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Nonlinear optical effects and third-harmonic generation in superconductors: Cooper pairs versus Higgs mode contribution

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The recent observation of a transmitted THz pulse oscillating at three times the frequency of the incident light paves the way to a powerful protocol to access resonant excitations in a superconductor. Here we show that this nonlinear optical process is dominated by light-induced excitation of Cooper pairs, while the collective amplitude (Higgs) fluctuations of the superconducting order parameter give in general a negligible contribution. We also predict a nontrivial dependence of the signal on the direction of the light polarization with respect to the lattice symmetry, which can be tested in systems such as, e.g., cuprate superconductors.

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The enormous technological advances made in the last two decades in time-domain spectroscopy [1,2] pose several challenges for our understanding of the interaction of light with matter. The use of low-energy THz waves [3] to first excite (pump) and then measure (probe) the system is particularly interesting for superconductors, since they can access the region $\omega < 2\Delta_0$ of the optical spectrum where linear-response absorption is suppressed by the opening of a superconducting (SC) gap Δ_0 in the quasiparticle spectrum. For example, recent [4–6] THz-pump–THz-probe experiments have shown that the probe field displays a periodic oscillation, whose possible connection to amplitude (Higgs) fluctuations of the SC order parameter has been investigated theoretically [7–11].

An interesting additional effect made possible by the use of intense electromagnetic (e.m.) THz field is the experimental observation [6] of the so-called third-harmonic generation (THG), i.e., the appearance below T_c in the transmitted pulse of a component oscillating three times faster than the incident light. This effect appears only below T_c with a maximum intensity at the temperature where the light frequency ω matches the SC gap value $\Delta_0(T)$, and has been attributed [6,12] to a resonant excitation of the Higgs mode. However, we show here that THG is dominated by the resonant excitations of Cooper pairs (CPs) (see Fig. 1), overlooked in previous theoretical work [6,12].

In contrast to pump-probe experiments [4,5], where the description of the intermediate relaxation processes of the photoexcited states becomes relevant [7–11], the THG effect can be understood as an equilibrium, nonlinear optical process. In this Rapid Communication we compute microscopically the nonlinear optical response of a superconductor and we show that the THG essentially measures lattice-modulated *density* correlations, which in the SC state diverge at the threshold $2\Delta_0$ above which CPs proliferate. This effect induces a resonant enhancement of the THG intensity when the frequency 2ω of the incoming electric field coincides with $2\Delta_0$, as observed experimentally. Once we identify the relevant nonlinear optical response function, we also find that the Higgs-mode contribution is largely subleading, due to symmetry reasons. Indeed, even if the Higgs mode can be excited by the THz field, as discussed previously [6,12], it essentially decouples from the optical probe. This is a consequence of the weak coupling between the SC amplitude and density fluctuations in BCS superconductors [13–16], as usually discussed in the context of Raman experiments [14,15,17]. The potential analogy with Raman experiments emerges also on the nontrivial dependence of the THG on the relative orientation between the e.m. field and the main crystallographic axes, due to the lattice symmetries of the band structure. This effect can be tested, e.g., in cuprate superconductors, where large monocrystals have been already studied by nonlinear spectroscopy [18]. Even though this polarization dependence can also be used to selectively excite the Higgs mode, its weak signal remains a major obstacle to its detection. Finally, by including the CP effects, missing in previous theoretical work [6,12] due to an incorrect computation of the nonlinear optical response, we reproduce very well the temperature dependence of the THG measured in Ref. [6].

