Electron diffusion induced valley Hall effect and nonlinear galvanodiffusive transport in hexagonal two-dimensional Dirac monolayer materials

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Diffusion currents are theoretically examined in two-dimensional Dirac materials, such as those of the transition metal dichalcogenides (TMD) family. The transversal effects are analogues of the valley Hall (VHE) and photogalvanic (PGE) transport phenomena in cases when the electron driving force is not an electric field but a gradient of electron density distribution in the sample. The latter can be created by a finite-sized laser spot or by the injection of electrons from other materials. We develop the theory of the diffusive VHE effect assuming the anisotropic electron-short-range-impurity skew scattering. The electron PGE-like transport caused by higher electron-density derivatives is analyzed assuming the trigonal warping anisotropy of electron valleys in a TMD monolayer. The nonlinear responses on an electron-density gradient are studied as well. The isotropic processes of electron scattering off the short-range and Coulomb centers are taken into account in the PGE-like transport theory.

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I. INTRODUCTION

Hexagonal two-dimensional monolayer structures, in particular, graphene and transition metal dicholcogenides (TMD) [1], are under active study nowadays. The research of their physical properties occurs in two main directions: optical properties [2,3] and transport phenomena [4–7]. Exceptional optical properties of TMD based on a specific structure of excitons and exciton-polaritons in TMDs have wide perspectives in optoelectronics [2].

The transport properties of monolayer TMD materials also demonstrate specific features due to their unique band structure [8,9]. TMD monolayers have the hexagonal lattice structure belonging to the D_{3h} point group producing the two-valley structure of their Brillouin zone having a strong valence band spin-orbit splitting. The specific optical interband selection rules, allowing for the driving of the valley degrees of freedom of the charge carriers producing very specific transport properties of these materials and the corresponding research direction called *valleytronics* [10] in modern literature have recently arisen.

The transport response of TMD materials under study can be split phenomenologically by the amplitude $\mathbf{E}$ of external electromagnetic wave into a linear and second order current density response. The linear transport phenomena, being first order with respect to $\mathbf{E}$, obey the relation $j_\alpha = \sigma_{\alpha\beta}E_\beta$, where j_α are current density components and $\sigma_{\alpha\beta}$ are kinetic coefficients being the components of generalized conductivity tensor, and α, β run in-plain coordinates x, y. The most intriguing linear transport phenomena in TMDs is the existence of valley Hall effect (VHE) [11–14]: the transverse current density rose as a linear response to the in-plane static electric field $\mathbf{E}$ and was determined by the nondiagonal components of conductivity tensor as $\sigma_H = \sigma_{xy} = \sigma_{yx}$. The specific feature of VHE is its valley-selective nature: the transverse VHE current densities flow in opposite directions in different valleys in such a way that the net transverse current, being summed over two valleys, vanishes. This is due to the time-reversal symmetry by which the valleys are coupled. The net VHE current can be viewed when the time-reversal symmetry is destroyed. The latter can be done either by the sample illumination by the circularly polarized electromagnetic field or by the external magnetic field. The experimental observation of VHE in a TMD-based transistor structure was made by the first method [13]. The microscopic mechanisms producing the VHE transport were intensively studied theoretically. Initially, VHE was predicted in Ref. [11], where the Berry-phase-induced anomalous contribution to the electron velocity was considered as the underlying microscopic mechanism of VHE [12]. Later on, the key role of anomalous electron scattering processes off the impurity potential in the theoretical understanding of VHE was recognized [15–17].

Recently, a detailed theoretical description for VHE was developed in Refs. [18,19] considering also the skew and side-jump electron scattering off the impurities in dirty samples. The authors considered also different electrons driving mechanisms, the standard one, due to the in-plane static electric field $\mathbf{E}$, and also due to the photon [20] and phonon electron dragging. It was shown that the dominant microscopic mechanism of VHE in disordered samples is determined by the skew-electron scattering off impurities. Later, the theoretical description of nonequilibrium VHE, including both impurity scattering and the interband valley-selective illumination, was developed [21]. The regime of strong interband

transitions, in the case of the Berry-phase-induced anomalous velocity VHE mechanism, was also analyzed [22]. It was shown that, in these cases, the skew-related electron scattering gives the dominant contribution to VHE, which made it possible to explain some observations of the VHE transport experiment [13].

The other transport phenomenon actively studied in TMD materials is the nonlinear effect, say, the photogalvanic effect (PGE) [23–26]. The PGE is the appearance of stationary (and also uniform in space) current density due to the external alternating electromagnetic field with a normal incidence to the TMD plane and, phenomenologically, can be written as $j_\alpha = \chi_{\alpha\beta\gamma} E_\beta E^*_\gamma$. The PGE effect is well known in 3D semiconductor physics [23,24] and it was well understood in conventional semiconductors [26–28]. In TMD materials, PGE may arise both due to the trigonal symmetry of electron valleys or also due to anomalous velocity (Berry-phase) and electron-impurity scattering. The PGE in TMD monolayers also has the valley-selective nature: the PGE current flow in opposite directions in different valleys [29,30]. Destroying the time-reversal symmetry produces the nonzero net PGE current in the sample [29,30].

In all transport effects discussed above, the driving force was the electric field: it is stationary in the plane field or an alternative electric field of electromagnetic wave. From the general point of view [31], any factor, which drives the system from equilibrium state, may produce the current. Any such a factor, called generalized force in statistical physics, may be a scalar, vector or even a tensor. As a vector, a generalized force may be an electric field, a temperature gradient, and particle density gradients, whereas a scalar generalized force may be a temperature difference [32] or particle concentration difference between different subsystems or their time derivatives. The tensor generalized force may be constructed from the second spatial derivatives of the scalar quantities, etc. Besides, the current can be excited by the higher orders of the vector forces together with their cross products. A general expression for currents caused by scalar F, vector F_j, and tensor F_{ij} forces is

$$j_i = \gamma_i^{(0)} F + \gamma_{ij}^{(1)} \partial_j F + \gamma_{ij}^{(2)} F_j + \gamma_{ijk}^{(3)} F_j F_k + \gamma_{ijk}^{(4)} F_{jk} + \dots$$

Besides, the cross terms due to different forces are possible in higher orders of generalized forces. The symmetry of the system restricts the possibility of coefficients $\gamma_i^{(0)}$, $\gamma_{ij}^{(1)}$, $\gamma_{ijk}^{(3)}$, and $\gamma_{ijk}^{(4)}$. The quantity $\gamma_i^{(0)}$ exists in pyroelectrics, the tensors $\gamma_{ijk}^{(3)} \neq 0$ or $\gamma_{ijk}^{(4)} \neq 0$ demand the absence of reflection symmetry.

