Parametric pumping of the two-dimensional quantum spin liquid

A. A. Zvyagin

Max-Planck-Institut fur Physik komplexer Systeme, Noethnitzer Strasse, 38, D-01187 Dresden, Germany ¨ and B. I. Verkin Institute for Low Temperature Physics and Engineering of the National Academy of Sciences of Ukraine,

Nauky Avenue, 47, Kharkov 61103, Ukraine

(Received 27 April 2016; revised manuscript received 11 January 2017; published 24 February 2017)

With the help of the exact solution of the Kitaev model the parametric pumping of the two-dimensional quantum spin liquid under the action of the ac magnetic field is studied. In the dynamical regime the field produces oscillations of the magnetization with the field's frequency, modulated by the Rabi-like oscillations. In the steady-state regime, the Rabi-like oscillations are damped. The absorption of the ac field by the Kitaev spin model is finite and manifests resonance features. Such a behavior is generic for quantum spin liquids with fermionic excitations, and it is different from the linear spin-wave response of magnetically ordered systems to such a parametric pumping.

DOI: [10.1103/PhysRevB.95.064428](https://doi.org/10.1103/PhysRevB.95.064428)

I. INTRODUCTION

Among the large variety of magnetic materials lowdimensional spin systems and systems with frustration demonstrate properties of quantum spin liquids [\[1\]](#page-5-0). The latter are characterized by strong spin-spin couplings; however, those interactions cannot yield a long-range magnetic order, either due to enhanced quantum fluctuations or due to frustrated bonds, and the emergent excitations there are fractionalized quasiparticles. The celebrated Kitaev spin model on the honeycomb lattice [\[2\]](#page-5-0) is one of the best examples of such a system. The relative simplicity of the model permits us to look for the new theoretical predictions for quantum spin liquids and experimental realizations in condensed-matter physics and ultracold atomic systems [\[3,4\]](#page-5-0). The realization of physics of Kitaev's model is related to magnetic materials, where spin and orbital degrees of freedom are strongly coupled. Electron spin resonance, in particular, the parametric excitation of a spin system by the external ac magnetic field, is a powerful tool to study dynamics of magnets. Taking into account constraints for the inelastic neutron scattering in some realizations of the Kitaev model (like in Ir oxides due to the strong neutron absorption), the spin-resonance method with very good energy resolution can be useful for the investigation of their dynamical magnetic properties. In quantum spin liquids, in particular, the parametric excitation by the ac magnetic field can permit us to understand the nature of spin quasiparticles.

In this work we study the response of the spin-1*/*2 two-dimensional system to the parametric action of the ac magnetic field (parametric pumping). It is a study of the two-dimensional quantum spin-liquid system affected by the parametric pumping that *exactly* takes into account the finite Hilbert space of spins. It is shown that the ac field-induced magnetization and absorption are *finite* in the considered system, unlike the response of spin waves of magnetically ordered systems to the parametric pumping.

II. PARAMETRIC PUMPING

Consider the spin-1*/*2 system on the honeycomb lattice with the Hamiltonian [\[2\]](#page-5-0)

$$
\mathcal{H}_0 = -\sum_{\alpha=x,y,z} J_\alpha \sum_{\alpha-\text{links}} S_j^\alpha S_{j'}^\alpha, \tag{1}
$$

where $J_{x,y,z}$ are exchange integrals (let us, for definiteness, consider the case with $J_x, J_y, J_z \ge 0$ and $S_j^{x,y,z}$ are the operators of the spin projections of the spins situated at the sites *j* of the lattice. Spins interact if they are situated at the neighboring sites. The special feature of the Kitaev model is that the interactions depend on the link type (i.e., along the links parallel to *z* axis only *z* projections of spins interact, etc.) [\[2\]](#page-5-0). Consider the parametric effect of the external ac magnetic field, polarized linearly, e.g., along the *z* axis, on the system. It can be described by the Hamiltonian

$$
\mathcal{H} = \mathcal{H}_0 - h \cos(\omega t) \sum_j S_j^z.
$$
 (2)

Here we denote $h = g\mu_B h_0$, where *g* is the *g* factor and μ_B is Bohr's magneton and $h_0 > 0$ and $\omega > 0$ are the magnitude and the frequency of the ac field. The magnitude of the ac field is considered to be much smaller than exchange constants and the frequency of the ac field, $h \ll J_{x,y,z} \sim \hbar \omega$. Dynamics of the spin system can be studied in the framework of the Liouville-like kinetic equation [\[5\]](#page-5-0) for the density matrix *ρ*, written as

$$
i\hbar \dot{\rho} = [\mathcal{H}, \rho] - i\hbar \gamma (\rho - \rho_0), \tag{3}
$$

where the overdot denotes the time derivative and the symbol [·*,*·] denotes the commutator. Here we suppose that the linear relaxation with the rate γ (caused by the interaction of the studied system with the environment, e.g., with phonons) is homogeneous and frequency independent and that possible oscillations relax to the state ρ_0 , which is the Gibbs distribution with \mathcal{H}_0 , natural for $h \ll J_{x,y,z}$.

First, let us use the substitution $\rho' = \rho \exp(-\gamma t)$; we get

$$
i\hbar \dot{\rho}' = [\mathcal{H}, \rho'] + i\hbar \gamma \rho_0 \exp(\gamma t). \tag{4}
$$

Then we use the unitary transformation $\rho' = U_1 \rho_1 U_1^{-1}$, where

$$
U_1 = \exp\left[i\frac{h}{\hbar\omega}\sin(\omega t)\sum_j S_j^z\right],\tag{5}
$$

with

$$
i\hbar \dot{\rho}_1 = [\mathcal{H}_1, \rho_1] + i\hbar \gamma U_1^{-1} \rho_0 U_1 \exp(\gamma t), \tag{6}
$$

2469-9950/2017/95(6)/064428(7) 064428-1 ©2017 American Physical Society

where

$$
\mathcal{H}_1 = -J_z \sum_{z-\text{links}} S_j^z S_{j'}^z - \frac{i}{4} \left[J_x \sum_{x-\text{links}} -J_y \sum_{y-\text{links}} \right]
$$

\n
$$
\times (S_j^+ S_{j'}^- + S_j^- S_{j'}^+) - \frac{1}{4} \left[J_x \sum_{x-\text{links}} -J_y \sum_{y-\text{links}} \right]
$$