We start from a microscopic SC model that captures the main ingredients of the problem:

$$H = \sum_{\mathbf{k},\sigma} \xi_{\mathbf{k}} c^\dagger_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma} - \frac{U}{N_s} \sum_{\mathbf{q}} \Phi^\dagger_\Delta(\mathbf{q}) \Phi_\Delta(\mathbf{q}), \qquad (1)$$

where $\xi_{\mathbf{k}} = \varepsilon_{\mathbf{k}} - \mu$ is the electronic dispersion with respect to the chemical potential μ, $U > 0$ is the SC coupling, and $\Phi_\Delta(\mathbf{q}) = \sum_{\mathbf{k}} c_{-\mathbf{k}+\mathbf{q}/2\downarrow} c_{\mathbf{k}+\mathbf{q}/2\uparrow}$. In mean-field approximation the Green's function in the usual basis of Nambu operators $\Psi^\dagger = (c^\dagger_{\mathbf{k}\uparrow}, c_{-\mathbf{k}\downarrow})$ reads $G_0^{-1} = i\omega_n \hat{\tau}_0 - \xi_{\mathbf{k}} \hat{\tau}_3 + \Delta_0 \hat{\tau}_1$, where $\hat{\tau}_i$

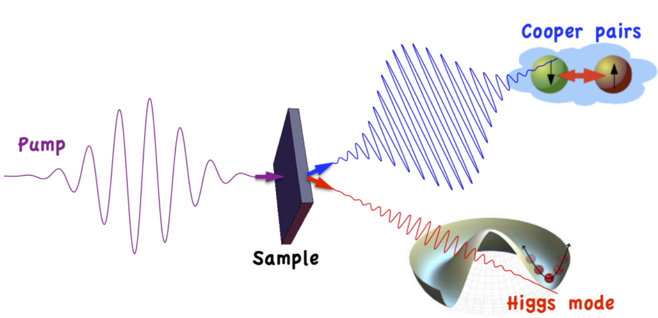

FIG. 1. Schematic of the THG. An intense THz pulse shining on the SC sample generates a transmitted component oscillating three times faster, due to the resonant excitations of Cooper pairs or Higgs fluctuations. The higher intensity of the former process can be modulated by changing the polarization of the incident light.

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are Pauli matrices, Δ_0 is the SC gap, and $E_{\mathbf{k}} = \sqrt{\xi_{\mathbf{k}}^2 + \Delta_0^2}$. The coupling to the gauge field $\mathbf{A}$ can be introduced by means of the Peierls substitution, $c^\dagger_{i+\hat{x}} c_i \rightarrow c^\dagger_{i+\hat{x}} c_i e^{ie\mathbf{A}\cdot\hat{x}}$. To derive the e.m. kernel we follow a standard procedure [19,20] to derive the action S_A written in terms of the gauge field $\mathbf{A}$ and SC collective modes. Since the coefficients of the effective action are given by fermionic susceptibilities this approach allows one to include both the quasiparticles and collective-mode contributions to the optical kernel, as has been proven already for the linear response [21,22]. As we shall see below, it turns out that the most relevant contributions to the nonlinear current $\mathbf{J}^{NL}$ can be written in compact notation as

$$J^{NL} \sim \mathbf{A}(\chi^{CP} + \chi^{H})\mathbf{A}^2, \tag{2}$$

$$\chi^{CP} \sim \langle \rho\rho \rangle, \qquad \chi^{H} \sim \langle \Delta\Delta \rangle, \tag{3}$$

where the CP contribution χ^{CP} probes lattice-modulated density fluctuations, while the Higgs contribution χ^{H} is proportional to the amplitude fluctuations. Even though both terms diverge at $\omega = \Delta_0$, the prefactor of χ^{H} turns out to be strongly suppressed by the particle-hole symmetry of the BCS solution, making it largely subdominant with respect to the CP one.

To make this argument quantitative we compute the nonlinear response by expanding the action S_A up to the fourth order in $\mathbf{A}$. For a uniform field the terms relevant for the THG are then

$$\begin{aligned} S[A] = \frac{1}{2} \sum_{\Omega_n} & e^4 A_i^2(\Omega_n) \chi_{ij}^{CP}(\Omega_n) A_j^2(\Omega_n) \\ & + X_{\Delta\Delta}(\Omega_n) |\Delta(\Omega_n)|^2 \\ & + 2e^2 A_i^2(\Omega_n) \chi_{A_i^2\Delta}(\Omega_n) \Delta(-\Omega_n), \end{aligned} \tag{4}$$

