities processes only, and we can disregard other sources of conducting electrons (such as the thermal ionization of shallow impurities). In this case, the (vertical) optical transitions occur at very small electron momenta \( p \).

**Intrinsic contribution.**— We find the electron Green’s function as a perturbative expansion up to the second-order with respect to the matrix elements of interband transitions,
\[
G(t, t') = G(t - t') + e \int_C dt_1 G(t - t_1) \nu \cdot A(t_1) \left\{ G(t_1 - t') + e \int_C dt_2 G(t_1 - t_2) \nu \cdot A(t_2) G(t_2 - t') \right\},
\] (13)

where in the matrix \( \nu \), we leave only interband elements, and \( G(t - t') \) is the free electron Green’s function defined by the equation
\[
\begin{pmatrix}
i\partial_t - \varepsilon_e & 0 \\
0 & i\partial_t - \varepsilon_v
\end{pmatrix} G(t - t') = \delta(t - t'),
\] (14)

which gives the retarded (advanced) Green’s functions \( G^R(A) (\varepsilon) = (\varepsilon - \varepsilon_\alpha \pm i\pi/2)^{-1} \) and the lesser Green’s function \( G^<_{\alpha}(\varepsilon) = f(\varepsilon_\alpha) [G^A(\varepsilon) - G^R(\varepsilon)] \) with \( \tau \) being the electron scattering time on impurities and \( \alpha = c, v \). We have also introduced a phenomenological relaxation time \( \tau \) in the framework of the Born approximation. Here, the scattering time \( \tau \) accounts for only intraband electron-impurity scattering processes.

Substituting Eq. (13) in (8), and making the Fourier transform over time, we keep only \( e^{-i\omega t_1}e^{i\omega t_2} \) and \( e^{i\omega t_1}e^{-i\omega t_2} \)-containing terms. The other terms describe fast \((\pm 2\omega)\)-oscillating currents, which vanish after time averaging. Then, we can find the stationary contribution \( Q_{xy}(\Omega, \omega) \) (see Supplemental Material [25]). It consists of several terms, shown in Fig. 2. For example, the term in Fig. 2(a) reads \( Q_{xy}^{(la)}(\Omega, \omega) = P_{xy}^{(la)}(\Omega, \omega, \sigma) + P_{xy}^{(la)}(\Omega, -\omega, -\sigma) \), where \( \sigma \) is the polarization of light, and
\[
\begin{align*}
\sigma_{\alpha}^{(l)} &= -\eta(\eta + \sigma)^2 \frac{2e^2}{\hbar} \left( \frac{e\nu_0 A}{\hbar\omega} \right)^2 \frac{\tau\Delta}{4\hbar} \frac{\theta\omega - \Delta}{\Delta}. 
\end{align*}
\] (17)

This formula is the first central result of this Letter. Let us return to it a bit later.

**The side-jump contribution.**— In order to calculate the conductivity due to the side-jump impurity processes, let us, first, introduce the impurity potential in the \( cv \)-basis in the elastic scattering approximation [16],
\[
u(p, p') = u_0(p, p') \left\{ 1 - \sin^2 \left( \frac{\theta}{2} \right) \left( 1 - e^{i\eta(\phi' - \phi)} \right) \right\}
\times \frac{\hat{s}_0 + \hat{s}_z}{2} + \left[ 1 - \cos^2 \left( \frac{\theta}{2} \right) \left( 1 - e^{i\eta(\phi' - \phi)} \right) \right] \frac{\hat{s}_0 - \hat{s}_z}{2}
\times \frac{1}{2} \sin \theta(1 - e^{i\eta(\phi' - \phi)}) \hat{s}_x,
\] (18)

where \( \hat{s}_0 \) is the unity matrix, the angle \( \theta \) corresponds to the momentum \( p; \phi \) and \( \phi' \) are the angles corresponding

