

Two-dimensional terahertz spectroscopy in electronic systems: A many-body diagrammatic approach

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The term “two-dimensional coherent spectroscopy” (2DCS) usually refers to experimental setups where a coherently generated electric field in a sample is recorded over many runs as a function of two time variables: the delay τ between two consequent excitation pulses and the time t over which the signal is emitted. Even if its implementation in the femtosecond time domain for investigation of vibrational molecular states was developed more than two decades ago, its experimental development in the terahertz domain with application to interacting electronic systems is still in its infancy. The present work aims at providing a theoretical framework for the description and interpretation of 2DCS by using the same many-body language based on a perturbative diagrammatic expansion that has been largely developed and applied in the literature to linear spectroscopy. By focusing on the case of centrosymmetric systems, we show how the problem of the theoretical interpretation of the 2D maps can be recast in the solution of two different but complementary problems. The first one is the evaluation of a third-order response function to the gauge field, whose derivation in the velocity gauge leads to a semianalytical expression for the computation of the 2D maps in the case of simple band dispersions and a Gaussianlike pulse envelope. This has the twofold advantage of reducing considerably the computational complexity and to guide the assignment of the spectral features to microscopic processes, as we demonstrate explicitly for a toy model of electrons in a semiconductinglike band structure. The second one is a careful description of multiwave propagation effects inside the material, that for bulk systems can completely screen the intrinsic properties of the nonlinear response, as we show for the case of soft superconducting Josephson plasmons. Our results provide a theoretical foundation prone to further extension to several interacting systems and offer a flexible method for modeling realistically the nonlinear responses across arbitrary spectral widths of the driving fields.

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I. INTRODUCTION

Recent experimental advances in the generation and control of phase-stable light pulses in the terahertz regime opened the way to the exploration of a wide range of phenomena connected to coherent driving of excitations in this energy window [1,2]. On the one hand, terahertz pulses can be used to selectively drive infrared active phonon modes, with the aim of inducing time-dependent modifications of the lattice structure that are not possible at equilibrium [3,4]. On the other hand, in the terahertz regime one can directly access the continuum of low-energy electronic excitations of metals, without inducing optical transitions to higher-energy

bands, as happens instead with light pulses in the visible range [5]. Such a perspective is particularly promising when the metallic state undergoes a phase transition to a lower symmetry state, as happens across a superconducting (SC) or charge-density wave (CDW) transition. In this case, not only the fermionic excitation spectrum is modified by the opening of a gap, but simultaneously new collective modes connected to the order-parameter fluctuations emerge [6–9]. Terahertz spectroscopy, and in particular its nonlinear counterpart, then arises as a preferential tool to coherently drive these collective excitations by avoiding the chain of relaxation processes occurring when the exciting pulse lies in the visible range [10,11].

From the theoretical side, the investigation of systems with a continuum of low-energy excitations, which eventually develop a gap in the terahertz range below a phase transition, has been so far based on effective models where only states near the Fermi level are considered. The nonlinear current generated by the intense terahertz field is computed in the vector-potential gauge $\mathbf{A}(t)$, coupled to the Hamiltonian via the minimal coupling shift $\mathbf{p} - e/c\mathbf{A}$ of the momentum operator. In centrosymmetric systems, the leading nonlinear current

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can then be expressed as

$$J^{(3)}(\omega) = \int d\omega_i K^{(3)}(\omega_1, \omega_2, \omega_3) A(\omega_1) A(\omega_2) A(\omega_3) \times \delta(\omega - \omega_1 - \omega_2 - \omega_3) \quad (1)$$

where space indices are omitted for simplicity, $A(\omega)$ is the Fourier transform of the perturbing terahertz field, and $K^{(3)}$ stands for the nonlinear optical kernel, obtained as a third-order response function to the perturbation $A(t)$. Despite the simplification of considering only low-energy electronic states, the computation of $K^{(3)}$ requires a careful account for processes of *diamagnetic* or *paramagnetic* nature, as the extensive literature on superconductors has shown [12–18]. The terminology relies on the fundamental light-electron coupling obtained via the minimal-coupling scheme, i.e.,

$$H_{\text{em}} = -\mathbf{j} \cdot \mathbf{A} + \rho \mathbf{A}^2 = H_{\text{para}} + H_{\text{dia}} \quad (2)$$

where $\mathbf{j}$ is the electronic current and ρ the electronic density. In the paramagnetic process one photon excites an electron-hole pair, while in the diamagnetic one this occurs via two photons coming at the same time. The main consequence for what concerns the nonlinear response $K^{(3)}$ is that diamagnetic interactions give rise to two-point (Kubolike) correlation functions, which share some analogies with those measured in the linear response, while paramagnetic interactions require one to compute up to four-point correlation functions, whose frequency properties are not accessible by linear spectroscopy.

As the benchmark case of superconductors has shown, the two classes of processes (diamagnetic vs paramagnetic) provide different and complementary information on the interactions at play in the systems. Theoretical predictions have been tested, for example, against experiments of high harmonic generation, where a single light pulse impinges on the sample and the nonlinear electric field generated by the nonlinear current at different harmonics is measured [11,19–21].

However, a complete experimental characterization of the different microscopic processes would require knowledge of the dependence of $K^{(3)}$ on *three* different frequencies, which is not possible simply reading $J^{(3)}$ on a single ω , as in Eq. (1). Recent experimental advances on the so-called two-dimensional terahertz spectroscopy (2DTS) made a significant step forward in this direction [22–24]. The experimental setup is based on the detection of the nonlinear signal induced by two strong light pulses delayed by an interval τ , by recording the response as a function of the acquisition time t and the delay τ [25–28]. When 2D temporal maps are transformed in the frequency domain, it is possible to read out different frequency combinations separately in the nonlinear kernel $K^{(3)}$.

In preliminary applications of 2DTS to electronic systems, glimpses of novel effects have been seen, including the possibility to experimentally disentangle signatures of superconducting amplitude modes [29–35], to address separately the role of disorder and inhomogeneity [36–38], to distinguish energy relaxation from momentum relaxation pathways [39,40], and to probe coupling between distinct modes in magnetic materials [41–43]. However, a general theoretical framework capable of providing a link between

these complex experimental protocols and the properties of the higher-order correlation functions involved in the measurements is still under construction [44–52]. This is in part due to the fact that 2DTS has traditionally a long history of applications to molecular systems [25–27,53], where the fundamental light-matter process encompasses a dipolarlike excitation of vibrational states, which is usually described with the density-matrix evolution in the time domain for a nondispersive two-level system.

The present manuscript aims to provide a completely different perspective, motivated by the preliminary results of the emerging field of 2DTS in electronic systems. Our goal is to provide a theoretical description of 2DTS protocols based on the computation of third-order correlation functions in the frequency domain via a diagrammatic approach, able to account for the features related to the existence of a continuum of excitations and their diamagnetic or paramagnetic coupling to light. This goal will be achieved by addressing two complementary aspects. The first one concerns the interplay between the spectral content of the pump field and the resonances of the nonlinear optical kernel. Such an interplay will be discussed within a paradigmatic toy model for a semiconductinglike band structure. We will show that diamagnetic and paramagnetic processes, connected to two-photon (diamagnetic) or single-photon (paramagnetic) excitation mechanisms, lead to markedly different signatures in the 2DTS maps, which can be used experimentally to test their relative importance.

The second aspect concerns the connection between the nonlinear response induced within the system and the nonlinear signal detected outside the sample, taking into account propagation effects [54–56]. For thin samples and materials with a featureless refractive index in the spectral range of the pump, such propagation effects play a minor role; conversely, for thick samples and in the presence of dipolar-active modes, they can even mask the intrinsic response of the system. This problem can be efficiently addressed by perturbatively solving Maxwell's equations in the presence of a nonlinear source current coming from Eq. (1), along the lines suggested in some recent publications [57–59]. In this manuscript, we will provide a general solution applicable to different experimental geometries once the frequency-dependent refractive index $n(\omega)$ of the material is known, and we will show the extreme sensitivity of 2DTS maps to such propagation effects. As a benchmark example of their relevance, we will discuss the case of soft Josephson plasmons, which represent the fundamental collective excitation of the phase degrees of freedom in layered superconductors, like cuprate superconductors. Josephson plasmons provide a natural source for nonlinear optical response, since their dynamics in the presence of a terahertz pump polarized along the stacking direction is described by an intrinsically anharmonic Hamiltonian:

$$H = J \cos \left(\phi - \frac{2e}{\hbar c} A \right). \quad (3)$$

Here ϕ denotes the interlayer phase difference between stacked planes coupled by a Josephson interaction J . In this context as well, the problem has been mostly studied in the past by solving in the time domain the nonlinear sine-Gordon equation for the current obtained from Eq. (3) [60–64]. However, recent measurements through a 2DTS setup [38,65]

turned the attention to the interpretation of the data in the 2D frequency maps. According to the same general principles outlined above, one should address the problem by taking into account that the Josephson model in Eq. (3) admits a diamagneticlike nonlinear coupling $\sim \phi^2 A^2$ of the plasma modes to the light pulses [66]. As we show below, this simple reasoning completely fails in providing an explanation for the experiments that crucially requires one to account for the modification of the driving pulse when it propagates inside the material. More specifically, we will show how simulations of the 2DTS maps generated by the nonlinear driving of Josephson plasmons in cuprates can be extremely sensitive to fine details of the modeling of the propagation effects. This finding suggests that some extra care should be used when the refractive index of the system is strongly frequency dependent in the spectral range covered by the pump, as it occurs for example in the case of infrared-active vibrational modes [54–56].

A. Synopsis of the paper

The present summary aims at providing the pipeline of the main results of the manuscript to guide the reader through the

various sections. The starting point will be the simple case of transmission through a nonabsorbing thin film, discussed in Secs. II and III, where the signal $E_{2D}(t, \tau)$ measured in 2D spectroscopy can be mapped into the nonlinear third-order current generated inside the material $J_{2D}^{(3)}(t, \tau)$:

$$E_{2D}(t, \tau) \sim J_{2D}^{(3)}(t, \tau). \quad (4)$$

As it is customary in the context of electronic systems, the current will be computed in response to an applied gauge potential $A(t)$. The Fourier transform $J_{2D}^{(3)}(\omega_t, \omega_\tau)$ will then be shown to depend on the convolution between the spectral content of the pump and probe pulses, denoted with $A_\tau(\omega)$ and $A_0(\omega)$, respectively, and the nonlinear (symmetric) kernel $K_S^{(3)}$, which encodes all the information on the resonant excitations of the system. The quantity $J_{2D}^{(3)}$ depends generically on the product of three electromagnetic fields: one can distinguish processes scaling as $A_\tau A_\tau A_0$ from processes scaling as $A_\tau A_0 A_0$. By making the dependence on the detection frequency ω_t and on the delay frequency ω_τ explicit, we show in Eqs. (14) and (15) that one can express these contributions as

$$E_{2D}(\omega_t, \omega_\tau) \sim A_0(\omega_t - \omega_\tau) \int d\omega K_S^{(3)}(\omega_t - \omega_\tau, \omega, \omega_\tau - \omega) A_\tau(\omega) A_\tau(\omega_\tau - \omega) \\ + A_\tau(\omega_\tau) \int d\omega K_S^{(3)}(\omega_\tau, \omega, \omega_t - \omega_\tau - \omega) A_0(\omega) A_0(\omega_t - \omega_\tau - \omega). \quad (5)$$

We notice that the first term of Eq. (5) would be also present in usual pump-probe experiments where the probe field is weaker than the pump one, while the second only arises when the two delayed pulses have the same strength.

Equation (5) will be used to discuss the outcome of 2D maps as one varies the bandwidth of the pulses, encoded in the $A_{\tau,0}(\omega)$ functions, and the nature of the excitations of the system, described by the kernel $K_S^{(3)}$. In Sec. II we discuss standard results for a monochromatic pulse where $A_{\tau,0}(\omega)$ only contains $\delta(\omega \pm \Omega)$. In this case the 2D maps present simple spots at multiples of the pulse frequencies $\pm\Omega$, as shown in Fig. 3. In the complementary case of an impulsive electric field, that can be approximated with a delta function in time, the 2DTS is dominated by the typical excitations processes described by the nonlinear kernel $K_S^{(3)}$. As a working example we discuss in Sec. II B the nondispersive two-level system, showing how one can reproduce within a diagrammatic language the standard results derived in the literature by means of a density-matrix formalism in real time, as shown in Fig. 6. At this stage, it should be emphasized that in realistic experiments the impulsive limit is rarely achieved. The first consequence of Eq. (5) is that one should preliminarily study the convolution spectra of the pump and probe pulses. For realistic single-cycle terahertz pulses the 2D map obtained by setting $K_S^{(3)} \sim \text{const}$ in Eq. (5) contains elongated features in correspondence of multiples of the central pulse frequency, as shown in Fig. 1(a). As a consequence, any feature of the kernel

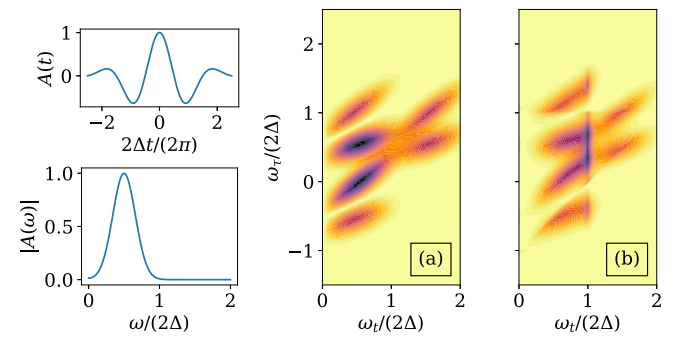


FIG. 1. Time-domain envelope of $A(t)$ representing the typical waveform for realistic few-cycles pulses used in the experiments (top). The Fourier transform (bottom) has a finite spectral width around the central frequency Ω , set in this example at $\Omega = \Delta$. (a) 2D map obtained from Eq. (5) for a frequency-independent $K_S^{(3)}$. In 2D experiments with pulses having a finite duration in time the convolution of the $A_{0,\tau}(\omega)$ spectra in Eq. (5) sets the region in the 2D map where one can access the system excitations, encoded in the nonlinear kernel $K_S^{(3)}$. The 2D features represent a broadened version of the spotlike response expected for a monochromatic pulse, and shown in Fig. 3 below. (b) When the full structure of $K_S^{(3)}$ is included in Eq. (5) the spectral weight gets redistributed in the 2D plane. In the present case we show the results for the semiconductinglike model derived in Sec. III where the main resonances follow the spectral gap at 2Δ . The parameters are the same used in Fig. 10(b) below.

which resides outside this convolution will not be accessible in the experimental situation of interest. When the resonances of the kernel overlap with the 2D map of the pulses the spectral weight is redistributed, and additional features highlighting the system excitations appear, as shown in Fig. 1(b).

In Sec. III we extend the formalism to dispersive electronic systems and consider as an example a toy model for a semiconductinglike band structure. We discuss the main steps needed to obtain a semianalytical form of the 2D signal, that are encoded in the set of Eqs. (56)–(66). The main goal of this section is to highlight the different signatures in the 2D maps that can be ascribed to diamagneticlike vs paramagneticlike processes. As we shall discuss in the following, these subsets of processes are interesting *per se* since they

can be relevant in contexts beyond the toy model considered here.

A final aspect addressed in the manuscript is the generalization of Eq. (4) to absorbing and bulk systems. In this case, one cannot simply approximate the 2D detected signal $E_{2D}(t, \tau)$ by the nonlinear current generated inside the material, since the latter depends on the local value of the gauge potential within the sample. We will show that one can extend Eq. (5) to include two ingredients: (i) the boundary conditions at the sample/vacuum interfaces, that connect the external field A_{ext} to the internal one via the refractive index $n(\omega)$ of the material, and (ii) the propagation effects inside the sample, that modify the four-wave mixing process behind the generation of a nonlinear current. Equation (5) can then be generalized to

$$E_{2D}(\omega_t, \omega_\tau) \sim \int d\omega K_S^{(3)}(\omega_t - \omega_\tau, \omega, \omega_\tau - \omega) M(\omega_t - \omega_\tau, \omega, \omega_\tau - \omega) A_{0,\text{ext}}(\omega_t - \omega_\tau) A_{\tau,\text{ext}}(\omega) A_{\tau,\text{ext}}(\omega_\tau - \omega) \\ + \int d\omega K_S^{(3)}(\omega_\tau, \omega, \omega_t - \omega_\tau - \omega) M(\omega_\tau, \omega, \omega_t - \omega_\tau - \omega) A_{\tau,\text{ext}}(\omega_\tau) A_{0,\text{ext}}(\omega) A_{0,\text{ext}}(\omega_t - \omega_\tau - \omega) \quad (6)$$

where $M(\omega_1, \omega_2, \omega_3)$ is a function of $n(\omega)$ that depends on the detection scheme (reflection or transmission), which can be thought of as a nonlinear generalization of the usual Fresnel coefficients. The explicit form of Eq. (6) to the case of reflection from a bulk sample is presented in Eq. (68), and its analogous in transmission is given in the Supplemental Material (SM) [67], where we also provide detailed steps for its derivation. As a case study of the relevance of this generalization we discuss in Secs. IV B–IV D the nonlinear response of superconducting Josephson plasmons in cuprate superconductors. In Sec. V we provide the final comments and an outlook on possible extensions of the present work. Since the manuscript aims at a pedagogical discussion of several relevant aspects, we provide further technical details, along with useful formulas for the computation of the 2D maps, in the SM [67].

II. GENERAL FORMALISM OF 2DTS

As a starting point, we need to connect the outcome of a 2D measurement to a third-order response function of the system. Let us consider the general 2D protocol sketched in Fig. 2. Here we denote with $E_0(s)$ the “fixed” field, whose envelope is centered at $s = 0$, and $\bar{E}_\tau(s)$ the “moving” one, which is centered at $s = -\tau$. Notice that we do not restrict here the time ordering of the two pulses, which can be varied according to the sign of τ : we thus have that either $\bar{E}_\tau$ or E_0 comes first for $\tau > 0$ or $\tau < 0$, respectively [68]. The experimental procedure consists in collecting the generated response at a time $s = t$ when both fields are present, $E_{0,\tau}^{\text{out}}(t, \tau)$ in Fig. 2(a), or when only one of them illuminates the system, $E_0^{\text{out}}(t)$ and $E_\tau^{\text{out}}(t, \tau)$ in Figs. 2(b) and 2(c), respectively. The generated radiation is collected as a function of (t, τ) and the differential signal is recorded as

$$E_{2D}(t, \tau) \equiv E_{0,\tau}^{\text{out}}(t, \tau) - E_0^{\text{out}}(t) - E_\tau^{\text{out}}(t, \tau). \quad (7)$$

In practice, the three signals entering Eq. (7) are extracted using a differential chopping scheme combined with

phase-resolved electro-optic sampling of the emitted terahertz field [31,37]. Here, the two excitation pulses are selectively modulated, and one can isolate the nonlinear contribution corresponding to the combined action of both fields, which is mathematically equivalent to the subtraction in Eq. (7). This equivalence holds provided that the system response is stationary across laser shots and that relaxation to equilibrium occurs on a timescale shorter than the pulse repetition rate, conditions typically satisfied in terahertz 2D spectroscopy experiments employing low to moderate fluences.

