# Theory of Two-Dimensional Nonlinear Spectroscopy for the Kitaev Spin Liquid

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Unambiguous identification of fractionalized excitations in quantum spin liquids has been a long-standing issue in correlated topological phases. Conventional spectroscopic probes, such as the dynamical spin structure factor, can only detect composites of fractionalized excitations, leading to a broad continuum in energy. Lacking a clear signature in conventional probes has been the biggest obstacle in the field. In this work, we theoretically investigate what kinds of distinctive signatures of fractionalized excitations can be probed in two-dimensional nonlinear spectroscopy by considering the exactly solvable Kitaev spin liquids. We demonstrate the existence of a number of salient features of the Majorana fermions and fluxes in two-dimensional nonlinear spectroscopy, which provide crucial information about such excitations.

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Quantum spin liquids (QSLs) are prominent examples of correlated topological paramagnets that may arise due to frustrating spin interactions in Mott insulators [1,2]. The long-range quantum entanglement and ground state degeneracy, which comprises the quantum order, differentiate QSLs from trivial paramagnets and symmetry-broken phases [3]. Important manifestations of the quantum order are the emergent gauge fields and quasiparticles carrying fractional quantum numbers [4]. Since the quantum entanglement is not directly observable, measuring these fractionalized excitations would be an important experimental footstep to identify quantum spin liquids. One of the most powerful probes in magnetism, the dynamical spin structure factor measured in inelastic neutron scattering, however, shows only a broad continuum as the spin-flip involves a multitude of fractionalized excitations. The absence of sharp signatures has hampered the progress in the discovery of quantum spin liquids.

In this Letter, we consider two-dimensional nonlinear spectroscopy as a tool to detect distinctive signatures of fractionalized quasiparticles in quantum spin liquids. The current work is motivated by a previous work that shows how the domain wall excitations in the transverse field Ising model can clearly be detected in two-dimensional THz spectroscopy [5]. Here we consider the exactly solvable Kitaev spin liquids on the honeycomb lattice [6] and investigate the signatures of Majorana fermions and fluxes in two-dimensional spectroscopy. We consider two magnetic-field pulses separated by time $\tau_1$ and measuring the nonlinear part of the induced transient magnetization at later time $\tau_2 + \tau_1$. The two-dimensional spectroscopy is represented by two frequencies corresponding to $\tau_1$ and $\tau_2$. The response consists of nonlinear susceptibilities, some of which correspond to the out-of-time-order correlators of the magnetization [7]. We show that the third-order nonlinear susceptibilities can give rise to clear signatures of the Majorana fermions and fluxes in the Kitaev spin liquids. We explain how one could obtain important information about such excitations from the output of the two-dimensional spectroscopy. Our main results are shown in Fig. 1.

*Model.*—On a honeycomb lattice with $2N$ number of sites, we consider Kitaev's spin-$\frac{1}{2}$ Hamiltonian with the isotropic strength of the bond-directional interactions [6],

$$\hat{H} = -\sum_{x\,\mathrm{bond}} \hat{\sigma}_j^x \hat{\sigma}_k^x - \sum_{y\,\mathrm{bond}} \hat{\sigma}_j^y \hat{\sigma}_k^y - \sum_{z\,\mathrm{bond}} \hat{\sigma}_j^z \hat{\sigma}_k^z, \tag{1}$$

where $\hat{\sigma}_j^{x,y,z}$ are the Pauli operators at site $j$. The model has a constant of motion $\hat{W}_p = \hat{\sigma}_1^x \hat{\sigma}_2^y \hat{\sigma}_3^z \hat{\sigma}_4^x \hat{\sigma}_5^y \hat{\sigma}_6^z = \pm 1$ for each hexagonal plaquette $p$, and we say there is a static $\mathbb{Z}_2$ flux at $p$ when $\hat{W}_p = -1$.

