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Giant ultrafast Kerr effect in superconductors

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We study the ultrafast Kerr effect and high-harmonic generation in superconductors by formulating a model for a time-varying electromagnetic pulse normally incident on a thin-film superconductor. It is found that superconductors exhibit exceptionally large $\chi^{(3)}$ due to the progressive destruction of Cooper pairs, and display high-harmonic generation at low incident intensities, and the highest nonlinear susceptibility of all known materials in the THz regime. Our theory opens up avenues for accessible analytical and numerical studies of the ultrafast dynamics of superconductors.

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I. INTRODUCTION

The discovery of superconductivity in 1911 by Kamerlingh Onnes introduced a startling new phenomenon to physics: that of systems exhibiting zero electrical resistance at low temperatures [1]. Although superconductivity is relatively easy to measure in the laboratory, a full microscopic explanation was not put forward for over 40 years, in the form of the Bardeen-Cooper-Schrieffer (BCS) theory [2]. Between the discovery of superconductivity and its satisfactory explanation there were several powerful attempts: including that of the London brothers in the 1930s [3], used to describe the observed expulsion of external magnetic fields; and the Ginzburg-Landau theory of the 1950s [4], a phenomenological model that was subsequently proved to be strictly linked to the BCS theory in certain limits, and was able to explain the existence of the spatial lattice distribution of flux vortices in type-II superconductors [5].

Research into superconductivity is currently very active, with recent work on high-temperature superconductors—reaching up to a critical temperature of 203 K—attracting considerable attention [6]. The finding that superconductors can act as very effective single-photon detectors also demonstrates their aptness in the field of photonics [7–9].

The effect of *static* external magnetic fields on a superconductor is one of the subject's most fascinating and studied aspects, and all the introductory and advanced textbooks contain many details on how superconductors behave under the influence of time-independent fields. However, somewhat surprisingly, a simple and intuitive study of the influence of a *time-dependent* electromagnetic pulse has been less fully developed. Early experimental work by Testardi [10] looked into the destruction of superconductivity by laser pulses; Gor'kov and Éliashberg [11,12] explored the theory of interaction of weak time-varying fields with superconductors and found tripled-frequency radiation generation—their derivations utilized Green's functions, complex analysis, and they employed a diagrammatic method. More recently, Matsunaga *et al.* experimentally observed the third-harmonic generation in NbN samples in the THz regime [13], which was explained theoretically by Cea *et al.*, who calculated the nonlinear currents by using the full microscopic model based on the BCS theory [14]. Theoretical and experimental work continues to be carried out in this area [15–18], however it is common for very advanced theoretical and computational tools to be used; in the research presented here we hope to capture much of the important physics using a much simpler approach.

In this work we present exhaustive analytical and numerical calculations based on the *time-dependent* Ginzburg-Landau equation, and show the appearance of a Kerr effect in a superconductor induced by an incident, arbitrarily short light pulse. We show that, due to progressive and steplike Cooper pair destruction, these superconducting materials display large nonlinear optical behavior, such as high harmonic generation, at extremely low laser intensities—typically on the kW/cm^2 scale in the THz regime—and may prove to be a key element in future photonics applications. Our approach is physically more transparent and much more intuitive than the approach based on the BCS Hamiltonian. We specialize our discussion to elemental niobium (Nb) thin films (a type-II superconductor).

II. GOVERNING EQUATIONS

We begin our study with the *time-dependent* Ginzburg-Landau (TDGL) equation [19] (setting the scalar potential ϕ to zero; see below for a justification of this):

$$\frac{\hbar^2}{2m^* D}\partial_t \psi + \frac{1}{2m^*}\left(\mathbf{p} - \frac{q}{c}\mathbf{A}\right)^2 \psi + \alpha\psi + \beta|\psi|^2\psi = 0, \tag{1}$$

where $\psi(x,y,t)$ is the complex order parameter ($|\psi|^2$ is proportional to the total number of Cooper pairs present in the sample: $\psi = \sqrt{n_c}e^{i\theta}$), n_c is the number density of Cooper pairs, x,y are the coordinates on the plane of the thin film, m^* is the mass of a Cooper pair (approximately equal to twice the mass of a free electron, when neglecting the binding energy), $q = -2e$ is its charge (equal to twice the free electron charge $-e$), D is the diffusion constant, $\mathbf{A}(x,y,t)$ is the vector potential describing the spatial and temporal structure of the incident electromagnetic pulse, $\mathbf{p}$ is the electron momentum operator, and c is the speed of light in vacuum. The phenomenological Ginzburg-Landau parameters α and β have units of energy and energy × volume, respectively and are explicitly given by $\alpha = \alpha_0(T - T_c)$, where α_0 and β are constants specific for the material used. T is the temperature of the system and T_c is the critical temperature below which superconductivity emerges [1].