In the simple case of nonabsorbing thin films the $E_{2D}(t, \tau)$ can be approximated with the (nonlinear) current $J_{2D}(t, \tau)$

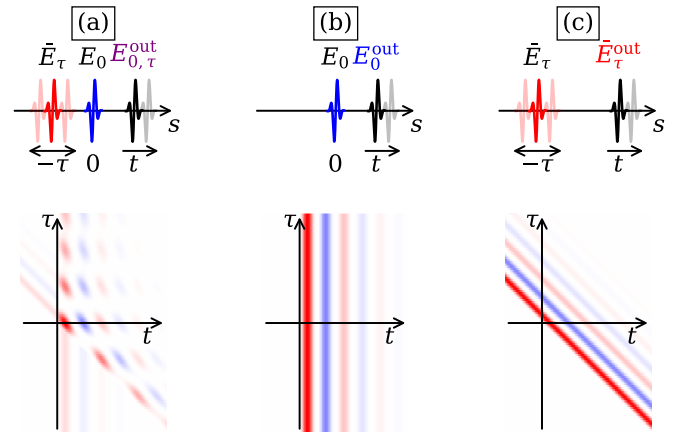


FIG. 2. Schematic representation of the three signals that generate the experimental 2D map according to Eq. (7). Top row: The field E_0 (blue) is kept fixed at $s = 0$, the field $\bar{E}_\tau$ (red) is centered at $s = -\tau$, and the outgoing signal is collected at time $s = t$. Both t and τ are scanned (light shading), with the latter being either positive (E_τ coming first) or negative (E_0 coming first). The signal is collected when both fields are present, $E_{0,\tau}^{\text{out}}$ in panel (a), or when only one of the two is present, E_0^{out} in panel (b) and E_τ^{out} in panel (c). Bottom row: Resulting signal for the three situations assuming a nonlinear polarization as generated by the two-level system discussed in Sec. II B.

induced in the system, $E_{2D}(t, \tau) \sim J_{2D}(t, \tau)$, neglecting thus any effect of the propagation of the electric fields inside and outside the sample, as detailed in the SM [67]. The analysis of Sec. IV will clarify the limits of such approximation. Consequently, one can *theoretically* compute the quantity:

$$J_{2D}(t, \tau) = J[A_{0,\tau}](t, \tau) - J[A_0](t) - J[\bar{A}_\tau](t, \tau) \quad (8)$$

where each term represents the induced current J in the presence of two ($A_{0,\tau}$) or one ($A_0, \bar{A}_\tau$) driving fields, here represented by their associated gauge potential, related by $E(t) = -\partial_t A$. The signal collected in time is usually Fourier transformed to the conjugated frequencies domain $(t, \tau) \rightarrow (\omega_t, \omega_\tau)$ and represented as a 2D map. Additional experimental data processing, such as phase untwisting [37,45] and time or frequency windowing [38,68], can in principle be incorporated within the present framework depending on the specific experimental realization.

Let us first perform the Fourier transform with respect to t of the 2D signal in Eq. (8) by explicitly writing the electric fields entering each term. For the first term we have the sum $A(\omega, \tau) = A_0(\omega) + \bar{A}_\tau(\omega)$, while $A(\omega, \tau) = A_0(\omega)$ or $\bar{A}_\tau(\omega)$ for the remaining two terms. Since the Fourier transform of $\bar{A}_\tau(t)$ is simply $\bar{A}_\tau(\omega) = e^{-i\omega\tau} A_\tau(\omega)$, where $A_\tau(\omega)$ is the spectrum of the delayed field for $\tau = 0$, we see that the τ dependence of the $J_{2D}(\omega_t, \tau)$ only enters via the phase factor $e^{-i\omega\tau}$ of the delayed pulse. Moreover, one can easily see that

the linear response cancels out in Eq. (8), since contributions to the current at first order in A , $J^{(1)}(\omega) = K^{(1)}(\omega)A(\omega)$, are simply additive. As a consequence, the lowest-order nonlinear process in a centrosymmetric material generating the 2D signal comes from a third-order current. Let us write this response in full generality as follows:

$$J^{(3)}(\omega_t, \tau) = \int d\omega_i \delta(\omega_t - \omega_1 - \omega_2 - \omega_3) K^{(3)}(\omega_1, \omega_2, \omega_3) \times A(\omega_1, \tau) A(\omega_2, \tau) A(\omega_3, \tau). \quad (9)$$

Here $K^{(3)}$ is again the nonlinear response kernel to the gauge potential A of Eq. (1), that depends in the most general case on the three frequencies ω_i , $i = 1, 2, 3$. We note that in principle in Eq. (9) there is a further summation over the Cartesian components of the kernel and gauge fields, that we keep aside for the moment to make the notation lighter. By replacing in Eq. (9) the $A(\omega, \tau)$ obtained for each term of Eq. (8) we note that the third-order terms proportional to A_0^3 and A_τ^3 drop out, leaving two contributions:

$$J_{2D}^{(3)} = J_{00\tau}^{(3)} + J_{0\tau\tau}^{(3)}. \quad (10)$$

The first corresponds to two interactions with field E_0 and one with field A_τ , which we will denote as $J_{00\tau}^{(3)}$. The other corresponds to two interactions with field A_τ and one with field A_0 , which we will denote accordingly as $J_{0\tau\tau}^{(3)}$. They read, as a function of ω_t and τ ,

$$J_{00\tau}^{(3)}(\omega_t, \tau) = \int d\omega_i K^{(3)}(\omega_1, \omega_2, \omega_3) \delta(\omega_t - \omega_1 - \omega_2 - \omega_3) [e^{-i\omega_1\tau} A_\tau(\omega_1) A_0(\omega_2) A_0(\omega_3) + \text{perm.}], \quad (11)$$

$$J_{0\tau\tau}^{(3)}(\omega_t, \tau) = \int d\omega_i K^{(3)}(\omega_1, \omega_2, \omega_3) \delta(\omega_t - \omega_1 - \omega_2 - \omega_3) [e^{-i(\omega_1+\omega_2)\tau} A_\tau(\omega_1) A_\tau(\omega_2) A_0(\omega_3) + \text{perm.}], \quad (12)$$

where by “perm.” we mean analogous terms where we permute the frequency indices $123 \rightarrow 231, 321$ in square brackets.

Let us focus on the first contribution, $J_{00\tau}^{(3)}(\omega_t, \omega_\tau)$. The Fourier transform $\tau \rightarrow \omega_\tau$ in Eq. (11) will produce a $\delta(\omega_i - \omega_\tau)$, which can be used to eliminate one integral over frequency and set one of the ω_i frequencies to ω_τ . Since there is still the energy-conservation delta to exploit in Eq. (11), we can use it to set the sum of the remaining two frequencies, both coming from A_0 , to $\omega_t - \omega_\tau$, remaining with only one integral over frequency. We notice that since this process is symmetric with respect to the frequency choice ω_i , since all of them appear in Eq. (11), we can introduce an explicitly symmetrized form of the nonlinear response kernel:

$$\begin{aligned} K_S^{(3)}(\omega_\tau, \omega, \omega_t - \omega_\tau - \omega) &= K^{(3)}(\omega_\tau, \omega, \omega_t - \omega_\tau - \omega) \\ &+ K^{(3)}(\omega, \omega_\tau, \omega_t - \omega_\tau - \omega) \\ &+ K^{(3)}(\omega, \omega_t - \omega_\tau - \omega, \omega_\tau). \end{aligned} \quad (13)$$

Since in the present framework the $K^{(3)}$ kernel is *already* symmetric with respect to *any* permutation of the incoming fields [25,69], as detailed in the SM [67], the previous expression reduces to $K_S^{(3)}(\omega_\tau, \omega, \omega_t - \omega_\tau - \omega) =$

$3K^{(3)}(\omega_\tau, \omega, \omega_t - \omega_\tau - \omega)$. We can then write out

$$J_{00\tau}^{(3)}(\omega_t, \omega_\tau) = A_\tau(\omega_\tau) \int d\omega K_S^{(3)}(\omega_\tau, \omega, \omega_t - \omega_\tau - \omega) \times A_0(\omega) A_0(\omega_t - \omega_\tau - \omega). \quad (14)$$

A similar analysis can be carried out for Eq. (12), $J_{0\tau\tau}^{(3)}$. In this case, one gets from the τ Fourier transform a $\delta(\omega_\tau - \omega_i - \omega_j)$, with $i \neq j$. But then the energy-conservation delta will require the only photon from A_0 to be at frequency $\omega_t - \omega_\tau$. As before we can then write out

$$J_{0\tau\tau}^{(3)}(\omega_t, \omega_\tau) = A_0(\omega_t - \omega_\tau) \int d\omega K_S^{(3)}(\omega_t - \omega_\tau, \omega, \omega_\tau - \omega) \times A_\tau(\omega) A_\tau(\omega_\tau - \omega). \quad (15)$$

We immediately notice that the two contributions can be transformed into each other by means of the exchange $\omega_\tau \leftrightarrow \omega_t - \omega_\tau$ and $A_0(\omega) \leftrightarrow A_\tau(\omega)$. In the following, we specifically analyze two limiting cases relevant to experimental situations: two time-delayed almost monochromatic pulses and two time-delayed almost impulsive fields.

A. Monochromatic limit

In the case of multicycle terahertz pulses one can approximately represent the field as monochromatic in order to get an intuition on the structure of the 2D map. We can then put $A_0(t) = \bar{A}_0 \cos(\Omega t)$ and $\bar{A}_\tau(t) = \bar{A}_\tau \cos(\Omega(t + \tau))$, using two different prefactors to distinguish the two classes of process. From the previous Eqs. (14) and (15) we obtain the analytical expressions

$$J_{2D}^{(3)}(\omega_t > 0, \omega_\tau) = \delta(\omega_t - \Omega)[2\delta(\omega_\tau - \Omega) + \delta(\omega_\tau + \Omega)]K_S^{(3)}(\Omega, \Omega, -\Omega)\bar{A}_0^2\bar{A}_\tau + \delta(\omega_t - 3\Omega)\delta(\omega_\tau - \Omega)K_S^{(3)}(\Omega, \Omega, \Omega)\bar{A}_0^2\bar{A}_\tau + \delta(\omega_t - \Omega)[2\delta(\omega_\tau) + \delta(\omega_\tau - 2\Omega)]K_S^{(3)}(\Omega, \Omega, -\Omega)\bar{A}_0\bar{A}_\tau^2 + \delta(\omega_t - 3\Omega)\delta(\omega_\tau - 2\Omega)K_S^{(3)}(\Omega, \Omega, \Omega)\bar{A}_0\bar{A}_\tau^2. \quad (16)$$

The final 2D map in the half plane $\omega_t > 0$ contains then six spots, four of them aligned along the vertical direction of the first harmonic $\omega_t = \Omega$ ($\omega_\tau = -\Omega, 0, \Omega, 2\Omega$), as shown in panel (a) in Fig. 3, and two of them aligned along the line $\omega_t = 3\Omega$ ($\omega_\tau = \Omega, 2\Omega$), as in panel (b) of the same figure. We note that various naming conventions are used in the literature to denote the spots in the two-dimensional plane, most whose originate from the time-domain evolution of systems driven by impulsive fields [23,37,53]. The terms “rephasing,” “nonrephasing,” “pump-probe,” and “two-quantum” acquire specific physical meanings only when the temporal ordering of the pulses can be uniquely determined. In the strictly monochromatic limit, where the pulses have infinite duration, the order of arrival cannot be inferred from the relative position of their envelopes. Nevertheless, since experimental narrowband pulses possess finite envelopes, we adopt in Fig. 3 the same convention [23,37,53] established in the impulsive limit for $\tau > 0$ according to Table I.

In panels (a) and (b) of Fig. 3 we also introduce a compact way of representing the nonlinear process that produces the various spots on the 2D map by means of a frequency vector scheme [37,70]. Since an oscillation in time at frequency Ω can be represented as a vector in the conjugated frequency domain oriented in the direction perpendicular to the wavefronts, we introduce two vectors $\vec{\Omega}_0 = (\Omega, 0)$ and $\vec{\Omega}_\tau = (\Omega, \Omega)$ in the (ω_t, ω_τ) plane to represent (positive frequency) photons

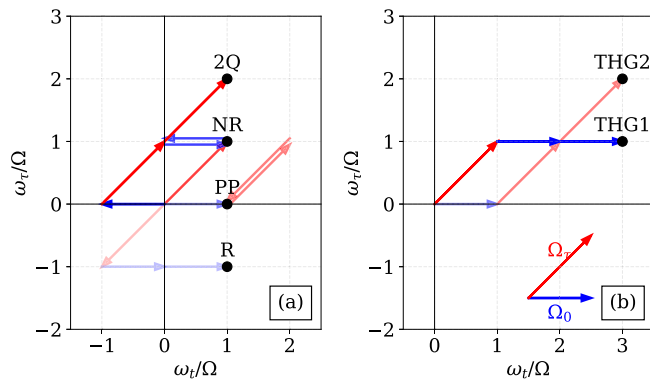


FIG. 3. Schematic outcome of a 2D experiment performed via monochromatic driving pulses $E(t) \sim \cos(\Omega t)$, obtained by using the frequency-vector representation introduced in the text for E_0 ($\vec{\Omega}_0$, blue arrow) and $\bar{E}_\tau$ ($\vec{\Omega}_\tau$, red arrow). (a) Representation of the processes providing an oscillation at $\omega_t = \Omega$, i.e. the nonlinear first harmonic. (b) Same representation for the third-harmonic signal at $\omega_t = 3\Omega$. Different shades of the arrows correspond to different “paths” to reach the six spots, which are named according to Table I.

coming from $A_0(t)$ or $\bar{A}_\tau(t)$, respectively. More specifically, for a monochromatic field $A_0(\tau) = \bar{A}_0 \cos(\Omega t)$ the Fourier components at $\pm\Omega$ are represented by the vectors $\pm\vec{\Omega}_0$, and analogously for the $A_\tau(t)$. The outcome of the nonlinear process can then be pictorially computed by the vector sum of two $\pm\vec{\Omega}_0$ and one $\pm\vec{\Omega}_\tau$ for the $A_0^2 A_\tau$ contribution, or one $\pm\vec{\Omega}_0$ and two $\pm\vec{\Omega}_\tau$ for the piece $A_0 A_\tau^2$.

In the perfectly monochromatic limit the spectral content of the 2D signal is uniquely set by the spectrum of the incoming field. In particular, at fixed $\omega_t = \Omega$ or 3Ω , the relative weight of the spots along the ω_τ direction only depends on the combinatorial prefactors and field amplitudes determined by Eq. (16). Here, the specific form of the third-order kernel only controls the relative weight between the first and third harmonic through $K^{(3)}(\Omega, \Omega, -\Omega)$ and $K^{(3)}(\Omega, \Omega, \Omega)$, respectively. It is worth noting that the third-harmonic kernel $K^{(3)}(\Omega, \Omega, \Omega)$ can also be extracted by single-pulse experiments, where one only measures the electric field transmitted or reflected from a sample [11,19–21], since higher harmonics of the incoming pulse can only arise from nonlinear processes. This configuration can be ultimately obtained as the $\tau = 0$ limit of the two-pulse protocol presented above, which in the frequency domain amounts to integrate in ω_τ the previous Eq. (16). On the other hand, in one-dimensional protocols the *nonlinear first harmonic*, weighted by $K^{(3)}(\Omega, \Omega, -\Omega)$, is superimposed to the linear response, that is usually much larger. As a consequence, even in the monochromatic limit the 2D protocol contains additional information as compared to single-pulse protocols [31,33].

B. Impulsive limit

A second typical approximation, used for experiments with single-cycle terahertz pulses [37,52], is the impulsive limit,

TABLE I. Summary of the spots obtained with monochromatic pulses. The labeling of the first-harmonic spots follows the usual convention appropriate in the impulsive limit for $\tau > 0$ for systems with discrete energy levels, with the correspondence $\Omega \leftrightarrow 2\Delta$, 2Δ being the level spacing [23,37,53].

Position	Name	Abbreviation
$(\omega_t, \omega_\tau) = (\Omega, 2\Omega)$	Two-quantum	2Q
$(\omega_t, \omega_\tau) = (\Omega, \Omega)$	Nonrephasing	NR
$(\omega_t, \omega_\tau) = (\Omega, 0)$	Pump-probe	PP
$(\omega_t, \omega_\tau) = (\Omega, -\Omega)$	Rephasing	R
$(\omega_t, \omega_\tau) = (3\Omega, \Omega)$	Third harmonic	THG1
$(\omega_t, \omega_\tau) = (3\Omega, 2\Omega)$	Third harmonic	THG2

modeled by deltalike fields in the time domain $E(t) \sim \delta(t)$, which are Fourier transformed to constants in frequency space. In this case, by direct inspection of Eqs. (14) and (15) it is understood that the behavior of the 2D map will mostly depend on the frequency structure of the nonlinear conductivity $\sigma^{(3)}(\omega_1, \omega_2, \omega_3) \sim K^{(3)}/(\omega_1\omega_2\omega_3)$, due to the relation $E \sim i\omega A$. The analysis of the 2D maps obtained for a representative dispersive electronic system will be the subject of the next section. However, to set the stage for this analysis we will first discuss a simpler and paradigmatic case, i.e., a nondispersive two-level system, which has been discussed in the literature within the time-dependent density-matrix formalism [25,26,37]. We will see that one can easily map these results into a collection of Feynman diagrams decorated with different pulse sequences, representing the various excitation pathways. As a starting model we then consider a collection of N noninteracting two-level systems ($|0\rangle, |1\rangle$) hosting N spinless electrons in the ground state at $T = 0$. Setting the zero of the energy at the chemical potential, the Hamiltonian can be written as

$$H = \sum_n (c_{n0}^\dagger, c_{n1}^\dagger) \begin{pmatrix} -\Delta & 0 \\ 0 & \Delta \end{pmatrix} \begin{pmatrix} c_{n0} \\ c_{n1} \end{pmatrix}, \quad (17)$$

where 2Δ is the level spacing and $n = 1 \dots N$ labels the electronic creation and annihilation operators $c_{n0,1}^\dagger$ and $c_{n0,1}$. The system can be coupled to the electromagnetic field through the dipolarlike interaction term

$$H_{\text{int}}(t) = \mu E(t) \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad (18)$$

where μ is the dipole moment of the transition. The diagrammatic representation of the light-electron coupling vertex is shown in Fig. 4.

