Rapid Communications

Mechanism for subgap optical conductivity in honeycomb Kitaev materials

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Motivated by recent terahertz absorption measurements in α-$RuCl_3$, we develop a theory for the electromagnetic absorption of materials described by the Kitaev model on the honeycomb lattice. We derive a mechanism for the polarization operator at *second order* in the nearest-neighbor hopping Hamiltonian. Using the exact results of the Kitaev honeycomb model, we then calculate the polarization dynamical correlation function corresponding to electric dipole transitions in addition to the spin dynamical correlation function corresponding to magnetic dipole transitions.

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Introduction. In Mott insulators, the electronic charge is localized at each site due to Coulomb repulsion, and the low-energy properties are described by the remaining spin and orbital degrees of freedom. Small charge fluctuations subsist due to virtual hopping of the electrons and generate the effective interaction. The same fluctuations can be responsible for a finite effective polarization operator [1–5]. As a result, some magnetic systems can respond to an external ac electric field in a nontrivial way [6–9]. In the case of the simple single-band Hubbard model, such an effect has been calculated at third order in the virtual hopping and is generally predicted only in frustrated lattices [1,2,5].

In this Rapid Communication, motivated by recent experiments of terahertz spectroscopy of α-$RuCl_3$ [10,11], we consider the case of Kitaev materials: multiorbital Mott insulators which are in close proximity to the Kitaev honeycomb model [12]. We show that by introducing additional on-site degrees of freedom, the restriction to frustrated lattices can be lifted, and we derive an effective polarization operator on each bond of the lattice.

The Kitaev honeycomb model is exactly solvable and possesses a quantum spin liquid (QSL) ground state. Its potential realization in real materials through the Jackeli-Khaliullin mechanism [13] attracted much attention in recent years. Exact analytical results for spin correlations have been derived for the Kitaev model [14] and used to predict the signatures of Majorana quasiparticles in inelastic neutron [15–18], Raman [18–20], and resonant x-ray scatterings [21]. As yet, potential Kitaev materials, such as Na_2IrO_3 [13,22–25] and α-$RuCl_3$ [26–31], all eventually reach a magnetically ordered state at sufficiently low temperatures [23–25,29–31], indicating significant deviations from the Kitaev model [32]. Nevertheless, in the case of α-$RuCl_3$, experimental observations of a residual continuum of excitations have been interpreted as remnants of the Kitaev physics [10,33–35].

In the terahertz absorption measurement [10], it is argued that the absorption continuum of α-$RuCl_3$ is too strong to be attributed to direct coupling to magnetic dipole (MD) moments so that there must be a contribution from electric dipole (ED) transitions. We study the response of low-energy excitations of Kitaev materials to an electromagnetic field by deriving a new microscopic mechanism. We show that the interplay of Hund's coupling, spin-orbit coupling (SOC), and a trigonal crystal field (CF) distortion results in a finite polarization operator up to *second order* in the nearest-neighbor hopping term, which is different from previous results obtained only at the third order. This is an important result as it sheds light on a new way to derive an electric polarization of pure electronic origin which is potentially relevant for various multiorbital Mott insulators. We then calculate the optical conductivity at $T = 0$ in the ideal case of a pure Kitaev model by combining analytical and numerical methods. We thus show that the fractionalized low-energy excitations, although emerging from an effective spin Hamiltonian, respond to an external *electric* field.

Model. The Hamiltonian of Kitaev materials has been discussed extensively in the literature [36–44]. The nearly octahedral ligand field strongly splits the e_g orbitals from the t_{2g} ones. For d^5 filling, one hole occupies the three t_{2g} orbitals per site with an effective angular momentum $L = 1$. We thus study a tight-binding model for the holes,

$$\mathcal{H} = \mathcal{H}_{\text{hop}} + \mathcal{H}_{\text{SOC}} + \mathcal{H}_{\text{CF}} + \mathcal{H}_{\text{int}}, \tag{1}$$

which is the sum of the kinetic hopping term, SOC, CF splitting among the t_{2g} orbitals, and the Coulomb and Hund interactions, respectively.

In the present Rapid Communication, we consider Hamiltonians with the full C_3 symmetry (which may be appropriate for α-$RuCl_3$ [39,42]). In addition, we only consider nearest-neighbor hopping processes.