Note the quite unconventional structure of Eq. (1): since ψ is strictly speaking not a wave function, there is a missing imaginary unit in the first term of this equation, which sets a strong difference from the nonlinear Schrödinger equation.

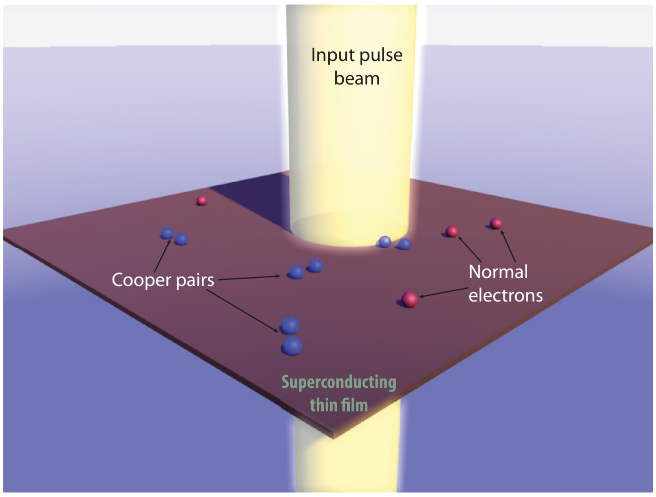

FIG. 1. Sketch of the system studied in this work. A superconducting thin film is excited by a normally incident, x-polarized electromagnetic pulse. The pulse destroys the Cooper pairs already present in the film, generating more "normal" electrons, which then diffuse and slowly recombine due to the electron-phonon interaction.

This difference is due to the thermodynamical interpretation of ψ as an order parameter, related to the free energy of the sample [4]. This makes Eq. (1) a nonlinear diffusion equation. The time-dependent Eq. (1), which is typically not discussed in basic textbooks on superconductivity (where its static version is usually treated), is the core equation of this work as it describes how the order parameter of a superconductor varies due to an electromagnetic field interaction and contains all of the important system parameters. The Ginzburg-Landau (GL) theory describes the emergence of superconductivity in terms of a phase transition and was shown to be derivable from the BCS theory in close proximity to the critical temperature [20], although the theory is used regularly and has been shown to be practically valid for temperatures well below T_c. It is a very general phenomenological model used in many areas of physics and avoids the use of many-body wave functions [1,21]. We choose to use the GL formalism instead of the microscopic BCS theory as we believe the former contains all of the necessary concepts and tools required—once combined with basic electrodynamics—to show the ultrafast optical nonlinearities inherent in superconductors. The original motivation for the development of GL theory was indeed to describe superconductivity without needing to delve into its microscopic structure. The gauge field can however be introduced in the BCS theory via a Peierls substitution in real space, as in Ref. [14].

The setup considered in this work is sketched in Fig. 1. An electromagnetic pulse is normally incident to the two-dimensional (2D) superconducting thin film. We make the simplifying assumption that the electric field of the incident light is polarized along the x direction only, as is the magnetic vector potential $\mathbf{A} = [A(x,y,t),0,0]$. Expanding Eq. (1) gives

$$\frac{\hbar^2}{2m^*D}\partial_t\psi - \frac{\hbar^2}{2m^*}\nabla^2\psi + \frac{i\hbar q}{2m^*c}(\nabla\cdot\mathbf{A})\psi + \frac{i\hbar q A}{m^*c}(\nabla\psi) + \frac{q^2A^2}{2m^*c^2}\psi + \beta|\psi|^2\psi + \alpha\psi = 0. \quad (2)$$