\n
$$
\times [S_j^+ S_{j'}^+ e^{i(2h/\hbar\omega)\sin(\omega t)} - S_j^- S_{j'}^- e^{-i(2h/\hbar\omega)\sin(\omega t)}]
$$
(7)

and $S_j^{\pm} = S_j^x \pm i S_j^y$. Then we can use the series

$$
\exp[iz\sin(\omega t)] = \sum_{n=-\infty}^{\infty} J_n(z)\exp(in\omega t),\tag{8}
$$

⎤

where $J_n(z)$ is the Bessel function. We obtain

$$
\mathcal{H}_1 = -J_z \sum_{z-\text{links}} S_j^z S_{j'}^z - \frac{i}{4} \left[J_x \sum_{x-\text{links}} -J_y \sum_{y-\text{links}} \right]
$$

$$
\times (S_j^+ S_{j'}^- + S_j^- S_{j'}^+) - \sum_{n=-\infty}^{\infty} J_n(2h/\hbar\omega)
$$

$$
\times \frac{1}{4} \left[J_x \sum_{x-\text{links}} -J_y \sum_{y-\text{links}} \right] e^{in\omega t} [S_j^+ S_{j'}^+ - S_j^- S_{j'}^-]. \quad (9)
$$

Now we take into account that $h \ll \hbar \omega$, i.e., $z \ll 1$. For small *z* we have $J_0 \approx 1$ and $J_{\pm 1}(z) \approx \pm z$. It follows that

$$
\mathcal{H}_1 \approx \mathcal{H}_0 \mp \frac{h}{2\hbar\omega} e^{\pm i\omega t}
$$

$$
\times \left[J_x \sum_{x-\text{links}} -J_y \sum_{y-\text{links}} \right] (S_j^+ S_{j'}^+ - S_j^- S_{j'}^-). \quad (10)
$$

III. SPIN WAVE APPROXIMATION

Now we need to diagonalize the H_0 part of H_1 . It can be realized in two ways. If the considered system is magnetically ordered, the standard way to study its dynamics is the spin-wave approximation. We can use, e.g., the Holstein-Primakoff representation of spin operators via bosonic ones [\[6\]](#page-5-0). Suppose $J_z > J_{x,y}$. The operators of spin projections can be approximately presented as

$$
S_j^z = (1/2) - b_j^{\dagger} b_j, \quad S_j^+ \approx b_j, \quad S_j^- \approx b_j^{\dagger}, \quad (11)
$$

with b_j and b_j^{\dagger} being bosonic operators of destruction and creation. After the Fourier and Bogolyubov transformations the Hamiltonian H_1 (up to a constant term) has the form

$$
\mathcal{H}_1 \approx \sum_{\mathbf{k}} \{ \varepsilon_{\mathbf{k}} a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}} + (h/\hbar \omega) f_{\mathbf{k}} [\exp(i \omega t) a_{\mathbf{k}} a_{-\mathbf{k}} + \text{H.c.}]\}, \quad (12)
$$

where $\varepsilon_{\mathbf{k}}$ is the energy of a magnon, $a_{\mathbf{k}}$ and $a_{\mathbf{k}}^{\dagger}$ destroy and create the magnon, respectively. and $f_{\bf k}$ is the coefficient for terms in the Hamiltonian \mathcal{H}_0 , which do not conserve $\sum_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}}$. Here we drop the terms like $[\exp(-i\omega t)a_k a_{-k} + \text{H.c.}]$, using the so-called resonance approximation [\[7\]](#page-5-0). This means that we consider exactly terms explicitly dependent on time which produce the nonzero contribution to the linear response. The remaining terms with an explicit time dependence can be omitted due to the smallness of the magnitude of the ac magnetic field (their contribution can be, in principle, calculated in the framework of perturbation theory).

It turns out that the same result, i.e., Eq. (12), can be obtained in the macroscopic approach for the magnetically ordered system using the spin-wave approximation [\[8\]](#page-5-0). Namely, suppose that the considered spin system is magnetically ordered. For the magnetically ordered system we can replace the operators of spin projections in Eq. (10) by their average values. This means that the quantum spins are replaced by the classical vectors of the site magnetic moments (due to the magnetic ordering), and the quantum Hamiltonian is replaced by the classical density of energy. For positive J_{α} such an ordering is ferromagnetic: All magnetic moments are parallel to each other. This approach contradicts the exact solution of the Kitaev model [\[2\]](#page-5-0), which is related to the spin liquid. Then, the dynamics of the classical vectors of the magnetic moments (i.e., of the order parameters of the magnetically ordered system) is described by the Landau-Lifshitz equation of motion. There magnetic moments move in the effective field, which is the variation of the density of the energy with respect to related magnetic moment. This approach is equivalent to the use of the mean-field approximation in quantum mechanics [\[9\]](#page-5-0). Suppose that due to $J_z > J_{x,y}$ the site magnetic moments of the considered ordered magnetic system are directed mostly along the *z* axis. Then, we can consider small deviations of magnetic moments (i.e., spin waves) and linearize the obtained equations of motion for those small deviations. The density of the energy, bilinear in such small deviations, can be diagonalized by the Fourier transform. Spin waves behave like bosons. They are often considered to be equivalent to magnons in magnetically ordered systems. Finally, using the resonance approximation, we get the density of the energy of spin waves in *the same form* as Eq. (12). Notice that for classical vectors of magnetic moments in magnetically ordered systems the number of states of their projections is infinite, unlike the finite number for a quantum spin. This is the reason why magnons in the magnetically ordered systems behave as bosons.

Now we use the unitary transformation $U_2 = \exp$ $[(-i\omega t/2)\sum_{\mathbf{k}} a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}}]$, with $\rho_1 = U_2 \rho_2 U_2^{-1}$, which yields

$$
i\hbar \dot{\rho}_2 = [\mathcal{H}_2, \rho_2] + i\hbar \gamma U_2^{-1} U_1^{-1} \rho_0 U_1 U_2 \exp(\gamma t),
$$

$$
\mathcal{H}_2 = \sum_{\mathbf{k}} \left[\left(\varepsilon_{\mathbf{k}} - \frac{\hbar \omega}{2} \right) a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + \frac{\hbar}{\hbar \omega} f_{\mathbf{k}} (a_{\mathbf{k}} a_{-\mathbf{k}} + \text{H.c.}) \right]. \tag{13}
$$

It is the quadratic form of Bose operators. It follows from the equations of motion for a_k and a_{-k}^{\dagger} that the increment (decrement) of the time dependence of a_k and a_{-k}^{\dagger} (and hence of the average with the density matrix or with the groundstate wave function at $T = 0$ of the number of magnons $a_k^T a_k$ depends on whether the value $-[\varepsilon_{\bf k} - (\hbar \omega/2)]^2 + |hf_{\bf k}/\hbar \omega|^2$ is larger (smaller) than $(h\gamma)^2$. In resonance we can neglect the term $[\varepsilon_{\bf k} - (\hbar \omega/2)]^2$. Then for any γ there exists the threshold value of the magnitude of the ac field h_c : For a magnitude of the

ac field larger than that threshold value, $h > h_c$, the number of magnons in the system grows with time exponentially (and the linear relaxation cannot limit such a growth). Such a parametric instability [\[8\]](#page-5-0) is observed in many magnetic systems [\[10\]](#page-5-0).

