

Two-dimensional nonlinear optical response of a spiral magnet

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We study the dynamical response function relevant for two-dimensional coherent nonlinear optical spectroscopy of the antiferromagnetic frustrated J_1 - J_3 Heisenberg model on the square lattice within its long-range-ordered, incommensurate diagonal spiral phase. We argue that in this phase effective dipole coupling to the electric field is important, with the spin-current coupling potentially being the dominant mechanism for spin $1/2$. For this setting, we use linear spin wave theory to evaluate the leading nonlinear polarization response which is of second order in the driving field. We show that the response function features a strong antidiagonal, galvanoelectric feature. The width of this feature is set by relaxation rates beyond the noninteracting magnon picture, thereby providing access to single-magnon lifetimes within the multimagnon continuum of the response function. Moreover, the response function is shown to display various structures in the two-dimensional frequency plane related to exceptional regions of the magnon dispersion.

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I. INTRODUCTION

Two-dimensional (2D) nonlinear coherent spectroscopy (CS) [1–3] has recently experienced an upsurge of interest as a dynamical probe of low-dimensional quantum magnets. This interest has been triggered in particular by the seminal suggestion that properties of single quasiparticles, which result from spin fractionalization, should be accessible by 2DCS [4]. This approach is different from the standard analysis of fractionalized spin dynamics by inelastic neutron scattering, where ubiquitous multiparticle continua mask the single-particle properties [5,6]. In turn a significant number of theoretical studies of nonlinear optical spectroscopy in quantum magnets, prone to fractionalization, have emerged. This pertains to spinons in quasi-one-dimensional quasi-(1D) spin-chain systems [7–14], as well as Majorana fermions and visons in Kitaev magnets [15–21] and fractons in X-cube and Haah code models [22].

In contrast to spin systems with a sought-for fractionalization, quantum magnets with long-range order (LRO) resulting from broken spin-rotational invariance and exhibiting magnon excitations have received much less attention regarding 2DCS. Existing early analysis of canted antiferromagnets [23] focused on 2D nonlinear electron spin resonance, driven by Zeeman coupling to external *magnetic* fields. In contrast, coupling magnets to *electric* fields, i.e., by the magnetoelectric effect (MEE) [24], is another important option [25–28].

While the MEE is of prime interest in the context of static phenomena, electromagnon excitations [29,30] show that dynamics is of equal importance [31] and may lead to observable consequences in optical probing [32–34].

In turn, the motivation for this work is to pursue a scenario in which the 2D nonlinear response of a quantum magnet is driven primarily by coupling to electric fields through the MEE. Moreover, we seek a situation in which only a single microscopic mechanism is relevant for the MEE. For that, we lay out the favorable conditions as follows: First, we require magnetic LRO. Then, the elementary excitations are magnons, which implies that any nonlinear dynamical response, driven by Zeeman coupling of the magnetization to magnetic fields, is necessarily an effect involving magnon-magnon interactions. We assume they are of subleading order and therefore drop coupling to magnetic fields. Second, we consider the most prominent microscopic mechanisms for MEE coupling more closely [26]. To begin, there is spin-dependent hybridization [35]. In this work, we will assume a spin- $1/2$ system for which this mechanism is strictly zero [36]. Next, we focus on the exchange-striction coupling [37]. This requires some sort of inversion symmetry breaking and is a relevant option for quasi-1D systems. In this work, however, we will stick with planar square lattices, for which inversion symmetry breaking is less likely. In turn we also dismiss exchange-striction coupling. Somewhat related to exchange striction are dynamic couplings to electric fields involving phonon assistance [38]. They require favorable optical vibrational modes which we waive for the present work. This leaves the spin-current coupling or inverse Dzyaloshinskii-Moriya interaction, introduced by Katsura, Nagaosa, and Balatsky (KNB) [39]. Quite generally, the KNB mechanism allows for a MEE, both in quasi-1D situations with chiral or spiral correlations (e.g., [40] and Refs. therein) and in systems with noncollinear LRO [41,42] in higher dimension. While for any specific material realization a nonzero coupling constant for

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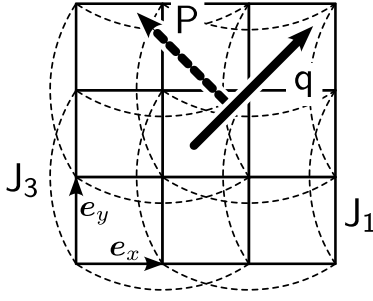


FIG. 1. Frustrated AFM J_1 - J_3 model on a square lattice including the ICS pitch vector $\mathbf{q}$ and classical KNB polarization $\mathbf{P}$, Eq. (14).

the KNB mechanism remains a question to be clarified, we focus on this form of light-matter coupling.

Summing up, we will consider a 2D quantum magnet which allows for spiral LRO. For that purpose we select the frustrated J_1 - J_3 antiferromagnetic (AFM) Heisenberg model on a square lattice (HSL). We subject it to a time-dependent electric field which couples to the spin system via the KNB mechanism. For this setting, we will evaluate the leading order 2D nonlinear response function (NRF). We analyze the spectral properties of the 2D NRF and show in particular that it displays a giant galvanoelectric effect, that it allows us to extract magnon lifetimes, and that it displays specific signatures of the magnon DOS. This paper is organized as follows: In Sec. II, we detail the model and its excitations, as well as the light-matter coupling. Section III describes the calculation of the 2D NRF. In Sec. IV, we discuss the central features of the 2D NRF. We summarize in Sec. V. The Appendix sketches an alternative calculational approach.

II. MODEL

In this section, we first detail the linear spin wave theory (LSWT) of the J_1 - J_3 HSL. Second, we explain the KNB polarization.