As a first attempt to understand the origin of the optical nonlinearity [not to be confused with the *electronic* nonlinearity, which is given by the $|\psi|^2\psi$ term in Eq. (2), and due to the phonon-mediated electron-electron interaction], we now use Eq. (2) to make a rough estimate of the third-order nonlinear optical coefficient exhibited by the thin film. In order to find an intensity scale around which nonlinear optical effects in the superconductor become important, we make the following assumptions: the order parameter ψ varies slowly in the spatial variables and we use the gauge given in Koyama [22]: $\nabla\cdot\mathbf{A} + \frac{\lambda_L^2}{\lambda_{\rm TF}^2}\frac{1}{c}\partial_t\phi = 0$, where λ_L is the London penetration depth, and $\lambda_{\rm TF}$ is the Thomas-Fermi screening length. We study niobium in this work, for which $\frac{\lambda_L^2}{\lambda_{TF}^2} \gg 1$, implying that the value of ϕ can be taken as negligible. In conjunction with the assumption of charge neutrality (which should hold in metallic superconductors unless they are extremely thin) [23] this enforces also a vanishing of $\nabla\cdot\mathbf{A}$ (using Gauss' law). Using the expanded TDGLE (2), and the assumptions detailed above, gives $\frac{\hbar^2}{2m^*D}\partial_t\psi + (\frac{q^2A^2}{2m^*c^2} + \alpha)\psi + \beta|\psi|^2\psi = 0$. We see that optical nonlinearities become important when the vector potential term in parentheses is dominant, and by using $\mathbf{E} = -(1/c)\partial_t\mathbf{A}$ to approximate the amplitudes $A \approx -Ec/\omega_0$, along with the usual light intensity equation $I = \epsilon_0 cE^2/2$ (assuming unity refractive index), we find a value for the nonlinear intensity scale given by $I_0 = c\epsilon_0 m^*|\alpha|\omega_0^2/q^2$, where ω_0 is the carrier frequency of the pulse. For light of intensity $I \geqslant I_0$ then the term proportional to $A^2\psi$ in (2), which is responsible for the nonlinear interplay between the field and the superconductor, cannot be ignored; and as we will see below a very powerful cubic nonlinearity is predicted.

The first term in Eq. (2) represents the rate of change of the order parameter with respect to time; the second term encodes the spatial diffusion dynamics of the Cooper pairs; the third term vanishes as shown above; the fourth term does not affect the system's nonlinear dynamics, only adding a linear phase; the fifth and sixth terms illustrate the inherent nonlinearities of the system manifested in the electronic cubic order parameter $|\psi|^2\psi$ and the photonic $A^2\psi$ cross terms; and the last term contributes to the "recovery" of the order parameter's original value, i.e., Cooper pair recombination after the superconductor has interacted with light.

The basic physics of the process giving rise to Kerr optical nonlinearity can be explained as follows: as the light pulse impacts the superconductor Cooper pairs are destroyed, increasing the number of normal electrons in the material. As each progressive peak and trough of the light pulse reaches the material the value of the order parameter drops and then begins to recover as Cooper pairs are destroyed—generating harmonics—and then reformed, due to the phonon-mediated potential. This mechanism is reminiscent of the ionization dynamics in plasma physics in which light strips electrons (free electrons are "created") from their atoms, see for instance a recent work on the subject [24].

The total current in the superconductor is composed of an Ohmic component and a supercurrent, the latter given explicitly by [19]

$$J_s = -\frac{q^2}{m^*c}|\psi|^2A - \frac{iq\hbar}{2m^*}(\psi^*\nabla\psi - \psi\nabla\psi^*), \quad (3)$$

where the second term is proportional to the spatial gradient of the phase θ of the order parameter, as can be seen if we rewrite (3) as $J_s = -\frac{q^2}{m^*c}|\psi|^2 A + \frac{q\hbar}{m^*}|\psi|^2\nabla\theta$. All time dependencies are implicit and vector notation has been removed as J_s has only an x component, since the input pulse is linearly polarized.