IV. ADAPTATION OF THE EXACT SOLUTION

The second (and, as we show below, the correct approach for spin liquids) way is to diagonalize the H_0 part of the Hamiltonian H_1 using the exact solution of the Kitaev honeycomb model [\[2\]](#page-5-0). We rewrite the Hamiltonian \mathcal{H}_1 exactly using the transformation to fermion operators of creation and destruction d^{\dagger} and *d* (for our purpose it is convenient to use the Dirac representation for fermion operators). It is the two-dimensional generalization [\[11,12\]](#page-5-0) of the Jordan-Wigner transformation [\[13\]](#page-5-0). To represent spin operators with spinless fermion operators we can use [\[11\]](#page-5-0) $S_{j,l}^z = d_{j,l}^{\dagger}d_{j,l}$ – $1/2$, $S_{j,l}^+ = d_{j,l}^{\dagger} \exp[\sum_{i,k < j} d_{i,k}^{\dagger} d_{i,k} + \sum_{i < l} d_{i,l}^{\dagger} d_{i,l}],$ and $S_{j,l}^- =$ $(S_{j,l}^+)^{\dagger}$. Here the indexes *j* and *l* denote the column and row of the brick lattice equivalent to the honeycomb lattice [\[11\]](#page-5-0). For each pair of Dirac fermion operators *d* and *d*† two Majorana fermion operators *a* and *f* can be defined as $a_{j,l} = i(d_{j,l}^{\dagger} - d_{j,l}^{\dagger})$ and $f_{j,l} = d_{j,l}^{\dagger} + d_{j,l}$ when $j + l$ is even and $a_{j,l} = d_{j,l}^{\dagger} + d_{j,l}$ and $f_{j,l} = i(d_{j,l}^{\dagger} - d_{j,l})$ when $j + l$ is odd. It turns out that the values $\alpha_j = i f_{j,l} f_{j,l+1}$ defined on each vertical bond are conserved [\[2,11\]](#page-5-0). The Hamiltonian \mathcal{H}_0 can be written (using more convenient enumeration of bonds instead of the column and rows) as the Hamiltonian of the Fermi gas on a brick lattice with the site-dependent chemical potential

$$
\mathcal{H}_0 = \sum_j [J_x (d_j^{\dagger} + d_j)(d_{j+\hat{x}}^{\dagger} - d_{j+\hat{x}}) + J_y
$$

$$
\times (d_j^{\dagger} + d_j)(d_{j+\hat{y}}^{\dagger} - d_{j+\hat{y}}) + 2J_z \alpha_j (2d_j^{\dagger} d_j - 1)], \quad (14)
$$

where *j* stands for the position of the *z* bond, \hat{y} connects two *z* bonds and crosses a *y* bond (a similar definition holds for \hat{x}), and $\alpha_j = \pm 1$ (α_j commutes with $d_{j'}$ and $d_{j'}^{\dagger}$ for any *j* and j'). Notice that a similar result (with different notations) was obtained in the original paper [\[2\]](#page-5-0); that is, the result does not depend on the numeration, as it must be. This transformation is exact (unlike the approximate Holstein-Primakoff one used above), and it is valid for any J_x , J_y , J_z . In the sectors with fixed α_j the diagonal form of the Kitaev model can be obtained after the Fourier and Bogolyubov transformations. It has a BCS-like form with the energy

$$
E_{\mathbf{k}} = \sqrt{\epsilon_{\mathbf{k}}^2 + \Delta_{\mathbf{k}}^2},\tag{15}
$$

where $\epsilon_{\mathbf{k}} = \pm J_z + J_x \cos k_x + J_y \cos k_y$ and $\Delta_{\mathbf{k}} = J_x \sin k_x +$ *J_y* sin *k_y*. The spectrum is gapless for $|J_x - J_y| \leqslant J_z \leqslant J_x +$ *Jy* and gapped otherwise.

Then our goal is to treat H_0 in Eq. [\(10\)](#page-1-0) exactly, i.e., to use the exact excitations of the Kitaev model in the fixed α_j sector (taking into account the change of sign). The Hamiltonian \mathcal{H}_1 can be written as

$$
\mathcal{H}_1 \approx \sum_{\mathbf{k}} \left\{ E_{\mathbf{k}} c_{\mathbf{k}}^{\dagger} c_{\mathbf{k}} + |\Delta_{\mathbf{k}}| \frac{h}{\hbar \omega} [c_{-\mathbf{k}} c_{\mathbf{k}} \exp(i \omega t) + \text{H.c.}] \right\}.
$$
 (16)

Here $c_{\mathbf{k}}^{\dagger}$ and $c_{\mathbf{k}}$ are Dirac fermion operators, in which \mathcal{H}_0 is diagonal for the fixed α_j sector. Notice that the pumping term related to the operator $\sum_j S_j^z$ can change homogeneously the sign of α_i of the original Hamiltonian [\[14\]](#page-5-0). The summation is over all **k** belonging to the subset of the Brillouin zone such that −**k** is out of that subset [\[12\]](#page-5-0). Again, we used the resonance approximation; that is, we consider exactly only terms explicitly dependent on time which produce the nonzero contribution to the linear response. Equation (16) is the Hamiltonian \mathcal{H}_1 ; that is, it is the Hamiltonian \mathcal{H}_0 plus the time-dependent term (linear in $h/\hbar\omega$). The Hamiltonian \mathcal{H}_0 is given by Eq. (14). We diagonalize the fermionic \mathcal{H}_0 in the sector with the fixed α_j by the Fourier and Bogolyubov transformation, i.e., consider H_0 exactly there. Then the time-dependent part of \mathcal{H}_1 is written in terms of normal modes of the Hamiltonian H_0 . After the Fourier and Bogolyubov transformation it is related to the nondiagonal quadratic form of operators $c_{-\mathbf{k}}c_{\mathbf{k}}$ and $c_{\mathbf{k}}^{\dagger}c_{-\mathbf{k}}^{\dagger}$ (considered resonance conditions). In the resonance approximation terms proportional to $[c_{-\mathbf{k}}c_{\mathbf{k}} \exp(-i\omega t) + \text{H.c.}]$ are dropped because their contribution is small (they are in the antiresonance). Then we use the unitary transformation $U_2 = \exp[(-i\omega t/2) \sum_{\mathbf{k}} c_{\mathbf{k}}^{\dagger} c_{\mathbf{k}}]$, with $\rho_1 = U_2 \rho_2 U_2^{-1}$, which yields

