

Nonlinear spectroscopy as a magnon breakdown diagnosis and its efficient simulation

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Identifying quantum spin liquids, magnon breakdown, or fractionalized excitations in quantum magnets is an ongoing challenge due to the ambiguity in interpreting excitation continua occurring in linear-response probes. Recently, it was proposed that techniques measuring higher-order response, such as two-dimensional coherent spectroscopy (2DCS), could resolve such ambiguities. Numerically simulating nonlinear response functions can, however, be computationally very demanding. We present an efficient Lanczos-based method to compute second-order susceptibilities $\chi^2(\omega_l, \omega_r)$ directly in the frequency domain. Applying this to extended Kitaev models describing α -RuCl₃, we find qualitatively different nonlinear responses between intermediate magnetic field strengths and the high-field regime. To put these results into context, we derive the general 2DCS response of partially polarized magnets within the linear spin-wave approximation, establishing that $\chi^2(\omega_l, \omega_r)$ is restricted to a distinct universal form if the excitations are conventional magnons. Deviations from this form, as predicted in our Lanczos-based simulations for α -RuCl₃, can hence serve in 2DCS experiments as direct criteria to determine whether an observed excitation continuum is of conventional two-magnon type or of different nature.

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Introduction. Nonlinear optics probes such as two-dimensional coherent spectroscopy (2DCS) [1] have wide applications in molecular chemistry [2,3], nanomaterials [4], and semiconductors [5,6]. In 2DCS, the time delays between two external field pulses and measurement are varied [Fig. 1(a)], which allows the investigation of higher-order susceptibilities. Recently, 2DCS has gained much attention in the field of frustrated magnetism [7–9] as a possibly powerful tool for distinguishing quantum spin liquids (QSLs) and other exotic states [10–12]. QSLs are generally characterized by absence of magnetic order and the presence of long-range entanglement and fractionalized excitations [13,14]. However, detecting and identifying them experimentally remains challenging due to the lack of smoking gun signatures. Moreover, most of their thermodynamic quantities are quite featureless [15]. Some studies have investigated low-energy fractionalized excitations in QSL candidates by transport measurements, the most prominent being a plateau in thermal Hall measurements in the Kitaev QSL candidate α -RuCl₃ [16,17], although such observations are still under debate [18–21]. In addition to thermal transport, especially the observations of excitation continua in linear-response experiments, have been taken as evidence for QSL behaviors [22–24]. However, such continua are difficult to distinguish from continua that can arise, for instance, from two-magnon states or static disorder [25].

2DCS in the terahertz frequency regime, on the contrary, promises to differentiate between different origins for scattering continua. Reference [10] demonstrated this by analytically investigating the exactly solvable transverse-field Ising chain model, for which higher-order susceptibilities distinctly differentiate between cases of dissipationless spinon excitations, spinon decay, and disorder. Further analytical studies were performed on different models [26,27], including on the Kitaev honeycomb model [8,28–31], where the possibility to probe fractionalized excitations in high-harmonic generation probes was shown. Recently, there have also been first numerical studies [32–37] of higher-order susceptibilities in quantum magnets, including matrix-product state [32–34] and exact diagonalization (ED) studies [35,37]. In these studies, nonlinear responses were simulated first in the time domain by explicit discretized time evolution, while the final results in the frequency domain were obtained by Fourier transforms of the two-dimensional time axes.

Such simulations can in principle follow the actual 2DCS measurement protocol [6,10,38]: As displayed in Fig. 1(a), two pulses with magnetic field along α at $t' = 0$ and along β at $t' = \tau$ are applied, and the magnetization along γ is measured at $t' = t + \tau$. Given the amplitudes of the two field pulses, B_0^α and B_τ^β , the time-dependent *nonlinear* magnetization along γ , $M_{\text{NL}}^\gamma = M_{B_0, B_\tau}^\gamma - M_{B_0}^\gamma - M_{B_\tau}^\gamma + M^\gamma$, corresponds to

$$M_{\text{NL}}^\gamma(t, \tau) = \chi_{\gamma\beta\alpha}^2(t, \tau + t) B_\tau^\beta B_0^\alpha + \mathcal{O}(B_t^3), \quad (1)$$

giving access to the leading-order nonlinear susceptibility $\chi_{\gamma\beta\alpha}^2(t, \tau + t)$ and higher-order susceptibilities contained in $\mathcal{O}(B_t^3)$. Explicitly simulating this time-dependent experiment is, however, rather computationally involved.