Performing a conventional perturbative expansion in powers of the electric field, one can obtain the third-order nonlinear susceptibility in real frequencies which reads

$$\begin{aligned} \chi^{(3)}(\omega_1, \omega_2, \omega_3) \sim & \frac{1}{\omega_{123} - 2\Delta} \frac{1}{\omega_2 + 2\Delta} \\ & \times \left(\frac{1}{\omega_1 - 2\Delta} + \frac{1}{\omega_3 - 2\Delta} \right) - (\Delta \rightarrow -\Delta) \\ & + \text{perm.}, \end{aligned} \quad (19)$$

where $\omega_{123} = \sum_i \omega_i$ and all real frequencies ω_i are meant to include a vanishingly small imaginary part $i\eta$ after the analytical continuation. Before proceeding further, we notice that from this nonlinear susceptibility we could immediately obtain the nonlinear kernel $K^{(3)}(\omega_1, \omega_2, \omega_3) \sim \omega_{123}\omega_1\omega_2\omega_3\chi^{(3)}(\omega_1, \omega_2, \omega_3)$, that we used in the previous derivations to connect the applied *gauge field* A to the experimentally relevant induced *current* J_{2D} . However, just for the rest of this section, we switch the focus to the induced *polarization* P_{2D} in response to the *electric field* to make a more explicit connection to the results of Ref. [37] obtained for the two-level system. As one can check, the conversion factor between the two in frequency space, $J_{2D}(\omega_t, \omega_\tau) \sim \omega_t P_{2D}(\omega_t, \omega_\tau)$, will not influence the position of the relevant resonances in the 2D plane. In the time domain, $J_{2D}(t, \tau) \sim$

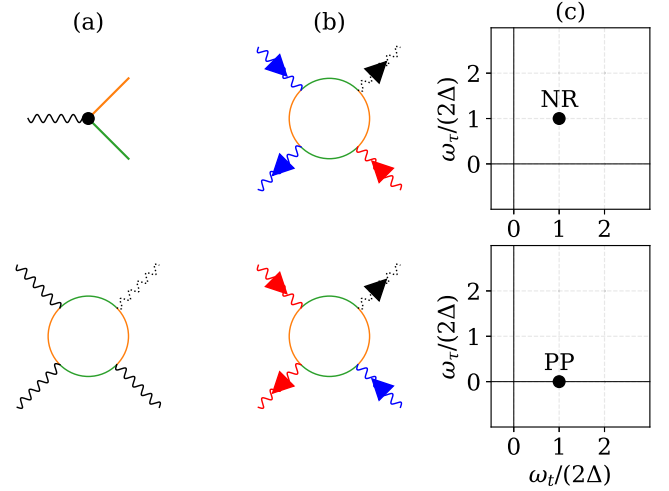


FIG. 4. Flowchart of the construction of the 2D map within the diagrammatic approach. (a) The starting building block (top) is the light-matter interaction vertex: for the two-level system of Eq. (17) it is the dipolar interaction of Eq. (18) where the electric field (wavy line) generates transitions between the lower (solid orange line) and upper (solid green line) level. The third order nonlinear response is then built from diagrams with four light-matter insertions (bottom). The dotted wavy line denotes the detection frequency $\omega_t = \sum_i \omega_i$, while each electromagnetic field with frequency ω_i appears as a solid wavy line. (b) To get the 2D map the latter black lines are colored as E_0 (blue) or E_τ (red), to form $P_{00\tau}^{(3)}$ in Eq. (21) (top) or $P_{0\tau\tau}^{(3)}$ in Eq. (22) (bottom), here exemplified for a specific field combination. In the impulsive limit the Fourier transforms of the fields are constant in frequency and the values on the external legs are set by the transition energy between $-\Delta$ and $+\Delta$. (c) The detection at $\omega_t > 0$ imposes $\omega_t = 2\Delta$, consequently the upper diagram carries $\omega_\tau = 2\Delta$ (red line) leading to the NR peak at $(\omega_t, \omega_\tau) = (2\Delta, 2\Delta)$; the lower diagram shows $\omega_t - \omega_\tau = 2\Delta$ (blue line), i.e. $\omega_\tau = 0$, generating the PP spot at $(\omega_t, \omega_\tau) = (2\Delta, 0)$.

$\partial_t P_{2D}(t, \tau)$, thus the time derivative will just change the phase of the oscillations.

To get an intuition for how this nonlinear response can be built up in the diagrammatic representation, we summarize in Fig. 4 the flowchart of its construction. Starting from the light-matter vertex, that in the case of the model in Eq. (18) is a dipolarlike interaction between the electric field E (wavy lines) and the two-level system (solid lines), one builds up Feynman diagrams with four external legs representing the perturbing fields at ω_i and the generated one at $\omega_t = \sum_i \omega_i$ (dashed wavy line). The loop accounts for the propagation of fermionic states at $\pm\Delta$ between two electric field pulses, whose explicit expression is the susceptibility of Eq. (19). Finally, we decorate the external legs with the detection frequency ω_t , that always occurs on the dashed wavy line, and with the combination of frequencies assigned to the E_0 (blue) and E_τ (red) pulses, in the spirit of the definitions (14) and (15) above.

A more complete representation of this process is provided in Fig. 5, where we accounted for all possible permutations of the external light legs. Notice that each of them leads to a different process, since excitations between the levels read out different frequency values. This representation helps us

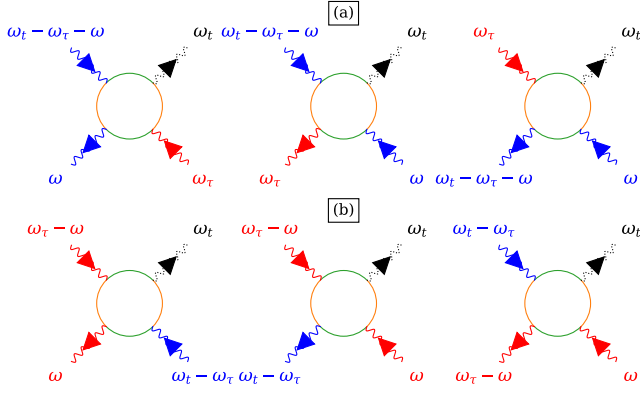


FIG. 5. Representation of all the possible ways of decorating the first term of the susceptibility in Eq. (19) with combinations of E_0 (blue) and E_τ (red). (a) Diagrams relevant for $P_{00\tau}^{(3)}$ in Eq. (21). (b) Those relevant for $P_{0\tau\tau}^{(3)}$ in Eq. (22).

to highlight the absorption/emission of photons at each interaction since we use different colors, orange or green, to distinguish propagators of the eigenstates at energy $-\Delta$ or $+\Delta$, respectively. Their alternation is a consequence of the

off-diagonal form of H_{int} in Eq. (18), while their ordering is forced by energy conservation if we set $\omega_t > 0$ for detection. We stress that the direction of the arrow in the blue and red photon lines (incoming or outgoing) denotes positive/negative frequencies, whose value in this impulsive limit is fixed by the difference ($+\Delta$ or $-\Delta$) between the energies of the propagators connecting each H_{int} insertion. Moreover, since in the impulsive limit $E_{0,\tau}(\omega) = \bar{E}_{0,\tau}$ are constant, the ω -integral in Eqs. (14) and (15) with the form of Eq. (19) can be performed by residues. We report, for instance, the contribution stemming from diagram (i), obtained by the assignment $\omega_{123} \rightarrow \omega_t, \omega_1 \rightarrow \omega_\tau, \omega_2 \rightarrow \omega, \omega_3 \rightarrow \omega_t - \omega_\tau - \omega$:

$$(i) \sim \int d\omega \frac{1}{\omega_t + i\eta - 2\Delta} \frac{1}{\omega + i\eta + 2\Delta} \times \left(\frac{1}{\omega_\tau + i\eta - 2\Delta} + \frac{1}{\omega_t - \omega_\tau - \omega + i\eta - 2\Delta} \right) \sim \frac{1}{\omega_t + i\eta - 2\Delta} \left(\frac{2}{\omega_t - \omega_\tau + i\eta} + \frac{1}{\omega_\tau + i\eta - 2\Delta} \right). \quad (20)$$

Summing all the pieces together, the resulting 2D map will be given by

$$P_{00\tau}^{(3)}(\omega_t, \omega_\tau) \sim \left(\frac{1}{\omega_t + i\eta - 2\Delta} - \frac{1}{\omega_t + i\eta + 2\Delta} \right) \left[\frac{2}{\omega_t - \omega_\tau + i\eta} + \frac{1}{\omega_\tau + i\eta - 2\Delta} + \frac{1}{\omega_\tau + i\eta + 2\Delta} \right] \bar{E}_0^2 \bar{E}_\tau, \quad (21)$$

$$P_{0\tau\tau}^{(3)}(\omega_t, \omega_\tau) \sim \left(\frac{1}{\omega_t + i\eta - 2\Delta} - \frac{1}{\omega_t + i\eta + 2\Delta} \right) \left[\frac{2}{\omega_\tau + i\eta} + \frac{1}{\omega_t - \omega_\tau + i\eta - 2\Delta} + \frac{1}{\omega_t - \omega_\tau + i\eta + 2\Delta} \right] \bar{E}_0 \bar{E}_\tau^2. \quad (22)$$

When $\eta \rightarrow 0$ the various fractions produce four peaks which for $\omega_t > 0$ are located at the intersections between the line $\omega_t = 2\Delta$, which accounts for the prefactor in round brackets, and the lines maximizing each addend in square brackets. For $P_{00\tau}^{(3)}$ in Eq. (21) this occurs at $\omega_\tau = \pm 2\Delta$ and $\omega_t - \omega_\tau = 0$, while for $P_{0\tau\tau}^{(3)}$ in Eq. (22) this leads to $\omega_\tau = 0$ and $\omega_t - \omega_\tau = \pm 2\Delta$. The four resulting spots are located at coordinates $(2\Delta, -2\Delta)$, $(2\Delta, 0)$, $(2\Delta, 2\Delta)$, and $(2\Delta, 4\Delta)$. These resonances can be attributed to the various combinations of fixed pulses E_0 (blue) and delayed pulses E_τ (red), as explicitly shown in Figs. 6(b) and 6(e) in the frequency vector scheme. Indeed, in analogy with Fig. 3(a), the 2D map can still be generated by a combination of frequency vectors, but, in the impulsive limit applied to the two-level system, the *length* of the arrow is dictated by the 2Δ energy, setting the scale of allowed interband transitions. In other words, in this case $\vec{\Omega}_0 = (2\Delta, 0)$ and $\vec{\Omega}_\tau = (2\Delta, 2\Delta)$: by locating the spots on the 2D map, one immediately reads out the resonance frequency of the system under investigation. As a further remark, we stress that although each diagram in Figs. 6(a) and 6(d) produces a different spot in Figs. 6(b) and 6(e), the inverse is not true, as evident for (i) and (iii) that both appear in $(2\Delta, 2\Delta)$ and $(2\Delta, 0)$. This is due to the fact that the integration over ω exemplified in Eq. (20) leads to the same frequency content for (i) and (iii).

In this regard, it is also instructive to compute the nonlinear signal in the time domain by going back from (ω_t, ω_τ) to (t, τ) . Due to the analytic continuation in Eqs. (21) and (22), the position of the poles in the upper or lower half plane for complex ω_t, ω_τ is extremely important to preserve the causality structure reflecting the ordering of the electric fields in time. Close inspection shows that the Fourier transform of terms containing $\omega_t - \omega_\tau$ leads to $\theta(t + \tau)\theta(-\tau)$, which produces a signal in the region where E_0 comes before E_τ , while from terms containing only ω_τ we get $\theta(t)\theta(\tau)$, which is nonzero when E_τ comes before E_0 . We stress that this information, although already contained in the frequency structure of the diagrams in Figs. 6(a) and 6(d), *cannot* be obtained at once by looking at the order of the field lines in the drawing. As an example, looking again at Eq. (20), we see that in this process both denominators $\omega_t - \omega_\tau$ and ω_τ are present, so that the process contributes to both time ordering $\tau > 0$ and $\tau < 0$. For this reason, to label the spots we follow the notation of Ref. [37], where both cases are covered.

Let us first compute the signal in time by Fourier transforms of Eqs. (21) and (22), which are shown in Figs. 6(c) and 6(d), respectively. For two interactions with E_0 and one with E_τ we have

$$P_{00\tau}^{(3)}(t, \tau) \sim \bar{E}_0^2 \bar{E}_\tau \{ \theta(t)\theta(\tau) \sin(2\Delta t) \cos(2\Delta \tau) + \theta(t + \tau)\theta(-\tau) \sin[2\Delta(t + \tau)] \}, \quad (23)$$

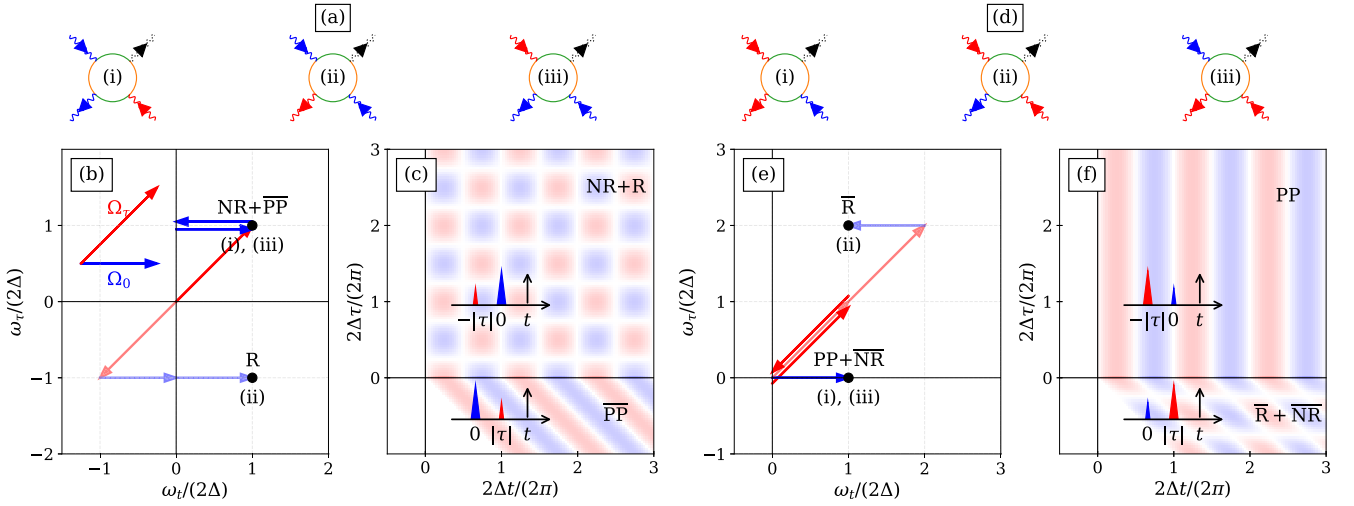


FIG. 6. Schematic outcome for the 2D experiment performed on a two-level system by using impulsive fields $E(t) \sim \delta(t)$, where in panels (a)–(c) we analyzed the contributions of $P_{00\tau}$ in Eq. (21) and in panels (d)–(f) those of $P_{0\tau\tau}$ [see Eq. 22]. (a), (d) Diagrammatic representation of the nonlinear processes obtained by distributing photons coming from E_0 (blue) and E_τ (red) in the arguments of $\chi^{(3)}$, which alternates propagators at energy $-\Delta$ (orange) and $+\Delta$ (green), following Figs. 5(a) and 5(b); each process is labeled as (i), (ii), and (iii). (b), (e) Corresponding frequency-vector scheme, where we indicated how diagrams on the upper row map in frequency space. The names of the peaks are given according to the order of the pulses; for $\tau > 0$ (E_τ first) we have R, PP, and NR, also used in Table I, and for $\tau < 0$ (E_0 first) we have $\bar{R}$, PP, and NR. (c), (f) Time-domain simulation of the 2D map, which shows the causality conditions embedded in the various processes, with indications of the corresponding contributions coming from the peaks in frequency space.

reported in Fig. 6(c). Two interactions with E_τ and one with E_0 produce instead

$$P_{0\tau\tau}^{(3)}(t, \tau) \sim \bar{E}_0 \bar{E}_\tau^2 \{ \theta(t) \theta(\tau) \sin(2\Delta t) + \theta(t + \tau) \theta(-\tau) \sin[2\Delta(t + \tau)] \cos(2\Delta\tau) \}, \quad (24)$$

presented in Fig. 6(f). As stated before, the notation of Table I refers to the processes obtained for $\tau > 0$, i.e., the first lines of Eqs. (23) and (24), and leads to the NR, R, and PP spots of Figs. 6(b) and 6(e). When $\tau < 0$ we are exchanging the role of the “pump” and “probe” fields with respect to the usual sense of pump-probe experiments. Then one sees that when the field amplitudes are exchanged, $\bar{E}_0 \leftrightarrow \bar{E}_\tau$, the 2D map in the time domain for $\tau < 0$ is connected to the one in $\tau > 0$ by the relation $P_{2D}^{(3)}(t, -|\tau|) = P_{2D}^{(3)}(t - |\tau|, |\tau|)$. This is equivalent to mapping the upper (lower) half plane of Fig. 6(c) into the lower (upper) half plane of Fig. 6(f), which is the time-domain equivalent of the transformation $\omega_t - \omega_\tau \leftrightarrow \omega_\tau$ in the frequency plane mentioned after Eqs. (14) and (15). Such a symmetry justifies the classification of the frequency spots originating from $\tau < 0$ as $\bar{PP}$, $\bar{NR}$, and $\bar{R}$ in Figs. 6(b) and 6(e). As a last remark, we notice that in the two-level system the two-quantum process at $\tau > 0$ never occurs, since we have no terms with structure $\sim \theta(t) \theta(\tau) \cos(4\Delta\tau)$ in the time domain, corresponding to a frequency structure $\sim (\omega_t - 4\Delta + i\eta)^{-1} (\omega_t - 2\Delta + i\eta)^{-1}$. To get such a resonance an excitation at energy 4Δ is needed, which is absent in the two-level system. We finally observe that Eqs. (23) and (24) are identical to the ones obtained in Ref. [37], by computing the nonlinear polarization via an explicit solution of the density-matrix evolution in the time domain. This equivalence is expected, since for a simple noninteracting and nondispersive system as the one of Eq. (17) an analytical solution can be easily

derived both in time and frequency space. As a consequence, the two theoretical approaches have the same predictive power as compared to experiments. The real issue is how to extend the calculations to more complex electronic systems, that is the purpose of the rest of the paper.

