Letter

Photoinduced anomalous supercurrent Hall effect

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We predict a photoinduced Hall effect in an isotropic conventional two-dimensional superconductor with a built-in supercurrent exposed to a circularly polarized light. This second-order with respect to the electromagnetic field amplitude effect occurs when the frequency of the field exceeds double the value of the superconducting gap. It reveals itself in the emergence of a Cooper-pair condensate flow in the direction transverse to the initial built-in supercurrent, which arises to compensate for the light-induced electric current of quasiparticles photoexcited across the gap. The initial supercurrent breaks both the time-reversal and inversion symmetries, while the presence of dilute disorder in the sample provides the breaking of the Galilean invariance. We develop a microscopic theory of the supercurrent Hall effect in the case of weak disorder and show that the Hall supercurrent is directly proportional to the quasiparticle recombination time, which can acquire large values.

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Introduction. The measurement of the optical response in superconductors is a powerful experimental technique to explore their quantum properties, which have been the focus of research for more than 50 years [1–9]. Despite such an extensive period of time, the study of the interaction of electromagnetic (EM) fields with superconductors remains a challenging topic since, as it is known, superconducting (SC) samples usually expel external EM fields [10]. Besides being of fundamental importance, the research on light-controlled transport of Cooper pairs is aimed at applications [11,12]. Examples of possible (but not yet implemented) light-matter interaction phenomena in superconductors include various nonlinear and higher-order response effects [13–15], in particular, the electric-field-induced enhancement of SC properties [16,17], giant second-harmonic generation under supercurrent injection [18], and light-mediated superconductivity [19–21], to name a few. Another route, which we inspect in this Letter, would be a photoinduced anomalous Hall effect—a possibility to manipulate a dc supercurrent flow by utilizing a Hall-like response.

The anomalous Hall effect in non-SC samples represents a stationary transport phenomenon, which constitutes the emergence of a transverse component of electric current in the absence of an external magnetic field [22]. The examples are the spin Hall effect, where the spin-orbit interaction plays the role of the magnetic field, the valley Hall effect [23–26] in two-dimensional (2D) Dirac materials [27–30], and the photoinduced anomalous Hall effect, actively studied in various systems [31]. Is it possible to find a setup for such an effect to involve Cooper pairs?

Before answering this question, let us briefly review the optical response theory in superconductors. As it is known, in clean single-band Bardeen-Cooper-Schrieffer (BCS) superconductors [32], the presence of particle-hole and inversion symmetries does not allow for momentum-conserving optical transitions [10,32,33]. The optical excitations under inversion symmetry can occur if they account for either impurity scattering [33,34] or a multiband structure [35,36]. The first theoretical analysis of the dynamical conductivity of superconductors exposed to EM fields with the frequency exceeding the SC gap was made by Mattis and Bardeen [34], who considered the "dirty case". They showed that in the absence of electron scattering on impurities ("clean case"), optical transitions across the SC gap exerted by a uniform light are forbidden. The reason is that the holelike and electronlike states are orthogonal to each other, and thus they give vanishing matrix elements describing the optical transitions across the SC gap. Soon, the Mattis-Bardeen theory was tested in a number of experiments [37]; it was also generalized to the case of strong electron-phonon interactions [38,39] and superconductors with an arbitrary electron mean free path [40].

Nevertheless, an optical transition can still take place in clean superconductors with broken inversion symmetry or in the presence of a spin-orbit interaction [35]. Inversion symmetry here can be broken in the presence of a built-in supercurrent [36,41,42]. However, the presence of a supercurrent is not a sufficient condition for optical transitions to

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occur since the Galilean invariance in parabolic single-band superconductors suppresses the transitions [4].

Two ways to break the Galilean symmetry are known: (i) by accounting for the nonparabolicity of electronic bands [41,43] and (ii) by accounting for the electron-impurity scattering. Both these scenarios have been realized for frequencies exceeding double the value of the SC gap. Recently, it became clear [44,45] that various relaxation time parameters may play an important role depending on the ratio between the EM field frequency and SC gap. It was shown that at low frequencies, the optical conductivity in the presence of a supercurrent is proportional to the inelastic electron relaxation time, and not to the elastic one. At low frequencies and temperatures close to the SC critical temperature T_c, the inelastic time is determined by the energy relaxation processes of quasiparticles. The energy relaxation time being much larger than the elastic one may result in giant optical conductivity and power absorption in the presence of a supercurrent-carrying state [44,45].

