Photogalvanic transport in fluctuating Ising superconductors

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In a two-dimensional noncentrosymmetric Ising superconductor in a fluctuating regime under the action of a uniform external electromagnetic field, there emerge two contributions to the photogalvanic effect due to the trigonal warping of the valleys. The first contribution stems from the current of the electron gas in its normal state, while the second contribution is of an Aslamazov-Larkin nature: It originates from the presence of fluctuating Cooper pairs when the ambient temperature approaches (from above) the temperature of the superconducting transition in the sample. The way to lift the valley degeneracy is the application of a weak out-of-plane external magnetic field producing a Zeeman effect. The Boltzmann equations' approach for the electron gas in the normal state and the time-dependent Ginzburg-Landau equations for the fluctuating Cooper pairs allow for the study of the photogalvanic current in two-dimensional transition-metal dichalcogenide Ising superconductors.

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I. INTRODUCTION

In two-dimensional (2D) materials, photoinduced transport phenomena, which are second order with respect to an external electromagnetic (EM) field, are in the focus of state-of-the-art research [1]. The majority of these effects fall into two categories. The first one includes the rectification effects that occur under an external uniform alternating EM illumination, which produces stationary uniform electric currents in the system. The second category encompasses all the effects characterized by the system response at a doubled field frequency, thus describing second-harmonic generation phenomena.

Second-order transport phenomena are usually sensitive to the polarization of the EM field and the symmetry of the system under study, namely, time-reversal symmetry and spatial inversion symmetry. The phenomenological relation between the photoinduced rectified electric current and the amplitude of the external EM field reads $j_\alpha = \zeta_{\alpha\beta\gamma} E_\beta E^*_\gamma$, where $\zeta_{\alpha\beta\gamma}$ is the third-order tensor acquiring nonzero components in noncentrosymmetric materials. In nongyrotropic semiconductor materials, the (rectified) photoinduced electric current occurs as a second-order response to the linearly polarized external EM wave. This constitutes the photogalvanic effect (PGE). This effect does not directly relate to either light pressure, photon-drag phenomena, or nonuniformity of either the sample or light field intensity, such as the photoinduced Dember effect. Instead, the microscopic origin of the conventional PGE lies in the asymmetry of the interaction potential or the crystal-induced Bloch wave function [2–4].

In modern van der Waals structures based on 2D monolayers of transition-metal dichalcogenides (TMDs) [5,6], the PGE current may arise due to the specific band structure of the material possessing two time-reversal-coupled valleys in the Brillouin zone. A typical example of these materials is molybdenum disulfide, MoS_2. It possesses the D_{3h} point group, and the presence of the C_3 axis results in the emergence of a trigonal warping of the electron dispersion in each valley, reflecting the noncentrosymmetry of the crystal structure. The theoretical analysis shows that the PGE current arises here in each valley (involving electrons residing in both valleys), and these currents have different signs in different valleys. As a result, net PGE current self-compensates and vanishes. A nonzero net current may only occur if the time-reversal symmetry is broken due to, e.g., the presence of an external magnetic field or illumination of the sample by a circularly polarized EM field causing interband transitions [7–9].

Furthermore, a recent discovery of the superconducting (SC) transition in TMDs [10–12] stimulated additional interest to the study of transport phenomena in 2D Dirac materials exposed to external EM fields at lower temperatures [13–15]. In the intermediate range of temperatures lying in between the normal and SC state of the electron gas, when $0 < T - T_c \ll T_c$ (where T_c is a SC critical temperature), the order parameter starts to experience fluctuations [16–18]. Moreover, large spin-orbit coupling in TMDs results in strong out-of-plane electron spin polarization and large in-plane critical magnetic fields beyond the Pauli limit. Thus, all the ingredients of an *Ising superconductor* possessing unique physical properties are available. When the time-reversal symmetry breaks by a weak magnetic field due to the Zeeman effect, and given the absence of spatial reversal symmetry, TMDs might demonstrate a pronounced nonreciprocal response in the regime of SC fluctuations [11,12,19–22].