![FIG. 2. Feynman diagrams for the intrinsic contribution to the photoinduced Hall electric conductivity \( \sigma_H = \sigma_{xy} \). Red helixes stand for the external circularly polarized light \( A(t) \); \( v_x \) and \( v_y \) are the velocity vertices; \( c \) and \( v \) mark the Green’s functions of electrons in conduction and valence bands.](image-url)
FIG. 3. Examples of Feynman diagrams for the side-jump contribution to conductivity. Blue dashed lines illustrate the impurity scattering processes. Blue vertices $\Upsilon_0$ are renormalized. Other notations are the same as in Fig. 2. In total, there are 120 diagrams of the kind (see Supplemental Material [25]). Some of them give strictly zero contributions. We also neglect some of the remaining diagrams due to their parametric smallness.

to $p$ and $p'$, respectively, $\langle |u|^2(p, p') \rangle^2 = n_i u_0^2$ with $n_i$ being the density of impurities and $m u_0^2 = (n_i \tau)^{-1}$. Following [16], we should, first, find the renormalized vertex (blue-colored in Fig. 3). The integral equation for the renormalized vertex reads

$$\langle \Upsilon_\beta(p) \rangle_{cc} = \langle v_\beta(p) \rangle_{cc}$$

$$+ \frac{d}{(2\pi)^2} \langle \Upsilon_\beta(p') \rangle_{cc} |u_{cc}(p, p')|^2 G_\beta^{R}(p', \varepsilon) G_\alpha^{A}(p, \varepsilon - \Omega),$$

where $\beta = x, y$ and from (18) we find

$$|u_{cc}(p, p')|^2 = n_i u_0^2 \left(1 - \sin^2 \left(\frac{\theta}{2}\right) \left(1 - e^{i\eta(\chi' - \phi)}\right)\right)^2.$$

Since we consider the transitions to the bottom of the conduction band, and thus $p \approx 0$, then we can safely neglect the terms $\sin^2 \left(\frac{\theta}{2}\right) \sim p^2$ as compared to 1. In this case, the integral in (19) equals zero. Hence, $\langle \Upsilon_x \rangle_{cc} \approx \langle v_x \rangle_{cc}$ with a good accuracy, which means that the vertex does not renormalize in the vicinity of the resonance of interband optical transitions.

Thus, the impurity loops only give averages of the kind,

$$u_{cc}(p, p') G_\alpha(p', \delta t) u_{cc}(p', p) \equiv \sum_{p'} \mathcal{F}(p, p') G_\alpha(p', \delta t),$$

$$\approx \frac{1}{m \tau} \int \frac{d p'}{(2\pi \hbar)^2} \left\{ \sin(\theta) - \frac{e^{i\eta(\chi' - \phi)}}{2} \right\} G_\alpha(p', \delta t),$$

and their complex conjugates. Performing the calculation of the diagrams depicted in Fig. 3 and similar (60 in total), we find (see Supplemental Material [25]) that each of the diagrams either gives zero contribution or proportional to $\omega - \Delta$:

$$\sigma_H^{(SI)} \propto \eta(\eta + \sigma)^2 \frac{2e^2}{\hbar} \left(\frac{e v_0 A}{\hbar \omega}\right)^2 \frac{\tau(h\omega - \Delta)}{4\hbar} \theta(h\omega - \Delta).$$

This formula is the second central result of this Letter.

Results and discussion.— Let us first discuss the significant distinctions between the equilibrium and nonequilibrium intrinsic contributions to VHE. The former is dissipation free. It does not depend on the electron relaxation time, and may exist in samples with ballistic transport of electrons [27–29]. In contrast, the nonequilibrium intrinsic contribution (17) is proportional to the electron relaxation time, and thus it is finite only in disordered samples. This is a consequence of the distinction between the equilibrium and nonequilibrium electron distribution functions. Indeed, the distribution function of photoexcited electrons satisfies the Boltzmann equation in the form, $\frac{\partial f_p}{\partial t} = g_p - (f_p - f_p^{(0)}/\tau)$, where $g_p$ is the probability of electron generation due to interband transitions, and the second term in the r.h.s. describes the relaxation processes given by the collision integral which we write here using the relaxation time approximation. In the steady state, and assuming that the conduction band is initially empty, $\frac{\partial f_p}{\partial t} = 0$ and $f_p^{(0)} = 0$. Then, the stationary but nonequilibrium distribution of photoexcited electrons reads $f_p = g_p - \frac{\tau}{\eta}$. Evidently, it depends on $\tau$ thus expressing the equality of generation and relaxation processes in the steady state. Furthermore, taking this distribution function and the expression for the anomalous velocity of electrons, and using the standard formula for the electric current density, we restore the result (17) (see Supplemental Material [29]).

