Letter

Theory for anomalous terahertz emission in striped cuprate superconductors

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Recent experiments in the doped cuprates $La_{2-x}Ba_xCuO_4$ have revealed the emission of anomalous terahertz radiation after impulsive optical excitation. Here, we theoretically investigate the nonlinear electrodynamics of such striped superconductors and explore the origin of the observed radiation. We argue that photoexcitation is converted into a photocurrent by a second-order optical nonlinearity, which is activated by the breaking of inversion symmetry in certain stripe configurations. We point out the importance of including umklapp photocurrents modulated at the stripe periodicity itself, which impulsively drive surface Josephson plasmons and lead to a resonant structure of outgoing radiation, consistent with the experiments. We speculate on the utility of the proposed mechanism in the context of generating tunable terahertz radiation.

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Electronic phases of quantum matter are typically distinguished by their signature electromagnetic responses [1–8]. For instance, in strongly anisotropic materials, the onset of superconductivity manifests as the appearance of Josephson plasmon (JP) edges in the terahertz reflectivity [9–12]. Likewise, states with an incommensurate charge density wave (CDW) exhibit a characteristic quasiparticle gap in the low-frequency conductivity, accompanied by resonance at the pinning frequency [13–15]. Recent experiment [16] in the doped single-layer cuprates $La_{2-x}Ba_xCuO_4$ (LBCO) demonstrated a unique nonlinear optical signature of the "superstripe" phase, i.e., when both superconducting and stripe orders are present and intertwined [17–22]. The discovered phenomenon can be summarized as follows: upon a strong optical pump pulse, outgoing radiation is observed at terahertz frequencies, with a spectrum peaked at the Josephson plasmon resonance (JPR). These findings are surprising because these materials are nominally centrosymmetric, and such radiation is not expected in the absence of a current or magnetic field bias. In this Letter, we provide a theoretical interpretation of these experimental observations. Crucially, this nonlinear effect cannot be understood from the perspective of either the superconducting or stripe orders individually and requires combining both types of symmetry breaking simultaneously.

The phenomenon of "radiating stripes" in LBCO provides several important insights into the nature of this striped superconductor. The presence of a subharmonic optical response constrains the symmetries of the stripe order [23] and, in particular, implies inversion symmetry is broken [24–28]. Consequently, the impulsive optical excitation is now coupled to both bulk and surface Josephson plasmons, which are otherwise symmetry-odd infrared-active modes [29]. We argue that the outgoing terahertz radiation originates from the surface Josephson plasmons and is sensitive to the stripe order, which outcouples these otherwise silent modes. From the observation that the terahertz radiation is sharply peaked at the Josephson plasmon resonance, we can infer that a photocurrent is being generated both at zero momentum and at momenta corresponding to the reciprocal vectors of the CDW lattice, yielding insight into the microscopic physics of the striped state.

The key finding of this Letter can be summarized as follows. The role of the CDW order is that it (i) makes the surface modes optically active (Fig. 1) and (ii) gives rise to a nonzero umklapp photocurrent [24]. We show that umklapp photocurrents resonantly drive the surface plasmons, which exhibit a large density of states near the Josephson plasma resonance $\omega_{JPR} \simeq 0.5$ THz, thereby resulting in sharp in frequency radiation (Fig. 2) consistent with the experiment of Ref. [16]. We point out that the proposed mechanism is generic and potentially useful for designing platforms for the generation of tunable terahertz radiation [30–34]. We remark that umklapp currents carry a large momentum of the CDW reciprocal lattice so that nominally, the resulting emitted light is expected to be far away from the light cone and, thus, decay

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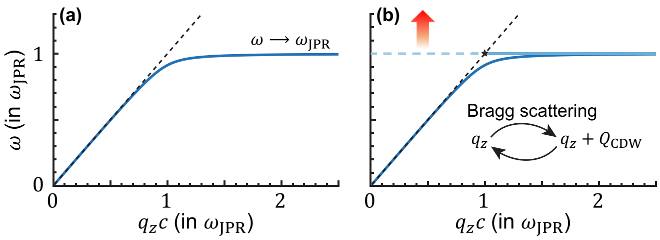