In this work, we introduce an alternative approach to calculate higher-order response functions efficiently in an ED

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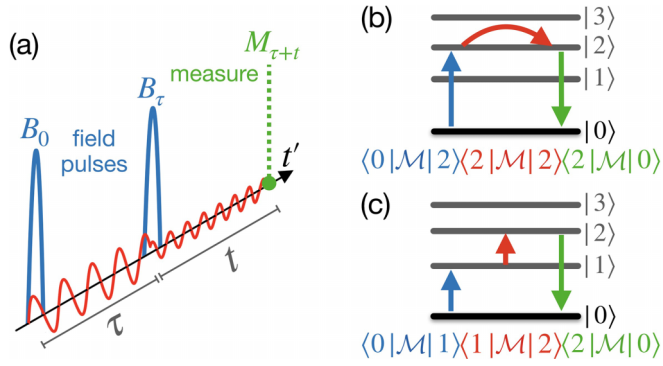


FIG. 1. (a) Sketch of a 2DCS measurement protocol. Two light pulses are applied with a time difference τ , and the measurement is performed at $t' = \tau + t$ with respect to the first pulse. (b), (c) Types of matrix elements contributing to zero-temperature second-order susceptibility $\chi_{\mathcal{M}\mathcal{M}\mathcal{M}}^2$.

framework, operating directly in frequency domain. We successfully benchmark the method with known results for the transverse-field Ising model. We then apply this method to extended Kitaev models under magnetic field, relevant to α -RuCl₃, and focus on the polarization channel “ $\chi_{\parallel}^2$ ”, whose corresponding linear-response ($\chi_{\parallel}^1$) features an excitation continuum [24]. Here, qualitatively different $\chi_{\parallel}^2$ responses are found between the intermediate-field and high-field regimes, corresponding to regimes where conventional magnons break down and are restored, respectively. We substantiate this analysis by showing that $\chi_{\parallel}^2$ is restricted to a distinct universal form for field-polarized magnets on the level of the linear spin-wave approximation. Deviations from this form, as predicted for α -RuCl₃ at intermediate-field strengths, can hence be used in 2DCS experiments as direct evidence for a breakdown of the conventional magnon picture.

Numerical method. We focus on the zero-temperature second-order susceptibility:

$$\begin{aligned} \chi_{ABC}^2(t, \tau + t) &= i^2 \Theta(t) \Theta(\tau) \langle [[\mathcal{A}(\tau + t), \mathcal{B}(\tau)], \mathcal{C}(0)] \rangle \\ &= -2\Theta(t) \Theta(\tau) \text{Re}[\langle \mathcal{A}e^{-i\mathcal{H}t} \mathcal{B}e^{-i\mathcal{H}\tau} \mathcal{C} \rangle \\ &\quad - \langle \mathcal{B}e^{i\mathcal{H}t} \mathcal{A}e^{-i\mathcal{H}(\tau+t)} \mathcal{C} \rangle], \end{aligned} \quad (2)$$

where without loss of generality the spectrum of $\mathcal{H}$ was shifted such that the ground state energy is $E_0 = 0$. $\mathcal{A}, \mathcal{B}, \mathcal{C}$ are operators of choice; examples are magnetization components along particular directions (relevant for terahertz 2DCS) or couplings to electrical polarization [28–30,39].