A. Spiral phase of the J_1 - J_3 antiferromagnet

Before starting the theoretical developments, we emphasize that our aim is not to add to the large body of work on the phases of J_1 - (J_2) - J_3 quantum antiferromagnets on a square lattice. This model came under scrutiny early on in the context of cuprate superconductors [43–46] and remains of great interest today (see [47] and references therein). Here, we instead use the J_1 - J_3 model as an established device, which allows us to safely claim the existence of a parameter region hosting an incommensurate spiral (ICS) state with LRO. With that in mind, we describe our LSWT of that ICS state. The Hamiltonian reads

$$H/J_1 = \sum_{\langle lm \rangle} \mathbf{S}_l \cdot \mathbf{S}_m + j \sum_{\langle\langle lm \rangle\rangle} \mathbf{S}_l \cdot \mathbf{S}_m, \quad (1)$$

where $\mathbf{l} = \mathbf{e}_x l_x + \mathbf{e}_y l_y$ and we set the lattice constant $a \equiv 1$, i.e., $\mathbf{e}_{x,y} = (1, 0), (0, 1)$ (see Fig. 1). $\mathbf{S}_l$ are spin operators with $\mathbf{S}_l^2 = S(S+1)$. As usual, S will be kept as a free parameter of the LSWT, although we have $S = 1/2$ in mind. We normalize all energies with respect to J_1 , i.e., $j = J_3/J_1$,

where J_1 (J_3) are nearest- (third-nearest-) neighbor exchange couplings. As for the classical phases of this model [43,45] and keeping $j \geq 0$, for $0 \leq j < 1/4 \equiv j_c$, the ground state is a standard nearest-neighbor AFM. For $j_c < j$, the system acquires an ICS with a fourfold degenerate 2D pitch angle of $\mathbf{q} = (\pm q, \pm q)$, where $q = \arccos[1/(4j)]$. While the analysis of the quantum phases of the more general J_1 - J_2 - J_3 model including a next-nearest-neighbor exchange J_2 has a long-standing history, its most recent status establishes that the ICS remains a robust feature of the model [47], even if quantum fluctuations shift the classical transition point substantially from $j_c = 1/4$ to larger values on the order of $j_c \sim 1$. Based on this, we will accept that ICS order is a valid assumption for the J_1 - J_3 model above a critical value of j and use LSWT to treat the elementary excitations in that regime, even if the value of j_c lacks quantum corrections and therefore is too small in LSWT. Before continuing, we mention that remaining within the J_1 - J_3 plane at $J_2 = 0$ is done to keep the algebra as simple as possible while incorporating the central conceptual ingredients for the 2DCS. These ingredients are the ICS, creating the finite polarization, and allowing for electric-field matter coupling. J_2 is not relevant for generating the ICS, and over a wide range of $0 \leq J_2 \lesssim 1$ it only slightly modifies j_c [47]. Furthermore, J_2 only complicates the magnon dispersion and dipole matrix elements quantitatively and creates more free parameters. That is not of interest to our main findings on the spectral structure of the NRFs.

For all calculations, we choose one out of the four degenerate pitch vectors, i.e., $\mathbf{q} = (q, q)$ (see Fig. 1), and rotate onto a locally ferromagnetic (FM) coordinate frame with spins $\tilde{\mathbf{S}}_l$,

$$\mathbf{S}_l = \begin{bmatrix} 0 & -\sin(\mathbf{q} \cdot \mathbf{l}) & \cos(\mathbf{q} \cdot \mathbf{l}) \\ 0 & \cos(\mathbf{q} \cdot \mathbf{l}) & \sin(\mathbf{q} \cdot \mathbf{l}) \\ -1 & 0 & 0 \end{bmatrix} \tilde{\mathbf{S}}_l. \quad (2)$$

The ICS is obvious, inserting a classical FM state $\tilde{\mathbf{S}}_l = S(0, 0, 1)$, which results in $\mathbf{S}_l = S(\cos(\mathbf{q} \cdot \mathbf{l}), \sin(\mathbf{q} \cdot \mathbf{l}), 0)$, i.e., an in-plane spin spiral with a diagonal pitch of (q, q) . Describing quantum fluctuations beyond this state by Holstein-Primakoff (HP) bosons requires only a *single-site* magnetic unit cell, with

$$\begin{aligned} \tilde{S}_l^z &= S - a_l^\dagger a_l, \\ \tilde{S}_l^+ &= (2S - a_l^\dagger a_l)^{1/2} a_l, \\ \tilde{S}_l^- &= a_l^\dagger (2S - a_l^\dagger a_l)^{1/2}. \end{aligned} \quad (3)$$

Performing the usual expansion of the Hamiltonian to leading $O(1/S)$ in terms of these HP boson, i.e., LSWT, we arrive at

$$H = \sum_k \mathbf{A}_k^\dagger \mathbf{h}_k \mathbf{A}_k + NS(S+1)e_{cl}, \quad (4)$$

where N is the number of sites and $e_{cl} = -[2j + 1/(4j)]$ is the classical energy per site for $S = 1$. $\mathbf{A}_k^\dagger = (a_k^\dagger, a_{-k}^\dagger)$ is a boson spinor, and

$$\mathbf{h}_k = \frac{S}{2} \begin{bmatrix} \mathcal{A}_k & \mathcal{B}_k \\ \mathcal{B}_k & \mathcal{A}_k \end{bmatrix}, \quad (5)$$

$$\begin{aligned} \mathcal{A}_k &= \frac{S}{8j} \{4 + 32j^2 + (8j - 2)[\cos(k_x) \\ &\quad + \cos(k_y)] + \cos(2k_x) + \cos(2k_y)\}, \end{aligned} \quad (6)$$

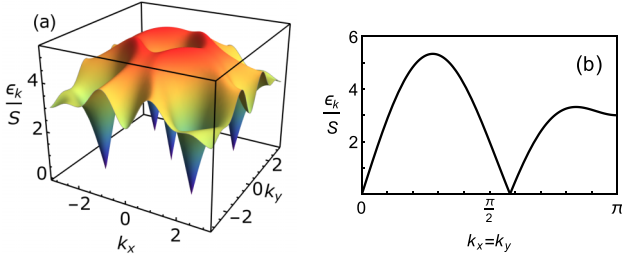


FIG. 2. LSWT magnon dispersion for $j = J_3/J_1 = 1$, normalized to J_1 . (a) The 3D view and (b) 2D cut along the diagonal $k_x = k_y$.

$$B_k = \frac{S}{8j} (4j+1) \{ (4j-1) [\cos(2k_x) + \cos(2k_y)] + 2\cos(k_x) + 2\cos(k_y) \}. \quad (7)$$