Next, let us compare the Hall photoconductivities (17) and (22) starting with the similarities. First, they both are proportional to the valley- and polarization-dependent coefficient $\eta(\eta + \sigma)^2$. Here $\eta = \pm 1$ dictates the direction of propagation of the Hall electric current, and $(\eta + \sigma)^2$ reflects the interband optical selection rules for 2D materials. Second, the conductivity is proportional to the conductance quantum $2e^2/\hbar$ (which also defines the dimensionality of each term in $\sigma_H$). Third, both the contributions are proportional to the intensity of external light $\sim A^2/\omega^2$, and the theta-function reflects the threshold-like behavior of the photo-absorption processes.

Let us now address the differences between the nonequilibrium intrinsic and side-jump terms. All the Feynman diagrams of the nonequilibrium side-jump effect can be split into two principally different kinds. In the diagrams of the first kind, the block related to the interband transitions (two spiral red lines in our illustrations) does not intersect with the block related to the scattering on impurities (dotted blue line, see a typical example in Fig. 3(a)). The contribution of such diagrams represents a product of probabilities of the interband generation and the side-jump scattering terms. Furthermore, the generation probability contains the energy conserva-
tion term, \( g_p \sim \delta(\omega - 2\epsilon_p) \). As a result, we find
\[
\sigma^{(SJ)}_H \propto \int dp \ p \sin^2(\theta) \delta(\omega - 2\epsilon_p) \propto (\omega - \Delta)(\omega - \Delta),
\]
where the factor \( \omega - \Delta \) results from the \( p \)-integration with account of \( \sin^2(\theta) \sim p^2 \). One of these splines emerges due to the vertex \( \langle Y_x(Y) \rangle \), and the other sine is coming from the impurity related term \( (21) \). Thus, the side-jump contribution gives a negligibly small impact close to the interband resonance, \( 0 < \omega - \Delta \ll \Delta \). This is in striking difference with the equilibrium conditions where the side-jump terms are dominating \( (17) \).

In the diagrams of the second kind, one (or more) of the light (red spiral) lines is under the impurity scattering (dotted) line, as it is shown in Fig. 3(b). Such terms describe the interference of interband and impurity scattering processes. These are purely quantum processes, which cannot be treated by semiclassical arguments and require a careful microscopic analysis (see Supplemental Material \( (25) \)). These diagrams also give the same factor \( \omega - \Delta \).

We see, that in the nonequilibrium situation of the interband valley-selective pumping, the intrinsic contribution \( (17) \) at the absorption edge is proportional to the large factor \( \tau\Delta \gg 1 \), whereas the side-jump term is negligibly small, and they relate as \( \sigma^{(SJ)} / \sigma^{(I)} \sim (\omega - \Delta) / \Delta \ll 1 \). It is important to note that this relation is universal and does not depend on the purity of the sample.

We should also comment on why we disregard the skew-scattering in this Letter. As it was reported in experimental works on the photoinduced VHE in MoS\(_2\), the skew-scattering can only be considerable in high-mobility samples \( (1, 3) \). Nevertheless, in other materials the skew-scattering contribution in the nonequilibrium situation might be important but we leave this question for future study.

In conclusion, we have developed a microscopic theory of the photoinduced valley Hall effect in two-dimensional Dirac materials employing the Keldysh nonequilibrium diagrams technique. We have demonstrated, that the main contribution to the Hall photoconductivity is stemming from the anomalous velocity terms (also called the intrinsic terms) while the side-jump contribution vanishes under the resonant light absorption conditions since it turns out proportional to the detuning between the frequency of external light and the bandgap.

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