In case of vector generalized force, the general expression for the current density may be written as

$$j_i = \alpha_{ij} F_j + \beta_{ijk} F_j F_k + \dots$$

The aim of the present paper is the theoretical description of VHE and PGE transport phenomena when electrons drive a vector generalized force given by the nonuniform electron density distribution in the sample, the particle density gradient, $\nabla \cdot n(\mathbf{r})$. The latter can be created by the sample illumination with a finite-in-plane laser spot [13] or by an injection of electrons from other materials [33]. From the phenomenological point of view, the diffusive-induced VHE reads as $j_\alpha(\mathbf{r}) = -eD_{\alpha\beta}\nabla_\beta n(\mathbf{r})$ with generalized diffusive coefficients, $D_{\alpha\beta}$ (e is an electron charge). The VHE will be given by their nondiagonal elements. The PGE-like current can be written as a nonlinear response to the density gradient, $j_\alpha(\mathbf{r}) = \chi_{\alpha\beta\gamma}\nabla_\beta\nabla_\gamma n(\mathbf{r}) + \zeta_{\alpha\beta\gamma}\nabla_\beta n(\mathbf{r})\nabla_\gamma n(\mathbf{r})$.

The paper has the following structure. In the next section, we derive the nondiagonal elements of $D_{\alpha\beta}$, considering the electron-impurity skew scattering as the dominating mechanism. The impurities are considered to be of the short-range type. The later sections are devoted to the derivation of $\chi_{\alpha\beta\gamma}$ and $\zeta_{\alpha\beta\gamma}$ tensors. As the microscopic mechanism of diffusive PGE-like transport, we consider the trigonal warping of the electron valley dispersion in TMDs; both short-range and Coulomb impurities are analyzed in that sections.

II. VALLEY HALL EFFECT DUE TO ELECTRON DIFFUSION

Here we consider the VHE effect due to the electron diffusion based upon the Boltzmann transport equation approach. We assume that the nonuniform electron density, which can be excited by the external electromagnetic field (or injected to the sample), acquires the fast energy relaxation and the resulting electron density is characterized by the quasiequilibrium Fermi distribution function with a chemical potential being the arbitrary function of coordinates. It reads $f(\mathbf{r}) = (1 + \exp[\varepsilon(\mathbf{p}) - \mu(\mathbf{r})]/T)^{-1}$, where $\varepsilon(\mathbf{p})$ is an electron dispersion in the given valley and $\mu(\mathbf{r})$ is an electron chemical potential. Here and below we measure the temperature in energy units, taking the Boltzmann factor $k_B = 1$. We assume that the spatial nonuniformity of the electron distribution function (and corresponding electron density $n(\mathbf{r})$) is weak in comparison with uniform electron density N in the sample, $|n(\mathbf{r}) - N| \ll N$. We assume also that uniform distribution function $f_0 = (1 + \exp[\varepsilon(\mathbf{p}) - \mu_0]/T)^{-1}$ corresponds to the uniform electron density value N in the sample. The Boltzmann transport equation in general form reads

$$\frac{\partial f}{\partial t} + \mathbf{v}\frac{\partial f}{\partial \mathbf{r}} + \mathbf{F}\frac{\partial f}{\partial \mathbf{p}} + Q\{f\} = 0, \tag{1}$$

where $\mathbf{v}$ is an electron velocity in a given valley, $\mathbf{F}$ is a possible external field, and Q is an electron-impurity collision integral. We further analyze the static ($\partial_t f = 0$) nonuniform electron density distribution in the absence of external fields $\mathbf{F} = 0$ in the sample. The nonuniform electric field will be considered later on.

The diffusion-like VHE current arises due to the nonuniform electron distribution and is determined by the first order correction $f^{(1)}(\mathbf{r})$ to the electron distribution function, with respect to gradients, satisfying the following Boltzmann equation:

$$\mathbf{v}\frac{\partial f(\mathbf{r})}{\partial \mathbf{r}} + Q\{f^{(1)}(\mathbf{r})\} = 0. \tag{2}$$

Further in this section we derive all expressions for one given valley. The electron dispersion is assumed in this section to have the parabolic form $\varepsilon(\mathbf{p}) = p^2/2m$, and $\mathbf{v} = \mathbf{p}/m$, respectively. Collision integral Q consists of two terms describing the isotropic Q^s and anisotropic Q^a electron-impurity scattering. The isotropic scattering is approximated by the

relaxation-time approach $Q\{f^{(1)}(\mathbf{r})\} = f^{(1)}(\mathbf{r})/\tau$. For simplicity, we set τ to be independent of the electron energy. The first order correction to the distribution function can be also split into symmetric and antisymmetric contributions, $f^{(1)} = f_s^{(1)} + f_a^{(1)}$, with respect to the electron momentum. The VHE current density is expressed via the antisymmetric part as

$$j_\alpha = e\int \frac{d^2\mathbf{p}}{(2\pi\hbar)^2} v_\alpha f_a^{(1)}. \tag{3}$$

Assuming the anisotropic scattering to be weak, the antisymmetric contribution to the electron distribution function can be found by a successive approximation. Thus we have the following set of equations:

$$\mathbf{v}\frac{\partial f_0}{\partial \mathbf{r}} + \frac{f_s^{(1)}}{\tau} = 0 \tag{4}$$

for the symmetric part and

$$\frac{f_a^{(1)}}{\tau} + Q^a\{f_s^{(1)}\} = 0 \tag{5}$$

for the antisymmetric one. Solving these equations, we find

$$j_\alpha(\mathbf{r}) = e\tau^2 \int \frac{d^2\mathbf{p}}{(2\pi\hbar)^2} v_\alpha Q^a\{v_\beta \nabla_\beta f(\mathbf{r})\}. \tag{6}$$