III. ESTIMATE OF $\chi^{(3)}$

The nonlinear electric susceptibility $\chi^{(3)}$ of the superconducting film can be derived as follows. The magnetic vector potential evolves due to the current density via the well-known wave equation:

$$\left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} - \nabla^2\right)\mathbf{A} = \frac{1}{\epsilon_0 c}\mathbf{J}, \tag{4}$$

where ϵ_0 is the vacuum permittivity. Taking the partial time derivative of both sides of Eq. (4) and using $E = -\frac{1}{c}\frac{\partial A}{\partial t}$ produces $(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} - \nabla^2)E = -\frac{1}{\epsilon_0 c^2}\partial_t J$, where the right-hand side is proportional to the material polarization. Assuming that the normal part of the current can be disregarded as it does not contribute to the optical nonlinearity (the metallic part of the system is simply absorptive), then the nonlinear polarization is seen to be of Kerr type, i.e., $P_{\rm NL} = \chi^{(3)}E^3$, by solving analytically Eq. (2) for a space-independent continuous wave $A(t) = A_0\cos(\omega_0 t)$ and expanding in Taylor series of A_0^2, stopping at the first order. One obtains

$$|\psi|^2 \simeq -\frac{a}{b} - \frac{d}{2b\omega_0^3}[a^2\omega_0\cos(2\omega_0 t) - a\omega_0^2\sin(2\omega_0 t)] + \cdots, \tag{5}$$

where we have defined $a \equiv 2m^*D\alpha/\hbar^2$, $b \equiv 2m^*D\beta/\hbar^2$, and $d \equiv q^2DA_0^2/(\hbar c)^2$. Inserting expression (5) into the supercurrent (3), one can derive the following GL form of the complex nonlinear coefficient (see full derivation in the Supplemental Material [25]):

$$\chi^{(3)}_{\rm GL} = -\frac{128k_B^2(T-T_c)^2e^4}{\epsilon_0\beta\omega_0^6\pi^2\hbar^2m_e^2}. \tag{6}$$

The value of $\chi^{(3)}_{\rm GL}$ in Eq. (6) is negative and depends on the input pulse frequency ω_0, and the parameters β and T_c which depend strongly on the specific material chosen, and of course on the temperature T, which must be below T_c. The frequency dependence ω_0^{-6} makes the superconductor very nonlinear for longer wavelengths, as it happens, for instance, in 2D Dirac materials like graphene [26]. Equation (6) is the first novel result of this work. It must be noted that the expression is valid only close to the critical temperature T_c (due to its emergence from TDGL theory) below which it becomes less accurate. Its reliability near T_c is shown theoretically in the Supplemental Material [25] where the result coincides with the full BCS theory in the limit which we derive for $T \to T_c$, and its reliability with respect to experiment is seen by comparing with the third-harmonic generation intensity as a function of temperature in Fig. 4B of Matsunaga *et al.* [13] and Fig. 3 C which qualitatively matches our results in the region of temperatures near T_c (our nonlinear response scales as ω_0^{-6}, so it peaks as $\omega_0 \to 0$; the resonance at $2\omega_0 = 2\Delta(T)$ seen in Matsunaga *et al.* [13] is not captured in our work as it cannot be derived from TDGL theory). Treatments of the temperature and frequency dependence of the nonlinearity extending down to $T = 0$ K, and a discussion of the resonance with the Higgs mode, have been discussed in the literature [13,14,27].

IV. SIMULATIONS AND RESULTS

Using the above TDGL Eq. (1) we perform time-dependent simulations in both one and two spatial dimensions to analyze how the order parameter of the superconductor evolves under the influence of an ultrashort pulse, which harmonics are produced, and the form of the supercurrent as a function of time. We analyze both cases as although the one-dimensional (1D) case is the simplest, and produces the essential results of the work, we find that the more realistic 2D results illustrate the spatial variation in order parameter with more clarity—we also expect future experimental tests of the theory to be carried out in a thin-film (i.e., quasi-2D) geometry. The time-dependent simulations are also necessary to check the ultrafast response of the thin superconductive film in realistic situations.

As a representative example, for our estimates we choose pure niobium (Nb) as it has the highest critical temperature T_c of all elemental superconductors [28]. The following results were taken for simulations of the system at temperature $T = 4$ K.