To efficiently calculate matrix elements of the form $\langle 0 | \mathcal{A}e^{-i\mathcal{H}t} \mathcal{B}e^{-i\mathcal{H}\tau} \mathcal{C} | 0 \rangle$ appearing in Eq. (2) ($|0\rangle$ being the exact ground state) using Lanczos routines, we first define the startvectors

$$|\phi_0^A\rangle, |\phi_0^B\rangle, |\phi_0^C\rangle, \text{ where } |\phi_0^O\rangle = \frac{\mathcal{O}|0\rangle}{\sqrt{\langle 0 | \mathcal{O}^\dagger \mathcal{O} | 0 \rangle}} = \frac{\mathcal{O}|0\rangle}{N_0^O}. \quad (3)$$

For each distinct one of these, a standard Lanczos routine [40] will generate a basis for the L -dimensional Krylov subspace:

$$\text{span}(\{|0\rangle, \mathcal{H}|0\rangle, \mathcal{H}^2|0\rangle, \dots, \mathcal{H}^{L-1}|0\rangle\}). \quad (4)$$

Fixing notation, we name the d -dimensional [$d = \dim(\mathcal{H})$] orthonormal basis vectors generated during the Lanczos routines as $|\phi_m^O\rangle$. Diagonalization of the tridiagonal matrix yields eigenvalues ϵ_m^O and L -dimensional eigenvectors v_m^O . The latter represent the d -dimensional vectors $|\psi_m^O\rangle = \sum_{l=0}^{L-1} v_{m,l}^O |\phi_l^O\rangle$ of the full Hilbert space.

Rewriting the first matrix element in Eq. (2) as

$$\begin{aligned} \langle 0 | \mathcal{A}e^{-i\mathcal{H}t} \mathcal{B}e^{-i\mathcal{H}\tau} \mathcal{C} | 0 \rangle \\ = N_0^A N_0^C \sum_{a,b=0}^{\infty} \frac{(-it)^a (-i\tau)^b}{a!b!} \langle \phi_0^A | \mathcal{H}^a \mathcal{B} \mathcal{H}^b | \phi_0^C \rangle, \end{aligned} \quad (5)$$

we emphasize that powers of $a, b < L$ in $(\langle \phi_0^A | \mathcal{H}^a)$ and $(\mathcal{H}^b | \phi_0^C)$ can be exactly reproduced within the respective Krylov subspaces [Eq. (4)]. Utilizing this, we insert projectors into the subspaces, $\mathcal{P}^O = \sum_{n=0}^{L-1} |\psi_n^O\rangle \langle \psi_n^O|$:

$$\begin{aligned} \langle \phi_0^A | \mathcal{H}^a \mathcal{B} \mathcal{H}^b | \phi_0^C \rangle &= \langle \phi_0^A | \mathcal{P}^A \mathcal{H}^a \mathcal{P}^A \mathcal{B} \mathcal{P}^C \mathcal{H}^b \mathcal{P}^C | \phi_0^C \rangle \\ &= \sum_{n,p=0}^{L-1} v_{n,0}^A v_{p,0}^{C*} (\epsilon_n^A)^a (\epsilon_p^C)^b \langle \psi_n^A | \mathcal{B} | \psi_p^C \rangle, \end{aligned} \quad (6)$$

valid for $a, b < L$, where we used that matrix elements of $\mathcal{H}$ within a Krylov space obey $\langle \psi_n^O | \mathcal{H}^a | \psi_m^O \rangle = \delta_{nm} (\epsilon_n^O)^a$ exactly for $a < L$, even when $|\psi_m^O\rangle$ are not converged to eigenvectors of $\mathcal{H}$ yet.

The approximation of the method is to insert Eq. (6) into Eq. (5) also for terms with $a, b \geq L$, i.e., $\langle \psi_n^O | \mathcal{H}^a | \psi_m^O \rangle \approx \delta_{nm} (\epsilon_n^O)^a$ for $a \geq L$. This insertion yields

$$\begin{aligned} \langle 0 | \mathcal{A}e^{-i\mathcal{H}t} \mathcal{B}e^{-i\mathcal{H}\tau} \mathcal{C} | 0 \rangle \\ \approx N_0^A N_0^C \sum_{n,m=0}^{L-1} v_{n,0}^A v_{m,0}^{C*} e^{-i(t\epsilon_n^A + \tau\epsilon_m^C)} \langle \psi_n^A | \mathcal{B} | \psi_m^C \rangle, \end{aligned} \quad (7)$$

