Coherent photogalvanic effect in fluctuating superconductors

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We develop a theory of the coherent photogalvanic effect (CPGE) in low-dimensional superconductors in the fluctuating regime. It manifests itself in the appearance of a stationary electric current of Cooper pairs under the action of two coherent electromagnetic fields of light with frequencies lying in the sub-terahertz range. We derive the general formula for the electric current density, study the particular cases of linear and circular polarizations of the external light fields, and show that the current might have a nonmonotonous spectrum at certain polarization angles and turns out very sensitive to the proximity of the ambient temperature to the critical temperature of superconducting transition: Approaching the critical temperature, the peak in the spectrum becomes narrower, its frequency experiences a redshift, and the intensity of the peak drastically grows.

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I. INTRODUCTION

In a broad sense, the photogalvanic (or photovoltaic) effect (PGE) consists in the emergence of an electric current or voltage in the sample under the action of electromagnetic (EM) field of light. Recent years demonstrate a growing interest in PGE, which has been studied in graphene [1], Weyl semimetals [2,3], mono- and dichalcogenides [4,5], ferroelectrics [6,7], and at terahertz frequencies [8]. There exist various types of PGE, including the bulk PGE [9,10], valve effect in semiconducting *p-n* junctions, photo-Dember effect [11], and photo-piezoelectricity, among others. All these phenomena are due to the presence of inhomogeneities either in the media (like a *p-n* junction) or in the light field itself.

There also exists PGE, which requires neither the inhomogeneity of optical excitation of electron-hole pairs nor the inhomogeneity of the sample [9,12–14]. Instead, the media lacking the inversion center is required. In this case, the stationary electric current represents the second-order response of the system to a uniform electric field $\mathbf{E}$ with frequency ω, $j_\eta = T_{\eta\lambda\gamma}E_{\omega\lambda}E^*_{\omega\gamma}$, where $\eta,\ \lambda,\ \gamma = x,\ y,\ z$. Thus, it is determined by the 3-rank conductivity tensor $T_{\eta\lambda\gamma}$ (which is nontrivial only in the media lacking the inversion center). The microscopic origin of this PGE is in the asymmetry of the interaction potential and the electron scattering processes or the crystal-induced Bloch wave function. Recently, other mechanisms of PGE related to the trigonal warping of valleys in transition metal dichalcogenides have also been suggested [15,16].

One of the types of PGE is the coherent photogalvanic effect (CPGE). It was predicted in works [17,18] and observed experimentally in [19,20]. It represents the emergence of a stationary electric current in a spatially homogeneous conducting sample exposed to two EM fields with frequencies ω and 2ω. Phenomenologically, the CPGE current can be written as [17] $j_\eta = \chi_{\eta\lambda\gamma\delta}E_{2\omega\lambda}E_{-\omega\gamma}E_{-\omega\delta} + \text{c.c.}$, where $\chi_{\eta\lambda\gamma\delta}$ is a 4-rank generalized conductivity tensor, and $\mathbf{E} = 2\text{Re}(\mathbf{E}_\omega e^{i\omega t} + \mathbf{E}_{2\omega}e^{2i\omega t})$, where $\mathbf{E}_{-\omega} = \mathbf{E}^*_\omega$. As compared with the traditional PGE, CPGE does not require the absence of the inversion symmetry and it depends on the phase difference between the EM fields.

Recently, photoinduced nonlinear transport phenomena in superconductors attracted growing interest of the community [21]. In particular, the photon drag effect [22] and the third harmonic generation [23] were proposed. PGEs have not been so far addressed in superconductors, to the best of our knowledge. It is known that the PGE in normal conductors at low temperatures exists due to the asymmetrical impurity scattering processes of electrons, like the skew- and side-jump effects, among others. These processes were investigated in view of recent research on the anomalous Hall effect in fluctuating superconductors [24], and it has been demonstrated that the Aslamazov-Larkin correction is not dressed by these asymmetric impurity scatterings.

In this paper, we study CPGE in a superconductor in the fluctuating regime [25,26] when the temperature is slightly above the critical temperature of superconducting (SC) transition T_c and in addition to normal (unpaired) electrons there start to emerge and collapse Cooper pairs called in this case the *SC fluctuations* (SFs) since the density of Cooper pairs fluctuates, according to the Aslamazov-Larkin (AL) effect. These SFs can dramatically change the conductivity of the system due to an additional *paraconductivity* term. As we showed in previous works [27], the presence of SFs can also drastically change the optical response of the system.

To describe the CPGE of SFs, we will use the Boltzmann transport equations approach [26], in the framework of which the Cooper pairs are described by the distribution function and an effective energy-dependent lifetime. This approach has proved sufficient if one considers the AL corrections to the conductivity [28–31], which we do in this paper.

II. GENERAL FORMALISM

The Boltzmann equation for SFs in the uniform external electromagnetic field reads

$$\frac{\partial f}{\partial t}+2e[\mathbf{E}_\omega(t)+\mathbf{E}_{2\omega}(t)]\cdot\frac{\partial f}{\partial \mathbf{p}}+\frac{f-f_0}{\tau_\mathbf{p}}=0, \qquad (1)$$

where f is the distribution function of fluctuating Cooper pairs, t is time, e is electron charge, $\mathbf{E}_\omega(t)=\mathbf{E}_\omega e^{i\omega t}+\mathbf{E}^*_\omega e^{-i\omega t}$ and $\mathbf{E}_{2\omega}(t)=\mathbf{E}_{2\omega}e^{2i\omega t}+\mathbf{E}^*_{2\omega}e^{-2i\omega t}$ are the first and second harmonics of the electromagnetic field of frequency ω, $\mathbf{p}$ is the center-of-mass momentum of the Cooper pair with the absolute value $p=|\mathbf{p}|$; $\tau_\mathbf{p}=\pi\alpha/(16\varepsilon_\mathbf{p})$ is the effective Cooper pair lifetime with α the parameter of the AL theory [26]; α is calculated using the relation $4m\alpha T_c\xi^2/\hbar^2=1$, where ξ is the correlation length given by

$$\xi^2=\frac{v_F^2\tau^2}{2}\left[\psi\left(\frac{1}{2}\right)-\psi\left(\frac{1}{2}+\frac{\hbar}{4\pi T\tau}\right)+\frac{\hbar\psi'(1/2)}{4\pi T\tau}\right]. \qquad (2)$$