The goal of this work is to develop a microscopic theory of a linear PGE effect in fluctuating Ising superconductors exposed to a linearly polarized EM field. The time-reversal symmetry here is waived due to the presence of a weak Zeeman field pointed across the monolayer [23,24]. Trigonal warping of the valleys K and K' characteristic of MoS_2 serves as a microscopic mechanism of the effect. Within the D_{3h} point symmetry group, the third-order conductivity (or transport coefficient) tensor possesses only one nonzero component. Thus, phenomenologically, the PGE current can be expressed as $j_x = \zeta(|E_x|^2 - |E_y|^2)$, $j_y = -\zeta(E_x E_y^* + E_x^* E_y)$. Therefore, the main task comes down to the calculation of the coefficient ζ and analyzing its behavior for various EM field frequencies and temperatures in the vicinity of T_c, taking into account the contribution of normal electrons and the corrections arising from the SC order parameter fluctuations.

II. EFFECTIVE ELECTRON DISPERSION IN CONDUCTION BAND

The superconducting transition in the MoS_2 monolayer occurs at electron densities exceeding 10^{14} cm^{-2} [10]. At such high densities, the Fermi level lies deeply in the conduction band. Thus, it is feasible to use a simplified electron energy dispersion. Then, according to the two-band model, the Hamiltonian reads (in $\hbar = k_B = 1$ units)

$$H = \frac{\Delta}{2}\sigma_z + v(\eta p_x\sigma_x + p_y\sigma_y) + \begin{pmatrix} 0 & \mu p_+^2 \\ \mu p_-^2 & 0 \end{pmatrix} + s\eta\frac{\lambda_c}{2}(\sigma_z + 1) - s\eta\frac{\lambda_v}{2}(\sigma_z - 1) + s\Delta_Z, \quad (1)$$

where Δ is the material band gap, σ_i are the Pauli matrices, v is the band parameter with the dimensionality of velocity, $\eta = \pm 1$ is the valley index, $\mathbf{p}$ is the electron momentum, $p_\pm = \eta p_x \pm i p_y$, μ is the band parameter describing the trigonal warping and nonparabolicity of electron dispersion, s is the z component of electron spin, $\lambda_{c,v}$ describe spin-orbit splitting of the conduction and valence bands, and $\Delta_Z \propto B$ is the Zeeman energy due to the external magnetic field applied across the monolayer plane.

The eigenvalues of the Hamiltonian (1) read

$$E_{s\eta}(\mathbf{p}) = s\Delta_Z + s\eta\frac{\lambda_c + \lambda_v}{2} \pm\sqrt{\left[\frac{\Delta - s\eta(\lambda_v - \lambda_c)}{2}\right]^2 + |h_\mathbf{p}|^2},$$
$$\frac{|h_\mathbf{p}|^2}{\Delta} = \epsilon_p + \eta w(p_x^3 - 3p_x p_y^2), \quad (2)$$

where $\epsilon_p = p^2/2m$ is the electron kinetic energy in the conduction band, $m = \Delta/(2v^2)$ is the electron effective mass, and $w = 2v\mu/\Delta$ is a warping amplitude. It should be mentioned, that in Eq. (2) we omitted the terms $\propto p^4$. Expression (2) can be further expanded using the inequality $|h_\mathbf{p}|^2/\Delta^2 \ll 1$,

$$E_{s\eta}(\mathbf{p}) \approx s(\Delta_Z + \eta\lambda_c) + \epsilon_p + \eta w(p_x^3 - 3p_x p_y^2), \quad (3)$$

counting the conduction band energy from the value $\Delta/2$.

III. NORMAL-STATE ELECTRON GAS CONTRIBUTION TO PGE

Let us first study the PGE current of normal-state electrons exposed to a uniform external EM field $\mathbf{E}(t) = \mathbf{E}e^{-i\omega t} + \mathbf{E}^* e^{i\omega t}$ with normal incidence to the monolayer, thus $\mathbf{E} = (E_x, E_y, 0)$. In the case $\omega \ll \epsilon_F$, where ϵ_F is the Fermi energy, the Boltzmann equation [25,26] represents a suitable tool to analyze the PGE transport [1,7]. In the framework of the (single) relaxation time approximation, the Boltzmann equation reads