FIG. 1. (a) Dispersion $\omega_s(q_z)$ of the surface Josephson plasmons. In strongly anisotropic superconductors, ω_s quickly saturates at $\omega_{\rm JPR}$. Dashed line corresponds to the light cone $\omega = cq_z$. (b) Schematic of the surface-plasmon spectrum in the presence of a weak stripes order, which couples momenta q_z to $q_z + Q_{\rm CDW}$. As such, the plasmon dispersion exhibits backfolding to the reduced Brillouin zone, defined by the CDW wave vector $Q_{\rm CDW}$. Notably, the surface plasmons that backfold inside the light cone can now radiate out.

on a length scale of the order of a few wavelengths at most. Therefore, the proposed mechanism of resonant coupling to the surface collective modes provides a pathway towards detecting umklapp currents with far-field optics.

Photocurrent generation. A crucial aspect of the experimental findings reported in Ref. [16] is that the frequency of the pump pulse $\Omega_{\rm pm} \simeq 375$ THz is at least two orders of magnitude larger than $\omega_{\rm JPR}$. This separation of energy scales suggests that the pump pulse drives the mobile electronic degrees of freedom that down-convert the large incoming frequency into a low-frequency photocurrent [8,35–40]. This is further bolstered by the expectation that inversion symmetry is broken for a realistic pattern of charge order [24]. The photocurrent is characterized by a rank-three conductivity tensor:

$$J^a_{\rm NL}(\omega) = \int \frac{d\omega'}{2\pi} \sigma^{(2)}_{abb}(-\omega; \omega', \omega - \omega') E^b_{\omega'} E^b_{\omega-\omega'}, \qquad (1)$$

where indices a, b represent the Cartesian directions. Following Ref. [16], we assume that the pump-pulse width

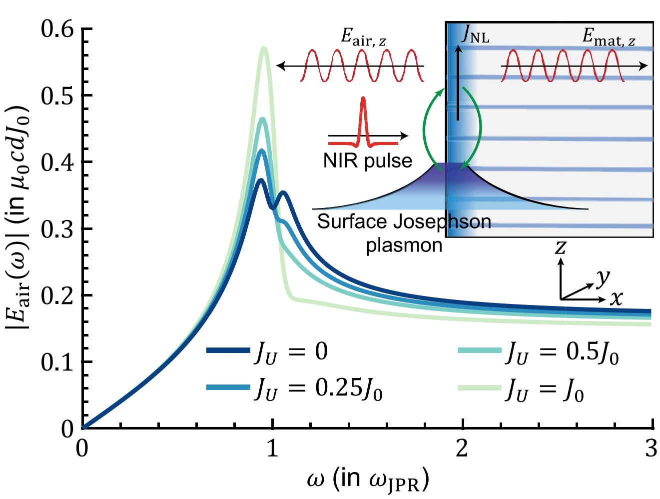

FIG. 2. Emission spectrum. We find that the outgoing radiation can be sharply peaked around $\omega_{\rm JPR}$ only in the presence of a nonzero umklapp photocurrent $J_U \neq 0$, Eq. (13). This umklapp component impulsively drives high momenta surface Josephson plasmons that can emit light [Fig. 1(b)]. Here we fixed $A = 0.1\varepsilon_{\infty,c}$.