$$
i\hbar \dot{\rho}_2 = [\mathcal{H}_2, \rho_2] + i\hbar \gamma U_2^{-1} U_1^{-1} \rho_0 U_1 U_2 \exp(\gamma t),
$$

$$
\mathcal{H}_2 = \sum_{\mathbf{k}} \left[\left(E_{\mathbf{k}} - \frac{\hbar \omega}{2} \right) c_{\mathbf{k}}^{\dagger} c_{\mathbf{k}} + \frac{\hbar}{\hbar \omega} |\Delta_{\mathbf{k}}| (c_{\mathbf{k}} c_{-\mathbf{k}} + \text{H.c.}) \right].
$$
(17)

The Hamiltonian H_2 has no explicit dependence on time. Hence, we can use the last unitary transformation $U_3 =$ $\exp[-it\mathcal{H}_2/\hbar]$, with $\rho_2 = U_3 \rho_0 U_3^{-1}$. The solution for the time dependence of the density matrix is

$$
\rho = V(t)\rho_0 V^{-1}(t) \exp(-\gamma t), \quad t \ll \gamma^{-1},
$$

\n
$$
\rho = \gamma V(t) \int_{-\infty}^{0} dt' V^{-1}(t') \rho_0 V(t') e^{\gamma(t'-t)} V^{-1}(t),
$$

\n
$$
t \gg \gamma^{-1},
$$
\n(18)

where $V(t) = U_1(t)U_2(t)U_3(t)$. The dynamics of the system in the dynamical regime for $t \ll \gamma^{-1}$ is determined by the first line of Eqs. (18), and in the steady-state regime for $t \gg \gamma^{-1}$ it is determined by the second line of Eqs. (18).

It is interesting to mention that one can obtain a similar result if instead of the unitary transformation U_1 one uses the diagonalization of the Hamiltonian H_0 . Then, writing the term $h \cos(\omega t) \sum_j S_j^z$ in terms of operators, in which \mathcal{H}_0 is diagonal, we use the resonance approximation. It produces a Hamiltonian H_2 similar to the one in Eq. (17) with the replacement $(h/h\omega) \rightarrow (h/2E_k)$. In resonance (where $\hbar \omega = 2E_{k}$) both approaches yield similar results.

V. RESULTS

Let us calculate the contribution to the magnetization per site of the model caused by the ac field in the ground state (for the sector with all $\alpha_i = -1$ [\[2,12,15\]](#page-5-0)). It is related to the positive sign in front of J_z in ϵ_k . The homogeneous change of the sign of α_j due to the pumping can produce the negative sign

Obviously, in the zero-temperature limit the density matrix ρ_0 is reduced to the ground-state wave function. To calculate the ac field-induced magnetization of the Kitaev system, we use the following relation. For spin operators in the neighboring sites j and j' of the lattice it is easy to show that $(1/2)(S_j^z + S_{j'}^z) = 2[\sqrt{2 + 2\alpha_j} + 2(\sqrt{2 - 2\alpha_j} \sqrt{2 + 2\alpha_j} d_j d_{j'}$]. The proof of the relation uses the definition of spin operators and properties of the fermionic Majorana and Dirac operators. We get $\Delta M^z = M_d^z \exp(-\gamma t)$ in the dynamical regime and $\Delta M^z = M_s^z$ in the steady-state regime, where $\Delta M^z = (g\mu_B/N) \sum_j \langle \Delta S_j^z \rangle$, *N* is the number of sites, the averaging with the density matrix (or with the ground-state wave function at $T = 0$) is used, and ΔM^z is the part of the magnetization of the Kitaev honeycomb spin-1*/*2 model, which depends on *h*. We obtain

$$
M_d^z = \frac{g\mu_B h}{\pi^2 \hbar^2 \omega} \int_0^{\pi} dk_x \int_{-\pi}^{\pi} dk_y
$$

$$
\times \frac{|\Delta_{\mathbf{k}}|^2}{E_{\mathbf{k}} \Omega_{\mathbf{k}}} \Biggl\{ \sin(\omega t) \sin(2\Omega_{\mathbf{k}} t) + \frac{2E_{\mathbf{k}} - \hbar \omega}{\hbar \Omega_{\mathbf{k}}} \cos(\omega t)
$$

$$
\times [1 - \cos(2\Omega_{\mathbf{k}} t)] - \frac{h \epsilon_{\mathbf{k}}}{2\hbar^2 \omega \Omega_{\mathbf{k}}} [1 - \cos(2\Omega_{\mathbf{k}} t)] \Biggr\}. \quad (19)
$$

Here the notation $\hbar\Omega_{\mathbf{k}} = [(E_{\mathbf{k}} - \hbar\omega/2)^2 + (h/\hbar\omega)^2 |\Delta_{\mathbf{k}}|^2]^{1/2}$ is used. In the steady-state regime we obtain

$$
M_s^z = \frac{g\mu_B h}{2\pi^2 \hbar^3 \omega} \int_0^\pi dk_x \int_{-\pi}^\pi dk_y \frac{|\Delta_{\mathbf{k}}|}{(\gamma^2 + \Omega_{\mathbf{k}}^2)}
$$

$$
\times \frac{|\Delta_{\mathbf{k}}|}{E_{\mathbf{k}}} \left[\frac{2E_{\mathbf{k}} - \hbar \omega}{2} \cos(\omega t) + \hbar \gamma \sin(\omega t) - \frac{h\epsilon_{\mathbf{k}}}{2\hbar \omega} \right]. \tag{20}
$$

It is also possible to calculate the power of the ac magnetic field, absorbed by the Kitaev spin model in the ground state. It is determined by

$$
Q \equiv \langle \dot{\mathcal{H}} \rangle_{av} = h_0 \omega \langle \sin(\omega t) \Delta M^z \rangle_{av}, \tag{21}
$$

where

$$
\langle A(t) \rangle_{av} = \lim_{\tau \to \infty} \int_0^{\tau} \langle A(t) \rangle dt. \tag{22}
$$