which introduced errors at the orders t^L and τ^L . Inserting Eq. (7) into Eq. (2) [and the analog of Eq. (7) for the second matrix element in Eq. (2)], and moving to the frequency domain, we arrive at

$$\begin{aligned} \chi_{ABC}^2(\omega_t, \omega_\tau) &= \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} dt d\tau \chi_{ABC}^{(2)}(t, \tau + t) e^{i\omega_t^+ t + i\omega_\tau^+ \tau} \\ &\approx \sum_{n,m=0}^{L-1} \frac{X_{n,m}}{(\epsilon_n^{A'} - \omega_t^+)(\epsilon_m^{C'} - \omega_\tau^+)} \\ &\quad + \frac{X_{n,m}^*}{(\epsilon_n^{A'} + \omega_t^+)(\epsilon_m^{C'} + \omega_\tau^+)} \\ &\quad - \frac{Y_{n,m}}{(\epsilon_m^{C'} - \epsilon_n^{B'} - \omega_t^+)(\epsilon_m^{C'} - \omega_\tau^+)} \\ &\quad - \frac{Y_{n,m}^*}{(\epsilon_m^{C'} - \epsilon_n^{B'} + \omega_t^+)(\epsilon_m^{C'} + \omega_\tau^+)}, \end{aligned} \quad (8)$$

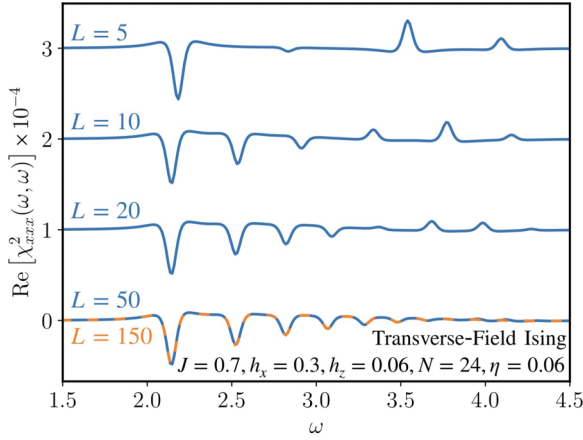


FIG. 2. Comparison of $\chi_{xxx}^2(\omega_t = \omega, \omega_\tau = \omega)$ for different employed Krylov space dimensions L for the transverse-field Ising model $\mathcal{H} = \sum_i^N -J\sigma_i^z\sigma_{i+1}^z - h_x\sigma_i^x - h_z\sigma_i^z$ with broadening $\eta = 0.06$ on $N = 24$ sites. For Eq. (2), the subscript of χ_{xxx}^2 denotes $\mathcal{A} = \mathcal{B} = \mathcal{C} = \sum_i \sigma_i^x$. Results for different L are offset by a constant, except $L = 150$ (shown dashed).

where $\omega_t^+ = \omega_t + i\eta$ and $\omega_\tau^+ = \omega_\tau + i\eta$ with a broadening $\eta > 0$, $\epsilon_a^{\mathcal{O}'} = \epsilon_a^{\mathcal{O}} - E_0$, and

$$X_{n,m} = N_0^{\mathcal{A}} N_0^{\mathcal{C}} v_{n,0}^{\mathcal{A}} v_{m,0}^{\mathcal{C}*} \sum_{l,p=0}^{L-1} v_{n,l}^{\mathcal{A}*} v_{m,p}^{\mathcal{C}} \langle \phi_l^{\mathcal{A}} | \mathcal{B} | \phi_p^{\mathcal{C}} \rangle, \quad (9)$$

$$Y_{n,m} = N_0^{\mathcal{B}} N_0^{\mathcal{C}} v_{n,0}^{\mathcal{B}} v_{m,0}^{\mathcal{C}*} \sum_{l,p=0}^{L-1} v_{n,l}^{\mathcal{B}*} v_{m,p}^{\mathcal{C}} \langle \phi_l^{\mathcal{B}} | \mathcal{A} | \phi_p^{\mathcal{C}} \rangle. \quad (10)$$