An asymmetric part of the collision integral responsible for the skew scattering, Q^a, for a TMD monolayer was found in Ref. [18] and reads

$$Q^a\{F(\mathbf{p})\} = W_0 \sum_{\mathbf{p}'} F(\mathbf{p}')[\mathbf{p}\times\mathbf{p}']_z \delta(\varepsilon_\mathbf{p} - \varepsilon_{\mathbf{p}'}), \tag{7}$$

where $F(\mathbf{p})$ is an arbitrary function of electron momentum. Parameter $W_0 = 2\pi u_0 v^2/\tau\Delta^2$ is expressed via the short-range impurity potential amplitude modelled by a Dirac delta function as $U(\mathbf{r}) = u_0\delta(\mathbf{r})$, v is the TMD monolayer band parameter having the velocity dimensionality, Δ is the TMD band gap and τ is the electron momentum relaxation time due to the electron-impurity scattering. A direct analysis based upon Eqs. (6) and (7) yields

$$j_y = 2e\tau^2 W_0 \rho N\langle\varepsilon\rangle\left(-\frac{\partial\mu}{\partial x}\right), \tag{8}$$

where $\rho = m/2\pi\hbar^2$ is an electron density of states, N is an equilibrium electron density, and

$$\langle\varepsilon\rangle = \frac{1}{N}\int_0^\infty d\varepsilon\rho\varepsilon f_0 \tag{9}$$

is an average electron energy. Taking into account the relation between density and chemical potential gradients

$$\nabla n(\mathbf{r}) = \nabla\mu(\mathbf{r})\int \frac{d^2\mathbf{p}}{(2\pi\hbar)^2}(-f_0') = \rho\left(1 - e^{-\frac{N}{\rho T}}\right)\nabla\mu(\mathbf{r}), \tag{10}$$

one finds a VHE diffusive coefficient

$$D_{yx} = \frac{2\tau^2 W_0 N\langle\varepsilon\rangle}{1 - e^{-\frac{N}{\rho T}}} = \frac{4\pi v^2\tau}{1 - e^{-\frac{N}{\rho T}}}\frac{N u_0\langle\varepsilon\rangle}{\Delta^2}. \tag{11}$$

In Eq. (10), we used the following approximation $\partial_\mu f(\mathbf{r}) = -f'(\mathbf{r}) \approx -f_0'$, where prime means derivative, with respect to the electron energy. A found expression Eq. (11) holds for a given valley; in the other valley, it has opposite sign.

III. GENERAL EXPRESSIONS FOR PGE-LIKE DIFFUSION CURRENTS

PGE-like current density, nonlinear with respect to electron density gradients, is phenomenologically expressed as

$$j_\alpha(\mathbf{r}) = \chi_{\alpha\beta\gamma}\nabla_\beta\nabla_\gamma n(\mathbf{r}) + \zeta_{\alpha\beta\gamma}\nabla_\beta n(\mathbf{r})\nabla_\gamma n(\mathbf{r}). \tag{12}$$

One can see, the current density is determined by the third-order tensors and, thus, it may occur in noncentrosymmetric systems. The TMD monolayer structures are described by the D_{3h} point group which is of noncentrosymmetric class. In the D_{3h} point group, the nonzero elements of any third-order tensor read

$$-\chi_{xxx} = \chi_{xyy} = \chi_{yxy} = \chi_{yyx}, \tag{13}$$

whereas other component are zero. The same relation holds for the $\zeta_{\alpha\beta\gamma}$ tensor. Thus it is enough to consider χ_{xxx} and ζ_{xxx} components only. We are interested in the PGE effect produced by the warping of the electron spectrum in TMD monolayers. Within the two-band model of electron dispersion of a TMD monolayer, the bare Hamiltonian accounting for the trigonal warping of electron valleys reads

$$H_0 = \frac{\Delta}{2}\sigma_z + v(\eta\sigma_x p_x + \sigma_y p_y) + \begin{pmatrix} 0 & \mu p_+^2 \\ \mu p_-^2 & 0 \end{pmatrix}, \tag{14}$$

where Δ is the TMD material bandgap, $v = p_{cv}/m_0$, μ is the warping strength band parameter, $p_\pm = p_x \pm ip_y$ is an electron momentum and $\eta - \pm 1$ is a valley index. In the effective mass approximation, the conduction band electron dispersion near K and $-K$ points of the Brillouin zone can be approximated as

$$\varepsilon_\mathbf{p} = \varepsilon_\mathbf{p}^0 + w_\mathbf{p},$$
$$\varepsilon_\mathbf{p}^0 = \frac{\mathbf{p}^2}{2m}, \quad w_\mathbf{p} = \eta w\left(p_x^3 - 3p_x p_y^2\right), \tag{15}$$

where $w_\mathbf{p}$ is a warping correction to the electron dispersion in the ηth valley, where its strength is $w \sim \mu$. It should be emphasized that the cubic-in-momentum $w_\mathbf{p}$ results from the quadratic terms in the Hamiltonian, Eq. (14), see Ref. [8]. The electron distribution function now has to be found up to the second order, with respect to electron density gradients. Thus the simple analysis of Boltzmann equation results in the PGE-like current density expression

$$j_\alpha = e\int \frac{d\mathbf{p}}{(2\pi\hbar)^2} v_\alpha \hat{Q}^{-1}(\mathbf{v}\nabla)\hat{Q}^{-1}(\mathbf{v}\nabla) f^w(\mathbf{r}), \tag{16}$$

where quasiequilibrium distribution function $f^w(\mathbf{r})$ contains now the electron dispersion with a warping correction, Eq. (15). The analytical theory can be developed assuming the smallness of the warping term $w_\mathbf{p}$ in the electron dispersion of Eq. (15). Thus we will find the current by Eq. (16) in the first order with respect to $w_\mathbf{p}$. The structure of Eq. (16) dictates that the warping correction may come from (i) the electron dispersion in distribution function $f^w(\mathbf{r})$, (ii) electron velocity

v_α and, finally, (iii) from the structure of collision operator $\hat{Q}$ [29]. The energy dispersion acquiring the warping correction in the first order is given by Eq. (15), the structure of the electron velocity with the first order warping correction can be easily found as

$$\mathbf{v} = \mathbf{v}^0 + \delta\mathbf{v},$$
$$\mathbf{v}^0 = \frac{\mathbf{p}}{m}, \quad \delta\mathbf{v} = 3\eta w\left(p_x^2 - p_y^2, -2p_x p_y\right), \tag{17}$$

whereas the correction to the collision operator caused by the warping term requires the careful analysis made below.