$\Delta\Omega_{\rm pm} \simeq 5$ THz is also much larger than $\omega_{\rm JPR}$, in turn implying that the photocurrent appears featureless for $\omega \lesssim \Delta\Omega_{\rm pm}, \Omega_{\rm pm}$:

$$J^a_{\rm NL}(\omega) \xrightarrow{\omega \lesssim \Delta\Omega_{\rm pm}, \Omega_{\rm pm}} \int \frac{d\omega'}{2\pi} \sigma^{(2)}_{abb}(0; \omega', -\omega') E^b_{\omega'} E^b_{-\omega'}.$$

This should be contrasted to the experiment [16], where the observed radiation is sharply peaked in frequency at $\omega_{\rm JPR}$. Intuitively, the broadband photocurrent acts like an impulsive drive to coherent Josephson plasma modes, both bulk and surface ones. Here we focus on the surface excitations because the scenario based on the bulk ones is inconsistent with the experimental data, as we discuss in the Supplemental Material [41] and further elaborate on below. In the following discussion, we investigate a semiphenomenological model, where we assume the presence of such a broadband drive and study how it interacts with Josephson plasmons.

To further appreciate the importance of the surface excitations, we argue that the photocurrent generation occurs at the surface of the sample. Indeed, the frequency of the incoming photoexcitation is large so that the skin depth of how light penetrates the sample is small. From the equilibrium optical properties [16], we estimate it to be $d \approx 200$ nm; this short electronic length scale is much shorter than any length scale associated with the collective excitations of the superconductor [42,43]. To illustrate this explicitly, we consider the bulk polariton dispersion in the vicinity of $q = 0$:

$$\omega^2_{\rm JP}(q_x) \approx \omega^2_{\rm JPR} + \frac{c^2 q_x^2}{\epsilon_{\infty,ab}}, \qquad (2)$$

where $\epsilon_{\infty,ab}$ is the high-frequency in-plane dielectric constant of the medium. Equation (2) allows defining the plasmon coherence length as $l_{\rm JP} = c/(\omega_{\rm JPR}\sqrt{\epsilon_{\infty,ab}}) \sim 100$ μm $\gg d$. Hence, from the perspective of collective modes, one indeed can consider the photocurrent as a surface phenomenon. We remark that in the experiment, the beam-spot size is large, $d_{\rm beam} \sim 500$ μm $\gtrsim l_{\rm JP}$, so that the pump can be approximately described as uniform along the interface.

For future reference, the electromagnetic response of layered materials, such as cuprates, is encoded into the anisotropic dielectric tensor $\hat{\varepsilon}(\omega) = {\rm diag}(\varepsilon_{ab}, \varepsilon_{ab}, \varepsilon_c)$. Motivated by the two-fluid model of anisotropic superconductors [41,44–47], which accurately captures optical linear response of cuprate superconductors [45,46], we choose the following form:

$$\varepsilon_{\alpha=ab/c}(\omega) = \epsilon_{\infty,\alpha}\left(1 - \frac{\omega_\alpha^2}{\omega^2} + \frac{i\gamma_\alpha}{\omega}\right). \qquad (3)$$

This form so far does not include the charge order; we will return to this below. The second term in Eq. (3) describes the reactive response of the superconducting fluid so that $\varepsilon_\alpha(\omega) \sim \omega^{-2}$ for $\omega \to 0$; ω_{ab} and $\omega_c = \omega_{\rm JPR}$ are the in-plane and c-axis plasma frequencies. The strong anisotropy of cuprates implies $\omega_{ab} \gg \omega_c$. We also expect the in-plane plasmons to be strongly damped $\gamma_{ab} \gg \gamma_c$. The third term represents the normal fluid; γ_α is a phenomenological damping parameter, which is proportional to the corresponding normal-fluid conductivity [41,45]. Unless specified otherwise, we use the following parameters [16,46,48]: $\omega_c = 1$ THz, $\gamma_c = 0.1$ THz,

$\epsilon_{\infty,c} = 25$, and $\epsilon_{\infty,ab} = 4$. Our conclusions do not rely on the choice of ω_{ab} and γ_{ab} as long as $\omega_{ab} \gg \omega_c$.