III. 2DTS OF DISPERSIVE ELECTRONIC SYSTEMS: PARAMAGNETIC VS DIAMAGNETIC PROCESSES

As we have shown in the previous section, in the impulsive limit the 2D map is dominated by the frequency content of the nonlinear optical kernel, that in turn is dominated for the two-level system by the energy difference 2Δ between the two nondispersive eigenstates. We want now to generalize this notion to the case of a many-body electronic system. In particular, we will consider a toy model for a semiconductinglike band structure hosting an optically active gap, in order to establish analogies and differences with the standard two-level model [25–27,53]. As we shall see, two main differences must be considered.

(i) In the usual representation of the gauge potential, the light-matter interaction admits multiphoton vertices in addition to the single-photon one, the dipolelike interaction of Eq. (18), the first one being a diamagneticlike two-photon vertex present already in the continuum limit.

(ii) The energy dispersion of valence and conduction states leads to a modification of the 2D features found for the two-level case even for the single-photon processes.

To make a concrete example we will compute the $K^{(3)}$ third-order response to the gauge field $\mathbf{A}$ by focusing on a two-dimensional system displaying a band gap at 2Δ . The fermionic spectrum is equivalent to the one obtained at mean-field level for a perfectly nested CDW phase with nesting vector $\mathbf{Q}$ on a two-dimensional square lattice [71]. By

introducing the spinor

$$\Psi_{\mathbf{k}}^{\dagger} = (c_{\mathbf{k}}^{\dagger}, \quad c_{\mathbf{k}+\mathbf{Q}}^{\dagger}) \quad (25)$$

the system is described by the spinless Hamiltonian

$$H = \sum_{\mathbf{k} \in \text{RBZ}} \Psi_{\mathbf{k}}^{\dagger} \begin{pmatrix} \epsilon_{\mathbf{k}} & \Delta \\ \Delta & \epsilon_{\mathbf{k}+\mathbf{Q}} \end{pmatrix} \Psi_{\mathbf{k}}, \quad (26)$$

with a nearest-neighbor hopping $\bar{t}$ on a square lattice, such that the band dispersion is

$$\epsilon_{\mathbf{k}} = -2\bar{t} \cos(k_x a) - 2\bar{t} \cos(k_y a) \quad (27)$$

where a is the distance between atomic sites, from now on set to unity. Since for $\mathbf{Q} = (\pi, \pi)$ the band structure is perfectly nested, $\epsilon_{\mathbf{k}+\mathbf{Q}} = -\epsilon_{\mathbf{k}}$, the $\mathbf{k}$ summation in Eq. (26) is restricted to the reduced Brillouin zone. As mentioned above, in the CDW case Δ can be thought of as a mean-field order parameter connected to the macroscopic average $\langle c_{\mathbf{k}}^{\dagger} c_{\mathbf{k}+\mathbf{Q}} \rangle$. The model in Eq. (26) is however used here to provide us with a pedagogical example, useful to highlight the role of dispersive excitations and the interplay between the frequency structure of the nonlinear kernel and that of the pulse. To this aim we will also introduce a phenomenological and constant damping of the electronic excitations, and we will neglect the additional contribution due to collective modes, present in a realistic CDW case [71]. As we shall see, even within these approximations the computation of the 2D maps for the arbitrary field spectrum can be cumbersome, and we will discuss possible semianalytical approximations for it. In addition, we will vary the ratio $\bar{t}/\Delta$ from low ($\bar{t}/\Delta = 0.1$) to large ($\bar{t}/\Delta = 2$) values in order to highlight the role of dispersive electronic excitations as compared to the two-level systems.

The fermionic Green's function in the Matsubara formalism for the Nambu spinor $\Psi_{\mathbf{k}}$ shows poles at $\pm E_{\mathbf{k}} = \pm \sqrt{\epsilon_{\mathbf{k}}^2 + \Delta^2}$ and reads

$$G_0(i\omega_n, \mathbf{k}) \equiv \begin{pmatrix} i\omega_n - \epsilon_{\mathbf{k}} & -\Delta \\ -\Delta & i\omega_n + \epsilon_{\mathbf{k}} \end{pmatrix}^{-1} \\ \equiv \mathcal{U}_{\mathbf{k}}^{\dagger} \tilde{G}_0(i\omega_n, \mathbf{k}) \mathcal{U}_{\mathbf{k}}, \quad (28)$$

where $i\omega_n$ are fermionic Matsubara frequencies. Here we introduced the usual Bogoliubov rotation matrices $\mathcal{U}_{\mathbf{k}} = u_{\mathbf{k}}\sigma_0 + i\sigma_2 v_{\mathbf{k}}$, with $u_{\mathbf{k}}^2 - v_{\mathbf{k}}^2 = \epsilon_{\mathbf{k}}/E_{\mathbf{k}}$ and $2u_{\mathbf{k}}v_{\mathbf{k}} = \Delta/E_{\mathbf{k}}$, which bring the Green's function G_0 to its diagonal form $\tilde{G}_0$:

$$\tilde{G}_0(i\omega_n, \mathbf{k}) = \begin{pmatrix} \frac{1}{i\omega_n - E_{\mathbf{k}}} & 0 \\ 0 & \frac{1}{i\omega_n + E_{\mathbf{k}}} \end{pmatrix}. \quad (29)$$

To compute the nonlinear response of the system we will follow the standard procedure of coupling the electrons to the gauge potential via the Peierls substitution, namely $\epsilon_{\mathbf{k}} \rightarrow \epsilon_{\mathbf{k}-e/c\mathbf{A}(t)}$. Such an approach, which extends to the lattice case the usual minimal coupling, leads to a set of coupling terms between the gauge field and electrons obtained by expansion in a power series of $\mathbf{A}$. It should be noted that the choice of using the gauge potential $\mathbf{A}(t)$ instead of the electric field $\mathbf{E}(t)$ corresponds to using the velocity instead of the length (or dipole) gauge in the standard nomenclature [72,73]. For dispersive electronic systems, this choice from one side avoids the computation of the dipole matrix elements, involving the position operator $\mathbf{r}$, which is cumbersome for electronic

systems [74,75]. On the other hand, it leads to a simpler frequency structure of the response function, which is rewritten in terms of simple poles. This is particularly relevant for numerical simulations with realistic pulses, since we can obtain a semianalytical expression by approximating the gauge field with a Gaussian spectrum, $A(t) \propto e^{-t^2/(2\tau_p^2)} \cos(\Omega t)$. The price to pay—as already happens in the linear response—is an increase in the number of diagrams needed to describe the optical properties of the material [76,77], as we shall discuss here.

Since the effective electronic model in Eq. (26) is quadratic, once the gauge field has been introduced by Peierls substitution the fermionic degrees of freedom can be integrated out exactly. This procedure can be efficiently carried out in a path-integral approach [12,78], where the quantum action $S[\mathbf{A}]$ for the electromagnetic field can be expressed by means of a perturbative series:

$$S[\mathbf{A}] = \sum_n \frac{1}{n} \text{Tr}(G_0 \Sigma_A)^n, \quad (30)$$

where the trace here means summing over Nambu, energy, and momenta degrees of freedom. Here Σ_A acts as a fermionic self-energy due to the coupling with the gauge field, that upon expansion in power series of $\mathbf{A}$ provides all the possible coupling terms between $\mathbf{A}$ and the electrons:

$$\Sigma_A(i\Omega, \mathbf{k}) = \sum_{n>0} \Sigma_A^{(n)}(i\Omega, \mathbf{k}) \sigma_3, \quad (31)$$

which for the semiconductinglike model under consideration has a σ_3 structure in Nambu space. A whole set of multiphoton vertices appears:

$$\Sigma_A^{(n)}(i\Omega, \mathbf{k}) = \frac{(-e)^n}{n!c^n} \sum_{\alpha_1, \dots, \alpha_n} \frac{\partial^n \epsilon_{\mathbf{k}}}{\partial k_{\alpha_1} \dots \partial k_{\alpha_n}} A_{\alpha_1 \dots \alpha_n}^n(i\Omega), \quad (32)$$

where $A_{\alpha_1 \dots \alpha_n}^n(i\Omega)$, with $i\Omega$ the bosonic Matsubara frequency, denotes the Fourier transform of $A_{\alpha_1}(t) \dots A_{\alpha_n}(t)$ in imaginary time, with $\alpha_i = x, y$ Cartesian components of the gauge field. The first two terms $n = 1$ and 2 represent the lattice equivalent of the paramagnetic and diamagnetic terms already present in continuum models [12], so that they describe single-photon and two-photon vertices in the diagrammatic representation, respectively. Since the current is obtained by a functional derivative with respect to the gauge field,

$$J_{\alpha}(t) = \frac{\delta S[\mathbf{A}]}{\delta A_{\alpha}(t)}, \quad (33)$$

we need to consider nonlinear processes up to the fourth order in the effective action for A to compute the generated current. The derivation of the nonlinear response in a Feynman diagram representation follows then the same logic outlined above while discussing the two-level system. However, in addition to the single-photon vertex already considered in Fig. 4, one must consider now multiphoton vertices, corresponding to the case where multiple light pulses at the same time drive a particle-hole excitation. As a consequence, fourth-order terms in the field A do not necessarily correspond to four-point susceptibilities, as it was the case for the two-level system shown in Fig. 4. Since in a lattice model vertices with an arbitrary number of A lines are allowed, as shown in Eq. (32),

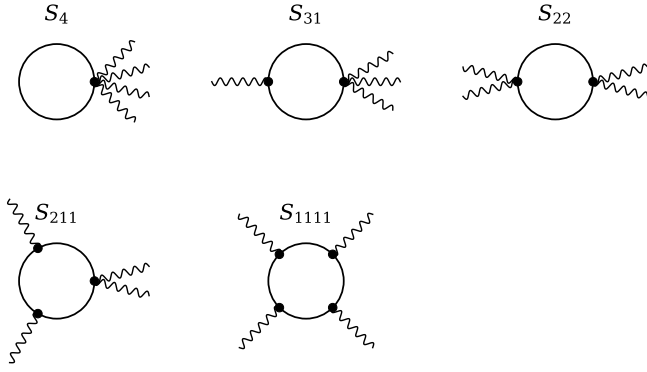


FIG. 7. Diagrams contributing to $S[A^4]$ in Eq. (34), obtained by combinations of the multiphoton vertices in Eq. (32). The wavy lines denote the gauge field, the bullets denote the σ_3 vertex of Eq. (31), and black lines denote the electronic Green's function in Nambu space [see Eq. (28)].

we have overall five contributions to the fourth-order effective action, represented by the Feynman diagrams of Fig. 7:

$$S[A^4] = S_4 + S_{22} + S_{13} + S_{112} + S_{1111}. \quad (34)$$

$$\begin{aligned} X^{(n)}(i\Omega_1, \dots, i\Omega_n, \mathbf{k}) &= \sum_{i\omega_m} \text{Tr}[G_0(i\omega_m, \mathbf{k})\sigma_3 G_0(i\omega_m + i\Omega_1, \mathbf{k})\sigma_3 \dots G_0(i\omega_m + i\Omega_{1\dots n}, \mathbf{k})\sigma_3] \\ &\equiv \sum_{i\omega_m} \text{Tr}[\tilde{G}_0(i\omega_m, \mathbf{k})\Lambda_{\mathbf{k}\mathbf{k}}^{(3)} \dots \tilde{G}_0(i\omega_m + i\Omega_{1\dots n}, \mathbf{k})\Lambda_{\mathbf{k}\mathbf{k}}^{(3)}], \end{aligned} \quad (35)$$

where we denote $i\Omega_{1\dots n} = i\Omega_1 + \dots + i\Omega_n$ and we also introduce the matrix $\Lambda_{\mathbf{k}\mathbf{k}}^{(3)} = \mathcal{U}_{\mathbf{k}}\sigma_3\mathcal{U}_{\mathbf{k}}^\dagger$ representing the light-matter coupling in the band basis. Due to the structure in Nambu space, we have $\Lambda_{\mathbf{k}\mathbf{k}}^{(3)} = \epsilon_{\mathbf{k}}/E_{\mathbf{k}}\sigma_3 - \Delta/E_{\mathbf{k}}\sigma_1$. As we shall see in the following, the σ_1 term is crucial to allow for optically active one-photon transitions between the occupied $-E_{\mathbf{k}}$ and the empty $E_{\mathbf{k}}$ band. To see this explicitly, let us examine the form of $X^{(2)}(i\Omega_{ij}, \mathbf{k})$:

$$X^{(2)}(i\Omega_{ij}, \mathbf{k}) = \sum_{i\omega_n} \text{Tr} \left[\begin{pmatrix} \frac{1}{i\omega_n - E_{\mathbf{k}}} & 0 \\ 0 & \frac{1}{i\omega_n + E_{\mathbf{k}}} \end{pmatrix} \Lambda_{\mathbf{k}\mathbf{k}}^{(3)} \begin{pmatrix} \frac{1}{i\omega_n + i\Omega_{ij} - E_{\mathbf{k}}} & 0 \\ 0 & \frac{1}{i\omega_n + i\Omega_{ij} + E_{\mathbf{k}}} \end{pmatrix} \Lambda_{\mathbf{k}\mathbf{k}}^{(3)} \right]. \quad (36)$$

As stated above, in Nambu space the matrix $\Lambda_{\mathbf{k}\mathbf{k}}^{(3)}$ is a linear combination of σ_3 (diagonal) or σ_1 (off-diagonal). Since the matrix $\tilde{G}_0$ is diagonal, the only contributions that survive in the trace are obtained from the product of the two terms containing σ_3 or the two terms containing σ_1 . The former contribution describes an *intraband* process involving scattering of electrons and holes belonging to the same bands, while the latter describes an *interband* process involving a scattering between the two copies of the semiconductinglike bands. By evaluating the Matsubara sum in Eq. (36), one sees that only the interband part survives, as expected for transitions at long wavelength and finite frequency.

In the following we will analyze first the contributions to the nonlinear kernel stemming from the two terms in the effective action, S_{22} (fully diamagnetic) and S_{1111} (fully paramagnetic), as shown in Figs. 8 and 9, respectively, and we will then show results obtained by including all the contributions

By close inspection of Fig. 7 one then sees that S_4 is a constant, independent of the incoming frequencies of the $A(\omega_i)$ fields. S_{22} and S_{31} are two-point correlation functions, which only depend on the sum of the frequencies of the external field lines, as it is the case for standard linear-response functions. Finally, S_{211} and S_{1111} are three-point and four-point correlation functions, respectively, with a separate dependence on the frequencies of the external legs. These different structures will be responsible for specific signatures in the two-dimensional coherent spectroscopy maps [67].

The overall derivation of the nonlinear current can be carried out in a similar way also for other types of ordered phases treated at mean-field level, as, e.g., the magnetic phases of Rice-Mele-Hubbard models [50], or excitonic insulators [79], as well as superconductors [12,13,15,16]. They are formally characterized by the same effective electronic gapped spectrum $\pm E_{\mathbf{k}} = \pm\sqrt{\epsilon_{\mathbf{k}}^2 + \Delta^2}$, but they generally produce a different structure in Nambu space for the gauge-field coupling of Eq. (31). Once these differences are considered, the strategy for computing the $K^{(3)}$ kernel remains the same.

To find the explicit expression of the diagrams in Fig. 7 we will need to compute n -point correlation functions of the form

in Fig. 10. The interest in discussing the different signatures in the 2D map of these two contributions is motivated by the fact that in other models the only relevant nonlinearities can be ascribed to a fully diamagnetic or a fully paramagnetic response. For example, in superconductors only diamagneticlike contributions are present in the clean limit [12], while the fully paramagnetic response dominates the nonlinear current at strong disorder. In addition, a diamagneticlike response appears whenever the interaction between light and matter can be mapped in an effective process where two photons excite simultaneously two collective modes. This is typically the case relevant for the nonlinear driving of infrared-active optical phonons [80–82], but also for the soft Josephson plasmons, discussed in Sec. IV below. For what concerns the fully paramagnetic term, it is interesting to compare its outcome with the response of the two-level Hamiltonian, where only single-photon dipolelike transitions are allowed. It is thus

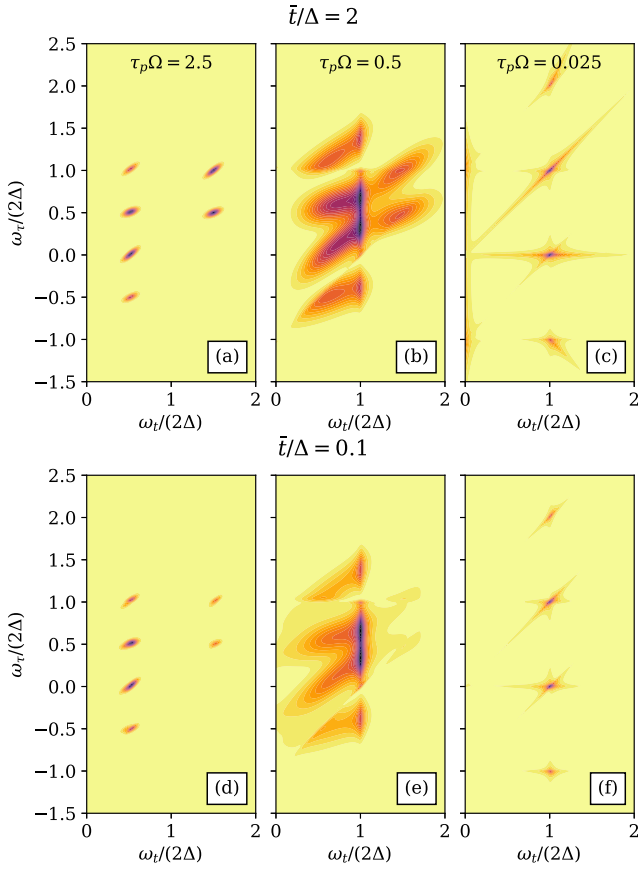


FIG. 8. (a)–(c) 2D map of $|J_{2D}^{para}(\omega_l, \omega_r)|$ obtained for the fully paramagnetic process setting $\bar{t}/\Delta = 2$, computed by integrating over energy E the sum of Eqs. (58) and (59) multiplied by $\rho^{\text{inter}}(E)$, plus the corresponding contribution from the mixed process. Here we assume a Gaussian pulse $A(t) = \bar{A}e^{-t^2/(2\tau_p^2)}\cos(\Omega t)$, by setting $\Omega = \Delta$ and using the values of $\tau_p\Omega$ reported in each column. We set $\eta/(2\Delta) = 0.05/(2\pi)$ for the analytic continuation. (d)–(f) 2D map for $\bar{t}/\Delta = 0.1$.

possible to make a direct comparison between the many-body situation where a continuum, dispersive set of excited states is present and the discrete, nondispersive case of the two-level system. In addition, previous work by several groups has shown that in disordered superconductors the nonlinear response is dominated by paramagneticlike terms [12–18]. Even though in this case a realistic computation should account for the effect of disorder on the energy eigenstates, the present results provide one with a hint on the expected 2D maps as a function of the pulse spectral width. Finally, we stress that, for what concerns the toy model in Eq. (26), in order to formally ensure the invariance between calculations in the two gauges, length $E(t)$ and velocity $A(t)$, one has to *sum* all the diagrams stemming from Eq. (34). For this reason, we provide in the SM [67] the calculation of all these terms, and we verify that gauge-invariance relations are satisfied [83]. These results are used to compute each term separately, also shown in the SM [67], and to obtain the full 2D map, whose differences with respect to the subset of fully paramagnetic and fully diamagnetic terms are discussed below.