where $A_j^2(\Omega_n)$ is the Fourier transform of $[A_j(t)]^2$ in Matsubara frequency $i\Omega_n = 2\pi n T$. The first term of Eq. (4) is the CP response, as given by

$$\chi_{ij}^{CP}(i\Omega_n) = \langle \rho_i \rho_j \rangle = \Delta_0^2 \sum_{\mathbf{k}} \partial_i^2 \varepsilon_{\mathbf{k}} \partial_j^2 \varepsilon_{\mathbf{k}} F_{\mathbf{k}}(i\Omega_n), \tag{5}$$

$$F_{\mathbf{k}}(i\Omega_n) = \frac{1}{N_s} \frac{\tanh(E_{\mathbf{k}}/2T)}{E_{\mathbf{k}}\left[(i\Omega_n)^2 - 4E_{\mathbf{k}}^2\right]}, \tag{6}$$

where we introduced the short notation $\partial_i^2 \varepsilon_{\mathbf{k}} \equiv \partial^2 \varepsilon_{\mathbf{k}} / \partial k_i^2$. Here $\langle \cdots \rangle$ denotes the correlation function for the operator $\rho_i(\mathbf{q}) = \sum_{\mathbf{k}} \partial_i^2 \varepsilon_{\mathbf{k}}\ c^\dagger_{\mathbf{k}+\mathbf{q}} c_{\mathbf{k}}$, showing that χ_{ij}^{CP} scales as the density-density correlation function [given in the BCS limit by $\chi_{\rho\rho} \equiv \Delta_0^2 \sum_{\mathbf{k}} F_{\mathbf{k}}(i\Omega_n)$], as anticipated in Eq. (2) above. Indeed, the band derivatives $\partial_i^2 \varepsilon_{\mathbf{k}}$ just represent in a lattice model the equivalent of the inverse mass $1/m$ for free electrons, and they always come along with a $A_i^2(\omega)$ term in the effective action [20,22]. The second term in Eq. (4) describes the collective fluctuations of the SC amplitude $\Delta, \langle |\Delta|^2 \rangle_{\mathbf{A}=0} = 1/X_{\Delta\Delta}$, given as usual [7,13,14,16,23] by

$$X_{\Delta\Delta}(\omega) = \left(4\Delta_0^2 - \omega^2\right) F(\omega), \quad F(\omega) = \sum_{\mathbf{k}} F_{\mathbf{k}}(\omega). \tag{7}$$

By analytical continuation $i\Omega_n \rightarrow \omega + i0^+$ one can easily see that both $\chi^{CP}(\omega)$ and $F(\omega)$ display a square-root divergence as $\omega \rightarrow 2\Delta_0$, which signals the proliferation of CP above the gap. As is well known, this effect makes the Higgs a nonrelativistic mode [16], i.e., amplitude fluctuations display an overdamped resonance at $\omega = 2\Delta_0$. Finally, the third term in Eq. (4) describes the coupling between the e.m. field and the Higgs mode, mediated by the function

$$\chi_{A_i^2\Delta}(i\Omega_n) = \langle \rho_i \Delta \rangle = 2\Delta_0 \sum_{\mathbf{k}} \left(\partial_i^2 \varepsilon_{\mathbf{k}}\right) \xi_{\mathbf{k}} F_{\mathbf{k}}(i\Omega_n). \tag{8}$$

Equations (5) and (8) define the basic correlation functions needed to compute the THG. They also explain why for BCS superconductors, where they are given explicitly by the right-hand sides of Eqs. (5)–(8), $\chi_{A_i^2\Delta}$ is very small. Indeed, in the continuum limit, where the band dispersion can be approximated with a parabolic one $\varepsilon_{\mathbf{k}} \simeq \mathbf{k}^2/2m$, so that $\partial_i^2 \varepsilon_k \simeq 1/m$, $\chi_{A_i^2\Delta}$ vanishes, since

$$\chi_{A_i^2\Delta}(\omega) \simeq \frac{N_F}{m} \int_{-\infty}^{\infty} d\xi \frac{\xi}{\sqrt{\xi^2 + \Delta_0^2}\left(\omega^2 - 4\Delta_0^2 - 4\xi^2\right)} \simeq 0,$$

where the integration range can be taken symmetric due to the approximate particle-hole symmetry of the BCS solution [13,16]. This result explains the suppression of the Higgs contribution to the THG.