All bold spinor operators in this work will also be referenced by their components, using the notation $A_{k\mu}^{(+)}$, with nonbold letters and subscripts $\mu = 1, 2$. The Hamiltonian can be diagonalized by means of a Bogoliubov transformation $A_k = U_k D_k$ onto diagonal bosons $D_k^+ = (d_k^\dagger, d_{-k})$ which create and destroy magnon excitations

$$H = \sum_k \epsilon_k d_k^\dagger d_k + E_0, \quad (8)$$

where

$$U_k = \begin{bmatrix} u_k & v_k \\ v_k & u_k \end{bmatrix}, \quad U_k^T h_k U_k = \frac{1}{2} \begin{bmatrix} \epsilon_k & 0 \\ 0 & \epsilon_k \end{bmatrix}, \quad (9)$$

with

$$u_k = \sqrt{\frac{A_k + \epsilon_k}{2\epsilon_k}}, \quad v_k = -\text{sgn}(B_k) \sqrt{\frac{A_k - \epsilon_k}{2\epsilon_k}}, \quad (10)$$

where ϵ_k is the magnon energy,

$$\begin{aligned} \epsilon_k &= \sqrt{A_k^2 - B_k^2} \\ &= \frac{S}{8j} [4 + 32j^2 + (8j-2)[\cos(k_x) + \cos(k_y)] \\ &\quad + \cos(2k_x) + \cos(2k_y)]^2 - \{(16j^2 - 1) \\ &\quad \times [\cos(2k_x) + \cos(2k_y)] + 2(4j+1) \\ &\quad \times [\cos(k_x) + \cos(k_y)]\}^2]^{1/2}, \end{aligned} \quad (11)$$

and $E_0 = NS(S+1)e_{cl} + \sum_k \epsilon_k/2$, where the second term is the usual zero-point energy of the harmonic Bose gas encoded in Eq. (8). Figure 2 displays the dispersion for a typical value of $j = 1$, which we will retain throughout this work. The diagonal cut in Fig. 2 clearly demonstrates the incommensurate pitch, which is at $q \simeq 0.58\pi$.

Apart from $j = 1$, we mention two special cases explicitly. The first is $j \rightarrow \infty$, where we obtain $\epsilon_k = 4jS\{1 - [\cos(2k_x) + \cos(2k_y)]^2/4\}^{1/2}$. This is exactly the dispersion of AFM magnons in LSWT on decoupled square lattices with unit cell $2a = 2$, which remains effective in that limit. The presence of four interpenetrating of these lattices is taken into account by the Brillouin zone (BZ) still extending over $k_{x,y} = [-\pi, \pi]$. The second case

is $j = j_c = 1/4$, where $\epsilon_k = S\{[\cos(2k_x) + \cos(2k_y) + 6]^2 - 16[\cos(k_x) + \cos(k_y)]^2\}^{1/2}/2$. While it does already have its zeros only at $k_0 = (0, 0)$ and $(\pm\pi, \pm\pi)$, as in the square lattice AFM state for $j < j_c$, the dispersion at these wave vectors directly at the critical point is anisotropically quadratic, with $\epsilon_{k_0+p} \simeq \sqrt{2}S(p_x^4 + p_y^4)^{1/2}$ for $|p| \ll \pi$. An analysis of the critical region is beyond the scope of this work.

B. Minimal coupling from KNB polarization

For the J_1 - J_3 model, the spin-current-induced, or KNB, coupling [39] to an external dynamic electric field $E(t)$ reads

$$H_{\text{KNB}}(t) = -\mathbf{P} \cdot \mathbf{E}(t),$$

$$\mathbf{P}/\gamma = \sum_{\langle lm \rangle} \mathbf{R}_{lm} \times \mathbf{S}_l \times \mathbf{S}_m + g \sum_{\langle\langle lm \rangle\rangle} \mathbf{R}_{lm} \times \mathbf{S}_l \times \mathbf{S}_m, \quad (13)$$

where $\mathbf{P}$ is the electric polarization γ and γg are effective nearest- and next-next-nearest-neighbor coupling constants. For the remainder of this work, like for the unit of energy J_1 , we normalize $\mathbf{P}$ to γ to abbreviate the notation. $\mathbf{R}_{lm}$ are the nearest- and next-next-nearest-neighbor lattice vectors $(1, 0)$, $(0, 1)$ and $(2, 0)$, $(0, 2)$.

The classical KNB polarization $\mathbf{P}_{cl}$ results from inserting the classical ICS into Eq. (13). Since the spiral is planar, and the pitch is diagonal, the classical spin cross products in Eq. (13) are perpendicular to the lattice plane and are identical for the x and y directions within each of the sums of Eq. (13). Therefore, the triple products including the lattice vectors along x and y lie in the lattice plane, and their total is perpendicular to the diagonal, i.e., the pitch. We get

$$\mathbf{P}_{cl} = S^2 \frac{(g-j)\sqrt{16j^2-1}}{4j^2} (-1, 1, 0). \quad (14)$$

This is depicted in Fig. 1. In principle, from Eq. (14), $\mathbf{P}_{cl} = 0$ if $j = g$. However, such a fine-tuned situation is unphysical in view of the different mechanisms for Heisenberg and KNB coupling. For the remainder of this work, we assume $j \neq g$, and to plot NRFs we will set them to be of a similar magnitude. In the commensurate AFM for $j < j_c$, the polarization is zero. Also for $j \rightarrow j_c + 0$ from within the ICS, the polarization turns to zero. In the limit of decoupled square lattices for $j \rightarrow \infty$, setting $g = cj$ with some arbitrary constant $c \neq 1$, $\mathbf{P}_{cl}$ from Eq. (14) approaches a finite value. This does not imply a finite polarization and is merely a consequence of normalizing $\mathbf{P}_{cl}$ to γ . If it is considered on the scale of $g\gamma$, which is proper in that limit, the polarization vanishes, as expected.