We see that in the dynamical regime $t \ll \gamma^{-1}$ the magnetization of the spin chain is finite; it oscillates with the frequency *ω* of the ac field, modulated by the Rabi-like low frequencies $\Omega_{\mathbf{k}}$ (in resonance $\hbar \Omega_{\bf k} \sim h$). On the other hand, in the steady-state regime $t \gg \gamma^{-1}$, the relaxation "smears out" the Rabi-like oscillations, with only high-frequency oscillations (with *ω*) remaining. The average value, about which the pumpingcaused contribution to the magnetization oscillates, is small because it is proportional to $(h/\hbar\omega)^2$ in both the dynamical and steady-state regimes. In the dynamical regime the absorption is obviously zero because no energy is taken from the system to the environment, and spins only oscillate. On the other hand, in the steady-state regime, due to the relaxation, we get

$$
Q = \frac{h^2 \gamma}{8\pi^2 h^2} \int_0^{\pi} dk_x \int_{-\pi}^{\pi} dk_y \frac{|\Delta_k|^2}{E_{\mathbf{k}} (\gamma^2 + \Omega_{\mathbf{k}}^2)}.
$$
 (23)

One can see that the absorption (as well as the magnetization) of the honeycomb Kitaev spin-1*/*2 model is also finite for any parameters of the model and frequency of the ac field.

Several limiting cases can be considered. It is important to obtain the one-dimensional limit of the Kitaev honeycomb spin model. It is related to one of J_α equal to zero, in which case the Hamiltonian H_0 describes the set of noninteracting spin chains. For example, for $J_y = 0$ we get $\varepsilon_k = \pm J_z + J_x \cos k_x$ and $\Delta_{\mathbf{k}} = J_x \sin k_x$, with $E_{\mathbf{k}} = [J_x^2 + J_z^2 \pm 2J_x J_z \cos k_x]^{1/2}$, which is equivalent to the Ising spin chain in the effective transverse field $\pm J_z$. Then the integration over k_y in Eqs. (19) – (23) is trivial, and we obtain the result for the response of the Ising spin-1*/*2 chain to the ac field (cf. Refs. [\[16–18\]](#page-5-0)). Similar results can be obtained for the limiting cases $J_x = 0$ and $J_z = 0$.

Also, it is interesting to consider the case in which we average the time dependence of the change of the magnetization with respect to the high-frequency oscillations $\omega \gg \Omega_{\bf k}$. In that case we get for $\omega t \gg 1$ and $\Omega_{\mathbf{k}}t \sim 1$

$$
M_d^z = -\frac{g\mu_B h^2}{\pi^2 h^4 \omega^2} \int_0^\pi dk_x \int_{-\pi}^\pi dk_y |\Delta_\mathbf{k}| \frac{\epsilon_\mathbf{k} |\Delta_\mathbf{k}|}{E_\mathbf{k} \Omega_\mathbf{k}^2} \sin^2(\Omega_\mathbf{k} t) \quad (24)
$$

and

(¯*hγ*)→0

$$
M_s^z = -\frac{g\mu_B h^2}{2\pi^2 h^4 \omega^2} \int_0^\pi dk_x \int_{-\pi}^\pi dk_y \frac{|\Delta_{\mathbf{k}}|}{\left(\gamma^2 + \Omega_{\mathbf{k}}^2\right)} \frac{\epsilon_{\mathbf{k}} |\Delta_{\mathbf{k}}|}{E_{\mathbf{k}}}.
$$
 (25)

These expressions show that in the dynamical regime the lowfrequency addition to the magnetization of the Kitaev model due to the ac field oscillates with the Rabi frequency, while in the steady-state regime the oscillations are "smeared out" by the linear relaxation.

If we take into account that in the steady-state regime

$$
\lim_{\gamma \to 0} \frac{(\hbar \gamma)}{[(\hbar \Omega_{\mathbf{k}})^2 + (\hbar \gamma)^2]} = \pi \delta(\hbar \Omega_{\mathbf{k}}),\tag{26}
$$

one of the the integrations (e.g., in k_y) in Eq. (23) can be realized. Notice that the argument of the δ function is $\Omega_{\mathbf{k}}$, not **k**. This is why we need to use the change in variables for this integration. For small $h/\hbar\omega$ we can obtain the approximate value of the integral (the Jacobi determinant of the change in variables for the integration is equivalent to the density of states of the Hamiltonian of free fermions with the energy E_k). That density of states has features at the edges of the band. We get

$$
Q \approx \frac{h^2 \omega \left[(\hbar \omega)^2 - 4 \left(J_x^2 + J_y^2 - J_z^2 \right) \right]}{16 \pi \hbar J_z^2 |J_x^2 - J_z^2|}
$$

$$
\times \left[4 J_x J_z + \left(J_x^2 - J_z^2 \right) \ln \left(\frac{J_x - J_z}{J_x + J_z} \right)^2 \right]
$$

\n
$$
\Theta \left[2 (J_x + J_y + J_z) - \hbar \omega \right] \Theta \left[\hbar \omega - 2 \min(|J_x - J_y - J_z|, |J_z - J_x - J_y|, |J_y - J_x - J_z|) \right].
$$
\n(27)

Here $\Theta(x)$ are the Heaviside step functions. They show that the resonance absorption can happen only if the half frequency of the ac magnetic field is between the upper and lower boundaries of the zone of fermion excitations E_k of the Kitaev model. The asymmetry of the expression for *Q* with respect to $J_{x,y,z}$ is due to the ac field, which distinguishes the directions. Notice that expression (27) is valid only for $J_{\alpha} \neq 0$

FIG. 1. The contribution of band excitations to the normalized absorption of the ac magnetic field by the Kitaev honeycomb spin-1*/*2 model (the gapless case $|J_x - J_y| \leqslant J_z \leqslant J_x + J_y$) as a function of the frequency ω of the ac field and the coupling J_z . We used the parameters $J_x = 1, J_y = 0.5$.

due to the above approximations. We point out that the result does not depend on the sign of J_z , i.e., on the sign of α_j .

The behavior of the absorbed power is shown in Fig. 1 for the fixed values of two exchange constants as a function of the third one and the frequency of the ac field for E_k gapless. The main contribution to the absorption comes from the region of parameters $|J_z| \sim J_x$. On the other hand, the gapped situation is shown in Fig. 2. The absorption for the gapless case, unlike the gapped one, is possible downward to the lowest values of the frequency. It differs from the known earlier cases (see $[8,16–18]$, where the parametric pumping existed only in spin systems with gapped excitations.