Here $\psi(x)$ is the digamma function; $v_F=\hbar\sqrt{4\pi n}/m$ is the Fermi velocity; τ is the electron relaxation time, $\varepsilon_\mathbf{p}=\varepsilon_p=p^2/4m+\mu$ is Cooper pair energy with m the electron mass, $\mu=\alpha T_c\epsilon$, $\epsilon=(T-T_c)/T_c>0$ the reduced temperature [26]; $f_0=T/\varepsilon_p$ is the classical Rayleigh-Jeans distribution of Cooper pairs at temperature T in the absence of external perturbations.

Furthermore, we assume that the EM fields cause small perturbations of the local density of SFs and do the expansion [32,33], $f(t)=f_0+\sum_n f^{(n)}(t)$. The nth order correction obeys the equation

$$\left(\frac{\partial}{\partial t}+\frac{1}{\tau_\mathbf{p}}\right)f^{(n)}(t)=-2e[\mathbf{E}_\omega(t)+\mathbf{E}_{2\omega}(t)]\cdot\frac{\partial f^{(n-1)}(t)}{\partial\mathbf{p}}. \qquad (3)$$

The general formula for the CPGE current density is $j_\eta=2e\int d\mathbf{p}u_\eta f/(2\pi\hbar)^2$, where $\eta=x,\,y$ and $u_\eta=p_\eta/2m$ is a Cooper pair velocity. In our case, the lowest-order nonzero contribution to the stationary current reads

$$j_\eta=2e\int\frac{d\mathbf{p}}{(2\pi)^2}u_\eta\langle f^{(3)}(t)\rangle,$$
$$\langle f^{(3)}(t)\rangle=-2e\tau_\mathbf{p}\mathrm{Re}\left(\mathbf{E}^*_\omega\cdot\frac{\partial f^{(2)}_\omega}{\partial\mathbf{p}}+\mathbf{E}^*_{2\omega}\cdot\frac{\partial f^{(2)}_{2\omega}}{\partial\mathbf{p}}\right), \qquad (4)$$

where $\langle\ldots\rangle$ stands for the time-averaging and the second-order corrections satisfy

$$\left(i\omega+\frac{1}{\tau_\mathbf{p}}\right)f^{(2)}_\omega=-2e\left(\mathbf{E}_{2\omega}\cdot\frac{\partial f^{*(1)}_\omega}{\partial\mathbf{p}}+\mathbf{E}^*_\omega\cdot\frac{\partial f^{(1)}_{2\omega}}{\partial\mathbf{p}}\right),$$
$$\left(2i\omega+\frac{1}{\tau_\mathbf{p}}\right)f^{(2)}_{2\omega}=-2e\mathbf{E}_\omega\cdot\frac{\partial f^{(1)}_\omega}{\partial\mathbf{p}}, \qquad (5)$$

and the first-order corrections read

$$\left(i\omega+\frac{1}{\tau_\mathbf{p}}\right)f^{(1)}_\omega=-2e\mathbf{E}_\omega\cdot\frac{\partial f_0}{\partial\mathbf{p}}=-2e(\mathbf{u}\cdot\mathbf{E}_\omega)f_0',$$
$$\left(-i\omega+\frac{1}{\tau_\mathbf{p}}\right)f^{*(1)}_\omega=-2e\mathbf{E}^*_\omega\cdot\frac{\partial f_0}{\partial\mathbf{p}}=-2e(\mathbf{u}\cdot\mathbf{E}^*_\omega)f_0',$$
$$\left(2i\omega+\frac{1}{\tau_\mathbf{p}}\right)f^{(1)}_{2\omega}=-2e\mathbf{E}_{2\omega}\cdot\frac{\partial f_0}{\partial\mathbf{p}}=-2e(\mathbf{u}\cdot\mathbf{E}_{2\omega})f_0', \qquad (6)$$

where $f_0'=\partial f_0/\partial\varepsilon_p$. Integrating by parts in Eq. (4) and taking the integrals, introducing for convenience dimensionless variables $\kappa=\varepsilon_p/\mu$ and $\beta=\pi\omega/(16T_c\epsilon)$, and then using $\mathbf{E}_\omega=E_1\mathbf{e}_\omega$ and $\mathbf{E}_{2\omega}=E_2\mathbf{e}_{2\omega}$, where we introduce two unity vectors in the directions of electric field harmonics, and finally parametrizing the fluctuating Cooper pair velocity as $\mathbf{u}=u\mathbf{n}$, where $\mathbf{n}=(\cos\varphi,\sin\varphi)$, we can find the total current density (see the Appendix)

$$j_\eta=\chi_{\eta\lambda\gamma\delta}e^*_{\omega\lambda}e^*_{\omega\gamma}e_{2\omega\delta}+\chi^*_{\eta\lambda\gamma\delta}e_{\omega\lambda}e_{\omega\gamma}e^*_{2\omega\delta}$$
$$+\zeta_{\eta\lambda\gamma\delta}e^*_{2\omega\lambda}e_{\omega\gamma}e_{\omega\delta}+\zeta^*_{\eta\lambda\gamma\delta}e_{2\omega\lambda}e^*_{\omega\gamma}e^*_{\omega\delta}, \qquad (7)$$