Surface Josephson plasmons. We begin by reviewing the properties of surface Josephson plasmons in the absence of CDW order [46,49,50]. These are evanescent collective modes that are confined to the interface $x = 0$ (inset of Fig. 2), i.e., they decay both into the air and into the sample [46,51,52]. Of the most physical interest for us below are the modes with the magnetic field pointing along the y axis:

$$B_y = \begin{cases} B_a e^{k_a x + i q_z z - i\omega t}, & x < 0 \\ B_m e^{-k_m x + i q_z z - i\omega t}, & x > 0. \end{cases} \tag{4}$$

Note that because of the translational invariance along the z axis, we choose the same dependence on z inside and outside the sample. Here k_a and k_m are yet unknown wave vectors that depend on both ω and q_z. We find them by solving the Maxwell equations in each media:

$$k_a^2 = q_z^2 - \frac{\omega^2}{c^2}, \quad k_m^2 = \varepsilon_c\left(\frac{q_z^2}{\varepsilon_{ab}} - \frac{\omega^2}{c^2}\right). \tag{5}$$

To describe evanescent electromagnetic waves, we choose the roots with $\mathrm{Re}\, k_a, \mathrm{Re}\, k_m > 0$. By matching the Fresnel boundary conditions ($E_{\mathrm{air},z} = E_{\mathrm{mat},z}$ and $B_{\mathrm{air},y} = B_{\mathrm{mat},y}$ at $x = 0$), we obtain an implicit equation on the dispersion $\omega_s(q_z)$ of the surface Josephson plasmons:

$$q_z = \frac{\omega_s}{c}\sqrt{\frac{\varepsilon_{ab}(\omega_s)[1 - \varepsilon_c(\omega_s)]}{1 - \varepsilon_{ab}(\omega_s)\varepsilon_c(\omega_s)}}. \tag{6}$$

The spectrum of these excitations is shown in Fig. 1(a). We find that $\omega_s(q_z)$ quickly saturates at around ω_{JPR}. In other words, we expect a large density of states of these excitations near ω_{JPR}. It is worth pointing out that the fact that the saturation occurs near the bulk plasmon resonance, $\omega_s(q_z) \to \omega_{\mathrm{JPR}}$ for $cq_z \gtrsim \omega_{\mathrm{JPR}}$, is a consequence of the strong anisotropy $\omega_{ab} \gg \omega_c$. For instance, in isotropic superconductors with $\omega_{ab} = \omega_c$ and $\varepsilon_{\infty,ab} = \varepsilon_{\infty,c} = 1$, a similar saturation occurs but at a notably lower frequency $\omega_{\mathrm{JPR}}/\sqrt{2}$. We finally remark that the two other surface Josephson plasmons exhibit a similar saturation but at much higher frequency.

An immediate problem we encounter is that the surface excitations lie outside the light cone and, therefore, cannot radiate. This issue is evaded by the charge order, which couples the mode with wave vector q_z to the ones with wave vectors $q_z + nQ_{\mathrm{CDW}}$, where $n = 0, \pm1, \pm2, \ldots$ and $Q = Q_{\mathrm{CDW}}$ is the ordering wave vector along the z axis. As such, the entire surface-plasmon dispersion becomes backfolded to the reduced Brillouin zone defined by Q. Some modes backfold into the light cone (Fig. 1) and, therefore, they can emit photons. This insight is crucial in explaining the experiment. In essence, the stripes act as a natural diffraction grating, which then outcouples the otherwise silent surface modes, much like a nanofabricated corrugation would be engineered [30–34,44,53–56].