By representing the Pauli operators in terms of Majorana fermions, $\hat{\sigma}_j^\alpha \doteq i b_j^\alpha c_j$, we can rewrite the model as $\mathbb{Z}_2$ gauge theory coupled to itinerant Majorana fermions,

$$\tilde{H} = \sum_{\alpha\,\mathrm{bond}} i \hat{u}_{jk}^\alpha c_j c_k \Rightarrow \tilde{H}_u = \sum_p \varepsilon_p \left( a_p^\dagger a_p - \frac{1}{2} \right), \tag{2}$$

where $a_p$ represents the normal-mode complex fermions. The model is exactly solvable because the emergent $\mathbb{Z}_2$ gauge fields $\hat{u}_{jk}^\alpha \equiv i b_j^\alpha b_k^\alpha$ commute with $\tilde{H}$ and themselves so that the Hilbert space is factorized into the gauge (flux) sector and the matter fermion sector, $\mathcal{H} = \mathcal{H}_F \otimes \mathcal{H}_M$. Hence, for a given gauge configuration $|F\rangle = |\{u_{jk}^\alpha = \pm 1\}\rangle \in \mathcal{H}_F$, $\tilde{H}$ is reduced to a quadratic

 

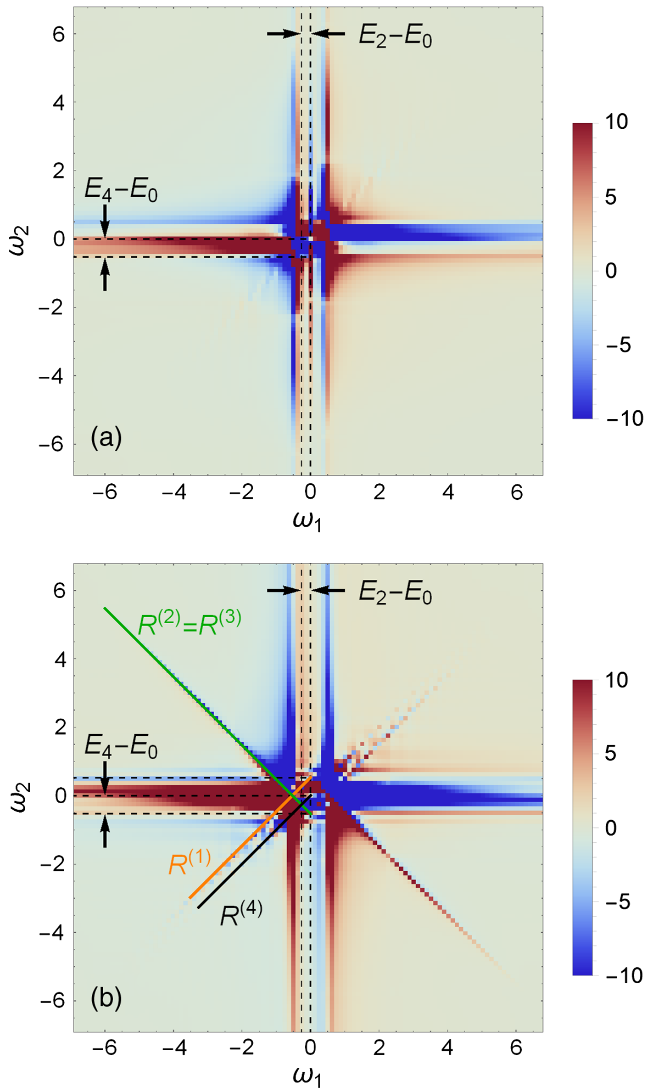


FIG. 1. Two-dimensional Fourier spectrum of the third-order susceptibilities (a) $\chi^{(3),z}_{zzz}(\omega_2, \omega_1, 0)$ and (b) $\chi^{(3),z}_{zzz}(\omega_2, 0, \omega_1)$ show the sharp vertical line signals at the two-flux gap, $\omega_1 = E_2 - E_0$. (b) $\chi^{(3),z}_{zzz}(\omega_2, 0, \omega_1)$ has one sharp diagonal signal for the Majorana fermions from $R^{(3),z}_{zzz}$ and two diagonal signals from $R^{(1),z}_{zzz}$ and $R^{(4),z}_{zzz}$. The two diagonal signals from $R^{(1),z}_{zzz}$ and $R^{(3),z}_{zzz}$ are extrapolated to finite $\omega_2$ intercept at the four-flux gap $\pm(E_4 - E_0)$, which hints at the four-flux intermediate states of the perturbative processes.

Hamiltonian $\tilde{H}_u$ for the itinerant Majorana fermion $c$, whose eigenstates $|M\rangle$ span the matter fermion sector $\mathcal{H}_M$.