To derive the nonlinear e.m. kernel we integrate out the amplitude fluctuations in Eq. (4), which is equivalent to compute the random-phase approximation vertex correction of the bare bubble χ_{ij}^{CP} [12,20]. One is then left with the action depending on the e.m. field only,

$$S^{(4)}[A] = \frac{e^4}{2} \int dt\, dt' \sum_{i,j} A_i^2(t) K_{ij}(t-t') A_j^2(t'), \tag{9}$$

$$K_{ij}(t-t') = \left[\chi_{ij}^{CP}(t-t') + \chi_{ij}^{H}(t-t')\right], \tag{10}$$

where the Higgs contribution χ_{ij}^{H} reads

$$\chi_{ij}^{H}(i\Omega_n) \equiv -\frac{\chi_{A_i^2\Delta}(i\Omega_n)\chi_{A_j^2\Delta}(i\Omega_n)}{X_{\Delta\Delta}(i\Omega_n)}, \tag{11}$$

and it is also diverging at $\omega = 2\Delta_0$ due to the vanishing of $X_{\Delta\Delta}$ [see Eq. (7)]. The nonlinear current J_i^{NL} follows by functional derivative of Eq. (9) with respect to $\mathbf{A}$:

$$\begin{aligned} J_i^{NL}(t) &= -\frac{\delta S^{(4)}[A]}{\delta A_i(t)} \\ &= -2e^4 A_i(t) \int dt' \sum_j K_{ij}(t-t') A_j^2(t'). \end{aligned} \tag{12}$$

Equation (12), with the definition (10) of the e.m. kernel, corresponds to Eq. (2) above. For a monochromatic incident field $\mathbf{A} = \bar{\mathbf{A}} \cos(\Omega t)$ it is given by

$$\begin{aligned} J_i^{NL}(t) = \frac{e^4 \bar{A}_i}{4} \sum_j & \{ e^{-3i\Omega t} K_{ij}(2\Omega) \\ & + e^{-i\Omega t}[2K_{ij}(0) + K_{ij}(2\Omega)] + \text{c.c.} \} \bar{A}_j^2, \end{aligned} \tag{13}$$

where one recovers the term oscillating at three times the incident frequency, with an amplitude controlled by the kernel K_{ij} evaluated at 2Ω. In the experiments of Ref. [6] the physical observable is the transmitted electric field $\mathbf{E}^{tr}$, which one expects to be proportional to the current (13). As a consequence, the intensity of the THG can be evaluated from Eq. (13) as $I_i^{THG}(\Omega) \propto |\int dt\, J_i^{NL}(t) e^{3i\Omega t}|^2$, which for a monochromatic wave gives

$$I_i^{THG}(\Omega) = I_0 e^8 A_i^2 \left| \sum_j K_{ij}(2\Omega) \bar{A}_j^2 \right|^2, \qquad (14)$$

where I_0 is an overall scale factor that depends on the geometry of the experiment.