The presented method becomes exact when $L = \dim(\mathcal{H})$, where $\epsilon_n^{\mathcal{O}}, |\psi_n^{\mathcal{O}}\rangle$ become the exact eigenpairs of $\mathcal{H}$. In practice, we expect $L \ll \dim(\mathcal{H})$ to suffice for well-converged results of χ^2 in most systems, much before the excited Lanczos eigenstates $|\psi_n^{\mathcal{O}}\rangle$ converge to eigenstates of $\mathcal{H}$ (see the Supplemental Material [41]). This can be understood intuitively from the fact that the method captures all contributions to χ^2 up to the L th order in t, τ exactly [cf. Eq. (5)] (and further orders approximately).

The method becomes computationally cheaper if some of $\mathcal{A}, \mathcal{B}, \mathcal{C}$ are equal to another. When $\mathcal{A} = \mathcal{B} = \mathcal{C}$, likely the most applied case, only a single Lanczos routine needs to be performed. An algorithm implementing that case is described in the Supplemental Material [41].

Benchmarking. We benchmarked the method against Ref. [35] on the transverse-field Ising model, which performed explicit time evolutions on two-dimensional time grids with subsequent Fourier transformation. At sufficient Krylov space dimension L , we find excellent agreement with Ref. [35] throughout the two-dimensional frequency plane [41]. Results for the frequency-plane diagonal ($\omega_t = \omega_\tau$), which hosts the most dominant intensity features for this model, are shown in Fig. 2 for different values of employed L , demonstrating the convergence behavior. In Fig. 2, the strongest features are found to set in already for very small $L \sim 5$ and $L \sim 10$. Satisfactory convergence sets in at circa $L \sim 50$, with no significant change compared to $L = 150$ (Fig. 2). Similar convergence behavior was observed for the results discussed below. For the

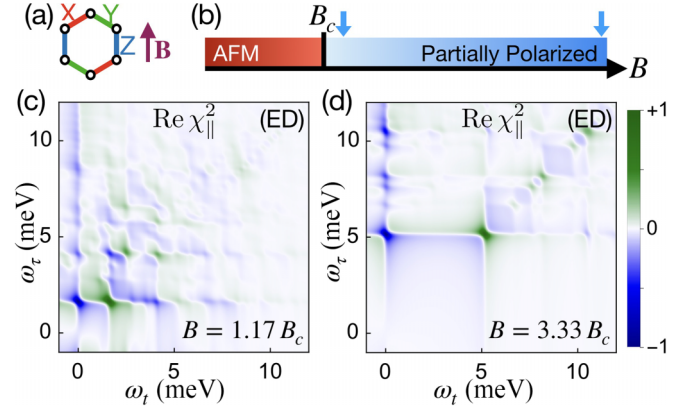


FIG. 3. (a) Bond definitions and chosen field direction $\mathbf{B}$. (b) $T = 0$ phase diagram for the α -RuCl₃ model. $B_c = 6$ T within ED. Second-order terahertz response $\text{Re} \chi_{||}^2$ for (c) $B = 1.17B_c$ and (d) $B = 3.33B_c$. Computed with $L = 150$, $\eta = 0.2$ meV on a 24-site C_3 -symmetric periodic cluster.

models considered in this study, the calculation of $\chi^2(\omega_t, \omega_\tau)$ for $L = 150$ was of similar computational cost as obtaining the ground state (via Ref. [49]), making the method rather cheap.