*Two-dimensional spectroscopy.*—To probe the fractionalized excitations of the Kitaev spin liquid, we consider a nonlinear magnetic resonance spectroscopy with two linearly polarized, spatially uniform pulses separated by time $\tau_1$,

$$\mathbf{B}(t) = B_0 \hat{z} \delta(t) + B_1 \hat{z} \delta(t - \tau_1), \tag{3}$$

where the two incident pulses $B_0$ and $B_1$ arrive at the system at $t = 0$ and $t = \tau_1$, respectively [5,8]. For simplicity, here we consider the case where the incident pulses are all polarized along the $\hat{z}$ direction. These magnetic fields linearly couple to the local moments $\hat{H}_{\mathrm{tot}}(t) = \hat{H} - \sum_j B^z(t) \hat{\sigma}^z_j = \hat{H} - B^z(t) \hat{M}^z$ and induce finite transient magnetization $\hat{M}^z_{01}(t)$ measured at later time $t = \tau_2 + \tau_1$. To discard the leading contributions from the linear response, two subsequent experiments measure $\hat{M}^z_0(t)$ and $\hat{M}^z_1(t)$ due to only a single pulse $B_0$ or $B_1$, respectively. The nonlinear induced magnetization defined as $\hat{M}^z_{\mathrm{NL}}(t) = \hat{M}^z_{01}(t) - \hat{M}^z_0(t) - \hat{M}^z_1(t)$ at later time $t = \tau_1 + \tau_2$ depends only on the nonlinear dynamical responses [5],

$$M^z_{\mathrm{NL}}(\tau_1 + \tau_2)/2N$$
$$= \chi^{(2),z}_{zz}(\tau_2, \tau_1) B^z_1 B^z_0 \tag{4}$$
$$+ \chi^{(3),z}_{zzz}(\tau_2, \tau_1, 0) B^z_1 B^z_0 B^z_0 \tag{5}$$
$$+ \chi^{(3),z}_{zzz}(\tau_2, 0, \tau_1) B^z_1 B^z_1 B^z_0 + \mathcal{O}(B^4), \tag{6}$$

where time-dependent perturbation theory gives the $n$th-order susceptibility [7] (we choose the unit $\hbar = 1$),

$$\chi^{(n),z}_{z,\ldots,z}(\tau_n, \ldots, \tau_1) = \frac{i^n}{2N} \langle [[\ldots[\hat{M}^z(\tau_n + \cdots + \tau_1), \hat{M}^z(\tau_{n-1} + \cdots + \tau_1)], \ldots], \hat{M}^z(0)] \rangle. \tag{7}$$

*Second-order susceptibility.*—The second-order susceptibility $\chi^{(2),z}_{zz}(\tau_2, \tau_1)$ can be calculated from the three-point correlation functions [7],

$$\chi^{(2),z}_{zz}(\tau_2, \tau_1) = \frac{i^2}{N} \sum_{l=1}^{2} \mathrm{Re}[Q^{(l),z}_{zz}(\tau_2, \tau_1)], \tag{8}$$

where

$$Q^{(1),z}_{zz}(\tau_2, \tau_1) = \langle \hat{M}^z(\tau_2 + \tau_1) \hat{M}^z(\tau_1) \hat{M}^z(0) \rangle, \tag{9}$$

$$Q^{(2),z}_{zz}(\tau_2, \tau_1) = -\langle \hat{M}^z(\tau_1) \hat{M}^z(\tau_2 + \tau_1) \hat{M}^z(0) \rangle. \tag{10}$$

Formally, we can insert the resolution of identity $\sum_P |P\rangle\langle P| = \mathbb{1}$ and decompose the three-point function into a sum of products of three matrix elements weighted by phase factors containing the dynamical information. In general,

$$\langle \hat{M}^z(t) \hat{M}^z(t') \hat{M}^z(0) \rangle = \sum_{jkl} \sum_{PQ} \langle G|\hat{\sigma}^z_j|P\rangle \langle P|\hat{\sigma}^z_k|Q\rangle \times \langle Q|\hat{\sigma}^z_l|G\rangle e^{i(E_G - E_P)t + i(E_P - E_Q)t'}, \tag{11}$$

where $|P\rangle$ and $|Q\rangle$ are the energy eigenstates, and $|G\rangle$ is the ground state. Since the spin operator $\hat{\sigma}_j^z$ at a $z$ bond anticommutes with $\hat{W}_p$ at the plaquettes sharing the $z$ bond, $|P\rangle$ has a pair of two adjacent fluxes. As $\hat{\sigma}_k^z$ can either annihilate the existing two-fluxes or create two new fluxes, $|Q\rangle$ has either zero flux or two nonadjacent fluxes or four fluxes, which cannot be connected to the zero-flux state $|G\rangle$ by the single spin operator $\hat{\sigma}_l^z$. Therefore, the second-order susceptibility should be zero under the $z$-polarized pulses.