The Hamiltonians are concisely expressed by using the hole operators,

$$\mathbf{c}_i^\dagger = (c_{i,yz,\uparrow}^\dagger, c_{i,yz,\downarrow}^\dagger, c_{i,xz,\uparrow}^\dagger, c_{i,xz,\downarrow}^\dagger, c_{i,xy,\uparrow}^\dagger, c_{i,xy,\downarrow}^\dagger). \tag{2}$$

The kinetic term is $\mathcal{H}_{\text{hop}} = -\sum_{\langle ij \rangle} \mathbf{c}_i^\dagger (\mathbf{T}_{ij} \otimes \mathbb{I}_{2\times 2}) \mathbf{c}_j$, where $\mathbb{I}_{2\times 2}$ is the 2×2 identity matrix and $\mathbf{T}_{ij}$'s are the hopping

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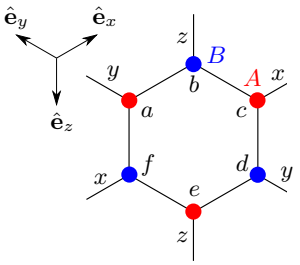

FIG. 1. One hexagon of the honeycomb lattice. The different bond types (x,y,z) are indicated along with their respective unit vectors $\hat{\boldsymbol{e}}_x$, $\hat{\boldsymbol{e}}_y$, and $\hat{\boldsymbol{e}}_z$. The A and B sublattices are colored in red and blue, respectively.

matrices among the d_{yz}, d_{xz}, and d_{xy} orbitals,

$$\mathbf{T}^x = \begin{pmatrix} t_3 & t_4 & t_4 \\ t_4 & t_1 & t_2 \\ t_4 & t_2 & t_1 \end{pmatrix}, \quad \mathbf{T}^y = \begin{pmatrix} t_1 & t_4 & t_2 \\ t_4 & t_3 & t_4 \\ t_2 & t_4 & t_1 \end{pmatrix},$$
$$\mathbf{T}^z = \begin{pmatrix} t_1 & t_2 & t_4 \\ t_2 & t_1 & t_4 \\ t_4 & t_4 & t_3 \end{pmatrix}, \tag{3}$$

where x, y, and z refer the type of the bond considered [see Fig. 1] and $t_{1\text{–}4}$ are the different hopping integrals. The SOC Hamiltonian is given by $\mathcal{H}_{\text{SOC}} = \lambda/2 \sum_{i,a} \mathbf{c}_i^\dagger (L^a \otimes \sigma^a)\mathbf{c}_i$ with $\lambda > 0$, where $(L^a)_{bc} = -i\epsilon_{abc}$ and σ^a are the Pauli matrices. The C_3 symmetric CF splitting of the t_{2g} orbitals corresponds to a trigonal distortion along the axis perpendicular to the plane of the honeycomb lattices $\mathcal{H}_{\text{CF}} = \Delta \sum_i \mathbf{c}_i^\dagger [(\mathbf{L}\cdot\hat{\boldsymbol{n}}_{\text{CF}})^2 \otimes \mathbb{I}_{2\times 2}]\mathbf{c}_i$ with $\hat{\boldsymbol{n}}_{\text{CF}} = [111]$. The interaction Hamiltonian $\mathcal{H}_{\text{int}}$ is the Kanamori Hamiltonian [45–47] with intraorbital Coulomb repulsion U, interorbital repulsion $U' = U - 2J_H$, and Hund's coupling J_H,

$$\begin{aligned}\mathcal{H}_{\text{int}} = {} & U \sum_{i,a} n_{i,a,\uparrow} n_{i,a,\downarrow} + (U' - J_H) \sum_{i,a<b,\sigma} n_{i,a,\sigma} n_{i,b,\sigma} \\ & + U'_{i,a\neq b} n_{i,a,\uparrow} n_{i,b,\downarrow} - J_H \sum_{i,a\neq b} c^\dagger_{i,a,\uparrow} c_{i,a,\downarrow} c^\dagger_{i,b,\downarrow} c_{i,b,\uparrow} \\ & + J_H \sum_{i,a\neq b} c^\dagger_{i,a,\uparrow} c^\dagger_{i,a,\downarrow} c_{i,b,\downarrow} c_{i,b,\uparrow}. \end{aligned} \tag{4}$$

In the limit $\lambda \gg t^2/U$, it is well known (see, e.g., Refs. [39,40]) that at second-order perturbation theory in $\mathcal{H}_{\text{hop}}$, the effective Hamiltonian is the KH$\Gamma\Gamma'$ model which includes the Heisenberg (H) and anisotropic (Γ and Γ') interactions (not shown here) in addition to the Kitaev model (K) described by

$$\mathcal{H}_K = -4J_K \sum_{\langle ij\rangle_\gamma} S_i^\gamma S_j^\gamma, \tag{5}$$

for the effective spins 1/2. The trigonal distortion is usually small, and we treat it as a perturbation ($|\Delta| \ll \lambda$) unless stated otherwise.

Polarization. The on-site Hamiltonian breaks the particle-hole symmetry as $\mathcal{H}_{\text{SOC}}$ and $\mathcal{H}_{\text{CF}}$ are both antisymmetric under the particle-hole transformation. Therefore, in contrast to the single-band Hubbard model [1], a finite polarization operator at second order in $\mathcal{H}_{\text{hop}}$ is not forbidden, even though the lattice is bipartite.