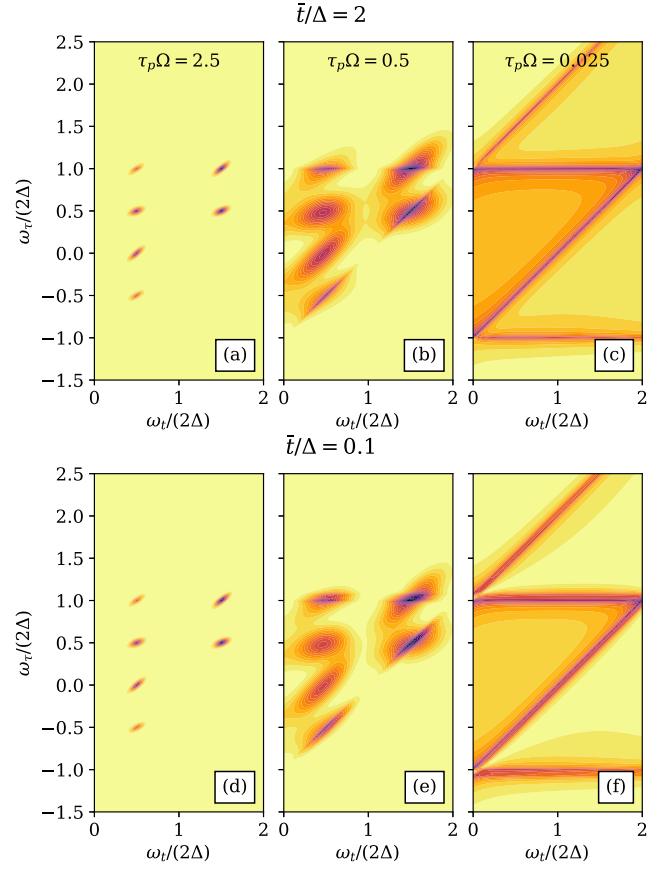


FIG. 9. Analogous of the previous Fig. 8 for the fully diamagnetic process, $|J_{2D}^{\text{dia}}(\omega_l, \omega_r)|$, under the same driving conditions. In panels (a)–(c) we consider $\rho^{\text{dia}}(E)$ derived from Eq. (54) for $\bar{t}/\Delta = 2$; we multiply it by the sum of Eqs. (62) and (63) and integrate the result over energy E . In panels (d)–(f) we consider instead $\bar{t}/\Delta = 0.1$.

A. Fully paramagnetic contribution

Let us start by analyzing the fully paramagnetic term, which is derived from S_{1111} in Fig. 7 as

$$\begin{aligned} J_{\alpha}^{(3),\text{para}}(i\Omega) &= \frac{\delta S_{1111}[A^4]}{\delta A_{\alpha}(-i\Omega)} \\ &\equiv \sum_{i\Omega_i} K_{\alpha\beta\gamma\delta}^{(3),\text{para}}(i\Omega_1, i\Omega_2, i\Omega_3) \\ &\quad \times A_{\beta}(i\Omega_1)A_{\gamma}(i\Omega_2)A_{\delta}(i\Omega_3)\delta(i\Omega - i\Omega_{123}), \end{aligned} \quad (37)$$

where

$$\begin{aligned} K_{\alpha\beta\gamma\delta}^{(3),\text{para}}(i\Omega_1, i\Omega_2, i\Omega_3) &= \sum_{\mathbf{k}, i\omega_n} \frac{\partial \epsilon_{\mathbf{k}}}{\partial k_{\alpha}} \frac{\partial \epsilon_{\mathbf{k}}}{\partial k_{\beta}} \frac{\partial \epsilon_{\mathbf{k}}}{\partial k_{\gamma}} \frac{\partial \epsilon_{\mathbf{k}}}{\partial k_{\delta}} \\ &\quad \times X^{(4)}(i\Omega_1, i\Omega_2, i\Omega_3, \mathbf{k}). \end{aligned} \quad (38)$$

In this case, one realizes that the trace will give nonzero results whenever one selects in $\Lambda_{\mathbf{k}\mathbf{k}}^{(3)}$ four σ_3 , two σ_3 and two σ_1 , or four σ_1 contributions. The first comes from fully intraband scattering processes and it vanishes after the Matsubara summation, as happened for $X^{(2)}$. The second one admits a mixture of interband and intraband scattering processes, with

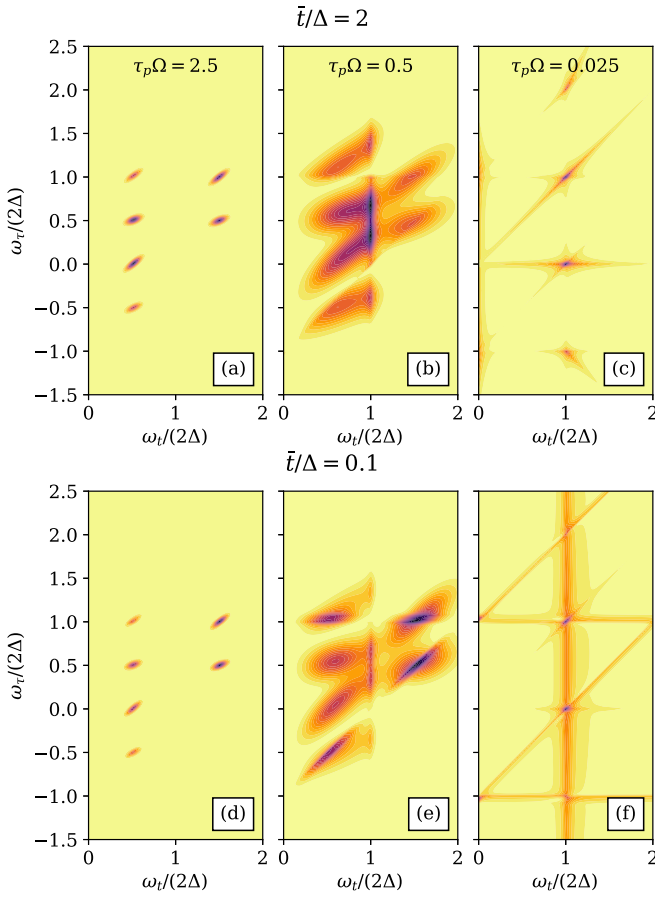


FIG. 10. Fully gauge-invariant 2D map of $|J_{2D}(\omega_l, \omega_r)|$ containing all the processes foreseen by Eq. (34), under the same driving conditions of Figs. 8 and 9. In panels (a)–(c) we consider $\tilde{t}/\Delta = 2$, while in panels (d)–(f) we consider instead $\tilde{t}/\Delta = 0.1$.

the various terms differing in the ordering of the scattering events. The third is equivalent to purely interband transitions between the CDW bands, mediated by four σ_1 vertices. After lengthy algebra, one can recollect [67] the various terms in a fully symmetric fashion under the exchange of $i\Omega_i$:

$$X^{(4)}(i\Omega_1, i\Omega_2, i\Omega_3, \mathbf{k}) = \tanh(\beta E_{\mathbf{k}}/2) \times \left[\frac{\Delta^4}{E_{\mathbf{k}}^4} K^{\text{inter}}(E_{\mathbf{k}}, i\Omega_1, i\Omega_2, i\Omega_3) + \frac{2\Delta^2 \epsilon_{\mathbf{k}}^2}{E_{\mathbf{k}}^4} K^{\text{mi}}(E_{\mathbf{k}}, i\Omega_1, i\Omega_2, i\Omega_3) \right]. \quad (39)$$

Here we define the interband contribution

$$\bar{K}^{\text{inter}}(E_{\mathbf{k}}, i\Omega_i) = \left(\frac{1}{i\Omega_{123} - 2E_{\mathbf{k}}} - \frac{1}{i\Omega_{123} + 2E_{\mathbf{k}}} \right) \times \sum_{i \neq j} \left(\frac{1}{i\Omega_i - 2E_{\mathbf{k}}} \frac{1}{i\Omega_j + 2E_{\mathbf{k}}} + \frac{1}{i\Omega_i + 2E_{\mathbf{k}}} \frac{1}{i\Omega_j - 2E_{\mathbf{k}}} \right), \quad (40)$$

which has the same frequency structure of Eq. (19). We also have the mixed one

$$\bar{K}^{\text{mi}}(E_{\mathbf{k}}, i\Omega_i) = \left(\frac{1}{i\Omega_{123} - 2E_{\mathbf{k}}} - \frac{1}{i\Omega_{123}} \right) \sum_{i \neq j} \frac{1}{i\Omega_{ij} - 2E_{\mathbf{k}}} \times \left(\frac{1}{i\Omega_i - 2E_{\mathbf{k}}} + \frac{1}{i\Omega_j - 2E_{\mathbf{k}}} \right) - (E_{\mathbf{k}} \rightarrow -E_{\mathbf{k}}). \quad (41)$$

The effects of the band dispersion and of the light polarization can be encoded in a tensorial density of states defined as

$$\tilde{\rho}_{\alpha\beta\gamma\delta}^{\text{para}}(\epsilon) \equiv \sum_{\mathbf{k}} \delta(\epsilon - \epsilon_{\mathbf{k}}) \frac{\partial \epsilon_{\mathbf{k}}}{\partial k_{\alpha}} \frac{\partial \epsilon_{\mathbf{k}}}{\partial k_{\beta}} \frac{\partial \epsilon_{\mathbf{k}}}{\partial k_{\gamma}} \frac{\partial \epsilon_{\mathbf{k}}}{\partial k_{\delta}}. \quad (42)$$

In this way, it is possible to rewrite the full paramagnetic term as an integral over the energy $E = \sqrt{\epsilon^2 + \Delta^2}$, reading

$$K_{\alpha\beta\gamma\delta}^{(3),\text{para}}(i\Omega_i) \equiv \int dE [\rho_{\alpha\beta\gamma\delta}^{\text{inter}}(E) \bar{K}^{\text{inter}}(E, i\Omega_i) + \rho_{\alpha\beta\gamma\delta}^{\text{mi}}(E) \bar{K}^{\text{mi}}(E, i\Omega_i)], \quad (43)$$

where we defined for later convenience

$$\rho_{\alpha\beta\gamma\delta}^{\text{inter}}(E) = \frac{E}{\sqrt{E^2 - \Delta^2}} \tanh(\beta E/2) \frac{\Delta^4}{E^4} \times \tilde{\rho}_{\alpha\beta\gamma\delta}^{\text{para}}(\sqrt{E^2 - \Delta^2}), \quad (44)$$

$$\rho_{\alpha\beta\gamma\delta}^{\text{mi}}(E) = \frac{E}{\sqrt{E^2 - \Delta^2}} \tanh(\beta E/2) \frac{2(E^2 - \Delta^2)\Delta^2}{E^4} \times \tilde{\rho}_{\alpha\beta\gamma\delta}^{\text{para}}(\sqrt{E^2 - \Delta^2}). \quad (45)$$

In the case of a square lattice and with the nearest-neighbor band dispersion in Eq. (27) one can obtain analytical expressions for the tensorial density of states Eq. (42), as shown in the SM [67].

B. Fully diamagnetic contribution

The fully diamagnetic contribution S_{22} leads to the nonlinear current

$$J_{\alpha}^{(3),\text{dia}}(i\Omega) = \frac{\delta S_{22}[A^4]}{\delta A_{\alpha}(-i\Omega)} \equiv \sum_{i\Omega'} K_{\alpha\beta\gamma\delta}^{(3),\text{dia}}(i\Omega') A_{\beta}(i\Omega - i\Omega') A_{\gamma}^2(i\Omega'), \quad (46)$$

where we defined as $A_{\gamma}^2(i\Omega)$ the Fourier transform of $A_{\gamma}(\tau)A_{\delta}(\tau)$ in Matsubara frequencies, namely,

$$A_{\gamma}^2(i\Omega) = \sum_{i\Omega'} A_{\gamma}(i\Omega') A_{\delta}(i\Omega - i\Omega'). \quad (47)$$

By means of a change of variable, the expression in Eq. (46) can also be rewritten in the completely symmetric form

$$J_{\alpha}^{(3),\text{dia}}(i\Omega) = \sum_{i\Omega_i} K_{\alpha\beta\gamma\delta}^{(3),\text{dia}}(i\Omega_{ij}) A_{\beta}(i\Omega_1) A_{\gamma}(i\Omega_2) A_{\delta}(i\Omega_3) \times \delta(i\Omega - i\Omega_{123}), \quad (48)$$

where $i\Omega_{ij}$ is the sum of any two incoming frequencies. Considering the structure of the S_{22} diagram in Fig. 7 one obtains

$$K_{\alpha\beta\gamma\delta}^{(3),\text{dia}}(i\Omega_{ij}) = \sum_{\mathbf{k}} \frac{\partial^2 \epsilon_{\mathbf{k}}}{\partial k_{\alpha} \partial k_{\beta}} \frac{\partial^2 \epsilon_{\mathbf{k}}}{\partial k_{\gamma} \partial k_{\delta}} X^{(2)}(i\Omega_{ij}, \mathbf{k}), \quad (49)$$

where $X^{(2)}$ has the typical structure of a two-point correlation function:

$$K_{\alpha\beta\gamma\delta}^{(3),\text{dia}}(i\Omega_{ij}) = \sum_{\mathbf{k}} \frac{\partial^2 \epsilon_{\mathbf{k}}}{\partial k_{\alpha} \partial k_{\beta}} \frac{\partial^2 \epsilon_{\mathbf{k}}}{\partial k_{\gamma} \partial k_{\delta}} \tanh(\beta E_{\mathbf{k}}/2) \times \frac{\Delta^2}{E_{\mathbf{k}}^2} \left(\frac{1}{i\Omega_{ij} - 2E_{\mathbf{k}}} - \frac{1}{i\Omega_{ij} + 2E_{\mathbf{k}}} \right). \quad (50)$$

As before, the frequency-dependent part of the response is encoded in the structure:

$$K^{\text{dia}}(E_{\mathbf{k}}, i\Omega_{ij}) = \frac{1}{i\Omega_{ij} - 2E_{\mathbf{k}}} - \frac{1}{i\Omega_{ij} + 2E_{\mathbf{k}}}, \quad (51)$$

while the effect of the electronic dispersion is accounted for by a tensorial density of states, defined for the diamagnetic vertex as

$$\tilde{\rho}_{\alpha\beta\gamma\delta}^{\text{dia}}(\epsilon) \equiv \sum_{\mathbf{k}} \delta(\epsilon - \epsilon_{\mathbf{k}}) \frac{\partial^2 \epsilon_{\mathbf{k}}}{\partial k_{\alpha} \partial k_{\beta}} \frac{\partial^2 \epsilon_{\mathbf{k}}}{\partial k_{\gamma} \partial k_{\delta}}. \quad (52)$$

In full analogy with Eq. (43), one can put

$$K_{\alpha\beta\gamma\delta}^{(3),\text{dia}}(i\Omega_{ij}) = \int dE \rho_{\alpha\beta\gamma\delta}^{\text{dia}}(E) \bar{K}^{\text{dia}}(E, i\Omega_{ij}), \quad (53)$$

where we also introduced the quantity

$$\rho_{\alpha\beta\gamma\delta}^{\text{dia}}(E) = \frac{E}{\sqrt{E^2 - \Delta^2}} \tanh(\beta E/2) \frac{\Delta^2}{E^2} \times \tilde{\rho}_{\alpha\beta\gamma\delta}^{\text{dia}}(\sqrt{E^2 - \Delta^2}). \quad (54)$$

Also in this case, for a square lattice and with the nearest-neighbor band dispersion in Eq. (27), one can obtain analytical expressions for the tensorial density of states Eq. (52), which is also shown in the SM [67].

C. Analytical continuation and 2D map

Once we compute the third-order kernel $K^{(3)}$ in Matsubara frequencies, we need to perform the analytical continuation to real frequencies, $i\Omega_i \rightarrow \omega_i + i\eta$, with $\eta > 0$ [69,84], to obtain the 2D map. The nonlinear current can then be expressed as a function of $(\omega_t, \omega_{\tau})$ by using the prescription in Eqs. (14)

and (15). Assuming for simplicity $A_{\tau}(\omega) = A_0(\omega) = A(\omega)$, we can write a contribution to the 2D map for each diagram in the effective action in Eq. (34):

$$J_{00\tau}^{(3)}(\omega_t, \omega_{\tau}) = A(\omega_{\tau}) \int d\omega K_S^{(3)}(\omega, \omega_{\tau}, \omega_t - \omega_{\tau} - \omega) \times A(\omega) A(\omega_t - \omega_{\tau} - \omega), \quad (55)$$

with an analogous expression for $J_{0\tau\tau}^{(3)}$, along the lines of Eq. (15).