where

$$\chi_{\eta\lambda\gamma\delta}=\frac{j_0}{2}\int_1^\infty\frac{(\kappa-1)d\kappa}{\kappa^2}\left(\frac{1}{\kappa-i\beta}+\frac{1}{\kappa+2i\beta}\right)\times\frac{\partial}{\partial\kappa}\left[\frac{\delta_{\eta\lambda}\delta_{\gamma\delta}/2-2\frac{\kappa-1}{\kappa}\overline{n_\eta n_\lambda n_\gamma n_\delta}}{\kappa(\kappa+i\beta)}\right],$$
$$\zeta_{\eta\lambda\gamma\delta}=\frac{j_0}{2}\int_1^\infty\frac{(\kappa-1)d\kappa}{\kappa^2(\kappa+i\beta)}\times\frac{\partial}{\partial\kappa}\left[\frac{\delta_{\eta\lambda}\delta_{\gamma\delta}/2-2\frac{\kappa-1}{\kappa}\overline{n_\eta n_\lambda n_\gamma n_\delta}}{\kappa(\kappa+2i\beta)}\right] \qquad (8)$$

are two auxiliary tensors, where the bar symbols stand for the averaging over the angle of the unity vector $\mathbf{n}$. In Eq. (8),

$$j_0=\frac{(2e)^4}{2\pi\hbar^2m}\frac{T\beta^3}{\mu\omega^3}E_1^2E_2, \qquad (9)$$

and

$$\overline{n_xn_xn_yn_y}=\overline{n_yn_yn_xn_x}=\overline{n_xn_yn_yn_x}=\overline{n_yn_xn_xn_y}$$
$$=\overline{n_xn_yn_xn_y}=\overline{n_yn_xn_yn_x}=\frac{1}{8};\quad\overline{n_x^4}=\overline{n_y^4}=\frac{3}{8}, \qquad (10)$$

whereas the other components (containing single x or y index such as $\overline{n_yn_xn_xn_x}$) vanish. Expressions (7) to (10) describe the general case of CPGE at any polarization and represent the main result of this paper.

III. LINEAR AND CIRCULAR POLARIZATIONS

Let us consider the most interesting cases from experimental point of view, presented in Fig. 1. Choosing $\mathbf{e}_\omega=(1,0)$ and $\mathbf{e}_{2\omega}=(\cos\theta_{2\omega},\sin\theta_{2\omega})$, which corresponds to Fig. 1(a), we find from Eqs. (7) to (10)

$$j_x=2\cos\theta_{2\omega}\mathrm{Re}\,(\chi_{xxxx}+\zeta_{xxxx}), \qquad (11)$$

$$j_y=2\sin\theta_{2\omega}\mathrm{Re}\,(\chi_{yxxy}+\zeta_{yyxx}). \qquad (12)$$

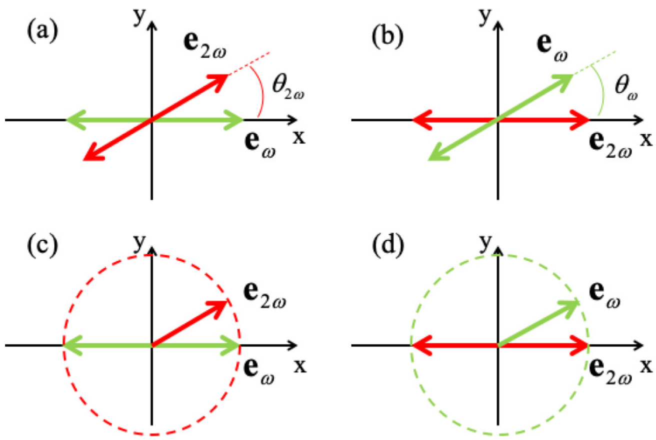

FIG. 1. Geometry of incident fields: Panels (a) and (b) correspond to linear polarization of both the fields, whereas (c) and (d) correspond to the cases of circular polarization of one of the fields (see text for details).

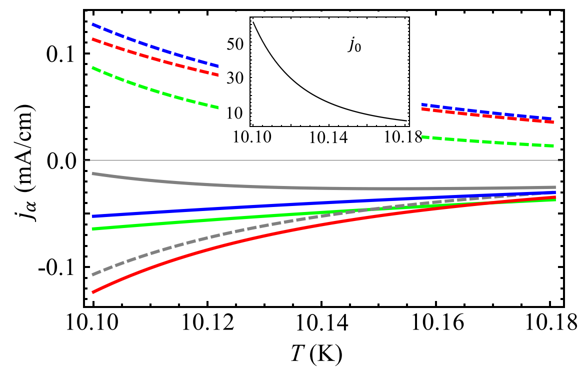

FIG. 2. Components of electric current densities, j_x (solid) and j_y (dashed), as functions of temperature. The green, gray, blue, and red curves correspond to Figs. 1(a), 1(b), 1(c), and 1(d) , respectively. Inset shows the dependence of j_0 on temperature. We used [34,35] $T_c = 10$ K, $n = 10^{14}$ cm^{-2}, $\tau = 0.1$ ps, $m = 0.5\,m_0$, where m_0 is free electron mass. We fixed $\omega = 2 \times 10^{11}\mathrm{s}^{-1}$, $\theta_\omega = \theta_{2\omega} = \pi/6$, $E_1 = 2$ V/cm, and $E_2 = 0.25$ V/cm.