In this Letter, we show that even in a single-band BCS superconductor [32], the breaking of both inversion and time-reversal symmetries by means of a built-in supercurrent, and the breaking of Galilean invariance by (weak) electron-impurity scattering results in the photoinduced transport of a Cooper-pair condensate in the direction transverse to the built-in supercurrent. Hereby we define the photoinduced anomalous supercurrent Hall effect. At temperatures $k_BT \ll \Delta$ with Δ the SC order parameter, the equilibrium density of quasiparticles above the gap is negligibly small in the absence of external radiation. Therefore, we expect that the photoinduced Hall current should be proportional to the inelastic relaxation time τ_R associated with the recombination processes of quasiparticles across the gap. Then, large τ_R at low temperatures will provide large values of the EM field-induced supercurrent, opening a way for the experimental verification of its existence.

The idea behind the photoinduced supercurrent Hall effect can be roughly explained using phenomenological arguments. Let us consider a 2D layer with a built-in stationary supercurrent generated either by a transport current or an external applied magnetic field. The supercurrent is a consequence of the nonzero supermomentum $\mathbf{p}_s$ of the Cooper pairs, associated with the phase difference of the condensate at the edges of the sample. Furthermore, if an isotropic 2D superconductor in a supercurrent-carrying regime is normally illuminated by an external EM radiation characterized by the in-plane vector potential $\mathcal{A}(t) = \mathcal{A}\exp(-i\omega t) + \mathcal{A}^*\exp(i\omega t)$, the emerging photoinduced stationary current of quasiparticles excited across the SC gap in the most general form reads

$$\mathbf{j} = a_\omega|\mathcal{A}|^2\mathbf{p}_s + b_\omega[\mathcal{A}(\mathcal{A}^*\cdot\mathbf{p}_s) + (\mathcal{A}\cdot\mathbf{p}_s)\mathcal{A}^*] + ic_\omega[\mathbf{p}_s\times[\mathcal{A}\times\mathcal{A}^*]]. \quad (1)$$

The first term in Eq. (1) gives the longitudinal (aligned along the supercurrent) photoexcited current density; the second term contains both the longitudinal and transverse quasiparticle currents; the third term gives only the transverse response [46]. Anisotropic contributions characterized by the coefficients b_ω and c_ω are induced by linearly and circularly polarized radiation, respectively.

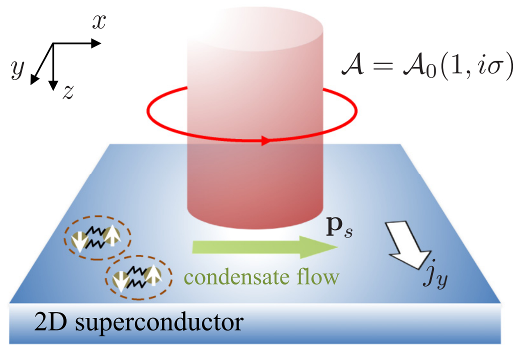

FIG. 1. System schematic: A two-dimensional superconductor with a built-in supercurrent exposed to an external EM field normally incident to the 2D plane. The EM field has a circular polarization, characterized by the vector potential $\mathcal{A}$. The built-in supercurrent, which represents a Cooper-pair condensate flow, is aligned along the x axis, and it is described by the momentum $\mathbf{p}_s$. A photoinduced quasiparticle current j_y emerges in the transverse direction.