Finally, Fig. 3 describes the frequency dependence of the absorbed power of the parametric pumping by the band states of the Kitaev spin model for the point $J_x = J_y = 1$ and $J_z = 0.5$. It is different from the situation studied in Refs. [\[16–18\]](#page-5-0), where the absorption of the parametric pumping is possible only for $J_x \neq J_y$ [\[14\]](#page-5-0). Spin-spin interactions in the Kitaev model in that sense are similar to the magnetic dipoledipole interaction, where the conservation of the projections of the total spin moment depends on the spatial orientation of bonds between spins. The dependence of the absorption on frequency is nonmonotonic, with two maxima, unlike the case of the gapped excitations.

It is interesting to notice that the absorption is maximal at the upper edge of the band both for the gapped and gapless cases.

In the present study we have taken into account only fermion excitations of the Kitaev honeycomb model. However, it is known that other (topological) excitations can exist at nonzero temperatures. In the ground-state *dynamical* response, the higher-energy (topological) states can also contribute. The analysis of the dynamical structure factors [\[19\]](#page-5-0) implies that

FIG. 2. The frequency dependence of the absorbed power of parametric pumping by band states of the Kitaev honeycomb model for the gapped case with the coupling $J_z > J_x + J_y$ with the parameters $J_x = 1, J_y = 0.5$, and $J_z = 2$.

there can be a contribution from the bound state [\[20\]](#page-5-0) caused by topological excitations with the change of the sign of α_i . The local level of such a bound state is situated much lower [\[19\]](#page-5-0) than the lower edge of the band E_k for the gapped E_k . If the half frequency of the ac field is equal to the energy

FIG. 3. The frequency dependence of the normalized absorbed power of the parametric pumping by the band states of the Kitaev honeycomb model for the case $J_x = J_y = 1$ and $J_z = 0.5$ (gapless excitations).

of that state, the resonance absorption due to that bound state, generally speaking, can contribute at low frequencies to the total absorption of the Kitaev system with gapped fermions. However, the absorption due to that local level also cannot produce parametric instability in the Kitaev spin model. Similar "low"- (with low frequency) and "high"-frequency modes were observed in electron-spin-resonance experiments (although in the standard circular polarization geometry of the ac field and in the ordered phase) in Ir oxide, which has the essential spin-orbit coupling [21], like in Kitaev's model. It turns out that in the one-dimensional limit with one of the exchange constants equal to zero, there is no such bound state for the change in the sign of α_i .

VI. SUMMARY

In summary, we have shown that the response of the two-dimensional quantum spin-liquid system with fermionic excitations to the parametric pumping of ac magnetic field is finite. The result is obtained by adapting the exact integrability of the Kitaev honeycomb model, which permits us to take into account exactly the finite Hilbert space of quantum spins there. Namely, fermionic excitations of the quantum spin liquid, which distinguishes the latter from the magnetically ordered system, are the reason for the absence of accumulation of excitations of the studied system in resonance under parametric pumping. It is different from the linear spin-wave approach within the macroscopic description for magnetically ordered systems [7,8], where the absorption grows with time exponentially because of the accumulation of spin waves (bosons) in resonance under the parametric pumping; that is, the spin-wave approximation cannot be used directly for the description of the parametric pumping of the Kitaev model. We expect the finite response to the parametric pumping to be generic for two-dimensional quantum spin-liquid systems with fermionic excitations, as well as for one-dimensional [16–18,22] and even three-dimensional [\[23\]](#page-6-0) systems with the quantum spin-liquid behavior with fermionic excitations. This prediction can be checked in experiments with the parametric effect of the ac magnetic field on quantum spin systems with Kitaev-like features of their behavior [\[24\]](#page-6-0).

ACKNOWLEDGMENTS

I thank very much R. Moessner for helpful discussions, and the unknown referee for pointing out that not all resonance terms were taken into account in the first version of the work. The support from the Deutsche Forschungsgemeinschaft via SFB 1143, and from the Department for Chemistry of V. N. Karazin Kharkov National University is acknowledged.