Instead, taking $\mathbf{e}_\omega = (\cos\theta_\omega, \sin\theta_\omega)$ and $\mathbf{e}_{2\omega} = (1, 0)$ we find for Fig. 1(b)

$$j_x = 2\cos^2\theta_\omega \mathrm{Re}\,(\chi_{xxxx} + \zeta_{xxxx}) + 2\sin^2\theta_\omega \mathrm{Re}\,(\chi_{xyyx} + \zeta_{xxyy}), \tag{13}$$

$$j_y = \sin(2\theta_\omega)\mathrm{Re}\,(\chi_{yxyx} + \chi_{yyxx} + \zeta_{yxxy} + \zeta_{yxyx}). \tag{14}$$

Furthermore, taking $\mathbf{e}_\omega = (1, 0)$ and $\mathbf{e}_{2\omega} = (1, i\sigma)/\sqrt{2}$, where $\sigma = \pm 1$ indicates left/right circular polarization, we find for Fig. 1(c)

$$j_x = \sqrt{2}\mathrm{Re}\,(\chi_{xxxx} + \zeta_{xxxx}),$$
$$j_y = \sqrt{2}\sigma\,\mathrm{Im}\,(\zeta_{yyxx} - \chi_{yxxy}). \tag{15}$$

Taking $\mathbf{e}_\omega = (1, i\sigma)/\sqrt{2}$ and $\mathbf{e}_{2\omega} = (1, 0)$ for Fig. 1(d) we have

$$j_x = \mathrm{Re}\,(\chi_{xxxx} + \zeta_{xxxx} - \chi_{xyyx} - \zeta_{xxyy}),$$
$$j_y = \sigma\,\mathrm{Im}\,(\chi_{yxyx} + \chi_{yyxx} - \zeta_{yxxy} - \zeta_{yxyx}). \tag{16}$$

IV. RESULTS AND DISCUSSION

The theory which we developed is applicable to generic two-dimensional superconducting materials. Let us choose a specific material, MoS_2. Its superconducting temperature ranges from around $T_c = 8$ to 18 K; the typical densities of the carrier of charge take the values from $n = 5 \times 10^{13}$ to 1.5×10^{14} cm^{-2}, and realistic electron scattering time is of the order $\tau \sim 0.1$ ps [34,35].

Figure 2 shows the temperature dependence of the electric current density. The components j_x and j_y corresponding to different cases in Fig. 1 exhibit a decay once the temperature increases as compared with the critical temperature (compare also with Fig. 3). It can be explained by the enhancement of the influence of SFs once we approach the critical temperature. In general, the temperature dependence of the current is mainly (although not fully) inherited from the factor j_0, that goes as T^{-2} at large temperatures, $T \gg T_c$, whereas it has a singularity $(T - T_c)^{-3}$ at temperatures approaching T_c (see inset in Fig. 2). This behavior is more singular than the one of the conventional paraconductivity, where the singularity is $(T - T_c)^{-1}$ [26].

Figure 3 shows the spectra of electric current density corresponding to different geometries of incident fields, shown in Fig. 1. In the case of linearly polarized light [Figs. 3(a) and 3(b)], it looks as if the x and y components of the current density show similar behavior: The magnitude of the current density decreases with the increase of frequency. However, analytical analysis (of the derivatives of the current density) demonstrates that this dependence is not monotonous in Fig. 3(a) for the current density j_y. Indeed, at some frequency, j_y for Fig. 3(a) crosses zero and changes its sign, thus it starts to increase. Later, it crosses a maximum and then decreases again. All the components of the current density saturate at high frequencies, independent of temperature.

In the case of circularly polarized light [Figs. 3(c) and 3(d)], the x and y components of electric current density show different behavior. While the x component behaves similar to that of linearly polarized light, the y components behave differently and reveal a nonmonotonous behavior. They also grow and then decay after overcoming a peak value. This nonmonotonous behavior can be explained from the analytical analysis of Eq. (15). In particular, the analytical expressions of $\chi_{\eta\lambda\gamma\delta}$ and $\zeta_{\eta\lambda\gamma\delta}$ on $\beta \propto \omega/(T - T_c)$ determine the behavior of the spectrum presented in Fig. 3. At small β, $j_x/j_0 \propto$ const. $+\, o(\beta)$ and $j_y/j_0 \propto \beta + o(\beta^3)$. In the limit of large β, both the components of current vanish. We also analyzed the behavior of the derivative j_y'/j_0 on frequency. It crosses zero manifesting the extremum of $j_y(\omega)$ dependence. It happens at $\beta \approx 0.44$ for Fig. 3(c), which corresponds to $\omega \approx 2.9 \times 10^{10}$ s^{-1} for $T = 10.10$ K. With the decrease of ϵ, the magnitudes of the currents grow since $\beta \propto \omega/(T - T_c)$. We can also see that once the ambient temperature approaches T_c, the dashed curves (j_y) in Figs. 3(c) and 3(d) get narrower, and the peak frequency experiences a redshift.