The phenomenological expression (1) is similar to the equation which describes the stationary photoinduced transport phenomena in non-SC 2D systems [47]. These effects are stationary since the corresponding response is proportional to $\mathcal{A}_\alpha\mathcal{A}^*_\beta$ [see Eq. (1)]. In non-SC systems, the role of $\mathbf{p}_s$ is played by an external in-plane static electric field $\mathbf{F}$. The corresponding longitudinal current is known as *photoconductivity*, whereas the transverse part is referred to as *the photovoltaic Hall effect* [47]. That transverse response is the result of the emergence of a photoinduced correction to the distribution function of photoelectrons $\delta f(\mathbf{v}, \mathbf{E}, \mathbf{F})$. Indeed, the theoretical analysis based on the Boltzmann transport equation shows [47] that there emerge third-order corrections to the electron distribution function, $\delta f(\mathbf{v}, \mathbf{E}, \mathbf{F}) \propto (\mathbf{vE})(\mathbf{vE}^*)(\mathbf{vF})$. These corrections are of an anisotropic form reflecting the phenomenon called *the anisotropic alignment of photoelectrons* [47], which constitutes the physical reason for the photovoltaic Hall effect in the case of a circularly polarized EM field.

As we are interested in the Hall-like transport in superconductors, we focus on the transverse component of the current (1), $j_y = b_\omega(\mathcal{A}_x\mathcal{A}^*_y + \mathcal{A}^*_x\mathcal{A}_y)p_s + ic_\omega(\mathcal{A}_x\mathcal{A}^*_y - \mathcal{A}^*_x\mathcal{A}_y)p_s$, by choosing the direction of condensate flow along the x axis, $\mathbf{p}_s = (p_s, 0)$, as in Fig. 1. Using the terminology of the two-fluid model, we call j_y the photoexcited current of quasiparticles contributing to the normal-state component of electron fluid. It provides an accumulation of carriers of charge at the transverse boundaries of the sample. In the case of a non-SC material, such an accumulation results in the emergence of the Hall electric field. In the case of a SC material, instead, the electric field cannot penetrate the SC sample. Therefore, the transverse quasiparticle current j_y should be accompanied by an induced *transverse* condensate flow j_s in such a way that the Hall electric field is compensated and thus

$$j_s + j_y = 0. \quad (2)$$

This current-compensation relation reflects a general property of superconductors [48]. It is based on the fact that the electric field inside the bulk of a SC sample vanishes, which

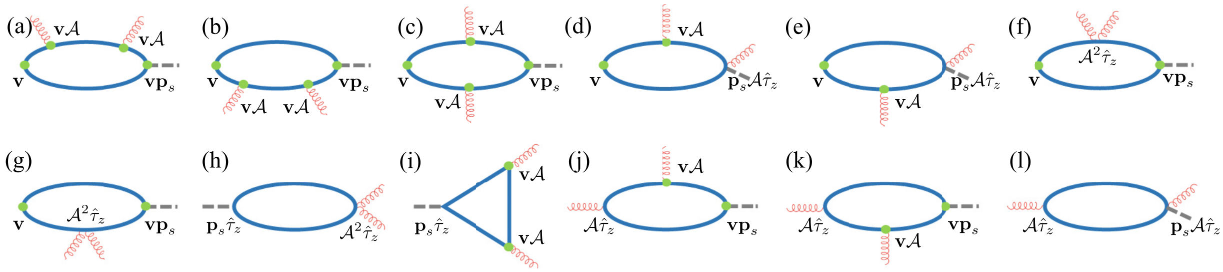

FIG. 2. Feynman diagrams describing the photoinduced quasiparticle electric current. Blue lines stand for the Green's functions of quasiparticles, red wavy lines indicate the external EM field $\mathcal{A}$, green circles represent the quasiparticle velocity vertex $\mathbf{v}$, and dashed lines denote the supercurrent momentum $\mathbf{p}_s$.

is commonly used in theory and experiments on thermoelectric, acoustoelectric, and photoelectric transport phenomena in superconductors [18,49–51]. A more precise microscopic study shows that the electric field does penetrate into the SC sample up to a distance λ, which is of the same order as the SC coherence length at low temperatures $T \ll T_c$, but at higher temperatures, $T - T_c \ll T_c$, λ is determined by the quasiparticle energy relaxation processes and it may reach an extremely large value, much greater than the SC coherence length [52,53]. Here, we study the former case, $T \ll T_c$, and thus the only requirement is that the width of the sample should be much bigger than λ.

Even though the net transverse current vanishes in the volume of a 2D SC sample, the emergence of j_s produces a Hall-like condensate phase difference on the transverse boundaries of the sample, $\Delta\phi_{\rm H} \propto -j_y w$, where w is the width of the sample in the y direction. This phase difference is directly related to the coefficients b_ω and c_ω, which determine the quasiparticle optical response across the SC gap.