*Third-order susceptibilities.*—With the vanishing second-order susceptibility, the third-order responses determine the outcome of the nonlinear spectroscopy. The third-order susceptibilities in Eqs. (5) and (6) are calculated from the four-point correlation functions $R_{zzz}^{(l=1,2,3,4),z}$, which are expanded from the nested commutators in Eq. (7) [7]:

$$\chi_{zzz}^{(3),z}(\tau_2,\tau_1,0) = \frac{1}{N}\sum_{l=1}^{4}\mathrm{Im}[R_{zzz}^{(l),z}(\tau_2,\tau_1,0)], \quad (12)$$

$$\chi_{zzz}^{(3),z}(\tau_2,0,\tau_1) = \frac{1}{N}\sum_{l=1}^{4}\mathrm{Im}[R_{zzz}^{(l),z}(\tau_2,0,\tau_1)], \quad (13)$$

where

$$R_{zzz}^{(1),z}(t_3,t_2,t_1) = \langle \hat{M}^z(t_1)\hat{M}^z(t_2+t_1)\hat{M}^z(t_3+t_2+t_1)\hat{M}^z(0)\rangle, \quad (14)$$

$$R_{zzz}^{(2),z}(t_3,t_2,t_1) = \langle \hat{M}^z(0)\hat{M}^z(t_2+t_1)\hat{M}^z(t_3+t_2+t_1)\hat{M}^z(t_1)\rangle, \quad (15)$$

$$R_{zzz}^{(3),z}(t_3,t_2,t_1) = \langle \hat{M}^z(0)\hat{M}^z(t_1)\hat{M}^z(t_3+t_2+t_1)\hat{M}^z(t_2+t_1)\rangle, \quad (16)$$

$$R_{zzz}^{(4),z}(t_3,t_2,t_1) = \langle \hat{M}^z(t_3+t_2+t_1)\hat{M}^z(t_2+t_1)\hat{M}^z(t_1)\hat{M}^z(0)\rangle. \quad (17)$$

Similar to the three-point function in Eq. (11), we can decompose the four-point functions using the resolution of identity. For example, $R_{zzz}^{(3),z}(\tau_2,0,\tau_1)$ becomes

$$\begin{aligned} R_{zzz}^{(3),z}(\tau_2,0,\tau_1) &= \langle \hat{M}^z(0)\hat{M}^z(\tau_1)\hat{M}^z(\tau_2+\tau_1)\hat{M}^z(\tau_1)\rangle \\ &= \sum_{jklm}\sum_{PQR}\langle G|\hat{\sigma}_j^z|P\rangle\langle P|\hat{\sigma}_k^z|Q\rangle\langle Q|\hat{\sigma}_l^z|R\rangle\langle R|\hat{\sigma}_m^z|G\rangle \\ &\quad\times e^{i(E_P-E_Q)\tau_1+i(E_Q-E_R)(\tau_2+\tau_1)+i(E_R-E_G)\tau_1}. \end{aligned} \quad (18)$$

Since each spin operator flips two adjacent fluxes, $|P\rangle$ and $|R\rangle$ must belong to the two-flux sectors while $|Q\rangle$ can be either the zero-flux or nonadjacent two-flux or four-flux state. The matrix elements for the spin operators can be calculated by rewriting $b_j^\alpha$ Majorana fermions in terms of the complex bond fermions [9–12]. The detailed calculations can be found in the Supplemental Material [13].

Although the above decomposition is exact, we cannot sum over an infinite number of energy eigenstates $|P\rangle$, $|Q\rangle$, $|R\rangle$. Hence, we approximate the correlation functions by truncating the summation up to intermediate states with single matter fermion [10,11]. Since each spin excitation accompanies one $c$ Majorana fermion, we consider the two-flux states $|P\rangle$ and $|R\rangle$ with one matter fermion and the matter vacuum four-flux state $|Q\rangle$ [14]. This single matter fermion approximation is known to be extremely successful to calculate the dynamical spin structure factor for the Kitaev spin liquid; 97.5% of the total weight of response can be captured by the one fermion response [10]. The approximation takes advantage of the vanishing density of states of the Kitaev spin liquid at zero energy. Small perturbations would not introduce dramatic reconfiguration of the matter fermions because only few states are accessible at low energy.