$$\frac{\partial f}{\partial t} + e\mathbf{E}(t)\cdot\frac{\partial f}{\partial \mathbf{p}} = -\frac{f - f_0}{\tau}, \quad (4)$$

where f is the electron distribution function, f_0 is the Fermi distribution, e is the elementary charge, and τ is the scattering time (on the pointlike impurities). In the expansion $f = f_0 + f_1(t) + f_2 + f_2(t) + \cdots$ with respect to the amplitude of the external electric field, the first-order correction depends on time, $f_1(t) = f_1 e^{-i\omega t} + f_1^* e^{i\omega t}$, whereas the second-order correction consists of the stationary, f_2, and alternating, $f_2(t)$, part. Summing up all the first-order terms yields

$$f_1 = -e\tau_\omega \mathbf{E}\cdot\frac{\partial f_0}{\partial \mathbf{p}} = e\tau_\omega \mathbf{v}\cdot\mathbf{E}(-f_0'), \quad (5)$$

where $\tau_\omega = \tau/(1 - i\omega\tau)$, $f_0' = \partial f_0/\partial E_{s\eta}$, and the electron velocity reads $\mathbf{v} = \partial_\mathbf{p} E_{s\eta}(\mathbf{p})$.

The stationary part of the second-order correction reads

$$f_2 = -e\tau\left(\mathbf{E}\cdot\frac{\partial f_1^*}{\partial \mathbf{p}} + \mathbf{E}^*\cdot\frac{\partial f_1}{\partial \mathbf{p}}\right), \quad (6)$$

which determines the PGE current,

$$j_\alpha = e\int\frac{d\mathbf{p}}{(2\pi)^2} v_\alpha f_2, \quad \alpha = x, y. \quad (7)$$

Combining Eqs. (5) and (6), and integrating by parts in Eq. (7), yields the expression for the PGE current density in the form

$$j_\alpha = e^3\tau(\tau_\omega^* E_\beta E_\gamma^* + \tau_\omega E_\beta^* E_\gamma) \times\sum_{s,\eta}\int\frac{d\mathbf{p}}{(2\pi)^2}\frac{\partial^2 v_\alpha}{\partial p_\beta \partial p_\gamma} f_0[E_{s\eta}(\mathbf{p})],$$
$$\frac{\partial^2 v_\alpha}{\partial p_\beta \partial p_\gamma} \equiv \frac{\partial^3 E_{s\eta}(\mathbf{p})}{\partial p_\beta \partial p_\gamma \partial p_\alpha}, \quad (8)$$

where $f_0[E_{s\eta}(\mathbf{p})] = \theta[\epsilon_F - E_{s\eta}(\mathbf{p})]$ for a degenerate electron gas with $\theta[x]$ the Heaviside step function. Considering that $\partial^2 v_\alpha/\partial p_\beta \partial p_\gamma = \pm 6\eta w$ (here, "+" stands for the xxx component, while "−" stands for the xyy, yxy, yyx components, the others are zero), from Eq. (8) it follows that the PGE current is proportional to the differences between electron densities $n_\pm$ in both valleys, $j_\alpha \propto \sum_{s,\eta}\eta n_\eta$. Therefore, the normal-state electron gas does not contribute to the nonreciprocal current in the framework of this model, as it is also claimed in Ref. [23]. The reason for such behavior is that the Zeeman field only redistributes the electrons between spin-resolved subbands in each valley, keeping the total electron density in the valley unchanged.

In order to have a finite PGE response, it is necessary to modify the original model, Eqs. (3) and (4), by introducing energy-dependent relaxation time τ_ε. In a particular case of electron scattering on Coulomb impurities in a 2D system, the relaxation time is proportional to the electron energy, $\tau_\varepsilon = \tau_0 \varepsilon_{\mathbf{p}}$, where τ_0 is a coefficient. Henceforth, in Eqs. (4)–(7), τ should be replaced by $\tau_\varepsilon = \tau_0\varepsilon_{\mathbf{p}} = \tau_0[\epsilon_p + \eta w(p_x^3 - 3p_xp_y^2)]$. Then, the PGE current density in the static limit $\omega\tau_\varepsilon = \tau_0\omega\epsilon_F \ll 1$ (which additionally provides the relation between ω and the doping) reads

$$j_\alpha = 2e^3 E_\beta E_\gamma^* \sum_{s,\eta} \int \frac{d\mathbf{p}}{(2\pi)^2} v_\alpha \tau_\varepsilon \frac{\partial}{\partial p_\beta}\left\{\tau_\varepsilon \frac{\partial}{\partial p_\gamma} f_0\right\}. \quad (9)$$

To derive (9), we also assumed the absence of intervalley scattering [27] and neglected the spin-flip processes transferring the electrons between spin-resolved subbands in a given valley.