- [1] P. A. Lee, [Science](https://doi.org/10.1126/science.1163196) **[321](https://doi.org/10.1126/science.1163196)**, [1306](https://doi.org/10.1126/science.1163196) [\(2008\)](https://doi.org/10.1126/science.1163196); L. Balents, Nature (London) **[464](https://doi.org/10.1038/nature08917)**, [199](https://doi.org/10.1038/nature08917) [\(2010\); Y. Zhou and P. A. Lee,](https://doi.org/10.1038/nature08917) Phys. Rev. Lett. **[106](https://doi.org/10.1103/PhysRevLett.106.056402)**, [056402](https://doi.org/10.1103/PhysRevLett.106.056402) [\(2011\); L. Savary and L. Balents,](https://doi.org/10.1103/PhysRevLett.106.056402) Rep. Progr. Phys. **[80](https://doi.org/10.1088/0034-4885/80/1/016502)**, [016502](https://doi.org/10.1088/0034-4885/80/1/016502) [\(2017\)](https://doi.org/10.1088/0034-4885/80/1/016502).
- [2] A. Kitaev, [Ann. Phys. \(NY\)](https://doi.org/10.1016/j.aop.2005.10.005) **[321](https://doi.org/10.1016/j.aop.2005.10.005)**, [2](https://doi.org/10.1016/j.aop.2005.10.005) [\(2006\)](https://doi.org/10.1016/j.aop.2005.10.005).
- [3] Y. Singh, S. Manni, J. Reuther, T. Berlijn, R. Thomale, W. Ku, S. Trebst, and P. Gegenwart, [Phys. Rev. Lett.](https://doi.org/10.1103/PhysRevLett.108.127203) **[108](https://doi.org/10.1103/PhysRevLett.108.127203)**, [127203](https://doi.org/10.1103/PhysRevLett.108.127203) [\(2012\)](https://doi.org/10.1103/PhysRevLett.108.127203); R. Schaffer, S. Bhattacharjee, and Y. B. Kim, [Phys. Rev. B](https://doi.org/10.1103/PhysRevB.86.224417) **[86](https://doi.org/10.1103/PhysRevB.86.224417)**, [224417](https://doi.org/10.1103/PhysRevB.86.224417) [\(2012\)](https://doi.org/10.1103/PhysRevB.86.224417); K. A. Modic, T. E. Smidt, I. Kimchi, N. P. Breznay, A. Biffin, S. Choi, R. D. Johnson, R. Coldea, P. Watkins-Curry, G. T. McCandless, J. Y. Chan, F. Gandara, Z. Islam, A. Vishwanath, A. Shekhter, R. D. McDonald, and J. G. Analytis, [Nat. Commun.](https://doi.org/10.1038/ncomms5203) **[5](https://doi.org/10.1038/ncomms5203)**, [4203](https://doi.org/10.1038/ncomms5203) [\(2014\)](https://doi.org/10.1038/ncomms5203); L. J. Sandilands, Y. Tian, K. W. Plumb, Y.-J. Kim, and K. S. Burch, [Phys. Rev. Lett.](https://doi.org/10.1103/PhysRevLett.114.147201) **[114](https://doi.org/10.1103/PhysRevLett.114.147201)**, [147201](https://doi.org/10.1103/PhysRevLett.114.147201) [\(2015\)](https://doi.org/10.1103/PhysRevLett.114.147201); T. Takayama, A. Kato, R. Dinnebier, J. Nuss, H. Kono, L. S. I. Veiga, G. Fabbris, D. Haskel, and H. Takagi, *[ibid.](https://doi.org/10.1103/PhysRevLett.114.077202)* **[114](https://doi.org/10.1103/PhysRevLett.114.077202)**, [077202](https://doi.org/10.1103/PhysRevLett.114.077202) [\(2015\)](https://doi.org/10.1103/PhysRevLett.114.077202).
- [4] L. M. Duan, E. Demler, and M. D. Lukin, [Phys. Rev. Lett.](https://doi.org/10.1103/PhysRevLett.91.090402) **[91](https://doi.org/10.1103/PhysRevLett.91.090402)**, [090402](https://doi.org/10.1103/PhysRevLett.91.090402) [\(2003\)](https://doi.org/10.1103/PhysRevLett.91.090402); G. Jackeli and G. Khaliullin, *[ibid.](https://doi.org/10.1103/PhysRevLett.102.017205)* **[102](https://doi.org/10.1103/PhysRevLett.102.017205)**, [017205](https://doi.org/10.1103/PhysRevLett.102.017205) [\(2009\)](https://doi.org/10.1103/PhysRevLett.102.017205); J. Chaloupka, G. Jackeli, and G. Khaliullin, *[ibid.](https://doi.org/10.1103/PhysRevLett.105.027204)* **[105](https://doi.org/10.1103/PhysRevLett.105.027204)**, [027204](https://doi.org/10.1103/PhysRevLett.105.027204) [\(2010\)](https://doi.org/10.1103/PhysRevLett.105.027204).
- [5] R. Karplus and J. Schwinger, [Phys. Rev.](https://doi.org/10.1103/PhysRev.73.1020) **[73](https://doi.org/10.1103/PhysRev.73.1020)**, [1020](https://doi.org/10.1103/PhysRev.73.1020) [\(1948\)](https://doi.org/10.1103/PhysRev.73.1020).
- [6] T. Holstein and H. Primakoff, [Phys. Rev.](https://doi.org/10.1103/PhysRev.58.1098) **[58](https://doi.org/10.1103/PhysRev.58.1098)**, [1098](https://doi.org/10.1103/PhysRev.58.1098) [\(1940\)](https://doi.org/10.1103/PhysRev.58.1098).
- [7] F. R. Morgenthaller, [J. Appl. Phys.](https://doi.org/10.1063/1.1984617) **[31](https://doi.org/10.1063/1.1984617)**, [S95](https://doi.org/10.1063/1.1984617) [\(1960\)](https://doi.org/10.1063/1.1984617); E. Schlömann, J. J. Green, and U. Milano, *[ibid.](https://doi.org/10.1063/1.1984759)* [31](https://doi.org/10.1063/1.1984759), [S386](https://doi.org/10.1063/1.1984759) [\(1960\)](https://doi.org/10.1063/1.1984759).
- [8] [V. E. Zakharov, V. S. L'vov, and S. S. Starobinets,](https://doi.org/10.1070/PU1975v017n06ABEH004404) Sov. Phys. Usp. **[17](https://doi.org/10.1070/PU1975v017n06ABEH004404)**, [896](https://doi.org/10.1070/PU1975v017n06ABEH004404) [\(1975\)](https://doi.org/10.1070/PU1975v017n06ABEH004404).
- [9] [A. A. Zvyagin, Fiz. Nizk. Temp.](https://doi.org/10.1063/1.4931784) **41**, 938 (2015) [Low Temp. Phys. **[41](https://doi.org/10.1063/1.4931784)**, [730](https://doi.org/10.1063/1.4931784) [\(2015\)](https://doi.org/10.1063/1.4931784)].