*Results.*—We compute the real-time four-point correlation functions on a periodic lattice with $125\times125$ unit cells. Two-dimensional Fourier transform of the third-order susceptibilities (Fig. 1) and the four-point correlation functions (Fig. 2) are the main results of our work. Here we exclude the case $|Q\rangle = |G\rangle$ in Eq. (18) where the four-point function becomes nothing but a product of the two two-point functions, e.g., $\langle\hat{\sigma}_j^z\hat{\sigma}_k^z\hat{\sigma}_l^z\hat{\sigma}_m^z\rangle = \langle\hat{\sigma}_j^z\hat{\sigma}_k^z\rangle\langle\hat{\sigma}_l^z\hat{\sigma}_m^z\rangle$, which can yield physically inconsistent results within the single matter fermion approximation.

There are three distinctive features in the third-order susceptibilities in Fourier space (Fig. 1). First, both $\chi_{zzz}^{(3),z}(\omega_2,\omega_1,0)$ and $\chi_{zzz}^{(3),z}(\omega_2,0,\omega_1)$, which are the Fourier transforms of $\chi_{zzz}^{(3),z}(\tau_2,\tau_1,0)$ and $\chi_{zzz}^{(3),z}(\tau_2,0,\tau_1)$, respectively, exhibit sharp vertical line signals at the two-flux gap, $\omega_1 = E_2 - E_0$. Second, $\chi_{zzz}^{(3),z}(\omega_2,0,\omega_1)$ has three extended diagonal signals. Third, if we extrapolate these diagonal signals to $\omega_1 = 0$, two of the three have an $\omega_2$ intercept equal to the four-flux gap $\pm(E_4 - E_0)$, i.e., there are overall shifts in these two diagonal signals.

While the results in Figs. 1 and 2 are the direct Fourier transforms of the real-time correlation functions [13], we can identify which processes are responsible for these distinctive signals in the susceptibilities from the formal analytic expressions of the Fourier transformed correlation functions. For example, the Fourier transformation of $R_{zzz}^{(3),z}(\tau_2,0,\tau_1)$ [Eq. (18)] can be written as