Expanding Eq. (9) in the lowest order in w and restoring dimensionality yields (see the Supplemental Material [28])

$$\boldsymbol{j} = 108\frac{e^3\tau_0^2\Delta_Z\lambda_c w n_e}{\hbar^3}\mathbf{F}(\mathbf{E}), \quad (10)$$

where $\mathbf{F}(\mathbf{E}) = (|E_x|^2 - |E_y|^2, -E_xE_y^* - E_yE_x^*)$, and $n_e = n_+ + n_-$ is a total electron density in both valleys. The PGE current of the normal-state electron gas, Eq. (10), represents the first important result of this paper: The nonreciprocal PGE response is finite in the case of electron scattering off Coulomb impurities in 2D samples.

Taking the electron density $n_e \sim 10^{14}$ cm^{-2}, the external magnetic field $B = 1$ T, the amplitude of the EM field $E_0 = 1$ V/cm, $\tau_0 = 10$ ps/eV (which is the highest possible value found from the relation $\tau_0\omega\epsilon_F \ll 1$ for $n_e = 10^{14}$ cm^{-2} and $\omega = 0.1$ ps^{-1}), and typical parameters for MoS_2 [23,29], $\lambda_c = 3$ meV and $w = -3.4$ eV Å^3, we find that a typical magnitude of the PGE current due to the normal 2D electron gas contribution amounts to $j \sim 10$ nA/cm.

IV. SUPERCONDUCTING FLUCTUATIONS' CONTRIBUTION TO PGE

The electric current density operator due to the presence of SC fluctuations reads

$$j = \frac{e^*}{2}\{\Psi^*\mathbf{v}(\hat{\mathbf{p}})\Psi + \Psi\mathbf{v}(-\hat{\mathbf{p}})\Psi^*\}, \quad (11)$$

where $e^* = 2e$ is a charge of a Cooper pair, $\mathbf{v}(\hat{\mathbf{p}})$ is a Cooper pair velocity operator, $\hat{\mathbf{p}} = -i\nabla$ is a momentum operator, and the superconducting order parameter $\Psi(\mathbf{r}, t)$ satisfies the time-dependent Ginzburg-Landau (TDGL) equation with accounting of the trigonal warping contribution to the kinetic energy of a Cooper pair,

$$\left[\gamma\frac{\partial}{\partial t} + \varepsilon(\hat{\mathbf{p}}) + 2ie\gamma\varphi(\mathbf{r}, t)\right]\Psi(\mathbf{r}, t) = f(\mathbf{r}, t). \quad (12)$$

In Eq. (12), $\gamma = \pi\alpha/8$, α is the parameter of GL theory, which is inversely proportional to the effective mass m and square of the coherence length ξ, $4m\alpha T_c\xi^2 = 1$, thus $\varepsilon(\mathbf{p}) = p^2/4m + \alpha T_c\epsilon + \Lambda(p_x^3 - 3p_xp_y^2) \equiv \alpha T_c(\epsilon + p^2\xi^2) + \Lambda(p_x^3 - 3p_xp_y^2)$ is the Cooper pair kinetic energy, and $\epsilon = (T - T_c)/T_c$ is the reduced temperature. The coherence length in 2D reads

$$\xi^2 = \frac{v_F^2\tau^2}{2}\left[\psi\left(\frac{1}{2}\right) - \psi\left(\frac{1}{2} + \frac{1}{4\pi T\tau}\right) + \frac{\psi'\left(\frac{1}{2}\right)}{4\pi T\tau}\right], \quad (13)$$

where $\psi(x)$ is the digamma function, and $v_F = \sqrt{4\pi n_e}/m$ is the Fermi velocity. Furthermore, in Eq. (12), $\varphi(\mathbf{r}, t) = \varphi e^{i\mathbf{kr} - i\omega t} + \varphi^* e^{-i\mathbf{kr} + i\omega t}$ is the scalar potential, which obeys standard correspondence with the external uniform EM field, $\mathbf{E} = -\nabla\varphi$.