To quantify explicitly the lattice effects we compute the nonlinear response for a nearest-neighbor tight-binding model on the square lattice $\varepsilon_{\mathbf{k}} = -2t(\cos k_x + \cos k_y)$, and we will consider first the half-filled case $n = 1$ ($\mu = 0$), where only the SC amplitude mode contributes to the nonlinear response. By making the replacement $\partial_i^2 \varepsilon_{\mathbf{k}} = 2t \cos k_i$ in Eqs. (5) and (8) one sees that $\chi_{A^2\Delta}$ is independent of the direction, while the CP part (5) is a tensor. Let us first consider the case of a field applied along the x axis, so that I_x^{THG} is controlled by the longitudinal K_{xx} kernel. The two separate CP $[I_x^{CP}(\Omega) \propto |\chi_{xx}^{CP}(2\Omega)|^2]$ or Higgs $[I_x^H(\Omega) \propto |\chi^H(2\Omega)|^2]$ contributions to the THG intensity for a monochromatic field are shown in Fig. 2(a). As one can see, even if the functional form is similar for the two terms, the CP contribution is much larger, and one can roughly estimate $I_x^H \sim (\Delta/U)^4 I_{xx}^{CP}$. The predominance of the CP response implies also a nontrivial dependence of the THG intensity on the direction of the incoming applied field with respect to the crystallographic axes. In the general case of

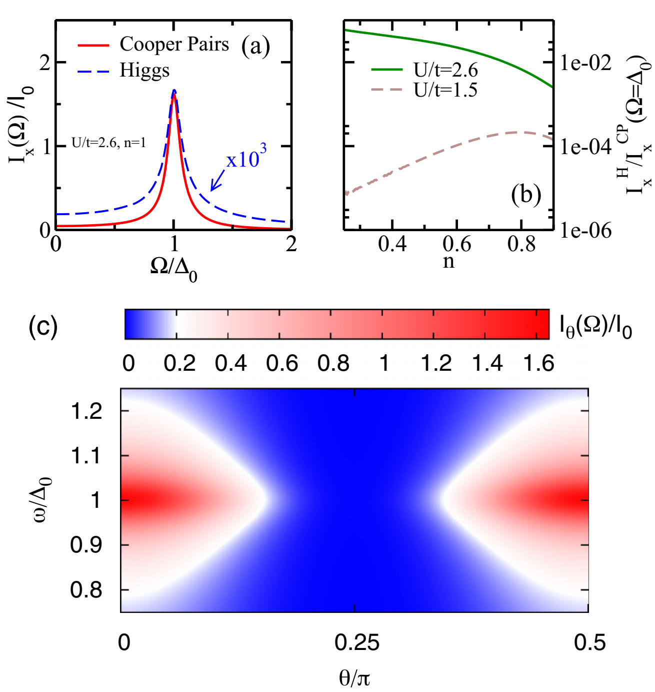

FIG. 2. (a) Comparison between the CP and Higgs contribution to the THG intensity at $T = 0$ as a function of the frequency Ω for a field along x and a residual broadening $\omega + i\delta$, $\delta = 0.1\Delta_0$. (b) Relative intensity of the Higgs and CP processes at $\omega = \Delta_0$ as a function of the density for two values of the SC coupling. Here the Coulomb screening from Eqs. (20) and (21) has been included. (c) Map of the frequency dependence of the THG intensity, Eq. (15), for a field at arbitrary angle θ with respect to the x direction.

$\bar{\mathbf{A}} = A_0 \cos(\Omega t)(\cos\theta, \sin\theta)$, θ being the angle with respect to the x axis, the intensity of the transmitted pulse in the field direction is

$$I_\theta^{THG}(\Omega) = I_0 e^8 A_0^6 |K_\theta(2\Omega)|^2, \qquad (15)$$

$$K_\theta = \chi_{xx}^{CP}(\cos^4\theta + \sin^4\theta) + 2\chi_{xy}^{C} \sin^2\theta \cos^2\theta + \chi^H, \qquad (16)$$

where we used the fact that $\chi_{yy/yx}^{CP} = \chi_{xx/xy}^{CP}$. When $\bar{\mathbf{A}}$ is applied along the diagonal ($\theta = \pi/4$) the longitudinal χ_{xx}^{CP} and transverse χ_{xy}^{CP} parts of the CP response are equally weighted. In this peculiar configuration one sees from the definitions (5)–(8) that $\chi_{xx}^{CP} + \chi_{xy}^{CP} = -\Delta_0 \chi_{A^2\Delta}/2$, i.e., the diverging CP contribution cancels out, and only the resonant Higgs response remains:

$$J_{\pi/4}^{NL}(t) = \frac{e^2 \Delta_0}{U} A(t) \langle \Delta(t) \rangle, \qquad (17)$$

where $\langle \Delta(t) \rangle$ is the average value of the amplitude fluctuations obtained from Eq. (4), i.e., $\langle \Delta(\omega) \rangle = e^2 \chi_{A^2\Delta}(\omega) \mathbf{A}^2(\omega) / X_{\Delta\Delta}(\omega)$ in the frequency domain. The angular dependence of $I_\theta^{THG}(\Omega)$ is shown in Fig. 2(c) as a color map: at $\theta = \pi/4$, where one probes only the Higgs mode, the intensity is strongly suppressed, in agreement with the result shown in Fig. 2(a). This prediction can be tested in systems like cuprate superconductors, where the band structure has in first approximation the symmetry discussed here and large monocrystals have been already used to probe SC resonances by pumping the system with near-infrared light [18]. It is worth noting that all these effects have been completely overlooked in the previous work [6,12] due to the incorrect replacement in the CP term (5) of the quantity $\partial_i^2 \varepsilon_{\mathbf{k}}$, which is *finite* at the Fermi surface, with $\xi_{\mathbf{k}}$, which is instead vanishing. This assumption removes both the divergence of the CP term $\chi_{ij}^{CP}(\omega)$ at $\omega = 2\Delta$ and its direction dependence, and leads always to the result (17), which is instead far from being generic. In addition, we also checked [20] that expression (10) can be obtained as well by means of the pseudospin formalism used in Refs. [6,12].

Since the response function (10) is dominated by the electronic states at the Fermi surface, the quantitative difference between the CP and the Higgs response depends in general on the electron density n. To quantify this effect away from half-filling one should retain in derivation (4) of the effective action also the terms [20] coupling the gauge field and the Higgs mode to the phase and density fluctuations, mediated by the response functions

$$\chi_{A_i^2\rho}(i\Omega_n) = \langle \rho_i \rho \rangle = \Delta_0^2 \sum_{\mathbf{k}} \partial_i^2 \varepsilon_{\mathbf{k}} F_{\mathbf{k}}(i\Omega_n), \qquad (18)$$

$$\chi_{\rho\Delta}(i\Omega_n) = \langle \rho \Delta \rangle = \Delta_0 \sum_{\mathbf{k}} \xi_{\mathbf{k}} F_{\mathbf{k}}(i\Omega_n). \qquad (19)$$

These terms, which vanish by particle-hole symmetry in the half-filled case, are crucial to account for the screening effects of the long-range Coulomb potential, in analogy again with the known result for the Raman response function [17]. By means of straightforward but lengthly calculations one can then show [20] that the nonlinear response function retains the structure (10) with the replacements

$$\chi_{ij}^{CP} \rightarrow \chi_{ij}^{CP,sc} = \chi_{ij}^{CP} - \frac{\chi_{A^2\rho}^2}{\chi_{\rho\rho}}, \tag{20}$$

$$\chi^H \rightarrow \chi^{H,sc} = -\frac{(\chi_{A^2\Delta} - \chi_{A^2\rho}\chi_{\rho\Delta}/\chi_{\rho\rho})^2}{X_{\Delta\Delta} - \chi_{\rho\Delta}^2/\chi_{\rho\rho}}, \tag{21}$$

where we used the fact that for the lattice model under consideration the function (18) is isotropic in the spatial indexes. While the mixing to the density and phase modes does not affect [16] the pole of the Higgs mode, identified now by the vanishing of the denominator of Eq. (21), it is crucial to screen both the CP and Higgs response as one moves away from half-filling. In Fig. 2(b) we show the ratio $I_x^{CP}(\Delta)/I_x^H(\Delta)$ as a function of the electron density n for two values of the SC coupling U. As we can see, even for the large value $U/t = 2.6$ of the SC coupling, where the deviations from the BCS (approximate) particle-hole symmetric case become more prominent, inducing a larger coupling of the Higgs to the light, the CP part remains the predominant one for the longitudinal response even in the low-density regime.