The critical temperature of bulk Nb is $T_c = 9.25$ K [29], and the zero-temperature coherence length $\xi(0) = 0.74\sqrt{\chi}\xi_0$, where ξ_0 is the intrinsic BCS coherence length and χ is the Gor'kov parameter [30]. This parameter depends on whether the superconductor is in the "dirty limit" or the "clean limit": a mean free electron path satisfying $l \gg \xi_0$ (i.e., the clean limit) gives $\chi \approx 1$; the other limit $l \ll \xi_0$ gives $\chi \approx 1.33(l/\xi_0)$. In the following we will assume our system is in the dirty limit as Nb has a mean free path $l = 9.5$ nm and $\xi_0 = 38$ nm (Fermi velocity $v_{\rm F} = 1.37 \times 10^6$ m s^{-1} and mean free electron flight time $\tau_{\rm free} = 7$ fs) [31,32]. In this dirty limit $\xi(0) = 16.2$ nm, and the diffusion parameter value is $D = 8.09 \times 10^{-4}$ m^2 s^{-1}, while the constants $\alpha_0 \simeq 1.2618 \times 10^{-24}$ J/K, and $\beta \simeq 1.256 \times 10^{-51}$ J m^3. We study the effects on the order parameter for sech-type pulses and for super-Gaussian pulses on the scale of $t_{\rm FWHM} = 9.55$ ps, which is available experimentally in the THz regime [33].

Using a laser pulse with carrier wavelength $\lambda_0 = 188$ μm and a Nb film for a target, we find that optical nonlinear effects should emerge at very low incident intensities of the order of kW/cm^2, and on ultrafast time scales of the order of picoseconds. Specifically, at a temperature $T = 4$ K, $I_0 \approx 3.1$ kW/cm^2. Our parameters used give the value of $\chi^{(3)} \approx -4.24 \times 10^{-7}$ m^2/V^2, which is to the best of our knowledge the highest theoretical value ever predicted in a $\chi^{(3)}$ material in the THz regime [34] as well as being many orders of magnitude above standard experimental and theoretical $\chi^{(3)}$ values in other regimes [35–38]—for comparison: carbon disulfide has a nonlinear optical response of $\chi^{(3)} = 3.1 \times 10^{-20}$ m^2/V^2 [36], various liquids exhibit a THz response of $\chi^{(3)} \approx 10^{-20}$ m^2/V^2 [39], and the semiconductor GaAs can exhibit a high value of $\chi^{(3)} \approx 10^{-12}$ m^2/V^2 in the THz regime [40]. Graphene also exhibits highly nonlinear behavior [41,42] as mentioned above; for example under the influence of a laser pulse with carrier wavelength $\lambda_0 = 188$ μm

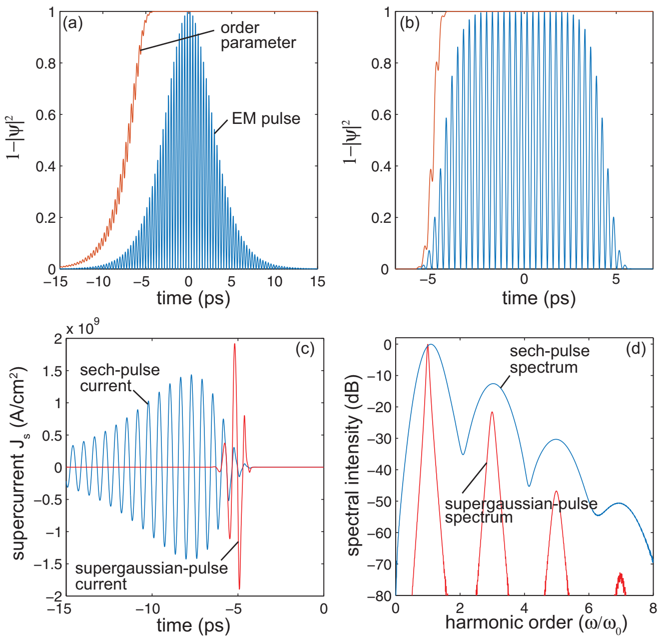

FIG. 2. (a) Effect of a sech-type pulse on order parameter, showing progressive steplike destruction of Cooper pairs. (b) Same is in (a) but for a super-Gaussian pulse with same duration and intensity. (c) Supercurrents for the two types of pulses used. (d) Final spectra showing the generation of odd harmonics. The pulse temporal profiles are normalized to unity. Note that in (a) and (b) $1-|\psi|^2$ is plotted, so when Cooper pairs are destroyed this quantity increases towards unity.