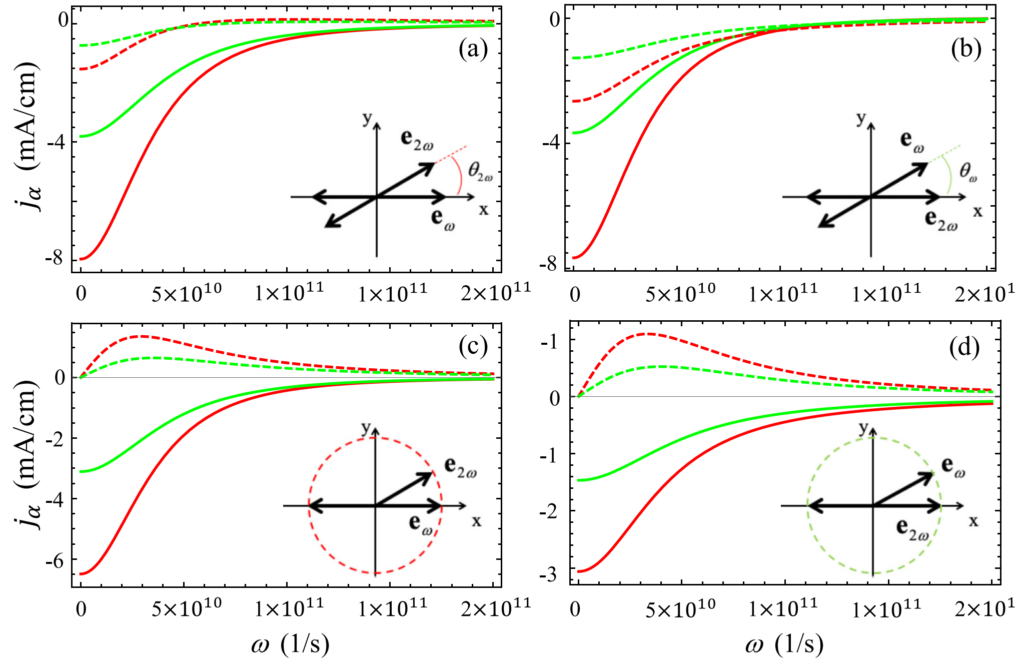

FIG. 3. Spectra of electric current density for different incident field geometries. The solid curves show x-projection of the current j_x as functions of frequency, while the dashed curves show j_y as functions of the same frequencies at two temperatures: 10.10 K (red) and 10.12 K (green) that are above the critical temperature $T_c = 10$ K. Other parameters were taken the same as in Fig. 2.

Figure 4 demonstrates the dependence of current densities on the polarization angles of the light fields. As it follows from Eqs. (11) to (16), the j_x and j_y components of the current depend on the angle only in the cases when both the incident fields have linear polarization. It corresponds to Figs. 1(a) and 1(b). As θ_ω or $\theta_{2\omega}$ varies from 0 to 2π, the magnitudes of current densities change their magnitudes and even sign. We want to note, that for Figs. 1(c) and 1(d) there is an extra factor σ, which reflects the chirality of the field but there is no dependence on the angle.

In this article, we considered one particular type of superconducting fluctuations: the Aslamazov-Larkin corrections. There also take place other contributions: the Maki-Thompson [36,37] and the "density of states" [38] corrections. However, the Boltzmann equations approach cannot be used for their description, and a quantum approach is required.

The Boltzmann equation approach in the case of a uniform alternating field is equivalent to the time-dependent Ginzburg-Landau approach, which can be used in the dirty-sample limit $T\tau \ll 1$ and when $\omega\tau \ll 1$ [39]. Thus, the theory developed in our paper is also limited by this inequality between the external EM field frequency ω and the electron momentum scattering time off impurities τ. In the same time, the parameter $\beta = \pi\omega/(16T_c\epsilon)$ is still arbitrary in the frameworks of the time-dependent Ginzburg-Landau approach (and in our consideration).

Another issue concerns the experimental observation of the predicted effect, which is sensitive to the relative phase between the E_ω and $E_{2\omega}$ fields. For the sake of stationarity and spatial homogeneity of the resulting CPGE electric current, mutual temporal and spatial coherence of the fields is required. Also, the angles of incidence of both the fields should

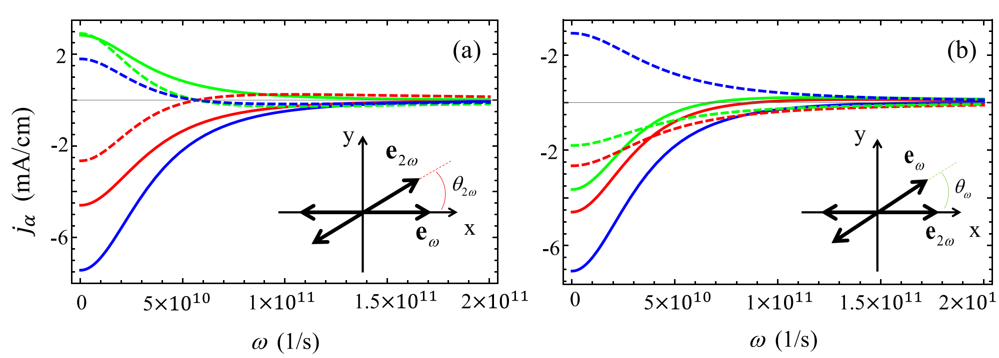

FIG. 4. Spectra of electric current densities corresponding to Figs. 1(a) and 1(b) for three different values of polarization angles (a) $\theta_{2\omega}$ and (b) θ_ω: $\pi/3$ (red), $7\pi/5$ (green), $9\pi/5$ (blue). We used $T = 10.10$ K. Other parameters were taken the same as in Fig. 2.

be smaller than the ratio of the wavelength to the sample size [40].

V. CONCLUSION

We studied the coherent photogalvanic effect in a two-dimensional superconductor in the fluctuating regime. We showed the emergence of a stationary electric current of Cooper pairs when the sample is exposed to two coherent electromagnetic fields of light with certain frequencies and different polarizations. We derived the general formula for the electric current density and investigated in detail the particular cases of linear and circular polarizations of the external light fields. We showed that the current might experience a non-monotonous dependence on frequency and it is very sensitive to the proximity of the temperature to the critical temperature of superconducting transition. In particular, the peak in the spectrum of the current becomes narrower, its frequency experiences a redshift, and the intensity of the peak grows once the temperature approaches T_c. These results capture the effects arising due to the interplay of the physics of superconducting (Cooper pair density) fluctuations and the polarizations of incident light fields.