The Cooper pair trigonal warping amplitude Λ, entering Eq. (12) through the term $\varepsilon(\hat{\mathbf{p}})$, in a clean superconductor ($\tau T_c \gg 1$) and for the s-wave singlet pairing can be expressed through the Zeeman field and the normal electron warping amplitude w [23],

$$\Lambda = \frac{93\zeta(5)\Delta_Z\lambda_c w}{28\zeta(3)(\pi T_c)^2}. \quad (14)$$

To estimate it, let us substitute typical parameters for MoS_2 (given in the last paragraph of the previous section) and the SC critical temperature $T_c = 10$ K: $|\Lambda| \approx 0.46$ eV Å^3 for $B = 1$ T.

The right-hand side of Eq. (12) is the Langevin force, describing SC fluctuations in the equilibrium. It satisfies the white-noise law,

$$\langle f^*(\mathbf{r})f(\mathbf{r}', t')\rangle = 2\gamma T\delta(\mathbf{r} - \mathbf{r}')\delta(t - t'), \quad (15)$$

which allows us to find an expression for the SC order parameter in equilibrium, $\langle|\Psi_{0\mathbf{p}}|^2\rangle = [\alpha(\epsilon + \xi^2\mathbf{p}^2)]^{-1}$ (here, $\Psi_{0\mathbf{p}}$ is the Fourier transform of the order parameter).

As concerns the applicability of the TDGL equation in the form (12), it is only valid in the low-frequency domain ($\omega\tau \ll 1$) given an arbitrary ratio between ω and T_c. In the range of moderate and high frequencies ($\omega\tau \gtrsim 1$), various nonlocality corrections emerge [30]. Treating them requires the usage of quantum-field theory approaches beyond the TDGL equation. Therefore, this theory is applicable to either clean superconductors, $\omega < \tau^{-1} < T_c$, or "dirty" superconductors obeying the relation $(\omega, T_c)\tau < 1$. Moreover, in addition to the Aslamazov-Larkin correction there exist other fluctuating contributions, such as the Maki-Tompson [31,32] and the "density of states" [33] ones, which are beyond the scope of the present paper. Treating them also requires the using of quantum-field theory approaches beyond the TDGL equation [17].

Let us start with a clean superconductor case. Expanding the order parameter with respect to the scalar potential, $\Psi(\mathbf{r}, t) = \Psi_0(\mathbf{r}, t) + \Psi_1(\mathbf{r}, t) + \Psi_2(\mathbf{r}, t) + \cdots$, and then substituting this expansion in Eq. (11) keeping only the second-order terms, gives two contributions to the electric current density,

$$j_\alpha^{\mathrm{I}} = e\{\Psi_1^* v_\alpha(\hat{\mathbf{p}})\Psi_1 + \Psi_1 v_\alpha(-\hat{\mathbf{p}})\Psi_1^*\}, \quad (16)$$

$$j_\alpha^{\mathrm{II}} = e\{\Psi_0^* v_\alpha(\hat{\mathbf{p}})\Psi_2 + \Psi_2^* v_\alpha(\hat{\mathbf{p}})\Psi_0\} + e\{\Psi_0^* v_\alpha(-\hat{\mathbf{p}})\Psi_2 + \Psi_2^* v_\alpha(-\hat{\mathbf{p}})\Psi_0\}, \quad (17)$$

where

$$\Psi_0(x) = \int dx' g(x-x') f(x'), \tag{18}$$

$$\Psi_{1(2)}(x) = -2ie\gamma \int dx' g(x-x')\varphi(x')\Psi_{0(1)}(x'), \tag{19}$$

with $x = (\mathbf{r}, t)$ the short-hand notation, and

$$g(\mathbf{r}, t) = \sum_{\varepsilon,\mathbf{p}} e^{i\mathbf{pr}-i\varepsilon t} g_{\mathbf{p}}(\varepsilon) \equiv \sum_{\varepsilon,\mathbf{p}} \frac{e^{i\mathbf{pr}-i\varepsilon t}}{-i\gamma\varepsilon + \varepsilon(\mathbf{p})} \tag{20}$$

the fluctuation propagator in standard form.