Next, we express the quantum corrections to $\mathbf{P}_{cl}$ in terms of HP bosons. For the remainder of this work, we focus on the components in the plane of the classical polarization, i.e., (P_x, P_y) . To begin, we note that in LSWT, apart from the classical contribution, $\mathbf{P}$ will not start at quadratic order in the bosons, but rather will also contain linear terms. Physically, they refer to a direct mixing between photons and magnons. Now, for all purposes of our analysis the wave vector of the light $\mathbf{q}_E$ satisfies $\mathbf{q}_E = \mathbf{0}$. This implies that all processes, possibly related to such linear mixing, need to occur at energies of $\epsilon_{k=\mathbf{q}_E} = 0$. Since we are interested only in finite frequencies of the external electric field, we discard linear boson contributions to $\mathbf{P}$.

Inserting the HP bosons into Eq. (13) and rotating into the locally FM frame is lengthy but straightforward. At quadratic order in $A_k^{(+)}$ we obtain

$$P_x = \sum_k A_k^+ p_{x,k} A_k, \quad (15)$$

$$p_{x,k} = -\frac{S}{2} \frac{\sqrt{16j^2 - 1}}{4j^2} \left(2(j-g) \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} + [j \cos(k_y) - g \cos(2k_y)] \begin{bmatrix} -1 & 1 \\ 1 & -1 \end{bmatrix} \right). \quad (16)$$

For P_y , an identical expression is obtained by substituting $p_{y,k} = -p_{x,k}|_{k_y \rightarrow k_x}$.

We emphasize that Eq. (16) is obtained from an expression which is valid at *any* pitch q after we insert its classical value in the ICS for $j \geq j_c$. Considering the same expression in a state with plain AFM order, i.e., at $q = \pi$ for $j < j_c$, simply yields $P_x = 0$. In other words, electric-field-induced bilinear coupling to magnons occurs only in the ICS.

III. SECOND-ORDER TWO-DIMENSIONAL RESPONSE FUNCTION

Formally, inversion symmetry forces $O(2)$ NRFs to be zero. However, the ICS breaks this symmetry. In turn, the leading order 2D response of the polarization $\langle \Delta P^\parallel \rangle(t)$, projected onto $E_{ac}(t)$, is $O(2)$. It is described by the Fourier transform into the 2D frequency plane of the retarded response function $\tilde{\chi}_2(t, t_1, t_2) = i^2 \Theta(t - t_1) \Theta(t_1 - t_2) \langle [P^\parallel(t), P^\parallel(t_1)] P^\parallel(t_2) \rangle$ [48]. The N -fold time integrations in the perturbative expansion of $\langle \Delta P^\parallel \rangle(t)$ at any order N in $\mathbf{P} \cdot \mathbf{E}_{ac}(t) \equiv P^\parallel E_{ac}(t)$ are totally symmetric with respect to any permutation of the N time arguments [48]. This is called intrinsic permutation symmetry. In turn, any N th-order contribution to $\langle \Delta P^\parallel \rangle(t)$ requires only the fully symmetrized response function $\chi_N(t, t_1, \dots, t_n) = \sum_M \tilde{\chi}_N(t, t_{M(1)}, \dots, t_{M(N)})/N!$ to be evaluated, where M labels all permutations. The Fourier transform of $\chi_N(t, t_1, \dots, t_n)$ can be obtained from analytic continuation to the real axis of the Matsubara frequency transform of the fully connected contractions of the imaginary time propagator $\chi_N(\tau_n, \dots, \tau_1) = \langle T_\tau (P^\parallel(\tau_n) \dots P^\parallel(\tau_1) P^\parallel) \rangle$ [49,50].

Diagrammatic calculations are performed in the diagonal HP boson basis. This involves calculation of contractions from operator groupings of the type

$$\sum_{\dots k_l \dots k_m \dots} \langle T_\tau (\dots D_{k_l}^+(\tau_l) s_{k_l} D_{k_l}(\tau_l) \dots D_{k_m}^+(\tau_m) s_{k_m} D_{k_m}(\tau_m) \dots) \rangle, \quad (17)$$

where $s_k = U_k^T \mathbf{p}_k^\parallel U_k$ is the transform to the diagonal boson representation of the polarization vertex $\mathbf{p}_k^\parallel = (p_{x,k}, p_{y,k}) \cdot \mathbf{e}_E$, projected onto the in-plane electric field. The time ordering allows for normal

$$-\langle T_\tau (D_{k\mu}(\tau) D_{k'\nu}^\dagger) \rangle = \delta_{\mu\nu} \delta_{kk'} G_\mu(\mathbf{k}, \tau), \quad (18)$$

as well as anomalous contractions

$$-\langle T_\tau (D_{k\mu}(\tau) D_{k'\nu}^\dagger) \rangle = -\langle T_\tau (D_{k\mu}(\tau) D_{-k'\bar{\nu}}^\dagger) \rangle = \delta_{\mu\bar{\nu}} \delta_{k,-k'} G_\mu(\mathbf{k}, \tau), \quad (19)$$

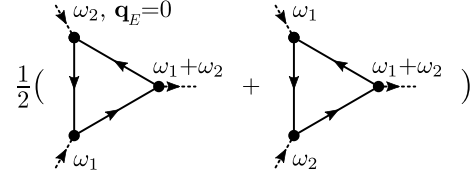


FIG. 3. Diagrams for the 2D NLRF $\chi_2(\omega_1, \omega_2)$ at order $O(2)$ in $E_{ac}(t)$. The solid lines carry an index $\mu = 1, 2$, referring to $G_{1,2}(\mathbf{k}, i\omega_n)$ of Eq. (20); the dots refer to the 2×2 polarization operator matrices $s_{k\mu\nu}$.

where $\mu = 1, 2$ and we define $\bar{\nu} = (\nu + 1 \bmod 2)$. Finally, the adjoint anomalous contractions satisfy $-\langle T_\tau (D_{k\mu}^\dagger(\tau) D_{k'\nu}^\dagger) \rangle = -\langle T_\tau (D_{-k\bar{\mu}}(\tau) D_{k'\bar{\nu}}^\dagger) \rangle = \delta_{\mu\bar{\nu}} \delta_{k,-k'} G_\mu(\mathbf{k}, \tau)$, using $\delta_{\bar{\mu}\nu} = \delta_{\mu\bar{\nu}}$.