In addition to the strong direction dependence of the THG intensity, a second check of the origin of the THG effect is its temperature evolution, measured in Ref. [6]. For a polycrystalline sample one should average the kernel Eq. (16) over θ, to account for the random direction of the e.m. field with respect to the crystallographic axes. One then finds that $\bar{J}_\theta^{NL} = [J_x^{NL} + J_{\pi/4}^{NL}]/2 \simeq J_x^{NL}/2$, so that one expects that the CP processes dominate. To check this we compute [20] the nonlinear current induced by an incoming electric field $\mathbf{A}(t)$ having a wave-packet profile similar to the one used in the experiments of Ref. [6] [see Fig. 3(a)]. In the frequency domain this wave packet corresponds to the power spectra shown in Fig. 3(b), centered at three possible values Ω_i of the incoming frequency. The temperature evolution of the THG intensity, i.e., $I_x(3\Omega_i)$, is then shown in Figs. 3(d)–3(f), where we compare the experimental data from Ref. [6] [panel (d)] with the theoretical calculations done including only the CP processes [panel (e)] or the Higgs contribution [(panel (f)]. Apart from the small overall intensity of the THG Higgs signal, which cannot be seen in the normalized data of Fig. 3, the excitation of the Higgs mode alone fails to reproduce the temperature dependence of the signal at the lowest frequency Ω_1.

In conclusion, we studied the nonlinear optical effects responsible for the THG in a superconductor. Since the relevant response function (10) measures lattice-modulated density fluctuations [see Eq. (5)], the optical process responsible for the THG is equivalent to a resonant excitation of CP. The Higgs-mode contribution is instead much smaller, since its coupling (8) to the optical probe is suppressed by symmetry, in analogy with standard Raman experiment [13–15,17], unless some additional channel makes the Higgs Raman visible [13,14,24]. In addition, also for THG experiments one can orient the light polarization with respect to the main crystallographic axes in order to modulate the THG intensity due to Cooper pairs. Even though this effect can be used in principle to selectively excite the Higgs signal, the weakness of its coupling to the light represents a major obstacle to its detection in optical experiments, in analogy with the results recently discussed in the context of linear optical spectroscopy [16]. The interplay between the nonlinear optical effects discussed here and the non-equilibrium processes [7–11] addressed in the context of pump-probe experimental protocols [1,2,4,5] remains an open question, which certainly deserves future experimental and theoretical work.

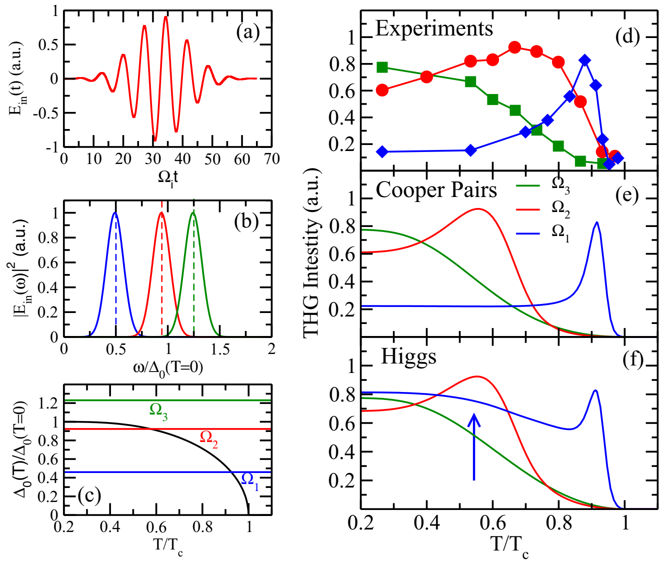

FIG. 3. (a) Profile of the incoming field, and (b) corresponding power spectra, used to simulate the experiments of Ref. [6]. The three central frequencies Ω_i of the power spectra are compared in panel (c) to the temperature dependence of the SC gap. For each Ω_i the resonant condition $\Omega_i = \Delta_0(T)$ occurs at a different temperature, in analogy with Ref. [6]. (d), (e) Temperature evolution of the THG intensity in the experiments (d), in the case of CP processes (e), and in the case of the Higgs processes alone (f), computed for $n = 1$, $U/t = 2.6$. Data computed at different Ω_i are normalized to have a similar overall scale, as done in panel (d). The excitation of the Higgs mode alone gives the wrong T dependence of the THG signal for the lowest frequency Ω_1, as marked by the arrow.