Here, we focus on the Hall-like response in the case of a circularly polarized external EM field $\mathcal{A} = \mathcal{A}_0(1, i\sigma)$, where $\sigma = \pm 1$ indicates the left/right polarization of the EM field. Then, the response is determined by the last term in Eq. (1) and thus the coefficient c_ω. In 2D superconductors, this term is also (as in the non-SC case) due to the anisotropic corrections to the distribution function of photoexcited quasiparticles. A theoretical description of such processes in superconductors cannot be treated via the Boltzmann equation approach, as they are only applicable in the case of small EM field frequencies, $\omega \ll 2\Delta$, and thus the processes of quasiparticle photoexcitations across the SC gap with $\omega > 2\Delta$, are beyond its applicability [54]. Instead, a microscopic theory based on the Green's functions technique is required.

The microscopic approach to the photoinduced current density. In the absence of relaxation processes, the Hamiltonian of a 2D superconductor with an isotropic s-type BCS pairing exposed to an external EM field reads (in $\hbar = k_B = c = 1$ units)

$$\hat{H} = \begin{pmatrix} \xi(\mathbf{p} - \mathbf{p}_s - e\mathcal{A}(t)) & \Delta \\ \Delta & -\xi(\mathbf{p} + \mathbf{p}_s + e\mathcal{A}(t)) \end{pmatrix}. \quad (3)$$

Here, $\xi(\mathbf{p}) \equiv \xi_p = \mathbf{p}^2/2m - E_F$ is the electron kinetic energy measured from the Fermi energy E_F, and we assume Δ to be real valued. The current density operator and the current density obey the standard relations,

$$\hat{\mathbf{j}} = -\frac{\delta \hat{H}}{\delta \mathcal{A}}, \quad \mathbf{j}(t) = -i\,\mathrm{Sp}\{\hat{\mathbf{j}}\,\hat{\mathcal{G}}^<(t,t)\}, \quad (4)$$

where $\hat{\mathcal{G}}^<(t,t)$ is a lesser component of the Green's function defined by the matrix equation $(i\partial_t - \hat{H})\hat{\mathcal{G}}(t-t') = \delta(t-t')$ in the Nambu and Keldysh representation. Expanding Eq. (4) up to first order with respect to $\mathbf{p}_s$ and up to second order with respect to $\mathcal{A}(t)$ yields a set of Feynman diagrams for the stationary current shown in Fig. 2.

In the absence of impurities, a single-band superconductor with a parabolic electron dispersion possesses Galilean invariance with or without a built-in supercurrent. Consequently, optical absorption vanishes in both cases. Meanwhile, the second-order stationary response is a consequence of photoabsorption across the gap. Thus, it vanishes in a clean case both in the absence and presence of the supercurrent. The inspection of all the diagrams in Fig. 2 confirms this statement (see Supplemental Material [55]), except one diagram shown in Fig. 2(l). The calculation of this diagram gives a nonzero current density, which seemingly violates the Galilean invariance of the theory. To restore the Galilean invariance, one has to account for additional terms reflecting the BCS electron-electron interaction-induced vertex corrections [43,55].

The optical absorption and the photoinduced electric current (1) acquire finite values when Galilean invariance is violated [43]. We consider the case when the violation occurs due to the presence of electron-impurity scattering in the sample, characterized by an effective lifetime τ_i. Another important ingredient for the effect to take place is by taking account of the relaxation processes of the photoexcited quasiparticles, characterized by τ_R. These processes restrict the accumulation of the photoexcited quasiparticles above the SC gap leading to a stationary regime with a stationary but nonequilibrium distribution function.