Any diagram at $O(n)$ is a closed loop of $n+1$ bilinear vertices, linked by a normal or anomalous contraction. Since the anomalous Green's functions are related to the normal ones by a mere shift of either the right or left matrix indices $\mu \rightarrow \bar{\mu}$ and a flip $k' \rightarrow -k'$ of the sign of the momentum, the necessary contractions can be reduced to only normal ones by relabeling the summation indices appropriately and by using a symmetrized vertex $s_{k\mu\nu} \rightarrow t_{k\mu\nu} = (s_{k\mu\nu} + s_{-k\bar{\nu}\bar{\mu}})/2$. With some algebra, one can show that $s_{-k\bar{\nu}\bar{\mu}} = s_{k\mu\nu}$, simplifying $t_{k\mu\nu} = s_{k\mu\nu}$. Therefore, all diagrams can be generated using the matrix vertices $s_{k\mu\nu}$ and a “single-arrow” normal Green's function $G(\mathbf{k}, i\omega_n)$ of the diagonal matrix shape, with entries

$$G(\mathbf{k}, i\omega_n) = \delta_{\mu\nu} G_\mu(\mathbf{k}, i\omega_n),$$

$$G_1(\mathbf{k}, i\omega_n) = \frac{1}{i\omega_n - \epsilon_k},$$

$$G_2(\mathbf{k}, i\omega_n) = G_1(-\mathbf{k}, -i\omega_n). \quad (20)$$

The diagrams for the $O(2)$ 2D NRF are shown in Fig. 3. Since each single-arrow line symbolizes two Green's functions, i.e., $G_{1(2)}(\mathbf{k}, i\omega_n)$, the diagram is a sum of $2^3 = 8$ expressions, each comprising one internal Matsubara frequency summation. We obtain

$$\chi_2(\omega_1, \omega_2) = \sum_k \left[\frac{8(1+2n_k)\epsilon_k^2 s_{k11} |s_{k12}|^2}{(z_1^2 - 4\epsilon_k^2)(z_2^2 - 4\epsilon_k^2)} \times \frac{(z_1^2 + z_1 z_2 + z_2^2 - 12\epsilon_k^2)}{((z_1 + z_2)^2 - 4\epsilon_k^2)} \right], \quad (21)$$

with the Bose function $n_k = 1/[\exp(\epsilon_k/T) - 1]$ and $z_{1,2} = \omega_{1,2} + i\eta$ complex frequencies, close to the real axis with $\omega_{1,2} \in \text{Re}$ and $0 < \eta \ll 1$. For the model at hand, since U_k and $\mathbf{p}_{x(y),k}$ are both real, we have $|s_{k12}|^2 = s_{k12}^2$. Equation (21) is one of the main results of this work, and it completes the evaluation of $\chi_2(\omega_1, \omega_2)$.

We mention that for the present bare LSWT approach, if we do not consider magnon interactions, $\chi_2(\omega_1, \omega_2)$ can also be evaluated by calculating commutators. The result is given in the Appendix. Including $1/S$ corrections, however, a diagrammatic approach is superior.

IV. DISCUSSION

We begin our discussion by recalling that in principle, due to the Mermin-Wagner-Hohenberg theorem, breaking continuous symmetry in $D \leq 2$ at $T \neq 0$ is not possible. In practice, for $D = 2$, this tends to manifest as weak finite- T logarithmic singularities arising from integration over poles $\sim 1/\epsilon_k^2$ from Goldstone zeros. To cure this, instead of either allowing for only $T = 0$ or including a third dimension in any calculation, a pragmatic remedy is to enforce a phenomenological cutoff ϵ_c , with $\epsilon_k > \epsilon_c > 0$. The energy gap ϵ_c mimics a finite correlation length, i.e., only quasi-long-range order. This procedure borrows from the square lattice Heisenberg antiferromagnet, for which a large body of theoretical and experimental work [51–60] clarified that the finite- T magnon propagation is overdamped beyond a correlation length ξ . In effect, the latter acts as the temperature-dependent size of an order-parameter domain for magnon propagation. It has been shown that this introduces an effective gap $\epsilon_c = v/2\xi$ into the magnon dispersion, where v is a low-energy magnon velocity [61–63]. We note that analyzing the dependence of ϵ_c on temperature and pitch angle in the ICS of our model is beyond the scope of this work, and we simply use some fixed $\epsilon_c \ll \max(\epsilon_k)$.

Implementing the cutoff in the LSWT for the ICS is straightforward. That is, we replace $\mathcal{A}_k \rightarrow \mathcal{A}_k + dS/8j$ in Eq. (6) with a free parameter d . This modifies ϵ_k from Eq. (12) such that the expression $4 + 32j^2$ in the first line is replaced by $d + 4 + 32j^2$. This opens a gap of $O(\sqrt{d})$ at $\mathbf{k} = \mathbf{0}$ and $\mathbf{q}$. The formal expressions for the Bogoliubov transformation remain unchanged, with only $\mathcal{A}_k$ and ϵ_k modified as just described.

Before continuing, we clarify that the preceding discussion is indeed relevant for $\chi_2(\omega_1, \omega_2)$, irrespective of any resonance conditions for $\omega_{1,2}$. That is, we count the powers of $1/\epsilon_k$ from the numerator of Eq. (21). The vertex $s_{k\mu\nu}$ comprises two Bogoliubov transforms (10), implying one factor of $1/\epsilon_k$ per vertex. Moreover, at any *finite* temperature and for $\epsilon_k \ll T$, the Bose function provides an additional factor of $1/\epsilon_k$. Therefore, in the vicinity of the Goldstone zeros, where $\epsilon_k \sim k$, with $k = |\mathbf{k}|$, the numerator of Eq. (21) scales $\sim 1/k$ for $\epsilon_k \gg T$ and $\sim 1/k^2$ for $\epsilon_k \ll T$. That is, at $T \neq 0$ and independent of $\omega_{1,2}$, the 2D momentum integration for $\chi_2(\omega_1, \omega_2)$ will be log singular for $\epsilon_c = 0$.