In analogy to what we found before in the case of the two-level system, we expect that in the impulsive limit the 2D map will be determined by the frequency resonances of the kernel. In the noninteracting electronic model that we are considering, the frequency dependence of the kernel is described by a combination of simple poles at $\pm 2E$, and the possible $2E$ values are weighted by a tensorial density of states $\rho(E)$, as shown in Eq. (43) for the paramagnetic contribution and Eq. (53) for the diamagnetic one. To get an analytical intuition on the structure of the 2D map, it is then convenient to recast the nonlinear signal by factorizing the energy integration from the signatures in the $(\omega_t, \omega_{\tau})$ planes obtained for each value of E . In other words, the integral one has to compute in Eq. (55) has the form

$$J_{00\tau}^{(3)}(\omega_t, \omega_{\tau}) \equiv \int dE \rho(E) \bar{J}_{00\tau}(E, \omega_t, \omega_{\tau}) \quad (56)$$

where for each value of E the quantity $\bar{J}_{00\tau}(E, \omega_t, \omega_{\tau})$ is obtained as

$$\bar{J}_{00\tau}(E, \omega_t, \omega_{\tau}) = A(\omega_{\tau}) \int d\omega \bar{K}_S(E, \omega, \omega_{\tau}, \omega_t - \omega_{\tau} - \omega) \times A(\omega) A(\omega_t - \omega_{\tau} - \omega). \quad (57)$$

Because $\bar{K}_S(E, \omega, \omega_{\tau}, \omega_t - \omega_{\tau} - \omega)$ is a product of simple poles, as shown in, e.g., Eqs. (40) and (41) for the paramagnetic term, when $A(\omega)$ has a Gaussian envelope the ω integration in Eq. (57) can be expressed in terms of Faddeeva functions. As we shall see below, the obtained *analytical* expression for each value of E remains to be numerically integrated over the tensorial density of states to provide the final result.

Let us first analyze the paramagnetic contribution. By following the procedure outlined above [67] one can write, for the fully interband contribution at a fixed E value, the expression

$$\begin{aligned} \bar{J}_{00\tau}^{\text{inter}}(E, \omega_t, \omega_{\tau}) &= A(\omega_{\tau}) \left(\frac{1}{\omega_t + i\eta - 2E} - \frac{1}{\omega_t + i\eta + 2E} \right) \\ &\times \left[\frac{F_{00\tau}^{\text{inter}}(2E)}{\omega_{\tau} + i\eta - 2E} + \frac{F_{00\tau}^{\text{inter}}(-2E)}{\omega_{\tau} + i\eta + 2E} + \frac{F_{00\tau}^{\text{inter}}(2E) + F_{00\tau}^{\text{inter}}(-2E)}{\omega_t - \omega_{\tau} + i\eta} \right], \end{aligned} \quad (58)$$

$$\begin{aligned} \bar{J}_{0\tau\tau}^{\text{inter}}(E, \omega_t, \omega_{\tau}) &= A(\omega_t - \omega_{\tau}) \left(\frac{1}{\omega_t + i\eta - 2E} - \frac{1}{\omega_t + i\eta + 2E} \right) \\ &\times \left[\frac{F_{0\tau\tau}^{\text{inter}}(2E)}{\omega_t - \omega_{\tau} + i\eta - 2E} + \frac{F_{0\tau\tau}^{\text{inter}}(-2E)}{\omega_t - \omega_{\tau} + i\eta + 2E} + \frac{F_{0\tau\tau}^{\text{inter}}(2E) + F_{0\tau\tau}^{\text{inter}}(-2E)}{\omega_{\tau} + i\eta} \right], \end{aligned} \quad (59)$$

where the function $F_{00\tau}^{\text{inter}}$ reads

$$F_{00\tau}^{\text{inter}}(2E) = \int d\omega \frac{A(\omega)A(\omega_t - \omega_\tau - \omega)}{\omega + i\eta + 2E}, \quad (60)$$

while the function $F_{0\tau\tau}^{\text{inter}}$ is obtained from $F_{00\tau}^{\text{inter}}$ by exchanging $\omega_t - \omega_\tau \leftrightarrow \omega_\tau$.

Let us now assume that the envelope of the incoming gauge potential can be well approximated by a Gaussian, i.e., $A(t) = \bar{A}e^{-t^2/(2\tau_p^2)}\cos(\Omega t)$. For sufficiently short-in-time pulses, where one can approximate $A(\omega) = \bar{A}$ as a frequency-independent constant over the energy scales of the system, the F^{inter} functions can be evaluated through residues and they simply reduce to a constant. This has the remarkable consequence that Eqs. (58) and (59) produce the same results obtained in the two-level system, described by Eqs. (21) and (22), respectively. In other words, we recover the same frequency structure of the two-level case, provided that Δ is replaced by E . We then expect, in analogy with Figs. 6(b) and 6(e), four peaks in the half plane $\omega_t > 0$ at coordinates $(2E, -2E)$, $(2E, 0)$, $(2E, 2E)$, and $(2E, 4E)$. The calculation of the contribution coming from $\bar{K}^{\text{mi}}$, reported in the SM [67], can instead be shown to lead to enhanced signals in the 2D map at the intercepts of the lines $\omega_t = 0, 2E$ with $\omega_\tau = 2E$ or $\omega_t - \omega_\tau = 2E$.

We stress here that the equivalence between the two-level case and the current lattice system needs to be properly framed. To be consistent with Ref. [37], in the two-level analysis we have shown the 2D map produced by a frequency-independent *electric field* $E(\omega) = \bar{E}$, which would correspond to a frequency-dependent gauge potential $A(\omega) \sim \bar{E}/\omega$ in Eqs. (58) and (59). In the following we want to analyze the 2D response to a short-in-time (broadband) and long-lasting (narrowband) electric field. Given the relation $E(t) \sim \partial_t A(t)$, we can realize such an effect by varying the envelope of our Gaussian $A(t)$ via $\tau_p\Omega$, even if the impulsive limit would not correspond to a $\delta(t)$ -like electric field. Within the Gaussian approximation for $A(t)$, but keeping a finite duration of the pulse, the functions F^{inter} can be reduced to an *analytical* expression, given by a combination of Faddeeva functions, as shown in the SM [67]. In practice, this implies a reduction in the numerical effort to calculate the 2D maps, whose result for the full paramagnetic contribution is shown in Fig. 8.

We simulate the signal for three values of $\tau_p\Omega$, which correspond to an incoming narrowband, intermediate, or broadband pulse, while setting the central frequency of the pump inside the gap $\Omega = \Delta$. As expected, for decreasing values of $\tau_p\Omega$, the 2D maps evolve from the six spots at multiples of Ω expected in the monochromatic limit, as shown in Figs. 8(a) and 8(d), to a more complex spectral distribution, in Figs. 8(c) and 8(f), dominated by the features of the kernel.

Since the latter strictly depends on the continuum of excitations, we consider two values of $\bar{t}/\Delta$. In Figs. 8(a)–8(c) we show the results setting $\bar{t}/\Delta = 2$, while in Figs. 8(d)–8(f) we consider $\bar{t}/\Delta = 0.1$, which intuitively corresponds to a less dispersive band structure. In the first case, panel (c), as the pump width increases, the spectral weight accumulates around four spots located at $(2\Delta, \pm 2\Delta)$, $(2\Delta, 0)$, and $(2\Delta, 4\Delta)$, i.e., the same positions appearing in the two-level simulation of Fig. 6. However, since the bands now have a

large dispersion, the peaks show a significant elongation along the lines that connect each one of them with the origin, namely $\omega_\tau = \pm\omega_t, 0, 2\omega_t$. On top of that, peaks along the line $\omega_t = 0$ are also visible in panel (c). As mentioned before, they purely arise from the mixed interband-intraband scattering mediated by $\bar{K}^{\text{mi}}$. Then it is possible to identify regions on the 2D map that are selectively sensitive to this specific nonlinear pathway, as also discussed in Ref. [49]. On the other hand, if $\bar{t}/\Delta = 0.1$ as in panel (f), we recover more similar results to Fig. 6. This can be understood by analyzing the behavior of the two functions $\rho^{\text{inter}}(E)$ and $\rho^{\text{mi}}(E)$, as shown in the SM [67]. While the former is singular for $E \rightarrow \Delta$, the latter vanishes in the same limit; in addition, there is a factor t^2/Δ^2 in the whole E interval suppressing $\rho^{\text{mi}}(E)$ with respect to $\rho^{\text{inter}}(E)$. Consequently, one can approximate $\rho^{\text{inter}}(E) \sim \delta(E - \Delta)$ and $\rho^{\text{mi}}(E) = 0$, so that the mixed scattering is now washed out and the elongation of the remaining four peaks is now concentrated in close proximity to 2Δ .

Let us now compute the result for the diamagnetic process. Here the calculation is simplified due to the dependence of the nonlinear kernel only on the sum of two incoming frequencies of the field, i.e., from Eq. (48) one has $\bar{K}^{\text{dia}}(\omega_{ij})$ as appropriate for two-photon vertices. For the $\bar{J}_{00\tau}^{\text{dia}}$ part of the 2D map, one needs then to write out all the possible combinations of two incoming frequencies that enter the kernel:

$$\omega_{ij} = \omega + \omega_\tau, \omega_t - \omega, \omega_t - \omega_\tau. \quad (61)$$

More explicitly, by considering the structure of Eq. (51), one finds

$$\begin{aligned} \bar{J}_{00\tau}^{\text{dia}}(E, \omega_t, \omega_\tau) = & 2A(\omega_\tau)[F_{00\tau}^{\text{dia}}(-2E) - F_{00\tau}^{\text{dia}}(2E)] \\ & + A(\omega_\tau)A^2(\omega_t - \omega_\tau) \left(\frac{1}{\omega_t - \omega_\tau + i\eta - 2E} \right. \\ & \left. - \frac{1}{\omega_t - \omega_\tau + i\eta + 2E} \right), \end{aligned} \quad (62)$$

$$\begin{aligned} \bar{J}_{0\tau\tau}^{\text{dia}}(E, \omega_t, \omega_\tau) = & 2A(\omega_t - \omega_\tau)[F_{0\tau\tau}^{\text{dia}}(-2E) - F_{0\tau\tau}^{\text{dia}}(2E)] \\ & + A(\omega_t - \omega_\tau)A^2(\omega_\tau) \left(\frac{1}{\omega_\tau + i\eta - 2E} \right. \\ & \left. - \frac{1}{\omega_\tau + i\eta + 2E} \right), \end{aligned} \quad (63)$$

where the function $F_{00\tau}^{\text{dia}}$ reads

$$F_{00\tau}^{\text{dia}}(2E) = \int d\omega \frac{A(\omega)A(\omega_t - \omega_\tau - \omega)}{\omega_\tau + \omega + i\eta + 2E}, \quad (64)$$

and $F_{0\tau\tau}^{\text{dia}}$ is obtained from $F_{00\tau}^{\text{dia}}$ by exchanging $\omega_t - \omega_\tau \leftrightarrow \omega_\tau$. As before, in the limit $A(\omega) = \bar{A}$ the F^{dia} functions are constant and the combination in the first lines of Eqs. (62) and (63) vanishes. The remaining terms then give

$$\bar{J}_{00\tau}^{\text{dia}}(E, \omega_t, \omega_\tau) \propto \frac{1}{\omega_t - \omega_\tau + i\eta - 2E} - (E \rightarrow -E), \quad (65)$$

$$\bar{J}_{0\tau\tau}^{\text{dia}}(E, \omega_t, \omega_\tau) \propto \frac{1}{\omega_\tau + i\eta - 2E} - (E \rightarrow -E). \quad (66)$$

From this expression we notice that the spectral weight on the 2D map in this approximation is spread along the lines $\omega_t - \omega_\tau = \pm 2E$ and $\omega_\tau = \pm 2E$.

The complete computation of the diamagnetic process is shown in Fig. 9. Here we used the same resonance condition $\Omega = \Delta$ and values $\tau_p \Omega$ and t/Δ adopted in Fig. 8. In the upper row, panels (a)–(c), we considered the more dispersive limit $\bar{t}/\Delta = 2$, while in the lower row, panels (d)–(f), we considered the $\bar{t}/\Delta = 0.1$ case. The diamagnetic process is much less effective in selecting specific spots in the 2D map compared to the paramagnetic ones, as one can observe in panels (c)–(f), where we show the results obtained in the impulsive limit, which reproduce the striplike phenomenology highlighted in Eqs. (65) and (66).

The set of Eqs. (56)–(66) represents the central result of the present work: it provides, indeed, a general expression for the computation of the 2D maps, where the details of the excitation spectrum of the system are fully encoded in the density of states $\rho(E)$ of Eq. (56). As an example, in the case of the 2D lattice model in Eq. (27), this is in turn given for paramagneticlike and diamagneticlike processes by the analytical expressions in Eqs. (44), (45), and (54), respectively. The present approach can be in principle extended to interacting electronic systems, once the spectrum E_α of the interacting eigenstates (labeled by a generic index α) and the matrix elements of light-matter couplings are known. In this case, some additional care should be taken to account for all the possible transitions triggered by the possible light-matter couplings in $K_S^{(3)}$, as shown in, e.g., Refs. [50,69].

In Fig. 10 we finally show the simulations of the fully gauge-invariant response $|J_{2D}^{(3)}(\omega_t, \omega_\tau)|$ for values $\bar{t}/\Delta = 0.1$ and $\bar{t}/\Delta = 2$, respectively, obtained by summing all the contributions to the nonlinear response kernel $K^{(3)}$ foreseen by Eq. (34). In the SM [67], we also provide separated plots for each class of processes to highlight their relative weight. As one can see, by varying either the spectral envelope, ranging from narrowband in panels (a),(d) to broadband in panels (c),(f), or the system's excitations, ranging from more (top row) to less (bottom row) dispersive bands, one gets quite different 2D maps, where contributions of different subsets can be dominant. As before, in the narrowband limit the spots are always controlled by the pulse frequency. As the pulse become broadband we can observe that for both choices of $\bar{t}/\Delta$ the fully paramagnetic contribution is always relevant. Signatures coming from two-point responses, arising either from the fully diamagnetic term S_{22} or from the mixed S_{31} one, as shown in the SM [67] for details, are visible in the fully gauge-invariant response as striplike features, whose relative strength depends on the ratio $\bar{t}/\Delta$.

As a final remark, we note that in the present discussion the nonlinear current is computed as a function of the pulse *inside* the sample. As mentioned at the beginning of Sec. II and explicitly shown in Sec. II B of the SM [67], this is a reasonable approximation for an experiment in transmission geometry from nonabsorbing thin films. How this has to be connected to the *incoming* pulse in different experimental geometries is a crucial point that will be discussed in the next section.

IV. PROPAGATION EFFECTS IN THE 2DTS MAPS: THE CASE OF JOSEPHSON PLASMONS

A. Screening and propagation effects connected to the linear response

In the previous section we focused on the different signatures obtained in the 2D maps when the nonlinear processes originate from paramagnetic or diamagnetic light-matter coupling terms at microscopic level. In both cases the possibility to disentangle different excitation pathways requires a broad pump spectrum, since in the opposite monochromatic limit the 2D spectrum has only isolated spots where the structure of the kernel cannot be resolved [see panels (a)–(d) of Figs. 8–10]. As a consequence, to really make a bridge between the theoretically-computed $K^{(3)}$, that can be carried out in principle in a controlled way within certain approximations, and the outcomes of the experimental measurements, one must be able to predict the spectral content of the pump field A_{in} *inside* the sample, where the nonlinear current is generated. In linear response, this is usually a simple problem to be solved via Maxwell's equations: indeed, when the induced current is linear in the gauge field A , Maxwell's equations are linear and all the eventual complications of the mixing of electromagnetic waves with a dipole-active mode can be accounted for by introducing the frequency-dependent refractive index $n(\omega)$ of the system. In addition, by properly using boundary conditions at the sample/vacuum interfaces, one can connect the field inside the material to the one outside. In the presence of nonlinear processes Maxwell's equations themselves become nonlinear and an alternative approach must be followed to answer two questions: (i) what is the spectrum of the terahertz fields driving the nonlinear current computed via Eq. (1); (ii) what is the connection between the nonlinear current generated inside the material and the nonlinear electric field in Eq. (7) detected experimentally to construct the 2D map.

To answer these questions we should go back to the definition in Eq. (7) of the experimentally measured 2D signal $E_{2D}(t, \tau)$, and discuss the limits of applicability of the approximation in Eq. (4), where $E_{2D}(t, \tau)$ is identified with the nonlinear current inside the material. As one can intuitively understand, the problem arises when the system hosts dipolar modes contributing to a frequency-dependent refractive index $n(\omega)$, that varies considerably on the range of frequencies set by the spectrum of the A pulse. Whenever this happens, we expect that the internal field A_{in} gets modified with respect to the *external* one A_{ext} . This effect can be accounted for macroscopically by the transmission coefficient

$$t(\omega) = \frac{2}{1 + n(\omega)}, \quad (67)$$

whose impact will be relevant whenever optically-active modes overlap with the pump/probe spectrum.

In addition to this, one has to account for the fact that the nonlinear current generated inside the material acts as a source for the Maxwell's equations, modifying the spectrum of the nonlinear field generated inside the system. This problem can be solved using an iterative perturbative approach [56,57,59,85], as detailed in the SM [67] for transmission and reflection geometries of a sample of any thickness. In summary, in the present case the pipeline of

the calculations is the following. Starting from the general assumption that $E_{2D}(t, \tau)$ in Eq. (7) represents the experimentally accessible quantity, one first computes the induced nonlinear current inside the system, Eq. (8), as a $J^{(3)}$ function of the gauge field A . Next, one accounts for propagation effects to connect a generic outgoing electric field E^{out} to the $J^{(3)}$ inside the system. This latter part is accomplished by solving the Maxwell's equations inside the system in the

presence of a nonlinear current, and accounting for both boundary conditions at the sample/vacuum interfaces, and for phase-matching conditions in the presence of multiwave mixing. All these steps are detailed in the SM [67]. Here we just report the final result for the third-order outgoing electric field in a reflection geometry from a bulk sample, i.e. the situation appropriate for the case of the Josephson plasmon measured in Ref. [38] that we will discuss in the following:

$$E^{\text{out}}(\omega_t) = \frac{4\pi i}{c} \frac{1}{1 + n(\omega_t)} \int d\omega_i K^{(3)}(\omega_1, \omega_2, \omega_3) A_{\text{ext}}(\omega_1) A_{\text{ext}}(\omega_2) A_{\text{ext}}(\omega_3) \frac{t(\omega_1)t(\omega_2)t(\omega_3)}{n(\omega_t)\omega_t/c + \sum_i n(\omega_i)\omega_i/c} \delta\left(\omega_t - \sum_i \omega_i\right). \quad (68)$$

In the case of a bulk system considered here, one only retains the generated reflected field on the left of the vacuum/material interface and the transmitted one on its right side, thus neglecting the effect of back-reflected fields from the opposite side of the sample. This is a reasonable approximation for a millimeter-scale superconducting sample probed with terahertz light, as it is the case for the experimental geometry in Ref. [38] discussed below. The transmission coefficients in Eq. (68) account then for the boundary conditions at the interface, while the denominator encodes the phase-matching condition ($\sum_i k_i - k_r = 0$) for the momenta $k_i = n(\omega_i)\omega_i/c$ of the generating pulses and the momentum $k_r = -n(\omega_t)\omega_t/c$ of the reflected outgoing field, where we also assume a local ($q = 0$) dielectric response via $n(\omega)$ appropriate for terahertz and optical probes. Further details on the derivation of the

formulas for the reflected/transmitted E^{out} in the bulk/thin film limit, along with their validation against known expressions, are provided in Secs. II A and II B of the SM [67], where all the needed conversion factors between the electric field and the gauge potential are carefully taken into account.