Altogether, the relaxation processes can be described by the effective inverse lifetime

$$\frac{1}{\tau_p} = \frac{1}{\tau_i}\frac{|\xi_p|}{\epsilon_p} + \frac{1}{\tau_R}, \quad (5)$$

where $\epsilon_p = \sqrt{\xi_p^2 + \Delta^2}$ is a quasiparticle dispersion with $\xi_p = (p^2 - p_F^2)/2m$. We will assume that under external EM radiation, photoexcited quasiparticles are generated at the gap edge, $2\Delta \gg \omega - 2\Delta > 0$, possessing the momentum $p \approx p_F$ ($|\xi_p| \to 0$). In this case, the dominant inelastic process is the recombination across the SC gap since $\tau_p = \tau_i\tau_R\epsilon_p/(\tau_R|\xi_p| + \tau_i\epsilon_p) \approx \tau_R$ at $|\xi_p| \to 0$.

At temperatures $T \ll \Delta$ and at the bottom of the quasiparticle branch, the recombination time reads [56,57]

$$\frac{1}{\tau_R} \propto \frac{T}{E_F} e^{-2\Delta/T} \max\{\Delta, 1/\tau_i\}. \tag{6}$$

This expression holds both for the clean $\Delta\tau_i \gg 1$ and dirty $\Delta\tau_i \lesssim 1$ cases. Since any recombination process represents a transition across the SC gap back to the SC condensate after forming a Cooper pair, the intensity of such a process is proportional to the thermal density of quasiparticles above the gap. This density is small at low temperatures, which is reflected in the exponential factors in Eq. (6). Thus, τ_R can acquire large values.

To analyze the current in a superconductor with impurities, we again address the Feynman diagrams in Fig. 2. Figures 2(f)–2(i) give only a longitudinal contribution, while Figs. 2(a)–2(c) result in both a longitudinal and transverse response if exposed to a linearly polarized EM field (which we also do not consider here). The remaining diagrams, Figs. 2(d), 2(e), and 2(j)–2(l), describe the longitudinal and transverse response in both cases of linear and circular light polarization. For circularly polarized light, Figs. 2(d) and 2(e) reveal the major contribution in the vicinity of the resonance $\omega \approx 2\Delta$ [58].

After some algebra, we find the transverse current density at tending to zero temperature [55],

$$j_y = \sigma\frac{8e^3}{m}p_s\mathcal{A}_0^2\sum_{\mathbf{p}}\gamma v_y^2\frac{u^2v^2(u^2-v^2)}{(2\epsilon_p-\omega)^2+\left(1/\tau_p^2\right)}, \tag{7}$$

where $\gamma^{-1} = 2\tau_i|\xi_p|$; $u^2 = (1+\xi_p/\epsilon_p)/2$ and $v^2 = (1-\xi_p/\epsilon_p)/2$ are Bogoliubov coefficients. The integration over the momentum $\mathbf{p}$ in Eq. (7) can be performed in a general form [55]. However, the formula can be additionally simplified using the substitution $[(2\epsilon_p-\omega)^2 + 1/\tau_p^2]^{-1} \to \pi\tau_p\delta(\omega - 2\epsilon_p)$, where $\tau_p \approx \tau_R$ at $p \approx p_F$. Restoring the dimensionality, we find the transverse photocurrent,

$$j_y = \sigma\frac{e^3}{2m\hbar^2}p_s\mathcal{A}_0^2\frac{\Delta^2}{\hbar^2\omega^2}\frac{\tau_R}{\tau_i}\Theta(\hbar\omega - 2\Delta), \tag{8}$$

which is determined by a large parameter τ_R/τ_i. Formulas (7) and (8) are the central results of this Letter. Figure 3 shows a normalized $c_\omega = j_y/\mathcal{A}_0^2$ for different parameters.

Discussion. First, Eq. (8) confirms that the Hall-like transport is absent in clean SC samples, as it is required by Galilean invariance. Indeed, in a clean limit, $\tau_i \to \infty$, the recombination time does not depend on the electron-impurity scattering time, Eq. (6), and thus in Eq. (8) the current is inversely proportional to τ_i, consequently vanishing in clean samples.