Here we describe the CDW order phenomenologically. Since the most dominant effect is expected to be due to the charge modulation along the z axis, we neglect the fact that stripes have a nontrivial structure along the x and y axes. Specifically, we assume that the CDW order parameter enters through the modulation of the c-axis plasmon frequency:

$$\omega_c \to \omega_c = \omega_{\mathrm{JPR}} + \delta\omega_c(z), \quad \delta\omega_c(z) \propto \cos(Qz). \tag{7}$$

This modulation, which we assume to be weak, modifies only the c-axis dielectric function, which we write as

$$\varepsilon_c(\omega) \to \varepsilon_c(\omega) + A\,\cos(Qz), \tag{8}$$

where A captures the strength of the stripes order. For simplicity, we assume that the stripe period along the z axis equals two lattice constants. This assumption is by no means crucial but allows us to make substantial analytical progress. Finally, to properly describe the momentum mixing, one should take into account the momentum dependence of the dielectric tensor (3). However, this dependence is expected to be nonessential at the scale of Q, as we show in [41], where we carefully consider the surface Josephson plasmons at large momenta.

To get the properties of surface Josephson plasmons in the medium with stripes, we proceed similarly as above but now take into account the Bragg mixing of the z-axis momenta. Specifically, we substitute the following evanescent-wave ansatz:

$$B_{\mathrm{air},y} = [\alpha_a e^{k_a x + i q_z z} + \beta_a e^{\tilde{k}_a x + i(q_z+Q)z}]e^{-i\omega t}, \tag{9}$$

$$\begin{aligned} B_{\mathrm{mat},y} = {} & [\alpha_m e^{-\lambda_1 x}(e^{iq_z z} + \gamma_1 e^{i(q_z+Q)z}) \\ & + \beta_m e^{-\lambda_2 x}(\bar{\gamma}_2 e^{iq_z z} + e^{i(q_z+Q)z})]e^{-i\omega t}, \end{aligned} \tag{10}$$

where $k_a(q,\omega) = \sqrt{q^2 - \omega^2/c^2}$ and $\tilde{k}_a = k_a(q+Q,\omega)$. The wave vectors λ_1 and λ_2, together with parameters γ_1 and $\bar{\gamma}_2$ that encode the mentioned mixing, are known functions of q_z and ω, which are obtained by solving the Maxwell equations inside the sample with stripes [41]. The four remaining unknown coefficients $\alpha_a, \beta_a, \alpha_m, \beta_m$ are related to each other via $B_{\mathrm{air},y} = B_{\mathrm{mat},y}$ and $E_{\mathrm{air},z} = E_{\mathrm{mat},z}$. These conditions, in turn, implicitly define the spectrum of surface plasmons through $\det \mathcal{M}(q_z,\omega) = 0$, where

$$\mathcal{M} \equiv \begin{bmatrix} 1 + \frac{\varepsilon_c}{\varepsilon_c^2 - A^2}\frac{k_m^2}{\lambda_1 k_a} & \left(1 + \frac{\varepsilon_c}{\varepsilon_c^2 - A^2}\frac{k_m^2}{\lambda_2 k_a}\right)\bar{\gamma}_2 \\ \left(1 + \frac{\varepsilon_c}{\varepsilon_c^2 - A^2}\frac{\tilde{k}_m^2}{\lambda_1 \tilde{k}_a}\right)\gamma_1 & 1 + \frac{\varepsilon_c}{\varepsilon_c^2 - A^2}\frac{\tilde{k}_m^2}{\lambda_2 \tilde{k}_a} \end{bmatrix}.$$

Schematic illustration of the modified by the stripes dispersion is shown in Fig. 1(b), where we reflect the backfolding of high momenta surface modes into the reduced Brillouin zone.