Now consider the warping correction to the collision operator $\hat{Q}$. An electron-impurity collision operator acting to the arbitrary function $\chi_{\mathbf{p}}$ in the lowest Born approximation yields

$$\hat{Q}\{\chi_{\mathbf{p}}\} = \frac{2\pi n_i}{\hbar}\sum_{\mathbf{p}'}|M_{\mathbf{p}\mathbf{p}'}|^2\delta(\varepsilon_{\mathbf{p}} - \varepsilon_{\mathbf{p}'})(\chi_{\mathbf{p}} - \chi_{\mathbf{p}'}), \tag{18}$$

where n_i is a short-range impurities density, and $M_{\mathbf{p}\mathbf{p}'}$ is a scattering matrix element of impurity potential. Following Ref. [29], we neglect here the warping-induced corrections to the scattering matrix elements, and thus, $\hat{Q}$ acquires the correction due to the presence of electron dispersion $\varepsilon_{\mathbf{p}}$ in the energy conservation law at the electron-impurity scattering. Thus one finds $\hat{Q}_0 + \hat{Q}_w$, where the action of bare collision operator $\hat{Q}_0$ on the nth harmonic of the distribution function reads

$$\lim_{\alpha\to 0}\left(\alpha + \hat{Q}_0\right)^{-1} e^{in\varphi} = e^{in\varphi}\lim_{\alpha\to 0}(\alpha + 1/\tau_n)^{-1},$$
$$\frac{1}{\tau_n} = \frac{2\pi n_i}{\hbar}\sum_{\mathbf{p}'}|M_{\mathbf{p}\mathbf{p}'}|^2\delta\left(\varepsilon^0_{\mathbf{p}} - \varepsilon^0_{\mathbf{p}'}\right) \times (1 - \cos n\theta), \tag{19}$$

whereas the action of $\hat{Q}_w$ is determined as

$$\hat{Q}_w\{\chi_{\mathbf{p}}\} = \frac{2\pi n_i}{\hbar}\sum_{\mathbf{p}'}|M_{\mathbf{p}\mathbf{p}'}|^2(w_{\mathbf{p}} - w_{\mathbf{p}'}) \times \delta'\left(\varepsilon^0_{\mathbf{p}} - \varepsilon^0_{\mathbf{p}'}\right)(\chi_{\mathbf{p}} - \chi_{\mathbf{p}'}). \tag{20}$$

Here prime means derivative, with respect to delta-function argument. Now express the inverse collision operator up to the first order with respect to the warping term as

$$(\hat{Q}_0 + \hat{Q}_w)^{-1} \approx \hat{Q}_0^{-1} - \hat{Q}_0^{-1}\hat{Q}_w\hat{Q}_0^{-1}, \tag{21}$$

the j_x PGE-like current density expression can be split into three contributions

$$j_x^{\mathrm{I}} = e\int\frac{d\mathbf{p}}{(2\pi\hbar)^2}v_x\hat{Q}_0^{-1}(v_x\partial_x)\hat{Q}_0^{-1}(v_x\partial_x)f^w(\mathbf{r}),$$
$$j_x^{\mathrm{II}} = -e\int\frac{d\mathbf{p}}{(2\pi\hbar)^2}v_x\hat{Q}_0^{-1}\hat{Q}_w\hat{Q}_0^{-1}(v_x\partial_x)\hat{Q}_0^{-1}(v_x\partial_x)f^w(\mathbf{r}),$$
$$j_x^{\mathrm{III}} = -e\int\frac{d\mathbf{p}}{(2\pi\hbar)^2}v_x\hat{Q}_0^{-1}(v_x\partial_x)\hat{Q}_0^{-1}\hat{Q}_w\hat{Q}_0^{-1}(v_x\partial_x)f^w(\mathbf{r}). \tag{22}$$

These expressions give the PGE-like current density. Below we analyze these expressions in case of electron scattering off the neutral short-range and charged Coulomb impurities, respectively.

A. PGE-like current density: short-range impurities

It is easy to show that if electrons scattered off the short-range impurity potential, the operator $\hat{Q}_w$ in Eq. (20) acting on the arbitrary function of electron momentum $\chi_{\mathbf{p}}$, gives zero

$$\hat{Q}_w\{\chi_{\mathbf{p}}\} = 0. \tag{23}$$

Thus both j^{II} and j^{III} current contributions in Eq. (A1) vanish. In the remaining term j^{I}, expanding the distribution function up to the first order, with respect to warping, is made with the following equation

$$f^w(\mathbf{r}) = f(\mathbf{r}) - w_{\mathbf{p}}\partial_\mu f(\mathbf{r}), \tag{24}$$

where $f(\mathbf{r})$ is a bare function with isotropic and parabolic $\varepsilon^0_{\mathbf{p}}$ electron dispersions. Keeping only $w_{\mathbf{p}}$-like first order terms, the PGE-like current density can be expressed via chemical potential gradients as

$$j_\alpha = A_{\alpha\beta\gamma}\nabla_\beta\nabla_\gamma\mu(\mathbf{r}) + B_{\alpha\beta\gamma}\nabla_\beta\mu(\mathbf{r})\nabla_\gamma\mu(\mathbf{r}), \tag{25}$$

where (for details, see Appendix)

$$A_{xxx} = e\eta wN\left[6\langle(\epsilon^2\tau_1\tau_2)'\rangle + 3\left\langle\left(\epsilon^2\tau_1^2\right)'\right\rangle - \langle(\epsilon^3\tau_1\tau_2)''\rangle\right],$$
$$B_{xxx} = e\eta wN\left[6\langle(\epsilon^2\tau_1\tau_2)''\rangle + 3\left\langle\left(\epsilon^2\tau_1^2\right)''\right\rangle - \langle(\epsilon^3\tau_1\tau_2)'''\rangle\right], \tag{26}$$

and τ_n is an electron relaxation time of nth harmonics; prime here means a derivative with respect to electron bare energy $\varepsilon \equiv \varepsilon^0_{\mathbf{p}}$, and the spatially uniform energy distribution averaging is determined by Eq. (9).