Next, we compare our results to a recent analysis of 2D NRFs in Kitaev magnets [19]. While these are fractionalized spin systems with Majorana fermion and vison elementary excitations, it is reassuring to realize that the 2D NRF and that in the present work are expressions with identical form, except for the difference in statistics. Therefore, similar to that in Ref. [19], $\chi_2(\omega_1, \omega_2)$ along the so-called rectification or galvanoelectric (GEE) line, $\omega \equiv \omega_1 = -\omega_2$, is anomalously singular. In fact, asymptotically for $\eta \ll 1$, Eq. (21) on the GEE line can be cast as

$$\chi_2(\omega, -\omega) \simeq \frac{\pi}{\eta} \sum_k \{ (1 + 2n_k) s_{11k} \times |s_{12k}|^2 [\delta(\omega + 2\epsilon_k) + \delta(\omega - 2\epsilon_k)] \}. \quad (22)$$

This function is purely real, consisting of a form factor weighted two-magnon density of states, multiplied by a prefactor $\propto 1/\eta$. The latter implies a globally singular NRF on the

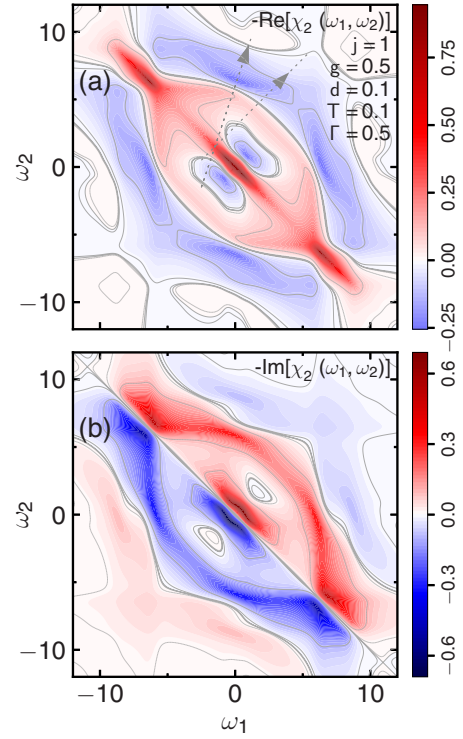


FIG. 4. Contours of (a) the real and (b) imaginary parts of the 2D NRF $\chi_2(\omega_1, \omega_2)$ at intermediate damping. Contour lines added at $\pm(0.1, 0.01, 0.001, 0)$ for better visibility of low-amplitude structures. $S \equiv 1$ to ease notation. Thin dashed lines with arrows refer to cuts in Fig. 6.

GEE line for $\eta \rightarrow 0^+$. This is consistent with similar behavior reported in the context of very different physical questions and for other systems [64–68]. We adopt the rationale of the latter works and replace the causal broadening η with a *physical scattering rate* Γ in order to render $\chi_2(\omega_1, \omega_2)$ finite along the GEE line. Higher-order $1/S$ corrections beyond LSWT, e.g., magnon self-energies, will allow for finite Γ but also extrinsic scattering from, e.g., lattice degrees of freedom. In the following, we keep Γ a free, momentum- and temperature-independent parameter.

Figure 4 displays contours of $\chi_2(\omega_1, \omega_2)$. We set $E_{ac}(t) = e_x E_{ac}(t)$ hereafter; i.e., only P_x is used. For the parameters chosen, $\Gamma/\max(\epsilon_k) \lesssim 0.1$. While this seems to be a realistic and not too small damping rate, it has been set primarily to keep the singular behavior along the GEE line on the same scale as other aspects of the plot. First, the overall dominant characteristic of $\chi_2(\omega_1, \omega_2)$ is its strong antidiagonal amplitude related to Eq. (22). Obviously, this feature is not confined strictly to $\omega_2 = -\omega_1$. In fact, as expected from the prefactor of $1/\Gamma$ in Eq. (22), smoothly continuing $\chi_2(\omega_1, \omega_2)$ into the 2D frequency plane, perpendicular to the GEE line, the prefactor can be viewed asymptotically as the limiting form of some Lorentzian $\sim i/(\omega_\perp + i\Gamma)$ with a frequency variable $\omega_\perp$ perpendicular to the GEE line. This captures both the real part $\sim 1/\Gamma$ and the imaginary part being strictly zero with a sign change across the GEE line, which is clearly visible in Fig. 4. Therefore, as another main result of this work, quasiparticle damping rates can be read off from 2D NRFs perpendicular to the GEE line. We emphasize that while

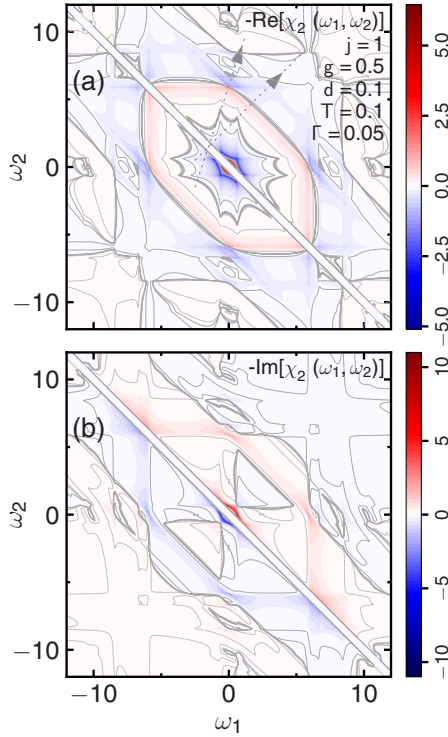


FIG. 5. Contours of (a) the real and (b) imaginary parts of the 2D NRF $\chi_2(\omega_1, \omega_2)$ at small damping. Contour lines added at $\pm(0.1, 0.01, 0.001, 0)$ for better visibility of low-amplitude structures. $S \equiv 1$ to ease notation. Thin dashed lines with arrows refer to cuts in Fig. 6.

we use a constant rate Γ , it is straightforward to extend the discussion of Eq. (22) to assume a momentum dependence Γ_k . In that case, the Lorentzian's FWHM in $\omega_\perp$ perpendicular to the antidiagonal at various locations $\omega = \pm\epsilon_k$ represents a momentum-resolved analysis.