An impulsive drive to the surface modes. Having established the structure of surface excitations, we turn to discuss the role of photocurrent. It lives on the surface of the material and results in the emission of radiation in both the air and sample (Fig. 2). Motivated by the experiment [16], we turn to evaluate the frequency dependence of the electromagnetic field emitted into the air. We assume that J_{NL} flows along the z axis so that the magnetic field is oriented along the y axis. Since J_{NL} is confined to the surface of the superconductor, we can incorporate this drive through the generalization of the Fresnel formalism, where as above we solve Maxwell's equations inside the two media independently and then match the solutions using appropriate boundary conditions. One of them is the continuity of the tangential component of the electric field (follows from Faraday's law): $E_{\mathrm{air},z} = E_{\mathrm{mat},z}$ at

$x = 0$. The other equation is obtained from integrating the fourth Maxwell equation:

$$\int_{0^-}^{d} dx(\partial_x B_y) = \int_{0^-}^{d} dx[\mu_0 J_{\rm NL} - i\omega\varepsilon_c(\omega)E_z/c^2]. \qquad (11)$$

The second term in Eq. (11) is parametrically small, $\int_{0^-}^{d} dx\, E_z \propto d/l_{\rm JP} \ll 1$, so we neglect it and obtain

$$B_{{\rm air},z} = B_{{\rm mat},z} - \mu_0 d J_{\rm NL}. \qquad (12)$$

Given the possibility of having a nonzero umklapp photocurrent [24], we write Eq. (1) as

$$J_{\rm NL}(z,\omega) = J_0 + J_U \cos(Q_{\rm CDW} z). \qquad (13)$$

This insight that one can have $J_U \neq 0$ is essential for understanding the experimental data, as we show below. Using the form (13), we turn to compute the spectrum of outgoing radiation. To this end, we employ the same ansatz as in Eqs. (9) and (10), except we now specialize on $q_z = 0$. By invoking the derived boundary conditions, we arrive at

$$\begin{bmatrix}\alpha_a\\ \beta_a\end{bmatrix} = \left(\begin{bmatrix}1 & \bar{\gamma}_2\\ \gamma_1 & 1\end{bmatrix}\mathcal{M}^{-1} - \begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}\right)\begin{bmatrix}\mu_0 d J_0\\ \mu_0 d J_U\end{bmatrix}. \qquad (14)$$

The coefficient α_a, in turn, gives the amplitude of the emitted into the air radiation $f(\omega)$ (see Fig. 2).

In the absence of the umklapp component $J_U = 0$, we find that $f(\omega)$ displays a double-peak structure, which comes from the splitting of the bulk JP resonance, encoded in the prefactors $\varepsilon_c/(\varepsilon_c^2 - A^2)$. It is worth pointing out that since the surface excitations exhibit saturation around $\omega_{\rm JPR}$, i.e., at the bulk resonance, it is not entirely clear that this splitting comes from the bulk rather than the surface. To resolve this question we consider in [41] isotropic superconductors, where the surface excitations are well separated from the bulk ones, and confirm that the double-peak structure originates from the bulk. We also further elaborate in [41] on the asymptotic behavior of $f(\omega)$ at both small and large frequencies. This double-peak splitting was not observed in the experiment [16]. In addition, the resulting spectral function is too broad for realistic parameters to explain the experimental data.

Most remarkably, provided $J_U \neq 0$, we find the emission spectrum $f(\omega)$ becomes sharply peaked at around $\omega_{\rm JPR}$. This peak originates from the fact that the umklapp component now resonantly drives the surface plasmons at wave vectors around $q_z = Q$; due to the backfolding into the light cone, these plasmons can now radiate photons, as illustrated in Fig. 1(b). We further elaborate on the surface origin of this effect in [41]. This result that the umklapp photocurrent can give rise to a sharp emission provides a natural interpretation of the experimental data.