The relation between tensors $A_{\alpha\beta\gamma}, B_{\alpha\beta\gamma}$ and $\chi_{\alpha\beta\gamma}, \zeta_{\alpha\beta\gamma}$ can be found in the general form for the arbitrary degeneracy of electron gas. It can easily be shown that

$$\chi_{\alpha\beta\gamma} = \frac{A_{\alpha\beta\gamma}}{\sum_{\mathbf{p}}(-f_0')},$$
$$\zeta_{\alpha\beta\gamma} = \frac{B_{\alpha\beta\gamma}\sum_{\mathbf{p}}(-f_0') - A_{\alpha\beta\gamma}\sum_{\mathbf{p}}f_0''}{\left[\sum_{\mathbf{p}}(-f_0')\right]^3}, \tag{27}$$

where

$$\sum_{\mathbf{p}}(-f_0') = \rho\left(1 - e^{-\frac{N}{\rho T}}\right),$$
$$\sum_{\mathbf{p}}f_0'' = \frac{\rho}{T}e^{-\frac{N}{\rho T}}\left(1 - e^{-\frac{N}{\rho T}}\right). \tag{28}$$

In systems where the electron dispersion is quadratic with respect to electron momentum, the relaxation time corresponding to the electron scattering off short-range impurities does not depend on the electron energy, $\tau_1 = \tau_2 = \tau = \text{const}$. Thus Eqs. (27) give

$$\chi_{xxx} = \eta\frac{12ewN\tau^2}{\rho}\frac{\langle\epsilon\rangle}{1 - e^{-\frac{N}{\rho T}}},$$
$$\zeta_{xxx} = \eta\frac{12ewN\tau^2}{\rho^2}\frac{1 - \frac{\langle\epsilon\rangle}{T}e^{-\frac{N}{\rho T}}}{\left(1 - e^{-\frac{N}{\rho T}}\right)^2}. \tag{29}$$

B. PGE-like current density: Coulomb impurities

Consider the PGE-like current in the case of electron scattering off Coulomb impurities. Expression (18) is valid with the matrix element given by the Fourier-transformed 2D Coulomb potential

$$M_{\mathbf{p}\mathbf{p}'} = \frac{2\pi e^2\hbar}{\kappa|\mathbf{p}-\mathbf{p}'|}, \tag{30}$$

where κ is the dielectric permittivity of surrounded media. Expressions (26), corresponding to the j_x^{I} current contribution, still hold with the only difference that now the electron momentum relaxation time Eq. (18) depends on the electron energy

$$\frac{1}{\tau_n} = \frac{|n|}{\tau(\varepsilon_{\mathbf{p}})}, \quad \frac{1}{\tau(\varepsilon_{\mathbf{p}})} = \frac{\pi e^4 n_i^C}{\hbar\kappa^2\varepsilon_{\mathbf{p}}}, \tag{31}$$

where n_i^C is a concentration of Coulomb centers. The other principal difference of the electron scattering off charged Coulomb centers is the nonzero warping-induced corrections to the collision integral Eq. (20) and the corresponding nonzero contributions to the current density given by j_x^{II} and j_x^{III} in Eq. (A1). The direct calculations of these contributions to the current density give (for details, see Appendix)

$$A^C_{xxx} = 12e\eta wN\frac{\kappa^4}{\pi e^8\left(n_i^C\right)^2}\langle\epsilon^3\rangle,$$
$$B^C_{xxx} = 36e\eta wN\frac{\kappa^4}{\pi e^8\left(n_i^C\right)^2}\langle\epsilon^2\rangle. \tag{32}$$

Now calculating expressions (26), accounting for relations (31), and combining the result with contributions (32), one finally finds

$$\chi_{xxx} = \eta\frac{wNk^4\hbar^2}{\pi^2e^7\left(n_i^C\right)^2}\frac{\langle\epsilon^3\rangle(14+12\pi)}{\rho\left(1-e^{-\frac{N}{\rho T}}\right)},$$
$$\zeta_{xxx} = \eta\frac{wNk^4\hbar^2}{\pi^2e^7\left(n_i^C\right)^2}\frac{\langle\epsilon^2\rangle(42+36\pi)-(14+12\pi)\frac{\langle\epsilon^3\rangle}{T}e^{-\frac{N}{\rho T}}}{\rho^2\left(1-e^{-\frac{N}{\rho T}}\right)^2}. \tag{33}$$

IV. NONUNIFORM ELECTRIC FIELD

Now consider the case when a nonuniform static electric field characterized by the scalar potential $\phi(\mathbf{r})$ is present in the sample. In equilibrium, the electrochemical potential $\mu(\mathbf{r})+e\phi(\mathbf{r})$ of the system must be constant and the current density should vanish. Thus the current should depend on the spatial derivatives of $\mu(\mathbf{r})+e\phi(\mathbf{r})$ only. Expanding the current with respect to the derivatives of electrochemical potential and its second-order powers, one obtains

$$j_i = a_{ij}\nabla_j(\mu+e\phi) + b_{ijk}\nabla_j(\mu+e\phi)\nabla_k(\mu+e\phi) + c_{ijk}\nabla_j\nabla_k(\mu+e\phi). \tag{34}$$

The equality to zero of the total current in equilibrium follows from these relations at $\mu(\mathbf{r})+e\phi(\mathbf{r}) = \text{const}$. Hence, the system responses to the chemical potential $\mu(\mathbf{r})$ and to the scalar potential $e\phi(\mathbf{r})$ are determined by the components of *the same* tensors. Let us consider this statement for the linear and nonlinear responses in details.

First consider the linear response, given by a_{ij} tensor. If the electric field $-\partial_x\phi$ and chemical potential gradient $\partial_x n$ are directed along the x axis, then the nondiagonal components $a_{xy} = a_{yx} = a_H$ desribe the VHE effect. The VHE transport being the response to the external static electric field, $j_y = \sigma_H E_x$, was studied in [18]. Coefficient a_H can be directly found by a comparison with expression Eq. (8). From Eq. (34), one finds the relation between kinetic coefficients describing the VHE under the electric field and chemical potential gradient as $e\,a_H = -\sigma_H$. The direct comparison of our result Eq. (8) and σ_H expression derived in Ref. [18] supports this relation as it should be. The relation between the linear response coefficients in the case of the external potential force and chemical potential gradient being applied to the VHE effect was proven for the excitonic VHE in Ref. [34].

Consider now the nonlinear transport given by the tensors b_{ijk} and c_{ijk} in Eq. (34). The tensor b_{ijk} determines the PGE effect and it is known for PGE caused by the electron valley traigonal warping mechanism [29] and it should coincide with the tensor B_{ijk} derived above, see Eq. (25). Let us prove this equality directly for the simple model of $\tau = \text{const}$ momentum relaxation time. If $\phi = 0$, substituting $\tau_1 = \tau_2 = \tau = \text{const}$ into Eq. (26), one finds the B_{xxx}-related current for a given valley

$$j_x = 12e\tau^2Nw(\nabla_x\mu)^2 \tag{35}$$

as a nonlinear response to the generalized force, $\nabla_x\mu$. It is constructive to compare this relation with the PGE current being the response to the electric field $j_x \propto (-e\nabla_x\phi)^2 = (eE_x)^2$. It can be found via the Boltzmann equation [35] and reads

$$j_x = 2e\tau^2(eE_x)^2\int\frac{d\mathbf{p}}{(2\pi)^2}\frac{\partial^2 v_x}{\partial p_x^2}f_0. \tag{36}$$

Taking into account the velocity expression, Eq. (17), we find the current for a given valley in the form $j_x = 12e\tau^2Nw(eE_x)^2$, with the same coefficient as in Eq. (35) as it should be from the symmetry arguments.