Combining Eqs. (16)–(20) and performing the averaging over the fluctuating Langevin forces gives a general expression for the PGE current:

$$\begin{aligned} j_\alpha &= 2(2e\gamma)^3 T \sum_{\varepsilon,\mathbf{q}} |g_{\mathbf{q}}(\varepsilon)|^2 \{ |g_{\mathbf{k}+\mathbf{q}}(\varepsilon+\omega)|^2 v_\alpha(\mathbf{q}+\mathbf{k}) \\ &\quad - v_\alpha(\mathbf{q})[g_{\mathbf{q}}(\varepsilon) g_{\mathbf{q}+\mathbf{k}}(\varepsilon+\omega) + g^*_{\mathbf{q}}(\varepsilon) g^*_{\mathbf{q}+\mathbf{k}}(\varepsilon+\omega)]\} |\varphi|^2 \\ &\quad + (\mathbf{k} \to -\mathbf{k}, \omega \to -\omega). \end{aligned} \tag{21}$$

Let us mention that in Eq. (21), only the static contribution to the product of two scalar potentials is accounted for, thus disregarding the 2ω harmonics.

After the integration over energy, Eq. (21) acquires a more compact form,

$$\begin{aligned} j_\alpha &= (2e)^3 \gamma^2 T \sum_{\mathbf{q}} \left\{ \frac{v_\alpha(\mathbf{q}+\mathbf{k})}{\varepsilon(\mathbf{q})\varepsilon(\mathbf{q}+\mathbf{k})} - \frac{v_\alpha(\mathbf{q})}{\varepsilon^2(\mathbf{q})} \right\} \\ &\quad \times \frac{\varepsilon(\mathbf{q}) + \varepsilon(\mathbf{q}+\mathbf{k})}{\gamma^2\omega^2 + [\varepsilon(\mathbf{q}) + \varepsilon(\mathbf{q}+\mathbf{k})]^2} |\varphi|^2 \\ &\quad + (\mathbf{k} \to -\mathbf{k}, \omega \to -\omega). \end{aligned} \tag{22}$$

Evidently, this current vanishes at $\mathbf{k} \to \mathbf{0}$. It motivates the need to expand the PGE current up to the second order over $\mathbf{k}$ using the correspondence between the electrostatic potential and components of the electric field, $(-ik_\beta)(ik_\gamma)|\varphi|^2 = E_\beta E^*_\gamma$. The first-order corrections vanish since the terms with opposite signs (directions) of $\mathbf{k}$ cancel each other out.

Furthermore, expanding $\varepsilon(\mathbf{p})$ in Eq. (22) up to the first order in warping Λ and integrating over the momentum $\mathbf{q}$, gives the paraconductivity contribution to the PGE as $\boldsymbol{j} = \zeta_S \mathbf{F}$, where after restoring dimensionality,

$$\begin{aligned} \zeta_S &= \frac{3e^3\Lambda m\pi}{16\hbar^3 k_B T_c \epsilon^2} \frac{1}{\tilde{\omega}^2} \left[1 + \frac{\log(1+\tilde{\omega}^2)}{\tilde{\omega}^2} + \frac{\pi}{2}\left(\frac{1}{\tilde{\omega}^3} - \frac{1}{\tilde{\omega}} \right) \right. \\ &\quad \left. - \frac{1}{\tilde{\omega}}\left(1 + \frac{1}{\tilde{\omega}^2}\right) \arctan\tilde{\omega} + \frac{1}{\tilde{\omega}}\left(1 - \frac{1}{\tilde{\omega}^2}\right) \arctan\frac{1}{\tilde{\omega}} \right], \\ \tilde{\omega} &= \frac{\pi\hbar\omega}{16 k_B (T - T_c)}, \end{aligned} \tag{23}$$

which represents the second important result of this paper.