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APPENDIX: DERIVATION OF THE ELECTRIC CURRENT DENSITY

Integrating by parts in Eq. (4) we find

$$\begin{aligned} j_\eta &= (2e)^2 \int \frac{d\mathbf{p}}{(2\pi)^2} \mathrm{Re}\left(\mathbf{E}^*_\omega f^{(2)}_\omega + \mathbf{E}^*_{2\omega} f^{(2)}_{2\omega}\right) \cdot \frac{\partial(u_\eta \tau_\mathbf{p})}{\partial \mathbf{p}}, \\ f^{(2)}_\omega &= -\frac{2e\tau_\mathbf{p}}{1+i\omega\tau_\mathbf{p}}\left(\mathbf{E}_{2\omega} \cdot \frac{\partial f^{*(1)}_\omega}{\partial \mathbf{p}} + \mathbf{E}^*_\omega \cdot \frac{\partial f^{(1)}_{2\omega}}{\partial \mathbf{p}}\right), \\ f^{(2)}_{2\omega} &= -\frac{2e\tau_\mathbf{p}}{1+2i\omega\tau_\mathbf{p}} \mathbf{E}_\omega \cdot \frac{\partial f^{(1)}_\omega}{\partial \mathbf{p}}. \end{aligned} \tag{A1}$$

Thus, we can split the current density on three terms

$$\begin{aligned} j^{(1)}_\eta &= -(2e)^3 \mathrm{Re} \int \frac{d\mathbf{p}}{(2\pi)^2} \mathbf{E}^*_\omega \cdot \frac{\partial(u_\eta \tau_\mathbf{p})}{\partial \mathbf{p}} \left(\frac{\tau_\mathbf{p}}{1+i\omega\tau_\mathbf{p}} \mathbf{E}_{2\omega} \cdot \frac{\partial f^{*(1)}_\omega}{\partial \mathbf{p}}\right), \\ j^{(2)}_\eta &= -(2e)^3 \mathrm{Re} \int \frac{d\mathbf{p}}{(2\pi)^2} \mathbf{E}^*_\omega \cdot \frac{\partial(u_\eta \tau_\mathbf{p})}{\partial \mathbf{p}} \left(\frac{\tau_\mathbf{p}}{1+i\omega\tau_\mathbf{p}} \mathbf{E}^*_{\omega} \cdot \frac{\partial f^{(1)}_{2\omega}}{\partial \mathbf{p}}\right), \\ j^{(3)}_\eta &= -(2e)^3 \mathrm{Re} \int \frac{d\mathbf{p}}{(2\pi)^2} \mathbf{E}^*_{2\omega} \cdot \frac{\partial(u_\eta \tau_\mathbf{p})}{\partial \mathbf{p}} \left(\frac{\tau_\mathbf{p}}{1+2i\omega\tau_\mathbf{p}} \mathbf{E}_{\omega} \cdot \frac{\partial f^{(1)}_{\omega}}{\partial \mathbf{p}}\right). \end{aligned} \tag{A2}$$

Again integrating by parts, we find

$$\begin{aligned} j^{(1)}_\eta &= (2e)^3 \mathrm{Re} \int \frac{d\mathbf{p}}{(2\pi)^2} f^{*(1)}_\omega \mathbf{E}_{2\omega} \cdot \frac{\partial}{\partial \mathbf{p}} \left(\frac{\tau_\mathbf{p}}{1+i\omega\tau_\mathbf{p}} \mathbf{E}^*_{\omega} \cdot \frac{\partial(u_\eta \tau_\mathbf{p})}{\partial \mathbf{p}}\right), \\ j^{(2)}_\eta &= (2e)^3 \mathrm{Re} \int \frac{d\mathbf{p}}{(2\pi)^2} f^{(1)}_{2\omega} \mathbf{E}^*_{\omega} \cdot \frac{\partial}{\partial \mathbf{p}} \left(\frac{\tau_\mathbf{p}}{1+i\omega\tau_\mathbf{p}} \mathbf{E}^*_{\omega} \cdot \frac{\partial(u_\eta \tau_\mathbf{p})}{\partial \mathbf{p}}\right), \\ j^{(3)}_\eta &= (2e)^3 \mathrm{Re} \int \frac{d\mathbf{p}}{(2\pi)^2} f^{(1)}_{\omega} \mathbf{E}_{\omega} \cdot \frac{\partial}{\partial \mathbf{p}} \left(\frac{\tau_\mathbf{p}}{1+2i\omega\tau_\mathbf{p}} \mathbf{E}^*_{2\omega} \cdot \frac{\partial(u_\eta \tau_\mathbf{p})}{\partial \mathbf{p}}\right). \end{aligned} \tag{A3}$$