The remaining tensor c_{ijk} components determine the unconventional PGE effect (with respect to the nonuniform electric field, $\nabla_j\nabla_k\phi(\mathbf{r}) = -\nabla_jE_k(\mathbf{r})$) in TMD materials. The corresponding tensor components are not known in literature, to the best of our knowledge.

V. ANISOTROPIC DIFFUSION

To illustrate the effect of c_{ijk} tensor components, consider the interband illumination of the TMD monolayer by an axially symmetric light source with circular polarization and study the charge spreading of photoexcited electrons. The light is absorbed in a single valley. In the approximation taking into account only ordinary diffusion, the diffusion spot will stay axially symmetric also. However, the inclusion of higher derivatives with respect to the electron density given by c_{ijk} tensor will result in the distortion of the spot. In our case, this distortion should be triangular, in accordance with the trigonal system symmetry. The luminescence from the illuminated spot due to the interband electron recombination should also

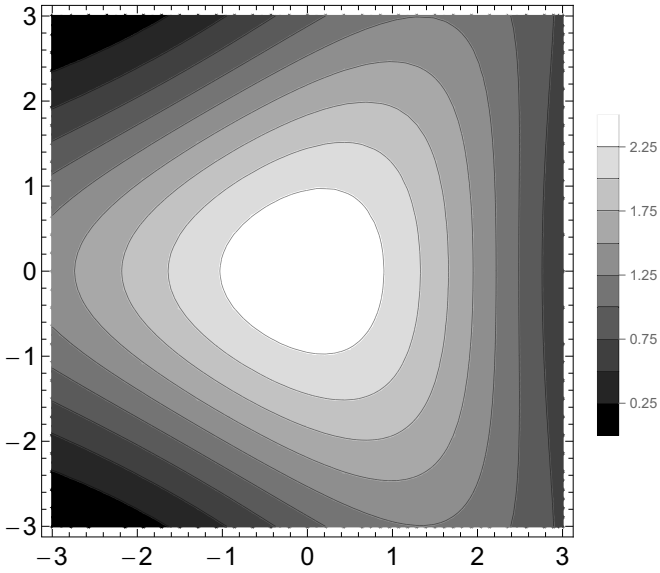

FIG. 1. The density plot of electron density distribution around spot illuminated by circular-polarized light. The length unite is χ/D. The distribution symmetry reflects the C_3 symmetry of the active valley.

repeat the trigonal symmetry diffusing electron spot. Let the TMD plane is illuminated locally creating the initial electron density distribution $n_0(\mathbf{r}, t)$, which reads

$$n_0(\mathbf{r}, t) = \begin{cases} n_1\delta(\mathbf{r}), & \text{for stationary illumination;} \\ n_2\delta(\mathbf{r})\delta(t), & \text{for flash illumination.} \end{cases} \quad (37)$$

Then, the spatial and temporary distribution of diffusing electrons yields

$$n(\mathbf{r}, t) = \int d\mathbf{r}' \int dt' g(\mathbf{r} - \mathbf{r}'; t - t') n_0(\mathbf{r}', t'), \quad (38)$$

where the Green function satisfy the equation

$$\left[\frac{\partial}{\partial t} - D\nabla^2 - \chi\frac{\partial}{\partial x}\left(\frac{\partial^2}{\partial x^2} - 3\frac{\partial^2}{\partial y^2}\right)\right] g(\mathbf{r} - \mathbf{r}'; t - t') = \delta(\mathbf{r} - \mathbf{r}')\delta(t - t'). \quad (39)$$

Here $\chi \equiv \chi_{xxx}/e$. Eq. (39) has C_3 symmetry in accordance with the valley symmetry. Thus, for the first case of Eq. (37), one finds

$$n(\mathbf{r}, t) = n(0, t) + n_1 \int \frac{d^2\mathbf{q}}{(2\pi)^2} \frac{e^{i\mathbf{qr}} - 1}{Dq^2 + i\chi_\mathbf{q}}, \quad (40)$$

where $\chi_\mathbf{q} = \chi q^3 \cos(3\phi_q)$. Assuming, that $\chi_\mathbf{q} \ll Dq^2$ is a small correction, one can find the density correction to the isotropic diffusion in analytical form as

$$\delta n(\mathbf{r}, t) = \frac{n_1\chi}{2\pi D^2} \frac{\cos(3\varphi_\mathbf{r})}{r}. \quad (41)$$

Figure 1 illustrates the stationary electron density distribution around the illuminated spot calculated numerically based on Eq. (40) at arbitrary $\chi_\mathbf{q}$.

In the second case of Eq. (37), the electron density distribution as a function of time and spatial coordinates reads

$$n(\mathbf{r}, t) = n_2 \int \frac{d^2\mathbf{q}}{(2\pi)^2} e^{i\mathbf{qr} - (Dq^2 - i\chi_\mathbf{q})t}, \quad (42)$$

where again for the case $\chi_\mathbf{q} \ll Dq^2$, the density correction can be easily found as

$$\delta n(\mathbf{r}, t) = -\chi \frac{n_2 t}{2\pi} \frac{r^3 \cos(3\varphi_\mathbf{r})}{(2Dt)^4} e^{-\frac{r^2}{4Dt}}. \quad (43)$$

This correction also have the C_3 rotational symmetry. Thus we show that the c_{ijk} tensor components in Eq. (34) describe the trigonal asymmetry of electron diffusion process in individually populated TMD valley.

VI. CONCLUSION

To conclude, here we report that the generalized forces given by the electron density gradient (rather than external electric field) lead to the transverse and longitudinal valley linear and nonlinear transport in 2D Dirac monolayer systems preserving the inversion center symmetry. We theoretically demonstrate that the skew electron scattering of diffusive electrons may result in the valley Hall transport as a first order response to the electron density gradient. A theoretical analysis of the second-order system response to the electron density gradients developed here shows the existence of a valley selective PGE-like transport due to the trigonal warping of electron dispersion in the valleys considering the electron scattering processes on both short-range and Coulomb-like impurity centers.