The third-order ac paraconductivity tensor ζ_S experiences its maximum at the static limit, $\zeta_S(\tilde{\omega} \ll 1) \approx \zeta_0(1 - 2\tilde{\omega}^2/5)$, whereas it decays with an increase of the frequency of the EM field as $\zeta_S(\tilde{\omega} \gg 1) \approx 6\zeta_0/\tilde{\omega}^2$, where $\zeta_0 = 2e^3\Lambda m\pi/64\hbar^3 T_c \epsilon^2$ is the paraconductivity tensor for the dc nonreciprocal current [23,24] (interestingly, using the Boltzmann kinetic equation gives the same result—see the Supplemental Material [28]).

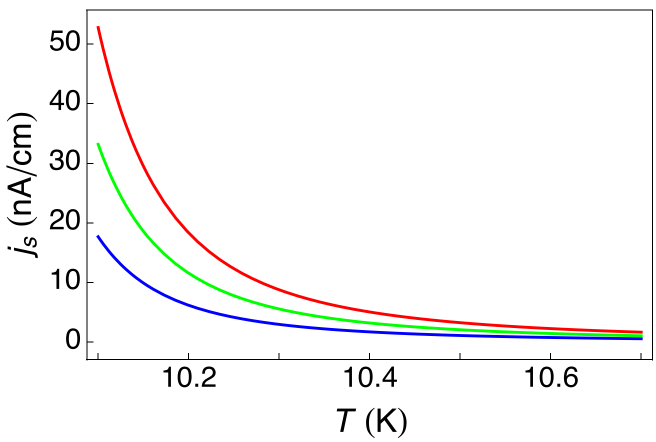

FIG. 1. Photogalvanic current of fluctuating Cooper pairs [Eq. (23) where Λ is replaced with Λ_τ] as a function of temperature for $\omega = 0.1$ ps^{-1} and various relaxation times: $\tau = 5$ ps (red), $\tau = 0.3$ ps (green), and $\tau = 0.1$ ps (blue). All parameters are for MoS_2: superconducting critical temperature $T_c = 10$ K, the warping amplitude $w = 3.4$ eV Å^3, $\lambda_c = 3$ meV, $B = 1$ T, and the amplitude of the electromagnetic field is $E_0 = 1$ V/cm.

Evidently, the dc component of ζ_S decays as $(T - T_c)^{-2}$ that is much faster than the conventional Aslamazov-Larkin correction in 2D, $\sigma^{\mathrm{AL}} \propto (T - T_c)^{-1}$ [16]. Moreover, the paraconductivity starts to decrease rapidly with an increase of frequency even for $\omega \gtrsim (T - T_c)$ while the power of this decrease coincides with the power of ϵ dependence of the dc conductivity component. Note, Eq. (23) is only valid for a linearly polarized EM field, while the PGE vanishes in the case of a circularly polarized light.

The next task is to generalize Eq. (23) for the case of an arbitrary impurity concentration by accounting for the relaxation time in the derivation of the Ginzburg-Landau free energy using the Green's function technique (see the Supplemental Material [28]). Indeed, the influence of SC fluctuations might be more prominent in dirty samples in accordance with the Ginzburg-Levanyuk criterion [34]. The calculations in the case of an arbitrary τT_c show that instead of the trigonal warping amplitude for the Cooper pairs Λ, which enters Eq. (23) in the clean limit, there comes into play an effective warping coefficient, $\Lambda_\tau = \Lambda \cdot f_\tau(2\pi T_c \tau)$, where

$$\begin{aligned} f_\tau(x) &= \frac{7\zeta(3)}{31\zeta(5)} \frac{\pi n_e}{m^2} \frac{x^3}{(\pi T_c \xi)^2} \left\{ -2\pi^2 + 4\psi'\left(\frac{1}{2} + \frac{1}{2x}\right) \right. \\ &\quad \left. + \frac{1}{x}\left[14\zeta(3) - \psi''\left(\frac{1}{2} + \frac{1}{2x}\right) \right] \right\}, \end{aligned} \tag{24}$$

which represents a monotonous function of τ, and $f_\tau(2\pi T_c \tau \gg 1) \to 1$ in the limit of a clean superconductor, whereas it vanishes linearly in the dirty case, $f_\tau(2\pi T_c \tau \ll 1) \to 0$. Interestingly enough, the trigonal warping term in the Ginzburg-Landau free energy and, as a consequence, in the photogalvanic current depends on the coherence length ξ and the relaxation time τ. Thus, the Cooper pairs in the fluctuating regime turn out sensitive to the presence of impurities in the sample, which is in contrast with the conclusions of