Application to α -RuCl₃. We apply the method to extended Kitaev models on the honeycomb lattice,

$$\begin{aligned} \mathcal{H} = & \sum_{\langle ij \rangle_\gamma} K S_i^\gamma S_j^\gamma + \Gamma (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) + J \mathbf{S}_i \cdot \mathbf{S}_j \\ & + \sum_{\langle\langle ij \rangle\rangle} J_3 \mathbf{S}_i \cdot \mathbf{S}_j - \sum_i \mu_B \mathbf{B} \cdot \mathbf{g} \cdot \mathbf{S}_i, \end{aligned} \quad (11)$$

where $\gamma = x, y, z$ accords to the bond type X, Y, Z [Fig. 3(a)] and $\{\alpha, \beta\} = \{x, y, z\} \setminus \{\gamma\}$. K corresponds to the Kitaev coupling, Γ to symmetric off-diagonal exchange, and J (J_3) to nearest-neighbor (third-neighbor) Heisenberg coupling. $\mathbf{B}$ is the static magnetic field and $\mathbf{g}$ the gyromagnetic tensor.

We focus on the Kitaev candidate material α -RuCl₃ under in-plane magnetic fields $\mathbf{B} \parallel (y - x)$ (parallel to a bond), for which we employ the minimal model from Refs. [50,51] as a representative one; $(K, \Gamma, J, J_3) = (-5, 2.5, -0.5, 0.5)$ meV and $g_{||} = 2.3$. It reproduces the material's unconventional *linear* response, in which conventional magnons can break down [50]. While α -RuCl₃ orders antiferromagnetically, in-plane magnetic fields of $B_c \approx 7$ T suppresses this order [Fig. 3(b)]. The nature of the phase(s) and of the excitations beyond B_c have been subject to significant debate due to various unconventional observations for $B \gtrsim B_c$, with the scenario of a field-induced Kitaev spin liquid and Majorana fermionic excitations under controversial scrutiny [16–20]. Nonetheless, undisputedly, for increasing field strengths $B \gg B_c$, the material asymptotically approaches the conventional polarized state [52]. We therefore investigate the higher-order response in the region out of magnetic order, $B > B_c$, focusing on potential differences between the regimes $B \gtrsim B_c$ and $B \gg B_c$.

For the operators $\mathcal{A}, \mathcal{B}, \mathcal{C}$ in χ_{ABC}^2 [Eq. (2)], we choose the magnetization $\mathcal{M}$, corresponding to the magnetic field pulses in Fig. 1(a), being parallel to the static external field $\mathbf{B}$, and to magnetic-dipole coupling with the light. This choice is motivated by the corresponding linear-response channel

$\chi_{\parallel}^1$ featuring an excitation continuum in α -RuCl₃, which has been discussed as evidence for a QSL [24]. We abbreviate $\chi_{\mathcal{M}\mathcal{M}\mathcal{M}}^2 \equiv \chi_{\parallel}^2$.

$\chi_{\parallel}^2$ results are shown in Figs. 3(c) and 3(d) for a low-field case ($B = 1.17B_c$) and a high-field case ($B = 3.33B_c$), computed with $L = 150$ on 24 sites. While we expect the poles on the diagonal in Fig. 3(d) to form a continuum in the thermodynamic limit, we choose here a cautious (small) broadening $\eta = 0.2$ meV that preserves their separate appearance, to convey the method's raw results.

We highlight the qualitatively different responses found between the two field regimes. In the *high-field* regime ($B \gg B_c$) shown in Fig. 3(d), $\chi_{\parallel}^2$ is dominated by poles located on two distinct lines within the frequency plane; the frequency diagonal ($\omega_t = \omega_r$, so-called “nonrephasing signal”) and the frequency vertical ($\omega_t = 0, \omega_r \neq 0$, “rectification signal”), which we abbreviate F_{diag} and F_{vert} , respectively. The nonzero intensity away from these lines primarily stems from the broadening of their poles: Broadening arises marginally from the artificial broadening η , but mostly from the inherent broadening $\sim \frac{1}{\omega_t \omega_r}$ of higher-order susceptibilities, related to phase twisting [2,35,53] as shown in detail in the Supplemental Material [41]. Contributions from poles located *outside* of F_{diag} and F_{vert} are present but subdominant.