Taking into account that

$$\frac{\partial(u_\eta \tau_\mathbf{p})}{\partial p_\lambda} = \frac{\tau_\mathbf{p}}{2m}\left(\delta_{\eta\lambda} - \frac{2m}{\varepsilon_p} u_\eta u_\lambda\right), \tag{A4}$$

we have

$$j_\eta^{(1)} = (2e)^4 \mathrm{Re} \int \frac{d\mathbf{p}}{(2\pi)^2} \frac{\tau_{\mathbf{p}}(-f_0')u_\gamma u_\delta}{2m(1-i\omega\tau_{\mathbf{p}})} \frac{\partial}{\partial \varepsilon_p}\left[\frac{\tau_{\mathbf{p}}^2(\delta_{\eta\lambda} - 2mu_\eta u_\lambda/\varepsilon_p)}{1+i\omega\tau_{\mathbf{p}}}\right] E_{\omega\lambda}^* E_{\omega\gamma}^* E_{2\omega\delta},$$

$$j_\eta^{(2)} = (2e)^4 \mathrm{Re} \int \frac{d\mathbf{p}}{(2\pi)^2} \frac{\tau_{\mathbf{p}}(-f_0')u_\gamma u_\delta}{2m(1+2i\omega\tau_{\mathbf{p}})} \frac{\partial}{\partial \varepsilon_p}\left[\frac{\tau_{\mathbf{p}}^2(\delta_{\eta\lambda} - 2mu_\eta u_\lambda/\varepsilon_p)}{1+i\omega\tau_{\mathbf{p}}}\right] E_{\omega\lambda}^* E_{\omega\gamma}^* E_{2\omega\delta}, \tag{A5}$$

$$j_\eta^{(3)} = (2e)^4 \mathrm{Re} \int \frac{d\mathbf{p}}{(2\pi)^2} \frac{\tau_{\mathbf{p}}(-f_0')u_\gamma u_\delta}{2m(1+i\omega\tau_{\mathbf{p}})} \frac{\partial}{\partial \varepsilon_p}\left[\frac{\tau_{\mathbf{p}}^2(\delta_{\eta\lambda} - 2mu_\eta u_\lambda/\varepsilon_p)}{1+2i\omega\tau_{\mathbf{p}}}\right] E_{2\omega\lambda}^* E_{\omega\gamma} E_{\omega\delta},$$

where $f_0' = \partial f_0/\partial \varepsilon_p$. Introducing dimensionless variables, $\kappa = \varepsilon_p/\mu$ and $\beta = \pi\omega/(16T_c\epsilon)$, then using $\mathbf{E}_\omega = E_0\mathbf{e}_\omega$ and $\mathbf{E}_{2\omega} = E_0\mathbf{e}_{2\omega}$, thus denoting two unity vectors in the directions of electric field harmonics, and parametrizing $\mathbf{u} = u\mathbf{n}$, where $\mathbf{n} = (\cos\varphi, \sin\varphi)$, we find from Eq. (A5)

$$j_\eta^{(1)} = j_0 \mathrm{Re} \int_1^\infty \frac{(\kappa-1)d\kappa}{\kappa^2(\kappa - i\beta)} \frac{\partial}{\partial\kappa}\left[\frac{\delta_{\eta\lambda}\overline{n_\gamma n_\delta} - 2\frac{\kappa-1}{\kappa}\overline{n_\eta n_\lambda n_\gamma n_\delta}}{\kappa(\kappa+i\beta)}\right] e_{\omega\lambda}^* e_{\omega\gamma}^* e_{2\omega\delta}, \tag{A6}$$

$$j_\eta^{(2)} = j_0 \mathrm{Re} \int_1^\infty \frac{(\kappa-1)d\kappa}{\kappa^2(\kappa + 2i\beta)} \frac{\partial}{\partial\kappa}\left[\frac{\delta_{\eta\lambda}\overline{n_\gamma n_\delta} - 2\frac{\kappa-1}{\kappa}\overline{n_\eta n_\lambda n_\gamma n_\delta}}{\kappa(\kappa+i\beta)}\right] e_{\omega\lambda}^* e_{\omega\gamma}^* e_{2\omega\delta}, \tag{A7}$$

$$j_\eta^{(3)} = j_0 \mathrm{Re} \int_1^\infty \frac{(\kappa-1)d\kappa}{\kappa^2(\kappa + i\beta)} \frac{\partial}{\partial\kappa}\left[\frac{\delta_{\eta\lambda}\overline{n_\gamma n_\delta} - 2\frac{\kappa-1}{\kappa}\overline{n_\eta n_\lambda n_\gamma n_\delta}}{\kappa(\kappa+2i\beta)}\right] e_{2\omega\lambda}^* e_{\omega\gamma} e_{\omega\delta}, \tag{A8}$$

where

$$\overline{n_\gamma n_\delta} = \int_0^{2\pi} \frac{d\varphi}{2\pi} n_\gamma n_\delta = \frac{1}{2}\delta_{\gamma\delta},$$

$$\overline{n_\eta n_\lambda n_\gamma n_\delta} = \int_0^{2\pi} \frac{d\varphi}{2\pi} n_\eta n_\lambda n_\gamma n_\delta.$$

Straightforward calculations give

$$\overline{n_x n_x n_y n_y} = \overline{n_y n_y n_x n_x} = \overline{n_x n_y n_y n_x} = \overline{n_y n_x n_x n_y} = \overline{n_x n_y n_x n_y} = \overline{n_y n_x n_y n_x} = \frac{1}{8}, \tag{A9}$$

$$\overline{n_x^4} = \overline{n_y^4} = \frac{3}{8}, \tag{A10}$$

whereas the other components (containing single x or y index such as $\overline{n_y n_x n_x n_x}$) vanish. Using Eqs. (A6) to (A10), we find the total current density described by Eqs. (7) and (8) in the main text.

[1] T. Oka and H. Aoki, Phys. Rev. B **79**, 081406(R) (2009).

[2] H. Ishizuka, T. Hayata, M. Ueda, and N. Nagaosa, Phys. Rev. Lett. **117**, 216601 (2016).

[3] E. J. König, H.-Y. Xie, D. A. Pesin, and A. Levchenko, Phys. Rev. B **96**, 075123 (2017).