The specific feature of the effects considered here is that the experimental study of these effects does not require two sources of external electromagnetic illuminations as it is for the conventional VHE and PGE transport based on the generalized force given by external electric field. The experimental observation of PGE or VHE transport requires unequal valley populations by additional circularly polarized light [13,21,29]. In case of the transport effect considered here, the electromagnetic illumination producing the unequal valley populations may simultaneously create the nonuniform electron density distribution, thus, producing the VHE and PGE-like current phenomena studied here.

The symmetry arguments being applied to the kinetic coefficients considered in the present paper and describing the linear and nonlinear responses to the nonuniform distribution of the electron density show that kinetic coefficients are directly coupled to the ones describing the linear and nonlinear responses to the electric field (VHE and PGE effects). We prove this directly and demonstrate this equivalence on the simple models. We also derived the tensor coefficients dealing with nonlinear (second-order) derivatives of electron chemical potential. These corrections responsible for the anisotropic electron diffusion processes of valley-selective photoexcited electrons. We considered several particular situations and demonstrated the presence of trigonal-symmetric diffusion in these cases.

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APPENDIX: DERIVATION OF CURRENT COMPONENTS

To find the expressions (26) and (32), we will combine Eqs. (16) and (21), this leads the current expression in the form $j_x = j_x^{\mathrm{I}} + j_x^{\mathrm{II}} + j_x^{\mathrm{III}}$, where

$$
\begin{aligned}
j_x^{\mathrm{I}} &= e\int \frac{d\mathbf{p}}{(2\pi\hbar)^2} v_x \hat{Q}_0^{-1}(v_x\partial_x)\hat{Q}_0^{-1}(v_x\partial_x) f^w(\mathbf{r}),\\
j_x^{\mathrm{II}} &= -e\int \frac{d\mathbf{p}}{(2\pi\hbar)^2} v_x \hat{Q}_0^{-1}\hat{Q}_w\hat{Q}_0^{-1}(v_x\partial_x)\hat{Q}_0^{-1}(v_x\partial_x) f^w(\mathbf{r}),\\
j_x^{\mathrm{III}} &= -e\int \frac{d\mathbf{p}}{(2\pi\hbar)^2} v_x \hat{Q}_0^{-1}(v_x\partial_x)\hat{Q}_0^{-1}\hat{Q}_w\hat{Q}_0^{-1}(v_x\partial_x) f^w(\mathbf{r}).
\end{aligned}
\tag{A1}
$$

First, we consider the short-range impurities. In this case, the action of the operator $\hat{Q}_w$ on the equilibrium function gives zero:

$$
\hat{Q}_w\{\chi_{\mathbf{p}}\} = 0. \tag{A2}
$$

Thus one finds that $j_x^{\mathrm{II}} = j_x^{\mathrm{III}} = 0$. Now, let's expand the distribution function up to the first order with respect to warping term as $f^w(\mathbf{r}) = f(\mathbf{r}) - w_{\mathbf{p}}\partial_\mu f(\mathbf{r})$. Using the relation $\partial_x f(\mathbf{r}) = \partial_\mu f(\mathbf{r})\partial_x\mu$, we can write the expression for j_x^{I} as

$$
j_x^{\mathrm{I}} = e\int \frac{d\mathbf{p}}{(2\pi\hbar)^2} v_x \hat{Q}_0^{-1}(v_x\partial_x)\hat{Q}_0^{-1} v_x\partial_x(f(\mathbf{r}) - w_{\mathbf{p}}\partial_\mu f(\mathbf{r})) \tag{A3}
$$

$$
= e\int \frac{d\mathbf{p}}{(2\pi\hbar)^2} v_x \hat{Q}_0^{-1}(v_x\partial_x)\hat{Q}_0^{-1} v_x\partial_x(f(\mathbf{r}) - w_{\mathbf{p}}\partial_\mu f(\mathbf{r})) \tag{A4}
$$

$$
\begin{aligned}
&= \underbrace{e\int \frac{d\mathbf{p}}{(2\pi\hbar)^2} v_x \hat{Q}_0^{-1} v_x \hat{Q}_0^{-1} v_x\left(\partial_\mu f_0 - w_{\mathbf{p}}\partial_\mu^2 f_0\right)}_{A_{xxx}} \nabla_x\nabla_x\mu\\
&\quad + \underbrace{e\int \frac{d\mathbf{p}}{(2\pi\hbar)^2} v_x \hat{Q}_0^{-1} v_x \hat{Q}_0^{-1} v_x \frac{\partial}{\partial\mu}\left(\partial_\mu f_0 - w_{\mathbf{p}}\partial_\mu^2 f_0\right)}_{B_{xxx}}\\
&\quad \times \nabla_x\mu\nabla_x\mu.
\end{aligned}
\tag{A5}
$$

Here we used the designations A_{xxx} and B_{xxx} from Eq. (25). Taking into account the expression for the electron velocity $v_x = \frac{\partial\epsilon}{\partial p_x} = \frac{p\cos\phi}{m} + 3\eta w p^2\cos 2\phi$, we get

$$
\begin{aligned}
A_{xxx} &= \eta w\int \frac{d\mathbf{p}}{(2\pi\hbar)^2}\left(\frac{p^4}{4m^2}\partial_\mu f_0\left(6\tau_1\tau_2 + 3\tau_1^2\right) - \frac{p^6\tau_1\tau_2}{8m^3}\partial_\mu^2 f_0\right)\\
&= \eta w N(6\langle(\epsilon^2\tau_1\tau_2)'\rangle + 3\left\langle\left(\epsilon^2\tau_1^2\right)'\right\rangle - \langle(\epsilon^3\tau_1\tau_2)''\rangle)
\end{aligned}
\tag{A6}
$$

and

$$
\begin{aligned}
B_{xxx} &= \eta w\int \frac{d\mathbf{p}}{(2\pi\hbar)^2}\left(\frac{p^4}{4m^2}\partial_\mu^2 f_0\left(6\tau_1\tau_2 + 3\tau_1^2\right) - \frac{p^6\tau_1\tau_2}{8m^3}\partial_\mu^3 f_0\right)\\
&= \eta w N(6\langle(\epsilon^2\tau_1\tau_2)''\rangle + 3\left\langle\left(\epsilon^2\tau_1^2\right)''\right\rangle - \langle(\epsilon^3\tau_1\tau_2)'''\rangle).
\end{aligned}
\tag{A7}
$$