Along the GEE line, Fig. 4 allows us to read off the bandwidth of $2\epsilon_k$. It is ≈ 10.7 for the parameters chosen, and indeed, along the antidiagonal the real part of the NRF approaches zero in that region. The intensity variations visible along the GEE line are a combination of matrix element and dispersion effects. Finally, Fig. 4 shows a strong ringlike pattern involving finite $\omega_{1,2}$. It is related to energies from the region of the BZ corners $\mathbf{k} \sim (\pi, \pi)$ (see Fig. 2). Since their magnitude varies only weakly for sufficiently large j , e.g., $\epsilon_{\pi,\pi} = 4 - 1/j$, this ring pattern remains located roughly within the same frequency range for all $j \gtrsim 1$.

In order to uncover finer details and for completeness, Fig. 5 displays the 2D NRF for a damping Γ which is 1 order of magnitude smaller than in Fig. 4. This implies a huge and very sharp GEE response, suppressing the visibility of all smaller intensities in Fig. 5. For visualization purposes, we therefore mask the NRF in a narrow strip along the GEE line to zero, which moves the NRF's fine structure to the front. First, at this damping, Fig. 5 demonstrates more clearly that the boundaries of the larger ringlike feature are set by the density of states at the energies from the region of the BZ corners $\mathbf{k} \sim (\pi, \pi)$. In fact, for the parameters chosen, $2\epsilon_{\pi,\pi} \approx 6$, which from the energy denominator in Eq. (21) leads to six exceptional frequency

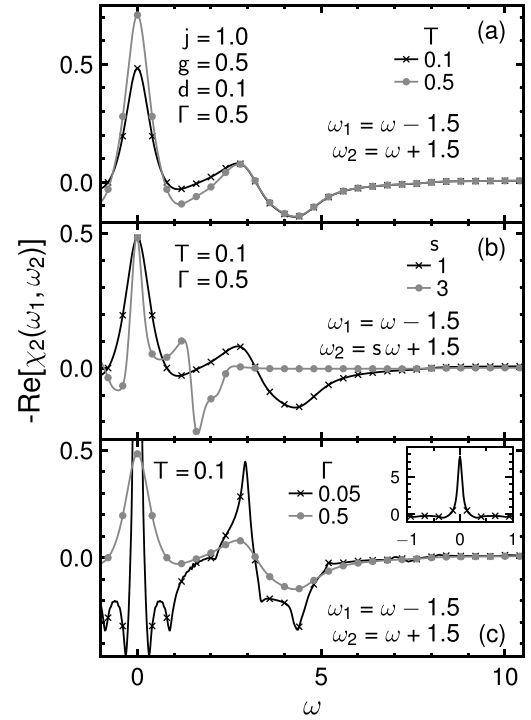


FIG. 6. Cuts of the real part of the 2D NRF $\chi_2(\omega_1, \omega_2)$ along the dashed directions in Figs. 4 and 5 versus two (a) temperatures, (b) directions, and (c) damping rates. $S \equiv 1$ to ease notation.

locations of $(\omega_1, \omega_2) \approx (\pm 6, 0), (0, \pm 6), (\pm 6, \mp 6)$, harboring sign changes of $\text{Re}[\chi_2(\omega_1, \omega_2)]$, which can be observed in Fig. 5 to set the ring feature. An additional location of a far weaker NRF amplitude with a sign change of $\text{Re}[\chi_2(\omega_1, \omega_2)]$ is visible at $(\omega_1, \omega_2) \approx (\pm 6, \pm 6)$. Second, the most intense feature is a small ringlike structure confined by exceptional energies $\omega_{1,2} \sim O(0.6)$. Part of it is masked to zero on the GEE line. These energies result from the correlation gap, primarily from $\mathbf{k} \sim \mathbf{q}$, where $2(\epsilon_q) \approx 0.6$. Finally, no obvious exceptional frequencies can be identified which relate to the maximum of the LSWT dispersion, i.e., $2\max(\epsilon_k) \approx 10.7$, which is clearly within axis limits of $|\omega_{1,2}| \leq 12$ in Fig. 5.

In Fig. 6, various cuts are depicted through the real part of the 2D NRF along the paths shown by dashed gray arrows in Figs. 4 and 5. To begin, these paths have been shifted from the region $\omega_{1,2} \ll 1$, so that they are not dominated by the correlation gap at small ω . Next, Fig. 6(a) shows typical behavior perpendicular to the GEE line at the damping Γ used in Fig. 4. Very clearly, the plot displays a quasi-Lorentzian with a FWHM $\sim \Gamma$ at $\omega = 0$. This was anticipated in the discussion of Eq. (22) and Fig. 4 and may potentially allow us to extract Γ from experiment. Moreover, Fig. 6(a) shows the impact of the Bose statistics of the magnons. That is, with increasing temperature, the low-energy amplitudes increase; however, the high-energy structure remains unchanged. Figure 6(b) shows the dependence of $\text{Re}[\chi_2(\omega_1, \omega_2)]$ on different cut directions and, in particular, the abrupt sign changes at exceptional frequencies in the context of the ring structures discussed for Figs. 4 and 5. That is, for the path with $(\omega_1, \omega_2) = (\omega - 1.5, 3\omega + 1.5)$, the exceptional point $(\omega_1, \omega_2) \approx (0, 6)$ is

reached at $\omega \approx 1.5$, where, indeed, Fig. 6(b) displays a rapid sign change of $\text{Re}[\chi_2(\omega_1, \omega_2)]$. The latter can be increased by decreasing Γ . Finally, Fig. 6(c) is primarily intended to show the scaling of the NRF amplitude with Γ close, but perpendicular, to the GEE line. For that purpose, the same values of Γ as used in Figs. 4 and 5 are chosen. For $\Gamma = 0.05$, we cut off $\text{Re}[\chi_2(\omega_1, \omega_2)]$ at low ω and redisplay it in the inset. *Cum grano salis*, the ratio of the peak intensities of $\text{Re}[\chi_2(\omega_1, \omega_2)]$ on the GEE line for $\Gamma_{\times}/\Gamma_{\bullet} = 0.1$ is approximately 10, in agreement with Eq. (22). The same can be read off for the ratio of the FWHMs. Apart from that, Fig. 6(c) obviously shows that $\Gamma = 0.5$ already leads to a significant smearing of fine structure of the NRF.