[4] T. Rangel, B. M. Fregoso, B. S. Mendoza, T. Morimoto, J. E. Moore, and J. B. Neaton, Phys. Rev. Lett. **119**, 067402 (2017).

[5] V. M. Kovalev and I. G. Savenko, Phys. Rev. B **99**, 075405 (2019).

[6] J. Seidel, D. Fu, S.-Y. Yang, E. Alarcón-Lladó, J. Wu, R. Ramesh, and J. W. Ager III, Phys. Rev. Lett. **107**, 126805 (2011).

[7] S. M. Young and A. M. Rappe, Phys. Rev. Lett. **109**, 116601 (2012).

[8] C. Somma, K. Reimann, C. Flytzanis, T. Elsaesser, and M. Woerner, Phys. Rev. Lett. **112**, 146602 (2014).

[9] B. I. Sturman and V. M. Firdkin, *The Photovoltic and Photorefractive Effects in Non-centrosymmetric Materials* (Gordon and Breach, Philadelphia, 1992).

[10] A. M. Glass, D. von der Linde, and T. J. Negran, Appl. Phys. Lett. **25**, 233 (1974).

[11] H. Dember, Phys. Z. **32**, 554 (1931).

[12] V. I. Belinicher and B. I. Sturman, Sov. Phys. Usp. **23**, 199 (1980).

[13] E. L. Ivchenko and G. E. Pikus, *Problemy Sovremennoi Fiziki* (Nauka, Leningrad, 1980).

[14] S. D. Ganichev and W. Prettl, *Intense Terahertz Excitation of Semiconductors* (Oxford University Press, Oxford, 2006).

[15] M. V. Entin, L. I. Magarill, and V. M. Kovalev, J. Phys.: Cond. Mat. **31**, 325302 (2019).

[16] A. V. Kalameitsev, V. M. Kovalev, and I. G. Savenko, Phys. Rev. Lett. **122**, 256801 (2019).

[17] M. V. Entin, Phys. Tech. Semicond. **23**, 1066 (1989).

[18] E. M. Baskin and M. V. Entin, Pis'ma Zh. Eksp. Teor. Fiz. **48**, 554 (1988) [JETP Lett. **48**, 601 (1988)].

[19] M. K. Balakirev and V. A. Smirnov, Pis'ma Zh. Eksp. Teor. Fiz. **61**, 537 (1995) [JETP Lett. **61**, 544 (1995)].

[20] M. K. Balakirev, L. J. Vostrikova, V. A. Smirnov, and M. V. Entin, Pis'ma Zh. Eksp. Teor. Fiz. **63**, 166 (1996) [JETP Lett. **63**, 176 (1996)].

[21] K. H. A. Villegas, V. M. Kovalev, F. V. Kusmartsev, and I. G. Savenko, Phys. Rev. B **98**, 064502 (2018); K. H. A. Villegas, F. V. Kusmartsev, Y. Luo, and I. G. Savenko, Phys. Rev. Lett. **124**, 087701 (2020).

[22] M. V. Boev, Phys. Rev. B **101**, 104512 (2020).

[23] M. Silaev, Phys. Rev. B **99**, 224511 (2019).

[24] S. Li and A. Levchenko, Ann. Phys. (NY) **417**, 168137 (2020).

[25] L. G. Aslamazov and A. I. Larkin, Fiz. Tverd. Tela **10**, 1104 (1968) [Sov. Phys. Solid State **10**, 875 (1968)].

[26] A. Larkin and A. Varlamov, *Theory of Fluctuations in Superconductors* (Oxford University Press, Oxford, 2005).

[27] V. M. Kovalev and I. G. Savenko, Phys. Rev. Lett. **124**, 207002 (2020).

[28] T. Mishonov and D. Damianov, Czech. J. Phys. **46**, 631 (1996).

[29] D. Damianov and T. Mishonov, Superlat. Microstr. **21**, 467 (1997).

[30] T. M. Mishonov, A. I. Posazhennikova, and J. O. Indekeu, Phys. Rev. B **65**, 064519 (2002).

[31] T. M. Mishonov, G. V. Pachov, I. N. Genchev, L. A. Atanasova, and D. Ch. Damianov, Phys. Rev. B **68**, 054525 (2003).

[32] C. Kittel, *Quantum Theory of Solid States* (Wiley, New York, 2004).

[33] A. A. Abrikosov, *Fundamentals of the Theory of Metals* (Dover, Mineola, NY, 2017).

[34] R. Wakatsuki, Y. Saito, S. Hoshino, Y. M. Itahashi, T. Ideue, M. Ezawa, Y. Iwasa, and N. Nagaosa, Sci. Adv. **3**, e1602390 (2017).

[35] Y. Saito, Y. Nakamura, M. S. Bahramy, Y. Kohama, J. Ye, Y. Kasahara, Y. Nakagawa, M. Onga, M. Tokunaga, T. Nojima, Y. Yanase, and Y. Iwasa, Nat. Phys. **12**, 144 (2016).

[36] K. Maki, Prog. Theor. Phys. **40**, 193 (1968).

[37] R. S. Thompson, Phys. Rev. B **1**, 327 (1970)

[38] L. G. Aslamazov and A. I. Larkin, Zh. Eksp. Teor. Fiz. **67**, 647 (1974) [Sov. Phys.-JETP **40**, 321 (1975)].

[39] A. G. Aronov, S. Hikami, and A. I. Larkin, Phys. Rev. B **51**, 3880 (1995).

[40] V. M. Kovalev, W.-K. Tse, and M. V. Entin, Zh. Eksp. Teor. Fiz. **106**, 549 (2017) [JETP Lett. **106**, 565 (2017)].