In contrast, at lower fields ($B \gtrsim B_c$), shown in Fig. 3(c), the majority of intensity stems from poles located away from F_{diag} and F_{vert} . Overall, these lead to an inhomogeneous continuum, spread across the two-dimensional frequency plane up to $\omega_t, \omega_r \lesssim 15$ meV.

To understand the origin of these two distinct responses, we first analyze the type of matrix elements contributing to second-order susceptibilities in general:

$$\chi_{\parallel}^2 \sim \langle 0 | \mathcal{M} | n \rangle \langle n | \mathcal{M} | m \rangle \langle m | \mathcal{M} | 0 \rangle, \quad (12)$$

where $|n\rangle, |m\rangle$ are excited states. The central matrix element $\langle n | \mathcal{M} | m \rangle$ contributes additional information compared to linear response $\chi_{\parallel}^1 \sim |\langle 0 | \mathcal{M} | n \rangle|^2$. Considering Eq. (12), it is instructive to distinguish between contributions with $n = m$ and those with $n \neq m$, pictured in Figs. 1(b) and 1(c). In the two-dimensional frequency plane, $n = m$ contributions [Fig. 1(b)] lead exclusively to poles along F_{diag} and F_{vert} , as those dominant in Fig. 3(d). If poles appear away from these locations [as in Fig. 3(c)], they can only stem from $n \neq m$ contributions, i.e., matrix elements between different excited states [Fig. 1(c)].

$\chi_{\parallel}^2$ for conventional magnons. To contrast our numerical results, we consider the $\chi_{\parallel}^1$ and $\chi_{\parallel}^2$ response expected on the level of linear spin-wave theory (LSWT) in the absence of magnon decay or interactions. While we will plot specific results for the α -RuCl₃ model, note that this discussion applies to the general LSWT response for any magnet's field-polarized phase.

Within LSWT, the magnetization operator $\mathcal{M}$ corresponds to a two-magnon operator, such that $\chi_{\parallel}^1$ (and $\chi_{\parallel}^2$) probes exclusively the two-magnon continuum. The resulting $\chi_{\parallel}^1(\omega)$ for the α -RuCl₃ model is shown versus B/B_c in Fig. 4(a) [54]. While the two-magnon gap grows monotonically with increasing field strength, the absolute $\chi_{\parallel}^1$ declines simultaneously, since the matrix elements $\langle 0 | \mathcal{M} | n \rangle$ rely on longitudinal

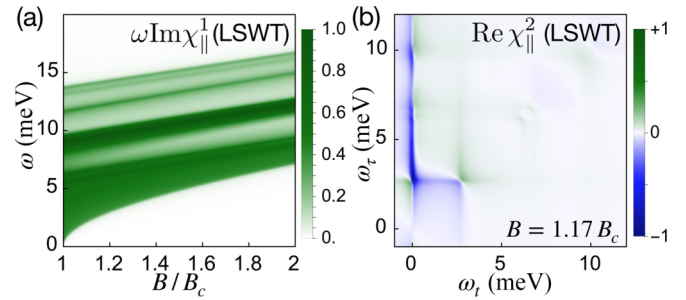


FIG. 4. (a) Linear terahertz response $\omega \text{Im} \chi_{\parallel}^1$ at LSWT level vs magnetic field for the partially polarized phase ($B > B_c$), accessing the two-magnon continuum. (b) Nonlinear response $\text{Re} \chi_{\parallel}^2$ at LSWT level at $B = 1.17B_c$.

fluctuations of the ordered moment, which are suppressed at high field.

Turning to $\chi_{\parallel}^2$, from inspecting the form of $\mathcal{M}$ in terms of magnon operators (see the Supplemental Material [41]), we find that contributions with $n \neq m$ [Fig. 1(c)] are *generally* strongly suppressed on the LSWT level: The only nonzero $n \neq m$ contributions relate to inter-magnon-band processes, which we conjecture to generally have tiny intensity compared to $n = m$ contributions, based on the fact that for Bravais lattices such contributions are completely forbidden, and no qualitative difference between Bravais and non-Bravais is expected. One non-Bravais example confirming this is discussed in the following.