Bulk plasmon scenario. We now comment on the role of bulk polaritons. Since they are also being impulsively driven by $J_0 \neq 0$, Eq. (14), they can produce characteristic radiation peaked in frequency near $\omega_{\rm JPR}$. While we cannot completely rule out the bulk scenario as a possible alternative interpretation of the experiment [16,41], there are two good reasons why the observed emission is dominated by the surface Josephson plasmons. The first one is that the resulting bulk emission spectrum turns out to be too broad for realistic parameters to explain the experiment [41]. In contrast, in the surface scenario, the umklapp photocurrent resonantly drives the surface modes resulting in sharp radiation without any fine-tuning of model parameters. The second argument invokes the experimental phenomenology that the observed radiation linewidth correlates with the out-of-plane CDW coherence length (once inversion symmetry is broken). This fact is inconsistent with the bulk scenario because the far-field reflectivity, which entirely determines the bulk emission properties, is known to be insensitive to the stripes [16,57]. At the same time, the observed phenomenology can naturally be explained within the surface scenario since the corresponding emission is acutely sensitive to the CDW order (for an additional discussion, see [41]) [see also Fig. 3(a)].

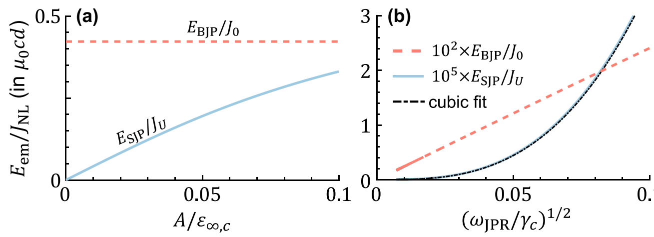

FIG. 3. Differences between the bulk and surface scenarios. (a) Emission amplitude $E_{\rm SJP}/J_U$ from the surface-plasmon scales linearly with the CDW amplitude A, Eq. (8), while the emission in the bulk case $E_{\rm BJP}/J_0$ is independent of A. (b) For $\omega_{\rm JPR} \ll \gamma_c$, we find that $E_{\rm SJP} \sim (\omega_{\rm JPR}/\gamma_c)^{3/2}$ for the surface scenario and $E_{\rm BJP} \sim (\omega_{\rm JPR}/\gamma_c)^{1/2}$ for the bulk one.

Finally, to further distinguish the two scenarios from each other, we theoretically predict the scaling of each response with temperature T. As T approaches T_c from below, the JPR softens to zero. As shown in Fig. 3, we find that the outgoing emission exhibits distinct behaviors for the two scenarios when $\omega_{\rm JPR} \ll \gamma_c$:

$$E_{\rm BJP} \propto J_0\left(\frac{\omega_{\rm JPR}}{\gamma_c}\right)^{1/2} \quad \text{and} \quad E_{\rm SJP} \propto J_U A\left(\frac{\omega_{\rm JPR}}{\gamma_c}\right)^{3/2}.$$

One important implication of this result is that the emission amplitude in the surface scenario $E_{\rm SJP}$ is expected to become suppressed compared to the bulk one $E_{\rm BJP}$ for $T \approx T_c$: $E_{\rm SJP}/E_{\rm BJP} \propto \omega_{\rm JPR}/\gamma_c$. Since according to Eq. (14) both scenarios contribute to the outgoing radiation, we conclude that the surface scenario dominates at lower temperatures, while the bulk one might play a role in the immediate vicinity of T_c.

Conclusion. To summarize, our interpretation of the observed terahertz emission in striped superconductors consists of two related arguments. The first one is the downconversion of optical fields by fermionic quasiparticles from high-frequency high-intensity pump pulse to low-frequency regular and umklapp photocurrents. Given that the emission is observed only in the striped phase, we expect that this downconversion arises from the CDW order [24]. The second argument is that photocurrents impulsively drive collective modes of the sample. In particular, the umklapp component resonantly couples to the surface Josephson plasmons, in turn resulting in spectrally sharp radiation at $\omega_{\rm JPR}$ and, thus, explaining the experiment [16]. In the electrodynamics of striped superconductors, both stripe and superconducting orders combine and play significant roles.

For outlook, an intriguing open question is the microscopic origin of the photocurrents and whether it can be related to a pair density wave [17]. The umklapp photocurrent is interesting on its own, even without superconductivity, and might be relevant, for instance, in moiré systems [24,58].

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