Once established a general expression for the nonlinear field propagating outside the sample we can proceed in the evaluation of the 2D map by following the same steps leading to Eqs. (14) and (15) in Sec. II. Let us consider e.g. the contribution with the double interaction with the $A_0(t)$ field and the single interaction with $A_\tau(t)$. One starts from Eq. (68) and replaces one frequency with ω_τ and the other two with ω and $\omega_t - \omega_\tau - \omega$, eliminating one frequency integration via the energy conservation and another one with the $\tau \rightarrow \omega_\tau$ Fourier transform. We end up with

$$E_{00\tau}(\omega_t, \omega_\tau) = \int d\omega K_S^{(3)}(\omega_\tau, \omega, \omega_t - \omega_\tau - \omega) A_{\tau, \text{ext}}(\omega_\tau) A_{0, \text{ext}}(\omega) A_{0, \text{ext}}(\omega_t - \omega_\tau - \omega) \times \frac{2\pi i t(\omega_t)t(\omega_\tau)t(\omega)t(\omega_t - \omega_\tau - \omega)}{n(\omega_t)\omega_t + n(\omega_\tau)\omega_\tau + n(\omega)\omega + n(\omega_t - \omega_\tau - \omega)(\omega_t - \omega_\tau - \omega)}. \quad (69)$$

In the spirit of Eq. (6), in this expression we have separated on the first line inside the integral the result that one would obtain neglecting the propagation effects inside the material, where the external fields are convoluted with the nonlinear third-order kernel. In the second line, instead, we see the explicit form of the function $M(\omega_\tau, \omega, \omega_t - \omega_\tau - \omega)$ introduced in Eq. (6), which has its own frequency dependence and can mask the genuine nonlinear effects included in $K^{(3)}$. In this way, we will see that even the simplest instantaneous kernel can show complicated signatures linked to the response due to the propagation of the generated signal at the interface between vacuum and material. An analogous result for $E_{0\tau\tau}(\omega_t, \omega_\tau)$ is obtained, as explained above, by switching $\omega_\tau \leftrightarrow \omega_t - \omega_\tau$ and $A_\tau \leftrightarrow A_0$, corresponding to the system interacting once with the field $A_0(t)$ and twice with $A_\tau(t)$.

Equation (69) and its counterpart for $E_{0\tau\tau}$ are general. However, to give a concrete example of the relevance of the propagation effects we apply this approach to a physically relevant case, that is the understanding of recent 2DTS experiments in Josephson plasmons [38]. The example is particularly relevant since, in contrast to the toy model of the insulator discussed in the previous Section, in this case the

same collective excitations contribute to screen the linear response to a terahertz pulse and to generate a nonlinear current. The understanding of 2DTS spectra of Josephson plasmon then offers a perfect benchmark to highlight the necessary steps required to extract from *measured* nonlinear electric field the relevant information on the *microscopic* processes at play in the system, encoded in the $K^{(3)}$ kernel.

B. Screening effects from Josephson plasmons

As we mentioned in the introduction, soft Josephson plasmons are the natural low-energy collective excitations in layered superconductors that emerge when the phase of the SC order parameter mixes with the electromagnetic waves propagating inside the material. Since layered systems, like SC cuprates, have a weak interlayer coupling, the low energetic cost for pair tunneling among planes leads to a soft (i.e. of the order of few terahertz) plasma mode, affected by a strong Landau damping by quasiparticles above T_c . However, below T_c the opening of a SC gap removes damping effects and a well-defined mode appears, whose signature in the linear response is a sharp plasma edge in the reflectivity of the sample

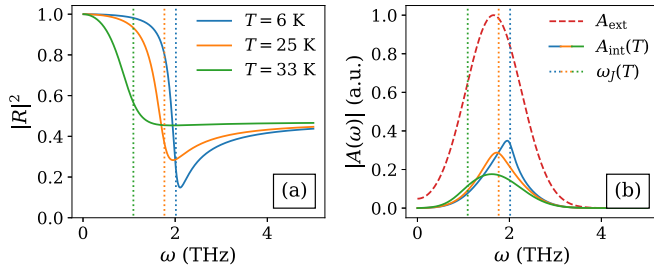


FIG. 11. (a) Simulated reflectance (left panel) of a LSCO sample at doping level $x = 0.17$ for three selected temperatures, using the interpolated parameters from Refs. [86,87] with the two-fluid model in Eq. (70). The dotted vertical lines denote the temperature-dependent value of $\omega_J(T)$. (b) Comparison between the spectrum of the gauge field outside the sample A_{ext} (dotted line), modeled as the experimental one used in Ref. [38], and inside the sample $A_{\text{in}}(\omega) = t(\omega)A_{\text{ext}}(\omega)$ (solid lines), where $t(\omega)$ is the transmission coefficient of Eq. (67). At low T the plasma edge (dotted vertical lines) produces a dramatic filtering of the incoming field around ω_J .

at the temperature-dependent value ω_J , as shown in Fig. 11(a). This is also the case for the nearly optimally-doped ($x = 0.17$) $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ sample measured by 2DTS in Ref. [38]. In this experiment the broadband external pump and probe fields have a central frequency Ω which matches the $T = 0$ value of ω_J , $\Omega \sim \omega_J(T = 0) = 2$ terahertz. As a consequence, the propagation of the light pulse *inside* the sample is modified by the formation of a plasmon-polariton, whose presence is encoded in the refractive index of the material. In the specific case of the SC Josephson plasmon a convenient description of the linear response can be achieved [88] by modeling the dielectric function for the transport along the stacking direction as the sum of a superfluid response and the normal dissipative term, namely

$$\epsilon(\omega) = \epsilon_\infty \left(1 - \frac{\omega_J^2}{\omega^2} + i \frac{\gamma}{\omega} \right). \quad (70)$$

Here γ is a phenomenological damping factor that accounts for the experimentally-observed smearing of the plasma edge, that also appears in the two-plasmon propagator in Eq. (73). The microscopic origin of this damping has been discussed in the literature in connection with a possible inhomogeneity of the Josephson couplings among different planes [89]. For the sake of the present discussion, the analysis of Ref. [88] suggests that the two-fluid model of Eq. (70) gives an excellent description of the measured linear response for the doping level of the sample of Ref. [38], that is the relevant aspect to quantitatively describe the refractive index $n(\omega) = \sqrt{\epsilon(\omega)}$ and the transmission coefficient in Eq. (67).

Here we considered for A_{ext} the pulse spectrum of Ref. [38], fitting the experimental applied field with $E(\omega) = i\omega A(\omega)$. We modeled the dielectric function in Eq. (70) by fitting the experimental data for LSCO samples at doping levels $x = 0.16$ [86] and $x = 0.18$ [87] at several temperatures, interpolating then the extracted values of ω_J and γ to estimate the ones appropriate for the $x = 0.17$ LSCO sample of [38]. As we can see from Fig. 11(b), at all temperatures the field gets strongly reshaped, with a narrowing of the peak and a shift from the (fixed) central frequency Ω of the pump

towards the temperature-dependent value of $\omega_J(T)$. In the next section, we will discuss how this effect influences the *nonlinear* response of the plasmon.

C. Nonlinear response of Josephson plasmons

A possible description of nonlinear effects emerging from plasma waves below T_c is provided by the Josephson model, describing a cosinelike energy dependence on the gauge-invariant phase difference among neighboring planes [60–64]

$$H = J \sum_n \cos \left(\theta_n - \theta_{n+1} - \frac{2e}{\hbar c} A \right), \quad (71)$$

where θ_n is the SC phase in layer n , J is the effective Josephson interaction between the phase degrees of freedom in neighboring layers, connected to the phase rigidity in the SC state. Here θ is a quantum variable whose conjugate field is the density: one can then show that the spectrum of the phase mode is the same of density fluctuations, which describe plasma waves with a value $\omega_J^2 \sim 4\pi e^2 J$ in the long-wavelength limit [9,90–92]. In addition, thanks to the minimal coupling of the phase difference to the gauge field, the *anharmonic* phase-only model of Eq. (71) provide a natural source for optical non linearity in the SC state [62–64,66]. While in the past these effects have been mainly described by solving the equation of motion for the phase variable $\phi_n \equiv \theta_n - \theta_{n+1}$ in the time domain, recent experimental measurements of high-harmonic generation and 2DTS with strong terahertz field triggered a direct theoretical computation of the nonlinear optical kernel in the frequency domain, along the lines of the approach discussed in the previous section. In full analogy with the discussion of the previous section, as shown in Eq. (34), the $K^{(3)}$ is obtained by deriving from Eq. (71) the terms of order A^4 . As one can intuitively understand, there is a first instantaneous (in time) contribution $\sim JA^4$ coming directly from the expansion of the cosine in Eq. (71), that has no structure in the frequency domain. In addition, one recovers a *diamagneticlike* coupling term $\sim A^2 \phi_n^2$ where two photons from the pump field are able to excite two plasma waves $\phi_n \equiv \theta_n - \theta_{n+1}$. The resulting contribution to the $K^{(3)}$ has the structure of a Kubo-like response, where two plasmons with opposite momenta are excited and then reabsorbed by the terahertz field. The computation of this kernel has been the subject of previous work [58,66], and we refer the reader to these papers for additional details in the derivation. For the purposes of the present discussion, we can then assume that the first, instantaneous contribution is represented by a constant kernel in the frequency domain, scaling with the Josephson coupling [58]

$$K^{\text{inst}} \sim J, \quad (72)$$

while for the two-plasmon kernel we can use the general expression [58]

$$K^{2p}(\omega) \sim J^2 \int dz \rho^{2p}(z) \frac{1}{z} \frac{\coth(\beta z/2)}{(2z)^2 - (\omega + i\gamma)^2}, \quad (73)$$

where $\rho^{2p}(z)$ is the density of states associated with the two-plasmon process. To get an intuition on the behavior of the two-plasmon kernel in Eq. (73), we observe that if one

neglects completely the momentum dispersion of the plasma waves in Eq. (73) then $\rho^{2p}(z) \propto \delta(z - \omega_J)$. As a consequence, one immediately recovers a maximum response of the system when the driving frequency ω matches $2\omega_J$. When the momentum dependence of the plasma waves is taken into account via the $\rho^{2p}(z)$ dependence, the resonances of the nonlinear kernel depend on the dispersion and the number of plasma branches, that is controlled by the number of layers per unit cells [58,90,92]. However, for systems with a single layer per unit cell, as the $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ sample measured in Ref. [38], the result remains qualitatively the same. Indeed, in the region in the proximity of ω_J , the density of states grows linearly as $\rho^{2p}(z) \sim z\theta(z - \omega_z)$. Moreover, near the critical temperature we can also approximate $\coth \beta z/2 \sim T/z$, so that the above kernel can be easily integrated and reads, neglecting constant prefactors,

$$K^{2p}(\omega) \sim \frac{T}{(\omega + i\gamma)^2} \log\left(1 - \frac{(\omega + i\gamma)^2}{4\omega_J^2}\right), \quad (74)$$

which almost coincides with the so-called plasmon squeezing contribution in Ref. [59]. The only effect of the plasmon dispersion is then to weaken the singularity from a delta-like to a log-like divergence for $\omega \approx 2\omega_J$. In the following, we will use the expression in Eq. (74) to simulate the 2D maps given by this process.

In full analogy with the diamagnetic electronic kernel in Eq. (53), also Eq. (73) depends on a single frequency, corresponding in the experiment to the sum of the frequencies of the two photons exciting the two plasmons. To compute the full 2D map we can then symmetrize the diamagnetic kernel in Eq. (73) exactly as we did in Eq. (61). We then obtain the kernel determining the nonlinear current originated from two interactions with A_0 and one with A_τ :

$$K_S^{(3),2p}(\omega_\tau, \omega, \omega_t - \omega_\tau - \omega) \sim K^{2p}(\omega + \omega_\tau) + K^{2p}(\omega_t - \omega) + K^{2p}(\omega_t - \omega_\tau). \quad (75)$$

Using again the symmetry in Eq. (14) upon replacement $\omega \rightarrow \omega_t - \omega_\tau - \omega$ we can show that the first two terms of the previous sum are actually equal. As it happened before, the form of the kernel $K_S^{(3),2p}(\omega_t - \omega_\tau, \omega, \omega_\tau - \omega)$ entering the response from one interaction with A_0 and two with A_τ is obtained by switching $\omega_\tau \leftrightarrow \omega_t - \omega_\tau$ in the previous expression.

In analogy with the discussion of the previous section, when the $A(\omega)$ spectrum is broad, one would expect that, in the absence of any modification of the pump spectrum inside the system, the processes mediated by the instantaneous kernel in Eq. (72) do not have clear structure in the frequency domain, while diamagnetic-like processes give rise to striplike features in the 2DTS maps, corresponding to the resonance frequencies of the kernel, as shown in Fig. 9(f). In Ref. [38] one is exactly in the impulsive limit for the driving A_{ext} terahertz field: assuming that this is also the case for A inside the system, the K^{2p} kernel leads to the map shown in Fig. 12(a). Here indeed the signal piles up along the lines $\omega_\tau \approx 2\omega_J$ and $\omega_\tau - \omega_t \approx 2\omega_J$, where $\omega_J = 2$ THz is the zero-temperature limit of the long-wavelength out-of-plane Josephson plasmon measured in Ref. [38]. Since the Josephson frequency $\omega_J(T)$ softens by increasing temperature, these

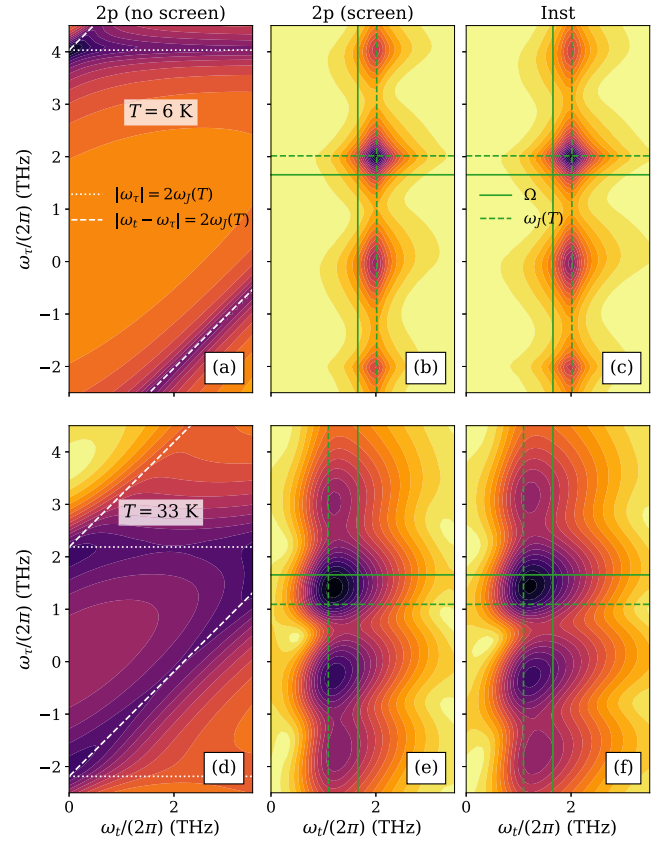


FIG. 12. The simulated 2D-terahertz response of LSCO ($x = 0.17$) probed with c -axis polarized electric field, with incoming spectrum similar to the one used in Ref. [38], for $T = 6$ K (a)–(c) and $T = 33$ K (d)–(f). In panels (a) and (d) we plot the behavior of the 2D signal produced by the two-plasmon kernel from Eqs. (73) and (75), considering the ideal situation of a constant in frequency field and neglecting propagation effects, which shows the typical signatures of the diamagnetic processes discussed in Sec. III. In panels (b) and (e) we consider the same two-plasmon excitation mechanism but we include propagation effects according to Eq. (68), while in panels (c) and (f) we compute the signal resulting from an instantaneous kernel. We superimpose as guides to the eye the positions of the central frequency of the pump (green dashed line) and of the plasma frequency at the various temperatures (blue dashed line).

signatures move in the 2D map when the system is warmed up, as shown in Fig. 12(d).

While the numerical results of Figs. 12(a)–12(d) are consistent with the general discussion of the previous section and with the analytical estimate of the kernel in Eq. (73), they are in striking contrast with the experimental findings of Ref. [38]. Indeed, the main outcome of these measurements is the appearance of four well-defined spots in the 2D map located at $(\omega_J, 0)$, $(\omega_J, \pm\omega_J)$, and $(\omega_J, 2\omega_J)$. These correspond to the response expected for a generic diamagnetic kernel when the pump field *inside* the sample can be approximated as a monochromatic one centered at $\Omega \sim \omega_J$, as shown in Fig. 9(a). As we shall see, this is indeed an effect of the plasmon contribution to the *linear* response, that acts as a “filter” on the external broadband pump, washing out any resonant effect connected to the kernel itself.

D. 2D maps including propagation effects

To account for the filtering effect shown in Fig. 11(b) we must resort to the full expression in Eq. (69), where the second line describes filtering by the $t(\omega)$ factors and propagation effects connected to multiwave mixing. We then computed the realistic 2D response by using the general expression in Eq. (69) and its related counterpart for $E_{0\tau\tau}(\omega_t, \omega_\tau)$ to the case of the Josephson plasmon, by adopting the previously discussed modeling of $n(\omega)$. The results are shown in Figs. 12(c)–12(f) for what concerns an instantaneous kernel, that just reduces to a constant, and in Figs. 12(b)–12(e) for what concerns the two-plasmon kernel in Eq. (73). Notice that, consistently with the physical origin of the $\epsilon(\omega)$, the same damping $\gamma(T)$ enters both the dielectric function via Eq. (70) and the two-plasmon kernel via Eq. (73). As a further remark, consistently with Ref. [38], we also want to only consider contributions from $t, \tau > 0$ in the 2D frequency map. To do so, we compute the inverse Fourier transform of Eq. (69) and its related $A_0 \leftrightarrow A_\tau$ counterpart, we select the relevant time quadrant, and then we Fourier transform back the signal to frequency space.