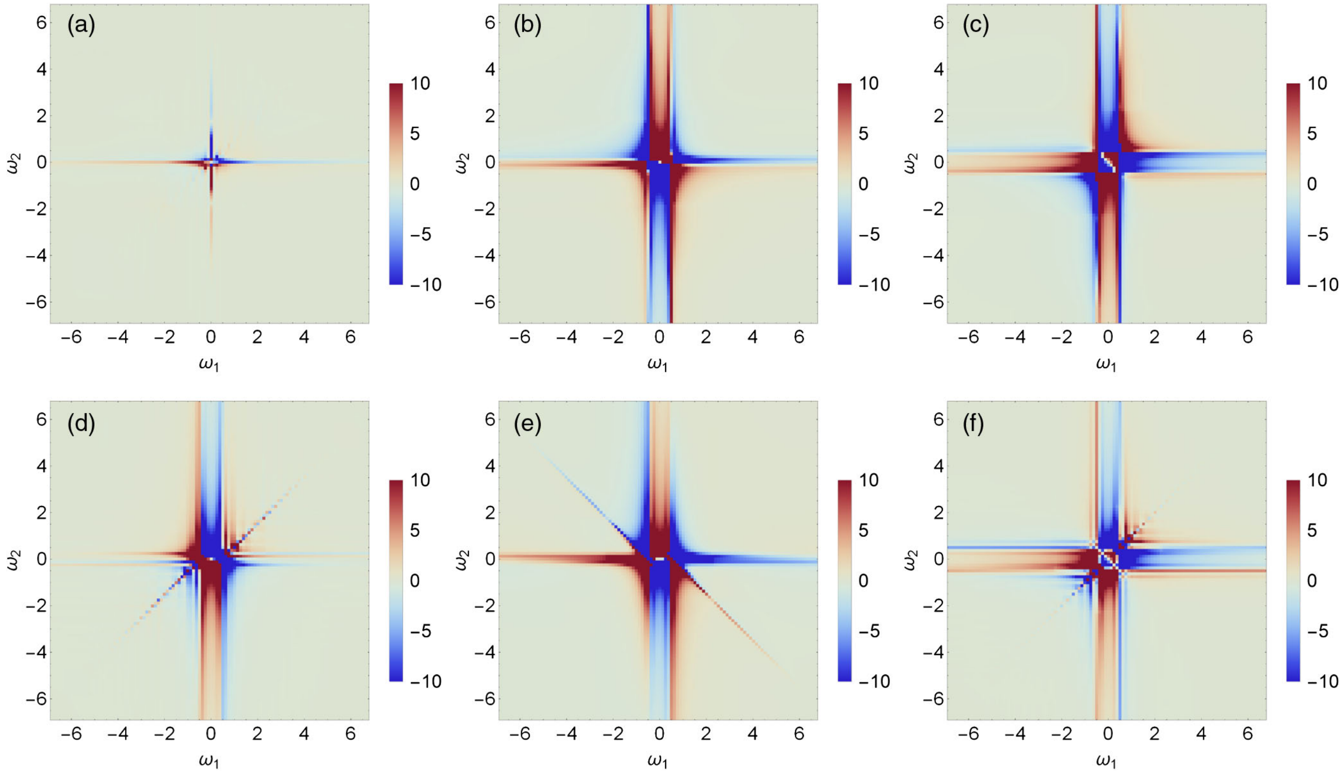

FIG. 2. Two-dimensional Fourier spectrum of the four point correlation functions. Here $\mathcal{F}$ is the Fourier transformation. (a) $\mathrm{Im}\mathcal{F}[\mathrm{Im}R^{(1,2),z}_{zzz}(\tau_2,\tau_1,0)]$ (b) $\mathrm{Im}\mathcal{F}[\mathrm{Im}R^{(3),z}_{zzz}(\tau_2,\tau_1,0)]$ (c) $\mathrm{Im}\mathcal{F}[\mathrm{Im}R^{(4),z}_{zzz}(\tau_2,\tau_1,0)]$ (d) $\mathrm{Im}\mathcal{F}[\mathrm{Im}R^{(1),z}_{zzz}(\tau_2,0,\tau_1)]$ (e) $\mathrm{Im}\mathcal{F}[\mathrm{Im}R^{(2,3),z}_{zzz}(\tau_2,0,\tau_1)]$ (f) $\mathrm{Im}\mathcal{F}[\mathrm{Im}R^{(4),z}_{zzz}(\tau_2,0,\tau_1)]$

$$\begin{aligned} &R^{(3),z}_{zzz}(\omega_2,0,\omega_1)\\ &\quad=\cdots+\sum_{j\neq k}\sum_{PQR}\langle G|\hat{\sigma}^z_j|P\rangle\langle P|\hat{\sigma}^z_k|Q\rangle\langle Q|\hat{\sigma}^z_k|R\rangle\langle R|\hat{\sigma}^z_j|G\rangle\\ &\qquad\times\frac{1}{4}\delta(\omega_1+E_2+\varepsilon_P-E_0)\delta(\omega_2+E_4-E_2-\varepsilon_R)\\ &\qquad+\sum_{j\neq k}\sum_{PQR}\langle G|\hat{\sigma}^z_k|P\rangle\langle P|\hat{\sigma}^z_j|Q\rangle\langle Q|\hat{\sigma}^z_k|R\rangle\langle R|\hat{\sigma}^z_j|G\rangle\\ &\qquad\times\frac{1}{4}\delta(\omega_1+E_2+\varepsilon_P-E_0)\delta(\omega_2+E_4-E_2-\varepsilon_R), \end{aligned} \tag{19}$$

where $E_n$ is the vacuum energy of the $n$-flux state, $\varepsilon_{P(R)}$ is the matter fermion energy, and the other contributions which cannot be written in terms of the delta functions are in $(\cdots)$. The delta function pieces would show no signal for $-(E_2-E_0)<\omega_1<0$. Similarly, the complex conjugate pair $R^{(3),z}_{zzz}(\omega_2,0,\omega_1)$ has no weight in $0<\omega_1<E_2-E_0$. This is nothing but the well-known spin gap for spin excitations [10]. If the first pulse $B_0$ does not transfer enough energy to excite two adjacent fluxes, the Kitaev spin liquid remains in the ground state. Hence, for finite nonlinear responses, the first pulse $B_0$ must transfer energy greater than the two-flux gap $E_2-E_0$.