Now we will evaluate anisotropic diffusion currents for the Coulomb impurities. Firs, let's calculate j_x^{I}, which has no explicit dependence of the matrix element $M_{\mathbf{pp}'}$, and thus the expressions for A_{xxx} and B_{xxx} have the same form as Eqs. (A6) and (A7) with the followoing relaxation times:

$$
\frac{1}{\tau_n} = \frac{|n|}{\tau(\varepsilon_{\mathbf{p}})}, \quad \frac{1}{\tau(\varepsilon_{\mathbf{p}})} = \frac{\pi e^4 n_i^C}{\hbar\kappa^2\varepsilon_{\mathbf{p}}}. \tag{A8}
$$

The expression for j_x^{II} reads

$$
j_x^{\mathrm{II}} = \underbrace{-e\int \frac{d\mathbf{p}}{(2\pi\hbar)^2} v_x \hat{Q}_0^{-1}\hat{Q}_w\hat{Q}_0^{-1} v_x\hat{Q}_0^{-1} v_x(\partial_\mu f_0)}_{A_{xxx}^{C,\mathrm{II}}} \nabla_x\nabla_x\mu \underbrace{-e\int \frac{d\mathbf{p}}{(2\pi\hbar)^2} v_x \hat{Q}_0^{-1}\hat{Q}_w\hat{Q}_0^{-1} v_x\hat{Q}_0^{-1} v_x\left(\partial_\mu^2 f_0\right)}_{B_{xxx}^{C,\mathrm{II}}} \nabla_x\mu\nabla_x\mu. \tag{A9}
$$

After a few transformations, one finds

$$
A_{xxx}^{C,\mathrm{II}} = -e\int \frac{d\mathbf{p}}{(2\pi\hbar)^2} v_x \hat{Q}_0^{-1}\hat{Q}_w\left[\frac{p^2\tau_1\tau_2\cos 2\phi}{2m^2}\partial_\mu f_0\right]. \tag{A10}
$$

To calculate this integral one needs to evaluate $\hat{Q}_w[p^2\tau_1\tau_2\cos 2\phi\partial_\mu f_0]$. Because $\tau \propto \epsilon = p^2/2m$, this leads to $\hat{Q}_w[p^6\cos 2\phi\partial_\mu f_0]$. Let's write the action of this operator explicitly:

$$
\hat{Q}_w[p^6\cos 2\phi\partial_\mu f_0(p)] = \frac{4\pi e^4\eta w m^2 n_i^C}{k^2\hbar p}\int_0^{2\pi} d\phi'\frac{d}{dp'}\left[(p^6\cos 2\phi\partial_\mu f_0(p) - p'^6\cos 2\phi'\partial_\mu f_0(p'))\frac{p^3\cos 3\phi - p'^3\cos 3\phi'}{p^2 + p'^2 - 2pp'\cos(\phi-\phi')}\right]_{p'=p}. \tag{A11}
$$

Simple, but cumbersome analysis allows us to transform Eq. (A12) to the following expression:

$$\hat{Q}_w[p^6\cos 2\phi\,\partial_\mu f_0(p)] = -\frac{4\pi e^4\eta w m^2 n_i^C}{k^2\hbar p}\frac{1}{2}\cos 3\phi\cos 2\phi\left[\left(5p^6\partial_\mu f_0 - \frac{p^8}{m}\partial_\mu^2 f_0\right)\int_0^{2\pi} d\theta\frac{(1-\cos 3\theta)\cos 2\theta}{1-\cos\theta}\right.$$
$$\left.+3p^6\partial_\mu f_0\int_0^{2\pi} d\theta\frac{(1-\cos 2\theta)\cos 3\theta}{1-\cos\theta} + p^6\partial_\mu f_0\int_0^{2\pi} d\theta\frac{1-\cos 3\theta}{1-\cos\theta}\right]$$
$$+\frac{4\pi e^4\eta w m^2 n_i^C}{k^2\hbar p}\frac{1}{2}\left(8p^6\partial_\mu f_0 - \frac{p^8}{m}\partial_\mu^2 f_0\right)\int_0^{2\pi} d\theta\frac{\sin 3\theta\sin 2\theta}{1-\cos\theta}\sin 3\phi\sin 2\phi. \tag{A12}$$

Substituting Eq. (A12) into Eq. (A10), one finds that only terms with $\cos\phi$ contribute to the current and thus

$$A_{xxx}^{C,\mathrm{II}} = e\frac{\eta w\kappa^2}{\hbar e^4 n_i^C}\int\frac{d\mathbf{p}}{(2\pi\hbar)^2}\tau_1\left(4\epsilon^3\partial_\mu f_0 - \epsilon^4\partial_\mu^2 f_0\right)$$
$$= e\frac{\eta w\kappa^2 N}{\hbar e^4 n_i^C}(-4\langle(\tau_1\epsilon^3)'\rangle - \langle(\tau_1\epsilon^4)''\rangle) \tag{A13}$$

As we can see from Eq. (A9), the expression for $B_{xxx}^{C,\mathrm{II}}$ will be the same, but with $\partial_\mu^2 f_0$ instead of $\partial_\mu f_0$:

$$B_{xxx}^{C,\mathrm{II}} = e\frac{\eta w\kappa^2 N}{\hbar e^4 n_i^C}(4\langle(\tau_1\epsilon^3)''\rangle - \langle(\tau_1\epsilon^4)'''\rangle). \tag{A14}$$

The same procedure can be applied to j_x^{III}. The final result reads

$$A_{xxx}^{C,\mathrm{III}} = e\frac{2\pi\eta w N}{\hbar^2}(-\langle(\tau_1\tau_2\epsilon^2)'\rangle + \langle\tau_1\tau_2\epsilon^3)''\rangle),$$
$$B_{xxx}^{C,\mathrm{III}} = e\frac{2\pi\eta w N}{\hbar^2}(-\langle(\tau_1\tau_2\epsilon^2)''\rangle + \langle(\tau_1\tau_2\epsilon^3)'''\rangle). \tag{A15}$$

Combining all contributions, one finds $A_{xxx}^C = A_{xxx}^{C,\mathrm{II}} + A_{xxx}^{C,\mathrm{III}}$ and $B_{xxx}^C = B_{xxx}^{C,\mathrm{II}} + B_{xxx}^{C,\mathrm{III}}$, where

$$A_{xxx}^C = 12e\eta w N\frac{\kappa^4}{\pi e^8\left(n_i^C\right)^2}\langle\epsilon^3\rangle,$$
$$B_{xxx}^C = 36e\eta w N\frac{\kappa^4}{\pi e^8\left(n_i^C\right)^2}\langle\epsilon^2\rangle. \tag{A16}$$

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