Concluding this discussion, for completeness, we mention that apart from the one-magnon self-energies, which contribute to Γ , another potential correction beyond LSWT could be vertex corrections in Fig. 3. While no feasible approach has been documented to treat such corrections for dynamical three-point correlation functions, their physics amounts to an effective light-matter coupling; i.e., they modify the dipole matrix elements. Perturbatively, the main conclusions of our work, the antidiagonal intensity and a 2D continuum background, should remain robust with respect to such modifications.

V. SUMMARY

To recap, we argued that spiral magnets are a promising test bed for coherent nonlinear optical spectroscopy. In such magnets, spin-current coupling can provide a dominant effective dipole moment which allows for minimal coupling to electric fields and generates a nonlinear response already at second order in the driving field. We detailed the consequences of this argument for the spiral phase of the square lattice J_1 - J_3 Heisenberg antiferromagnet using linear spin wave theory and evaluating the second-order response function. Apart from a rich landscape in the 2D frequency plane, we found the prime feature of this response function is a dominant antidiagonal structure which allows us to read off quasiparticle lifetimes. This may be of interest for experiments.

Speculating on potential candidate materials, we are unaware of a direct realization of the model we analyze. However, several type-II multiferroics exist which harbor similar physics, namely, spiral spin states or spiral correlations and KNB coupling. Among them are quasi-one-dimensional systems, e.g., MnWO_4 [69], LiCu_2O_2 [70], and LiCuVO_4 [71]; triangular delafossites, e.g., AgCrO_2 [72]; spinels, such as CoCr_2O_4 [73]; and several manganites [74]. Linear coupling to electric fields, i.e., electromagnons, have already been observed in some of them. Additionally, very recently, type-II multiferroicity was discovered down to the ultrathin limit in the van der Waals compound NiI_2 [75–78]. *Ab initio* modeling suggests this system has layer-number-dependent spiral phases, which, remarkably, result from third-nearest-neighbor spin frustration, and it displays in-plane polarization from KNB coupling [79], similar to that in the present work. NiI_2 shows second harmonics generation [75,76], i.e., a nonlinear response. Finally, spiral magnets induced by structure [80] might be of interest for 2DCS studies if, quantum chemically, they allow for sizable KNB coupling.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [81].

APPENDIX: COMMUTATOR APPROACH

It has become customary in part of the literature to discuss the 2D Fourier transform of the retarded response function $\tilde{\chi}_2(t, t_1, t_2) = i^2 \Theta(t - t_1) \Theta(t_1 - t_2) ([P^\parallel(t), P^\parallel(t_1)], P^\parallel(t_2))$ directly, discarding intrinsic permutation symmetry. As long as $P^\parallel(t)$ can be expressed in terms of known eigenstates and energies, e.g., for free quasiparticles as in the present case of noninteracting LSWT, calculating the commutators is an alternative to calculating the diagrams in Fig. 3. However, in the case of nondiagonal quasiparticle interactions, approximate approaches to evaluating the commutator correlation function are conceptually unclear, rendering the diagrammatic approach superior in principle at least.

For completeness, we list the result for $\tilde{\chi}_2(\omega_1, \omega_2)$ that we find from the commutator expression using the time-dependent $P^\parallel(t)$ of the diagonal boson representation of the LSWT:

$$\tilde{\chi}_2(\omega_1, \omega_2) = \sum_k \left[\frac{16(1+2n_k)\epsilon_k^2 s_{k11} |s_{k12}|^2}{(z_1 + z_2)(z_2^2 - 4\epsilon_k^2)} \times \frac{(z_1 + 2z_2)}{[(z_1 + z_2)^2 - 4\epsilon_k^2]} \right]. \quad (\text{A1})$$

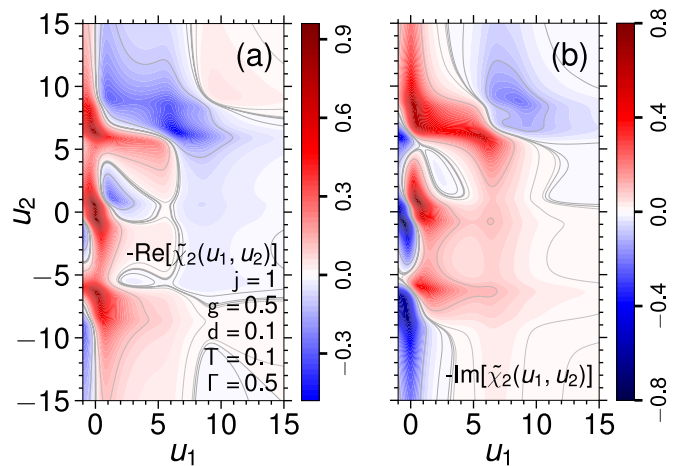


FIG. 7. Contours of (a) the real and (b) imaginary parts of the commutator correlation function $\tilde{\chi}_2(u_1, u_2)$ at intermediate damping. Contour lines added at $\pm(0.1, 0.01, 0.001, 0)$ for better visibility of low-amplitude structures. $S \equiv 1$ to ease notation.

Indeed, most importantly, the condition of $[\tilde{\chi}_2(\omega_1, \omega_2) + \tilde{\chi}_2(\omega_2, \omega_1)]/2 = \chi_2(\omega_1, \omega_2)$ being identical to Eq. (21) is satisfied.

Apart from considering $\tilde{\chi}_2$ instead of χ_2 , it has also become customary to Fourier transform the latter with respect to a different set of time coordinates, i.e., $t' + \tau = t - t_2$ and $\tau = t_1 - t_2$. This is motivated by a two- δ -function pulse sequence, separated by time τ and followed by time t' up to a measurement. This changes the frequency variables to $u_1 = \omega_1 + \omega_2$

and $u_2 = \omega_2$. While we feel that contours of $\tilde{\chi}_2(u_1, u_2)$ are less intuitive to interpret, we also display them in Fig. 7, using the parameters of Fig. 4. Typically, only the first and fourth quadrants in the u_1, u_2 plane are displayed. We follow this habit, but keep a small portion of the negative u_1 axis. The latter is to clarify that in $\tilde{\chi}_2(u_1, u_2)$ a strong GEE intensity is also observed but is located along the u_2 axis at $u_1 = 0$. Other central features can be “rediscovered” by comparing Figs. 4 and 7 side by side.

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