Another important feature, the shifted diagonal in Fig. 1(b), also comes from the $R^{(3),z}_{zzz}(\omega_2,0,\omega_1)$ [Fig. 2(e)]. When $\varepsilon_P=\varepsilon_R=\varepsilon$, note that the matrix element $\langle G|\hat{\sigma}^z_j|P\rangle\langle P|\hat{\sigma}^z_k|Q\rangle\times\langle Q|\hat{\sigma}^z_k|R\rangle\langle R|\hat{\sigma}^z_j|G\rangle=|\langle G|\hat{\sigma}^z_j|P\rangle\langle P|\hat{\sigma}^z_k|Q\rangle|^2\geq 0$. Hence, the summation over sites $\sum_{j\neq k}$, equivalently the summation over all different four-flux configurations $|Q\rangle$ excited by $\hat{\sigma}^z_j$ and $\hat{\sigma}^z_k$, results in only constructive interference. Therefore we get the strongly enhanced signal when

$$\omega_1=E_0-E_2-\varepsilon<0, \tag{20}$$

$$\omega_2=E_2-E_4+\varepsilon=-\omega_1-(E_4-E_0), \tag{21}$$

which corresponds to the shifted diagonal with the slope of $-1$ and the $\omega_2$ intercept $-(E_4-E_0)$. According to Eq. (20), the domain of the line is determined by the single matter fermion bandwidth and the two-flux gap, and this is confirmed by Figs. 1(b) and 2(e).

Following a similar logic, we can understand two-flux gaps and the other two coherent diagonal signals coming from $R^{(1),z}_{zzz}(\omega_2,0,\omega_1)$ and $R^{(4),z}_{zzz}(\omega_2,0,\omega_1)$, which have contributions with the constraints in the sum over intermediate states via $\delta(\omega_1+E_0-E_2-\varepsilon_R)\delta(\omega_2+E_4-E_2-\varepsilon_R)$ and $\delta(\omega_1+E_0-E_2-\varepsilon_R)\delta(\omega_2+E_0-E_2-\varepsilon_P)$, respectively. $R^{(1),z}_{zzz}(\omega_2,0,\omega_1)$ yields the shifted diagonal

$\omega_2 = \omega_1 - (E_4 - E_0)$, and $R^{(4),z}_{zzz}(\omega_2, 0, \omega_1)$ gives $\omega_2 = \omega_1$ from the constructive interference with $\varepsilon_P = \varepsilon_R$ for $\omega_1 \geq E_2 - E_0$.

*Conclusion.*—In this work, we have demonstrated how two-dimensional spectroscopy can be used to obtain useful information about fractionalized excitations in the Kitaev spin liquids, where the single spin-flip process excites a Majorana fermion and two fluxes in adjacent plaquettes. The spectroscopic signatures as a function of two frequencies, $\omega_1$ and $\omega_2$, corresponding to the delay time of two successive magnetic pulses and the time of measurement, offer a clear identification of both the Majorana fermions and flux excitations. We demonstrated that the two-flux gap appears in $\omega_1$ and the shifted diagonal signal in the $\omega_1 - \omega_2$ plane has an $\omega_2$ intercept at the four-flux gap. Most importantly, the presence of the sharp diagonal signals is the direct consequence of the itinerant Majorana fermions. The domain of finite response in the two-frequency $\omega_1 - \omega_2$ plane is determined by a number of stringent conditions, which makes it possible to identify clear signatures of fractionalized excitations. It will be interesting to extend our work to other theoretical models of quantum spin liquids, that are not exactly solvable. Furthermore, the results reported here may be tested in a number of candidate materials for the Kitaev spin liquids [15–21]. We expect our results will shed significant light on the identification of fractionalized excitations and quantum spin liquids.

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[14] Among $N-1$ translationally inequivalent flux configurations for $|Q\rangle$, two configurations have two nonadjacent fluxes instead of four fluxes. These states contribute to the weak diagonal signals in the first and third quadrants (guided by black line in Fig. 1) due to $R^{(4)}$ correlations, but they do not contribute to the other diagonal signals. Our calculations found no other qualitatively distinctive features from these two-flux states compared to the four-flux state contributions.
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