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[1] J. Orenstein, Phys. Today **65**(9), 44 (2012).

[2] C. Giannetti, M. Capone, D. Fausti, M. Fabrizio, and F. Parmigiani, arXiv:1601.07204 [Adv. Phys. (to be published)].

[3] For a review, see, e.g., T. Kampfrath, K. Tanaka, and K. A. Nelson, Nat. Photonics **7**, 680 (2013).

[4] Ryusuke Matsunga and Ryo Shimano, Phys. Rev. Lett. **109**, 187002 (2012).
[5] Ryusuke Matsunaga, Yuki I. Hamada, Kazumasa Makise, Yoshinori Uzawa, Hirotaka Terai, Zhen Wang, and Ryo Shimano, Phys. Rev. Lett. **111**, 057002 (2013).
[6] Ryusuke Matsunaga, Naoto Tsuji, Hiroyuki Fujita, Arata Sugioka, Kazumasa Makise, Yoshinori Uzawa, Hirotaka Terai, Zhen Wang, Hideo Aoki, and Ryo Shimano, Science **345**, 1145 (2014).
[7] A. F. Volkov and S. M. Kogan, Zh. Eksp. Teor. Fiz. **65**, 2038 (1973) [Sov. Phys. JETP **38**, 1018 (1974)].
[8] R. A. Barankov, L. S. Levitov, and B. Z. Spivak, Phys. Rev. Lett. **93**, 160401 (2004).
[9] R. A. Barankov and L. S. Levitov, Phys. Rev. Lett. **96**, 230403 (2006).
[10] H. Krull, D. Manske, G. S. Uhrig, and A. P. Schnyder, Phys. Rev. B **90**, 014515 (2014).
[11] T. Papenkort, T. Kuhn, and V. M. Axt, Phys. Rev. B **78**, 132505 (2008).
[12] N. Tsuji and H. Aoki, Phys. Rev. B **92**, 064508 (2015).
[13] P. Littlewood and C. M. Varma, Phys. Rev. B **26**, 4883 (1982).
[14] T. Cea and L. Benfatto, Phys. Rev. B **90**, 224515 (2014).
[15] D. Pekker and C. M. Varma, Ann. Rev. Condensed Matter Phys. **6**, 269 (2015).
[16] T. Cea, C. Castellani, G. Seibold, and L. Benfatto, Phys. Rev. Lett. **115**, 157002 (2015).
[17] T. P. Deveraux and R. Hackl, Rev. Mod. Phys. **79**, 175 (2007).
[18] B. Mansart, J. Lorenzana, A. Manna, A. Odehb, M. Scarongella, M. Cherguib, and F. Carbone, Proc. Natl. Acad. Sci. USA **110**, 4539 (2013).
[19] N. Nagaosa, *Quantum Field Theory in Condensed Matter Physics* (Springer, New York, 1999).
[20] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.93.180507 for more details.
[21] A. Paramekanti, M. Randeria, T. V. Ramakrishnan, and S. S. Mandal, Phys. Rev. B **62**, 6786 (2000).
[22] See, e.g., L. Benfatto, A. Toschi, and S. Caprara, Phys. Rev. B **69**, 184510 (2004).
[23] I. O. Kulik *et al.*, J. Low Temp. Phys. **43**, 591 (1981).
[24] M.-A. Méasson, Y. Gallais, M. Cazayous, B. Clair, P. Rodière, L. Cario, and A. Sacuto, Phys. Rev. B **89**, 060503(R) (2014).