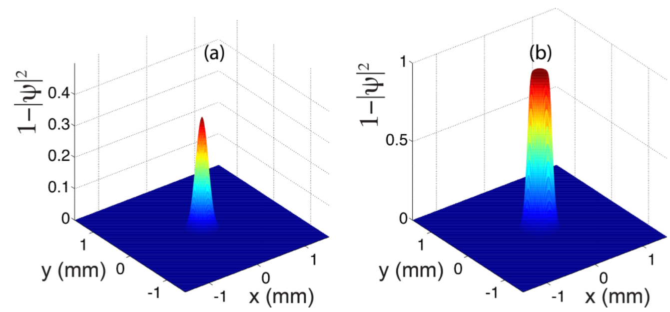

FIG. 3. (a) Early frame from the 2D simulation, taken at $t = -7.5$ ps [same horizontal scale used in Fig. 2(a)], showing the effect of a short sech-type pulse on the spatial distribution of order parameter. (b) Same as (a) but for a later time $t = -4.5$ ps, showing the saturation of the order parameter around the highest intensity region of the beam, where the pulse has locally destroyed all the available Cooper pairs.

(the same wavelength we study here for niobium) graphene has a predicted susceptibility of $\chi^{(3)} \approx 3 \times 10^{-11}$ m^2/V^2 [42], a very high value however still several orders of magnitude lower than predicted here for a niobium superconductor.

In the first simulation we show what happens in a simplified scenario, when taking into account one spatial dimension only (x). Figure 2 displays the results of simulations in the 1D case analyzing the effects of a short pulse of the form $A(x,t) \equiv A_0\text{sech}(t/t_0)\sin(\omega_0 t)e^{-(x/x_0)^2}$ at intensity $I = 16I_0 = 49.6$ kW/cm^2, and a beam width $x_0 = 200$ μm; as well as an eighth order super-Gaussian pulse with the same duration and peak intensity. Figure 2(a) shows the sech-type pulse intensity (normalized to unity, blue line) and its effect on the order parameter (quantity $1-|\psi|^2$, also normalized to unity, red line). Note that we show $1-|\psi|^2$ for clarity, so when this quantity increases the pulse is *destroying* Cooper pairs. It can be seen that the order parameter drops in steplike fashion with each peak and trough of the pulse, eventually recovering after interacting with the pulse. (Note: This recovery is not shown in the figure due to space constraints; depending on the parameters used, the order parameter can take a relatively long time to recover. The recovery of the order parameter can be seen in the videos provided in the Supplemental Material [25].) Figure 2(b) presents the same data in this case for the super-Gaussian pulse, also showing a very similar dynamics. The supercurrents generated by both types of pulse are given in Fig. 2(c), while Fig. 2(d) displays the emission spectra induced by both pulses, showing the formation of odd harmonics in both cases, as expected from our theory. We now consider two spatial dimensions (x,y) in Fig. 3, which displays the results of simulations in two spatial dimensions analyzing the effects of a short pulse of the form $A(x,y,t) = A_0\text{sech}(t/t_0)\sin(\omega_0 t)e^{-(x^2+y^2)/x_0^2}$, using the same parameters as in Fig. 2. Figures 3(a) and 3(b) show frames of the evolution of the order parameters at $t = -7.5$ ps and $t = -4.5$ ps using the same horizontal scale as in Fig. 2(a) (see Supplemental Material [25] for the full videos of the evolution): Fig. 3(a) showing the dip in order parameter as the pulse first interacts with the material, while Fig. 3(b) showing the plateau as superconductivity is completely destroyed in a localized region about which pulse intensity is highest. Results in the 2D case exactly match those of the 1D case when the spatial distribution of the pulse is approximated at its origin $x = y = 0$ value, and give the same spectra and supercurrents as in Fig. 2, confirming the validity and the accuracy of the simplified 1D model above.

V. CONCLUSIONS

It is found that superconducting thin films below the critical temperature show an exceptionally large Kerr effect at low light intensities with a fast response on ultrafast time scales in the THz regime. Our simulations show in real time the effects on the order parameter of Nb due to sech-type and super-Gaussian incident pulses and the efficient generation of odd harmonics. The approach used here is simple and effective, and is generally suited to the study of both type-I and type-II superconductors. We hope this work will stimulate further investigations into superconductor-based nonlinear optical materials for ultrafast applications, with these materials proving to be a key element in future nonlinear photonics systems.

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[25] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.95.214504 for a video showing the effect of a super-Gaussian pulse on order parameter (order_parameter_and_pulse_supergaussian.mp4); a video showing the effect of a hyperbolic secant pulse on order parameter (order_parameter_and_pulse_sech.mp4); and for derivations and useful definitions (Supplemental.pdf).
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