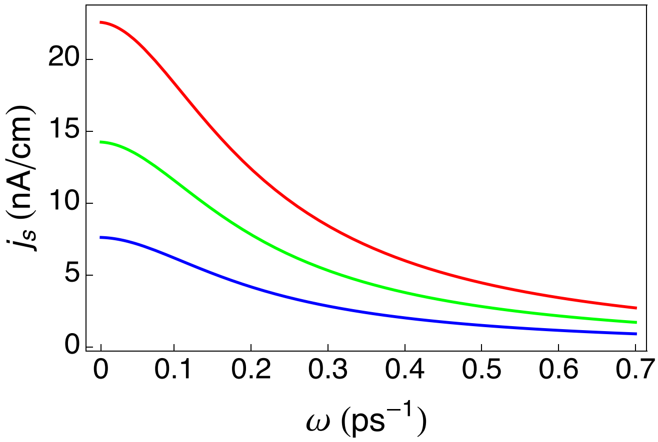

FIG. 2. Photogalvanic current of fluctuating Cooper pairs [Eq. (23) where Λ is replaced with Λ_τ] as a function of electromagnetic field frequency for $T = 10.2$ K and various relaxation times: $\tau = 5$ ps (red), $\tau = 0.3$ ps (green), and $\tau = 0.1$ ps (blue). All other parameters are the same as in Fig. 1.

the Aslamazov-Larkin theory being applied to the first-order response current.

Figures 1 and 2 show the temperature and frequency dependencies of the PGE current. Red curves correspond to the case of a clean superconductor, $\tau T_c \gg 1$. In terms of the EM field intensity, $I = c\epsilon_0|\mathbf{E}|^2/2$ with c the speed of light and ϵ_0 the vacuum permittivity, the estimation gives $j/I \approx 400\,\mathrm{nA\,m/W}$ for $T = 10.1$ K and $B = 1$ T. Green and blue curves correspond to the case of dirty superconductors, $\tau T_c \ll 1$, and demonstrate the effect of the pointlike impurities on the temperature and frequency dependencies of the PGE contribution due to SC fluctuations.

The above-developed theoretical description of fluctuating PGE transport shows the key role of the external magnetic field and corresponding Zeeman contribution to the GL functional. In the absence of a magnetic field, the warping parameter $\Lambda = 0$, and the Cooper pair dispersion becomes $\varepsilon(\mathbf{p}) = \alpha T_c(\epsilon + p^2\xi^2)$, that corresponds to an isotropic superconductor. In this case, the specific type of PGE effect, called coherent PGE [35,36], may exist: The rectified current appears as the third-order response to the EM field having the ground ω and double-frequency 2ω harmonics. The corresponding theory of coherent PGE in isotropic fluctuating superconductors was developed in our recent paper [37].

V. CONCLUSIONS

We conclude that in a two-dimensional noncentrosymmetric fluctuating Ising superconductor possessing trigonal warping of the valleys and exposed to a uniform external electromagnetic field, there emerge two contributions to the photogalvanic effect. The first contribution originates from the normal-state electron gas in the presence of Coulomb impurities in the sample. The second contribution stems from the presence of superconducting fluctuations. In order to lift the valley degeneracy and thus have a nonzero photogalvanic electric current in the system, it is sufficient to use a weak out-of-plane external magnetic field producing a Zeeman effect and breaking the time-reversal symmetry.

The photogalvanic effect thus possesses an Aslamazov-Larkin nature since it originates from the presence of fluctuating Cooper pairs when the ambient temperature approaches the temperature of the superconducting transition in the sample. The electric current, as a second-order response of the system, possesses, first, a more pronounced temperature divergence $(T - T_c)^{-2}$, as compared with the Aslamazov-Larkin correction to the Drude conductivity, and, second, the current density has no smallness related to the electron-hole asymmetry of the quasiparticle spectrum, as it takes place in other second-order response effects [38–40].

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