Let us first consider the $T = 6$ K case, reported in Figs. 12(b) and 12(c) for the instantaneous K^{inst} and two-plasmon K^{2p} kernel, respectively. We clearly observe the four spots located at the positions $(\omega_J, \pm\omega_J)$, $(\omega_J, 0)$, and $(\omega_J, 2\omega_J)$, with $\omega_J \sim 2$ THz, consistent with the experiments [38]. This result simply follows from the fact that at low temperature, where the plasma edge is particularly sharp, the filtering effect encoded in the $t(\omega_i)$ factors of Eq. (68) effectively converts the maps expected for a broadband pulse to the ones of a narrowband pulse, whose spectral content is controlled by the linear response. By increasing the temperature the peaks redshift along the ω_t axis following the softening of the $\omega_J(T)$ plasma edge, as shown in the dashed guiding lines in panels (e) and (f), while along the ω_τ axis the peak maxima are still sensitive to the central frequency of the pump. This behavior can be understood by close inspection of Eq. (69). Indeed, the overall $t(\omega_i)$ factor, which is only related to the propagation of the outgoing radiation, fixes the detection frequency of the 2D signal at the temperature-dependent plasma edge $\omega_J(T)$, while the remaining transmission factors integrated over ω contribute to spread out the ω_τ response in a range given by the overlap between A_{ext} , centered at Ω , and the transmission factor $t(\omega)$, centered at $\omega_J(T)$. A direct consequence of these findings is that it is unlikely that an experiment could resolve the difference between the instantaneous and diamagneticlike process. Indeed, both at $T = 6$ K [Figs. 12(b) and 12(c)] and at $T = 33$ K [Figs. 12(e) and 12(f)] the computed maps display essentially the same spectral features, due to a dominant effect of propagation, making it hard to distinguish minor differences in an experimental setup.

A somehow different conclusion on this respect has been reached recently in Ref. [59], where the authors argue that a different behavior between the instantaneous (named mean-field) and the two-plasmon (named squeezing) process can be observed for $T \sim T_c$. There are two differences between our approach and the one of Ref. [59]. First, the authors model the external pulse with a Lorentzian instead of a Gaussian profile, as shown in Fig. 13(e). To make the comparison with their

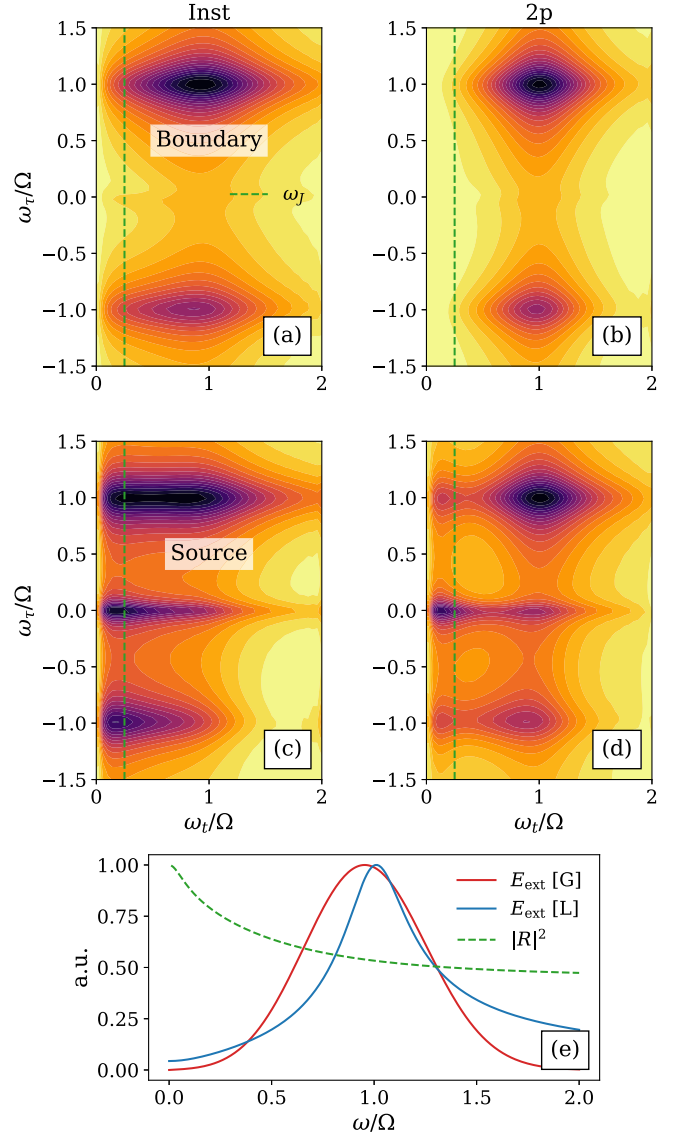


FIG. 13. 2D contour maps for the $E_{00\tau}$ signal, containing only the rephasing and nonrephasing contributions, obtained for the instantaneous (a), (c) and two-plasmon (b), (d) contributions using the screening procedure presented in this work (a), (b) and the one in Ref. [59] (c), (d). In the latter case a somehow qualitative difference between the two processes can be appreciated. (e) Comparison between the Lorentzian external pulse (blue line) used for the 2D maps of panels (a)–(d), that coincides with the model adopted in Ref. [59], and the Gaussian one (red line) used in the simulations of Fig. 12 (red). We also superimposed the reflectance (green dashed line) corresponding to a choice of parameters in Eq. (70) consistent with the one of Ref. [59], i.e. $\omega_J/\Omega = 0.25$, $\gamma/\Omega = 1$, and $\epsilon_\infty = 25$. Additional simulations for reasonable variations of the damping and pump spectrum are also shown in the SM [67].

result meaningful, we then repeat the computation of the 2D maps using the same pulse profile of Ref. [59]. The results are shown in Figs. 13(a) and 13(b). Apart from a sharpening of the features with respect to Fig. 12 due to the different choice of the pulse profile, the qualitative findings are the same. More specifically, the instantaneous and two-plasmon

responses remain almost spectroscopically indistinguishable, in contrast to what is stated in Ref. [59].

In addition to a different modeling of the external pulse, the authors of Ref. [59] propose a different description of the propagation effects, which are detailed in Sec. II C of the SM [67]. In short, the assumption there is to compute how the system responds to a homogeneous external current perturbation $J^{\text{ext}}(\omega)$ representing the applied electric field $E^{\text{ext}}(\omega)$, instead of solving the boundary-value problem leading to Eq. (68). This leads to a modified expression which does not contain the phase-matching denominator in Eq. (68) and screens the internal fields by $1/\epsilon(\omega)$ rather than $1/[1 + n(\omega)]$. To test the impact of this approximation as compared to the expression in Eq. (68) we then implement it and reproduce in Figs. 13(c) and 13(d) the 2D maps of Ref. [59]. Let us first focus on the comparison between panels (a) and (b), where the computation is done with the featureless instantaneous kernel, so that different spectral signatures can only be ascribed to propagation effects. In the approach based on the general formula 68 the spectral weight of the terahertz pulse inside the system remains localized around the central pump frequency, as shown in Fig. 13(a). In contrast, when one approximates screening effects as done in Ref. [59] the spectral weight of the internal field extends towards $\omega_l \approx 0$, as shown in Fig. 13(c). On the other hand, the two approaches to the screening give more similar results for what concerns the two-plasmon process, shown in Fig. 13(b) in the case of the simulation based on Eq. (68), and in Fig. 13(d) for the approximation used in Ref. [59]. Indeed, for the two-plasmon process the K^{2p} kernel filters out the spectral components of the driving fields around the resonance at $2\omega_l$, making the final result less sensitive to the modeling of the internal field.

The outcome of the comparison shown in Fig. 13 is that when screening effects are particularly pronounced some extra care should be taken in their modeling. Indeed, by following the approach of Ref. [59] one can in principle identify spectroscopically different features between the instantaneous and two-plasmon process, since the latter, in Fig. 13(d), shows well-defined spots, absent in the former, in Fig. 13(c). In other words, the experiments could discriminate among the two, as indeed suggested by the authors of Ref. [59]. However, this conclusion is at odds with the results obtained with the formula based on the boundary conditions in Eq. (68), where both processes lead to extremely similar maps, as shown in Figs. 13(a) and 13(b). At present, the most promising experimental route to distinguish the two processes relies on the identification of the frequency resonance of the two-plasmon kernel as a temperature enhancement [58], as observed, e.g., in Ref. [93] by third-harmonic generation measurement with multicycle pulses.

V. DISCUSSION AND CONCLUSIONS

The general formalism presented in Secs. III and IV aims to provide a theoretical framework that allows us to extract from the experimental 2DTS maps the crucial physical ingredients behind them. These include two complementary aspects: the first one is the theoretical computation of the nonlinear $\chi^{(3)}$ optical kernel, which for electronic systems

TABLE II. Summary of the maxima of the 2D (ω_l, ω_τ) map for the S_{1111} (para) and S_{22} (dia) processes studied in this section, in the broadband limit for the gauge field.

	Para	Dia
$J_{00\tau}$	$(\omega_l, \omega_\tau) = (2\Delta, 2\Delta)$ $(\omega_l, \omega_\tau) = (2\Delta, -2\Delta)$	$\omega_\tau = 2\Delta$ $\omega_\tau = -2\Delta$
$J_{0\tau\tau}$	$(\omega_l, \omega_\tau) = (2\Delta, 0)$ $(\omega_l, \omega_\tau) = (2\Delta, 4\Delta)$	$\omega_l - \omega_\tau = 2\Delta$ $\omega_l - \omega_\tau = -2\Delta$

amounts to computing correlation functions that in the diagrammatic language correspond to bubbles with multiple (up to four) photon vertices. These have been classified as paramagneticlike and diamagneticlike, referring to microscopic light-matter processes where the particle-hole excitations are driven by a single photon or two photons, respectively. As we have seen, when the pump pulse is short enough in time, so that its frequency spectrum is relatively broad on the energy scales relevant for microscopic excitations, the two classes of processes lead to specific and distinct signatures in the 2DTS. These general features, summarized in Table II, can be understood from the set of equations Eqs. (56)–(66), where we formulate the 2D problem by disentangling the detection structure, carried out by the dependence in (ω_l, ω_τ) , from the energy spectrum of the excitations, encoded in the form of the energy prefactors in Eqs. (44), (45), and (54). In the present manuscript we applied the formalism to a toy model reproducing a semiconductinglike band structure. We worked out several semianalytical expressions for the optical kernel and for the convolution with Gaussian light pulses. This approximation allowed us to make the numerical computation significantly faster, especially in the present case where the density of states of the fermionic excitations admits an analytical representation. Even though the experimental pulses are not necessarily Gaussian, and realistic band structures can be more complex, the present results provided us with a clear *interpretation* of the numerically obtained 2D maps, which had been already attempted by some authors [44,47,49–52,94]. An example of future applications of these results to realistic cases is provided, e.g., by the recent measurements of the nonlinear response of superconducting NbN in Ref. [31]. In that work the possible predominance of paramagnetic processes over the diamagnetic ones, that is expected on the basis of theoretical analysis of the nonlinear kernel, has been deduced from the temperature dependence of the nonlinear first-harmonic measurements, rather than from the structure of the 2D maps. The present formalism offers instead the possibility to infer this information directly from the analysis of the 2D maps at a given temperature, and possibly to distinguish *spectroscopically* pure electronic processes from those mediating collective modes, as, e.g., the Higgs mode. In fact, despite the fact that this issue has been extensively addressed theoretically, a clear experimental signature of the different excitation pathways is still lacking in single-pulse experiments, while recent experimental findings [30,31,33,34,95] suggest that it could be captured by two-pulse protocols. We observe that the present formalism can also be applied in the presence of the lower-order $\chi^{(2)}$ susceptibility, that is

the leading nonlinear process in systems lacking inversion symmetry, due to lattice or magnetic effects. Also in this case the response will be built upon paramagnetic and diamagnetic light-matter vertices, leading to potentially different spectroscopic signatures. Another interesting consequence is the possibility of extracting the *full* frequency dependence of $\chi^{(2)}(\omega_1, \omega_2)$ from the 2D map, since the procedure described in Sec. II would now saturate all the internal frequency integrals, while with $\chi^{(3)}$ there is always a residual ω integral to perform in Eqs. (14) and (15).

A second aspect concerns a proper treatment of the possible mixing of the electromagnetic waves with dipolar modes of the material. This is a well-known effect already present in linear response, that is usually described at the level of Maxwell's equations and Fresnel boundary conditions, which condense all the complications of the polariton formation in a frequency-dependent refractive index $n(\omega)$. However, for nonlinear response the same exact treatment is not possible, since Maxwell's equations themselves are not linear. To overcome this limitation we discussed the outcomes of an iterative procedure, based again on a perturbative approach to the linear response. The generic 2DTS map must then be computed using Eq. (68), or its generalization to transmission geometry. Such a tedious procedure turns out to be necessary, since propagation effects can dominate the features of the experimental response, masking microscopic mechanisms manifest via the $\chi^{(3)}$ optical kernel. As a case study we applied the formalism to the 2DTS of soft Josephson plasmons in layered superconductors, and we showed that in this case we cannot find a way to distinguish experimentally the two nonlinear contributions expected theoretically, corresponding to the presence or absence of a two-plasmon intermediate process. Such a conclusion is somehow at odds with previous theoretical results of Ref. [59], due to a different approach to the problem of the screening.

As we discussed in Sec. II B, the present diagrammatic approach can be used also to address the well-studied case of 2DTS in two-level systems, by reproducing the results usually obtained via a time-dependent density-matrix formalism. An interesting question is then how to incorporate in the many-body language the description of typical effects emerging within this context, the primary one being the role of *inhomogeneity*. Indeed, for molecular systems 2DTS can distinguish spectroscopically the signatures connected to the intrinsic lifetime $1/\gamma$ of a single mode from the ones due to the existence of a finite distribution ρ of possible values for the mode excitation energy ω_0 . As discussed in Refs. [23,96] the two effects appear as a broadening of the rephasing peak in different directions: while $\Delta\omega_0$ mostly controls the peak width along the diagonal of the lower half plane, $\omega_\tau = -\omega_t$, γ determines its width in the perpendicular direction as shown in Fig. 14. This effect can be reproduced by a phenomenological formula for the spectrum of the rephasing peak:

$$P^{\text{rep}}(\omega_t, \omega_\tau) \sim \int d\omega_0 \rho(\omega_0) \frac{1}{\omega_t - \omega_0 + i\gamma} \frac{1}{\omega_\tau + \omega_0 + i\gamma}, \quad (76)$$

where the choice of a Gaussian envelope for $\rho(\omega_0)$ leads to the same findings of Ref. [96]. A natural question is how

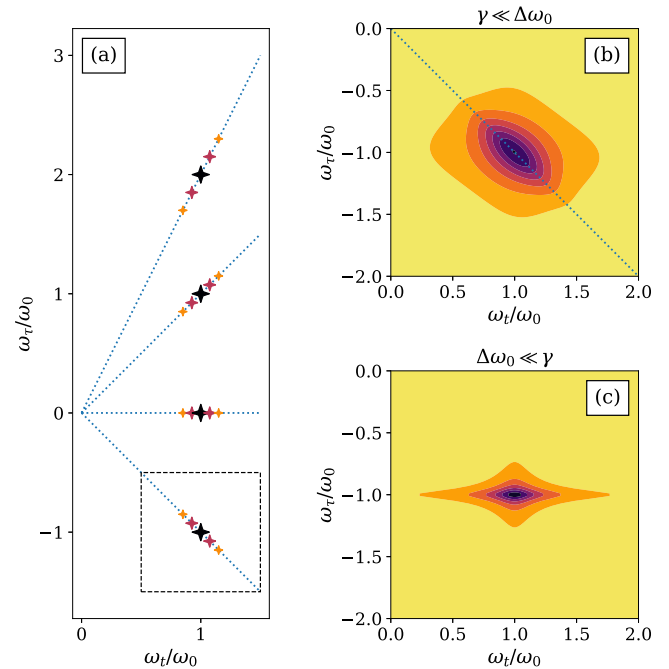


FIG. 14. A sketch showing the effect of inhomogeneous broadening. (a) A collection of noninteracting dipoles (orange stars) with an energy distribution centered around ω_0 produces a sliding of the four peaks foreseen in Fig. 6 along specific lines (blue dotted lines), with an envelope sensitive to the width of the distribution $\Delta\omega_0$. (b) In the case where the intrinsic mode damping γ is small with respect to $\Delta\omega_0$, an elongation of the rephasing peak along $\omega_\tau = -\omega_t$ is observed. (c) In the opposite limit $\Delta\omega_0 \ll \gamma$, the mode has a starlike shape [96].

much of this phenomenology, that can be easily understood within a single-mode picture, survives and can be observed in dispersive electronic systems and in realistic situations where also propagation effects must be taken into account. A somehow similar signature is indeed already present in our noninteracting electronic model considered in Sec. III. If we examine the difference between the nondispersive and dispersive limits of the paramagnetic process in Figs. 8(c) and 8(f), we see that the tensorial density of states ρ^{para} in Eq. (42) acts as an effective inhomogeneous broadening of the 2Δ resonance in momentum space, producing an elongation of the various peaks related to the finite bandwidth set by $\bar{\Gamma}$. In particular, the rephasing peak acquires an elongation in the antidiagonal direction analogous to the one of Fig. 14(b).

The renewed interest in this problem has been triggered by the 2DTS measurements of the Josephson plasmon discussed in Sec IV: indeed, the measured spectra show an elongation of the nonrephasing peak at $(\omega_J, -\omega_J)$ in the diagonal direction, which is not reproduced by the simulations of Fig. 12. At the level of the formalism of the present manuscript, one could easily account for this effect by phenomenologically mediating the 2DTS spectra for a single realization of the ω_J value over a Gaussian distribution of possible ω_J values with variance $\Delta\omega_J$. However, such a procedure completely misses the answer to a rather profound question: how a microscopic distribution of local plasma values ω_J impacts on the macro-

scopic propagation of electromagnetic waves in the medium, leading to signatures in the linear and nonlinear response. Valuable attempts have already been made to answer this question for the linear [88,89] and nonlinear [48] responses. Still, a clear understanding of the additional effects due to propagation in a dipolar inhomogeneous system is lacking, leaving open the question of the real potential of 2DTS to distinguish spectroscopically intrinsic mode lifetimes from emergent space inhomogeneities.

A related open question concerns the recent proposal [39,40] concerning the ability of 2DTS to identify energy-relaxation channels, absent in the linear optical response, which is only sensitive to momentum-relaxation processes. So far, the persistence of such slow-relaxation processes has been reported for a variety of conventional and unconventional metals, but a clear formulation of the problem in terms of microscopic processes relevant for this specific nonlinear optical effect is still in its infancy [97]. The results of the present manuscript represent a step in this direction, offering a route to bridge the experimental results based on multidimensional terahertz spectroscopy to the theoretical investigation of interaction processes not accessible in linear response, with the final goal of understanding the microscopic mechanisms responsible for the observed physical effects.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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