- [10] L. W. Hinderks and P. M. Richards, [J. Appl. Phys.](https://doi.org/10.1063/1.2163629) **[39](https://doi.org/10.1063/1.2163629)**, [824](https://doi.org/10.1063/1.2163629) [\(1968\)](https://doi.org/10.1063/1.2163629); M. H. Seavey, *[ibid.](https://doi.org/10.1063/1.1657790)* **[40](https://doi.org/10.1063/1.1657790)**, [1997](https://doi.org/10.1063/1.1657790) [\(1969\)](https://doi.org/10.1063/1.1657790); [Phys. Rev. Lett.](https://doi.org/10.1103/PhysRevLett.23.132) **[23](https://doi.org/10.1103/PhysRevLett.23.132)**, [132](https://doi.org/10.1103/PhysRevLett.23.132) [\(](https://doi.org/10.1016/0038-1098(72)90226-8)[1969](https://doi.org/10.1103/PhysRevLett.23.132)[\); W. Jantz, J. Schneider, and B. Andlauer,](https://doi.org/10.1016/0038-1098(72)90226-8) Solid State Commun. **[10](https://doi.org/10.1016/0038-1098(72)90226-8)**, [937](https://doi.org/10.1016/0038-1098(72)90226-8) [\(1972\)](https://doi.org/10.1016/0038-1098(72)90226-8); S. O. Demokritov, V. E. Demidov, O. Dzyapko. G. A. Melkov, A. A. Serga, B. Hillebrands, and A. N. Slavin, [Nature \(London\)](https://doi.org/10.1038/nature05117) **[443](https://doi.org/10.1038/nature05117)**, [430](https://doi.org/10.1038/nature05117) [\(2006\)](https://doi.org/10.1038/nature05117); T. Neumann, A. A. Serga, V. I. Vasyuchka, and B. Hillebrands, Appl. Phys. Lett. **94**, 195202 (2009).
- [11] X.-Y. Feng, G.-M. Zhang, and T. Xiang, [Phys. Rev. Lett.](https://doi.org/10.1103/PhysRevLett.98.087204) **[98](https://doi.org/10.1103/PhysRevLett.98.087204)**, [087204](https://doi.org/10.1103/PhysRevLett.98.087204) [\(2007\)](https://doi.org/10.1103/PhysRevLett.98.087204); H.-D. Chen and J. Hu, [Phys. Rev. B](https://doi.org/10.1103/PhysRevB.76.193101) **[76](https://doi.org/10.1103/PhysRevB.76.193101)**, [193101](https://doi.org/10.1103/PhysRevB.76.193101) [\(2007\)](https://doi.org/10.1103/PhysRevB.76.193101); H.-D. Chen and Z. Nussinov, [J. Phys. A](https://doi.org/10.1088/1751-8113/41/7/075001) **[41](https://doi.org/10.1088/1751-8113/41/7/075001)**, [075001](https://doi.org/10.1088/1751-8113/41/7/075001) [\(2008\)](https://doi.org/10.1088/1751-8113/41/7/075001).
- [12] M. Schmitt and S. Kehrein, [Phys. Rev. B](https://doi.org/10.1103/PhysRevB.92.075114) **[92](https://doi.org/10.1103/PhysRevB.92.075114)**, [075114](https://doi.org/10.1103/PhysRevB.92.075114) [\(2015\)](https://doi.org/10.1103/PhysRevB.92.075114).
- [13] P. Jordan and E. Wigner, [Z. Phys.](https://doi.org/10.1007/BF01331938) **[47](https://doi.org/10.1007/BF01331938)**, [631](https://doi.org/10.1007/BF01331938) [\(1928\)](https://doi.org/10.1007/BF01331938).
- [14] I thank the referee who pointed out this fact.
- [15] E. H. Lieb, [Phys. Rev. Lett.](https://doi.org/10.1103/PhysRevLett.73.2158) **[73](https://doi.org/10.1103/PhysRevLett.73.2158)**, [2158](https://doi.org/10.1103/PhysRevLett.73.2158) [\(1994\)](https://doi.org/10.1103/PhysRevLett.73.2158).
- [16] A. A. Zvyagin and V. M. Tsukernik, Fiz. Nizk. Temp. **11**, 88 (1985) [Sov. J. Low Temp. Phys. **11**, 47 (1985)].
- [17] A. A. Zvyagin, Fiz. Nizk. Temp. **16**, 80 (1990) [Low Temp. Phys. **16**, 41 (1990)].
- [18] A. A. Zvyagin, [Phys. Rev. B](https://doi.org/10.1103/PhysRevB.92.054405) **[92](https://doi.org/10.1103/PhysRevB.92.054405)**, [054405](https://doi.org/10.1103/PhysRevB.92.054405) [\(2015\)](https://doi.org/10.1103/PhysRevB.92.054405).
- [19] J. Knolle, D. L. Kovrizhin, J. T. Chalker, and R. Moessner, [Phys. Rev. Lett.](https://doi.org/10.1103/PhysRevLett.112.207203) **[112](https://doi.org/10.1103/PhysRevLett.112.207203)**, [207203](https://doi.org/10.1103/PhysRevLett.112.207203) [\(2014\)](https://doi.org/10.1103/PhysRevLett.112.207203).
- [20] G. Baskaran, S. Mandal, and R. Shankar, [Phys. Rev. Lett.](https://doi.org/10.1103/PhysRevLett.98.247201) **[98](https://doi.org/10.1103/PhysRevLett.98.247201)**, [247201](https://doi.org/10.1103/PhysRevLett.98.247201) [\(2007\)](https://doi.org/10.1103/PhysRevLett.98.247201).
- [21] S. Bahr, A. Alfonsov, G. Jackeli, G. Khaliullin, A. Matsumoto, T. Takayama, H. Takagi, B. Büchner, and V. Kataev, *Phys. Rev.* B **[89](https://doi.org/10.1103/PhysRevB.89.180401)**, [180401](https://doi.org/10.1103/PhysRevB.89.180401) [\(2014\)](https://doi.org/10.1103/PhysRevB.89.180401).
- [22] A. A. Zvyagin, *Quantum Theory of One-Dimensional Spin Systems* (Cambridge Scientific Publishers, Cambridge, 2010).
- [23] [J. Knolle, D. L. Kovrizhin, J. T. Chalker, and R. Moessner,](https://doi.org/10.1103/PhysRevB.92.115127) Phys. Rev. B **[92](https://doi.org/10.1103/PhysRevB.92.115127)**, [115127](https://doi.org/10.1103/PhysRevB.92.115127) [\(2015\)](https://doi.org/10.1103/PhysRevB.92.115127); A. Smith, J. Knolle, D. L. Kovrizhin, J. T. Chalker, and R. Moessner, *[ibid.](https://doi.org/10.1103/PhysRevB.93.235146)* **[93](https://doi.org/10.1103/PhysRevB.93.235146)**, [235146](https://doi.org/10.1103/PhysRevB.93.235146) [\(2016\)](https://doi.org/10.1103/PhysRevB.93.235146).
- [24] K. W. Plumb, J. P. Clancy, L. J. Sandilands, V. V. Shankar, Y. F. Hu, K. S. Burch, H.-Y. Kee, and Y.-J. Kim, [Phys. Rev. B](https://doi.org/10.1103/PhysRevB.90.041112) **[90](https://doi.org/10.1103/PhysRevB.90.041112)**, [041112](https://doi.org/10.1103/PhysRevB.90.041112) [\(2014\)](https://doi.org/10.1103/PhysRevB.90.041112); J. A. Sears, M. Songvilay, K. W. Plumb,

J. P. Clancy, Y. Qiu, Y. Zhao, D. Parshall, and Y.-J. Kim, *[ibid.](https://doi.org/10.1103/PhysRevB.91.144420)* **[91](https://doi.org/10.1103/PhysRevB.91.144420)**, [144420](https://doi.org/10.1103/PhysRevB.91.144420) [\(2015\)](https://doi.org/10.1103/PhysRevB.91.144420); A. Banerjee, C. A. Bridges, J. Q. Yan, A. A. Aczel, L. Li, M. B. Stone, G. E. Granroth, M. D. Lumsden, Y. Yiu, J. Knolle, S. Bhattacharjee, D. L. Kovrizhin, R. Moessner, D. A. Tennant, D. G. Mandrus, and S. E. Nagler, [Nat. Mater.](https://doi.org/10.1038/nmat4604) **[15](https://doi.org/10.1038/nmat4604)**, [733](https://doi.org/10.1038/nmat4604) [\(2016\)](https://doi.org/10.1038/nmat4604).