LSWT results for the (honeycomb lattice) α -RuCl₃ model are shown in Fig. 4(b) for $B = 1.17B_c$. As described before, the dominance of matrix elements as in Fig. 1(b) leads to relevant poles appearing only on F_{diag} and F_{vert} . This overall structure therefore represents the universal form expected for two-magnon continua in $\chi_{\parallel}^2$ in a magnet's partially polarized phase. For the present model at $B/B_c = 1.17$, the two-magnon continuum begins at ~ 2.6 meV [Fig. 4(a)], leading to the onset of the strong rectification signal at $(\omega_t, \omega_r) = (0, 2.6)$ meV and a characteristic node feature at $(\omega_t, \omega_r) = (2.6, 2.6)$ meV in $\text{Re}[\chi_{\parallel}^2]$. LSWT results at other field strengths retain this form but with accordingly shifted energies [cf. Fig. 4(a)].

Deviations from a shape of predominantly F_{diag} and F_{vert} poles in a measured $\chi_{\parallel}^2$ can hence be used to directly diagnose continua to arise from unconventional excitations, where standard LSWT does not capture the full physics. This case is found in the numerical α -RuCl₃ simulations at $B \gtrsim B_c$ [Fig. 3(c)], where the dominant intensity arises from non- F_{diag} and non- F_{vert} poles. As the high-field limit suppresses quantum fluctuations and restores conventional magnons, the according $B \gg B_c$ result of the same model [Fig. 3(d)] recovers the LSWT-expected form of dominant F_{diag} and F_{vert} poles. $\chi_{\parallel}^2$ therefore offers a direct measurement of the breakdown of conventional magnon excitations away from the high-field limit. Whether this unconventional response is caused by, e.g., decaying and interacting magnons or fractionalized excitations is an open question for this class of materials and goes beyond the scope of the current study.

Beyond α -RuCl₃, a similar analysis can be applied to $\chi_{\parallel}^2$ measurements in the high-field phase of other frustrated

TABLE I. General expected response in linear and nonlinear terahertz response in the (partially) polarized phase of quantum magnets, for the channel with photons' magnetic fields parallel to the ordered moment.

State and fluctuations	Linear $\chi_{\parallel}^1(\omega)$	Nonlinear $\chi_{\parallel}^2(\omega_l, \omega_r)$
Fully polarized (no quantum fluctuations)	Zero	Zero
Partially polarized, LSWT-type fluctuations	Continuum	Homogeneous continuum along F_{diag} and F_{vert}
Partially polarized, non-LSWT fluctuations	Continuum ^a	Inhomogeneous continuum ^a

^aNote that the types of possible unconventional fluctuations are diverse, and can (dependent on the material) lead to additional features, such as bound states appearing outside of the continuum.

magnets. A useful summary for the interpretation of such measurements is presented in Table I. All three cases are represented in the discussed α -RuCl₃ model under in-plane fields, where the table's rows correspond to $B \rightarrow \infty$, $B \gg B_c$, and $B \gtrsim B_c$, respectively.

Outlook. We showed that nonlinear spectroscopy can unveil crucial insights about the nature of excitations in highly frustrated spin systems: By analyzing the positions of poles in the two-dimensional frequency plane of the susceptibility $\chi_{\parallel}^2$ one can directly assess the breakdown of conventional magnon excitations. We predicted such an unusual $\chi_{\parallel}^2$ response to be observable in α -RuCl₃ in the highly discussed region of in-plane magnetic fields $B \gtrsim 7$ T. Experimental 2DCS measurements on α -RuCl₃ and other frustrated magnets are highly desirable.

With the newly introduced efficient numerical method, the calculation of such response functions is straightforward and can be applied to different classes of models

and materials, as well as to operators beyond magnetization, for example, to study nonlinear susceptibilities via coupling to the electric field of the light. The presented Lanczos-based approach can furthermore be extended to other higher-order susceptibilities, such as third-order response.

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Data availability. The data that support the findings of this article are openly available [55].

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