Next, let us estimate the magnitude of transverse photocurrent density and compare it with the built-in longitudinal supercurrent. For $p_s/m = j_s/(en_s)$, $\omega \sim 1$ THz, $n_s = 10^{13}$ cm^{-2} [59], $\tau_i = 10^{-11}$ s^{-1}, $\tau_R = 10^{-7}$ s^{-1}, and for the EM field intensity $I = 1$ W/cm^2, we find $j_y/j_s \sim 0.1$. Thus, the transverse current represents a correction to the built-in current, as expected, but a considerable correction. As concerns the longitudinal photocurrent, Figs. 2(d) and 2(e) give an estimation

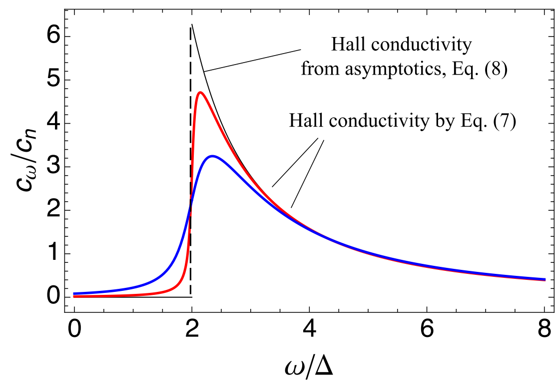

FIG. 3. Circularly polarized EM field-induced nonlinear transverse conductivity as a function of the normalized frequency of the EM field [coefficient c_ω from Eq. (1) normalized on $c_n = (e^3/4m\pi)(\tau_R/\tau_i)$ as a function of ω/Δ in $\hbar = 1$ units]. The calculations were performed using Fig. 2(d). [Figure 2(c) gives the same contribution.] Blue, red, and black curves correspond to the normalized relaxation times $\tau_R\Delta = 4, 20, \infty$, correspondingly.

$j_x \approx j_y/(\omega\tau_i)$, and for $\omega\tau_i \gg 1$, the longitudinal correction to current is smaller than the transverse current.

It is important to note that the developed theory is fully gauge invariant in the xy plane. In derivations, we used the gauge $\varphi = 0$ for the EM field with a normal incidence to the 2D plane, and considered the second-order response, which is transverse to the direction of EM wave propagation. None of the collective modes is excited in the SC sample in this case. However, in the case of an oblique incidence of the external EM field (not considered here), the long-range Coulomb forces appear in the sample, and then the gauge invariance requires taking into account the collective excitations of the order parameter [4,60].

Furthermore, we assumed that the given built-in supercurrent does not affect the characteristics of the relaxation processes, which play an essential role in the theory. This assumption is valid since we only consider linear in p_s photocurrent (1). In principle, the electron-impurity scattering time and the quasiparticle recombination time being scalars may only acquire corrections proportional to p_s^2 or higher orders, which is beyond our consideration.

As for the possible experiments, a typical setup to measure the transverse phase difference is by enclosing the transverse edges of the sample (in the y direction) by a superconducting loop. In this case, the Hall-like photoinduced phase difference produces a correction $\delta\Phi \propto j_y$ to flux through the loop, $\Phi = n\Phi_0 + \delta\Phi$, with Φ_0 the flux quantum and n an integer. Such a flux correction can be measured by a superconducting quantum interference device (SQUID) with high accuracy. A similar experimental technique was used to study the thermoelectric-induced condensate phase difference [50], and it was also proposed for the detection of acoustoelectric phenomena in superconductors [49].

Another method requires attaching a normal-metal probe to the transverse sample edges to measure directly the photoinduced quasiparticle current at the normal metal–insulator–superconducting (NIS) junction [61]. This method was also used to measure the thermoelectric-induced condensate phase difference in superconductors [62].

Finally, there exists the possibility of a measurement by optical methods [12,42], in particular, by the Kerr (or Faraday) effect. There, the Hall-like response can be detected through the change in polarization of the reflected or transmitted EM field [63,64].

Conclusion. We developed a theory of a photoresponse in a single-band 2D isotropic superconductor with a built-in supercurrent, exposed to an external circularly polarized electromagnetic field. This theory accounts for the presence of impurities in the sample (via a random impurity potential), which destroys the Galilean invariance for the transverse transport to take place. Using this theory, we predicted a photoinduced second-order transport phenomenon—the emergence of a transverse (Hall-like) photoinduced supercurrent—and demonstrated that its magnitude is primarily determined by the quasiparticle recombination time. This photoinduced supercurrent Hall effect opens a way to manipulate the direction of superconducting condensate flow via optical tools without external magnetic fields.

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