In the atomic limit ($t_{1\text{–}4} = 0$), the system has exactly one hole per site, i.e., $n_i = 1$ for all sites i, where $n_i = \sum_{\alpha=1}^{6} n_{i\alpha}$ (α labels the six t_{2g} states). The polarization operator measures the deviation from this configuration and is defined as $\mathbf{P} = e\sum_i \mathbf{r}_i \delta n_i$, where $\delta n_i = n_i - 1$ and $\mathbf{r}_i$ is the position of site i. Conservation of charge entails $\sum_i \delta n_i = 0$. In the following, we set $e = 1$.

We find the existence of a finite effective polarization operator at the second order in perturbation theory in $\mathcal{H}_{\text{hop}}$ if Δ and J_H are finite. The effective polarization can be written as

$$\mathbf{P}_{\text{eff}} = \sum_{\langle ij\rangle_\gamma} (P_{ij} - P_{ji})\hat{\boldsymbol{e}}_\gamma \equiv \sum_{\langle ij\rangle_\gamma} P_{\langle ij\rangle}\hat{\boldsymbol{e}}_\gamma, \tag{6}$$

where $\hat{\boldsymbol{e}}_\gamma$ is the unit vector along the γ bond connecting the sites $i \in A$ sublattice and $j \in B$ sublattice [see Fig. 1] and P_{ij} is given by perturbation theory. We can furthermore use the symmetry group of the bond $\langle ij\rangle$ to narrow down the possible terms in $P_{\langle ij\rangle}$ [48]. Due to the octahedral CF, the symmetry group of a γ bond of the honeycomb lattice is $\{e, i, C_2(\gamma), m_\perp(\gamma)\}$ whose elements are the identity element, the inversion transformation, the C_2 rotation around $\hat{\boldsymbol{e}}_\gamma$, and the reflection relative to the plane perpendicular to $\hat{\boldsymbol{e}}_\gamma$, respectively. Note that the trigonal CF distortion does not affect the structure of the group. Considering how $S_i^a S_j^b \hat{\boldsymbol{e}}_\gamma$ ($a,b \in \{x,y,z\}$) transforms under the different group elements, Eq. (6) reduces to

$$\mathbf{P}_{\text{eff}} = \sqrt{2}\mathbb{A} \sum_{\langle ij\rangle_\gamma} [\hat{\boldsymbol{e}}_\gamma \cdot (\mathbf{S}_i \times \mathbf{S}_j)]\hat{\boldsymbol{e}}_\gamma. \tag{7}$$

In the basis fixed by the octahedral CF (in which the Kitaev Hamiltonian is written), $\hat{\boldsymbol{e}}_x = (0,1,-1)/\sqrt{2}$, $\hat{\boldsymbol{e}}_y = (-1,0,1)/\sqrt{2}$, and $\hat{\boldsymbol{e}}_z = (1,-1,0)/\sqrt{2}$. Equation (7) is valid for any general real symmetric hopping matrices which preserve the C_3 symmetry. The unitless constant $\mathbb{A}$ is calculated at second order in $\mathcal{H}_{\text{hop}}$ by using the eigenstates of the 15×15 two-hole on-site Hamiltonian. In order to obtain an analytical result, we only keep terms linear in Δ. For $t_1 = t_3 = t_4 = 0$, we find

$$\mathbb{A} = t_2^2 \Delta \left[\frac{128(21\lambda + 8U)}{81\lambda(3\lambda + 2U)^4} J_H + O\left(J_H^2\right) \right], \tag{8}$$

which scales as $t_2^2 \Delta J_H/(U^3\lambda)$ for $U \gg J_H$, $\lambda \gg \Delta$. The full expression (exact in J_H) and the expression including all the hopping integrals $t_{1\text{–}4} \neq 0$ are included in the Supplemental Material [49] together with details about the perturbation theory and numerical calculations of $\mathbb{A}$ exact in Δ.

Only a few hopping processes are possible at second order in $\mathcal{H}_{\text{hop}}$ (see the Supplemental Material [49]). Even when $\Delta = 0$ or $J_H = 0$, different allowed processes contribute to $P_{\langle ij\rangle}$, but they interfere destructively, and their contributions overall vanish. The interference is not completely destructive only when both $\Delta \neq 0$ and $J_H \neq 0$.

Optical conductivity. The spin dynamics of the pure Kitaev model have been investigated thoroughly. However, for Kitaev materials, additional integrability breaking terms are indispensable. In particular, they explain the magnetic ordering

at low temperatures. In this case, even the calculation of the spin structure factor becomes a challenge. Very recent works indicate that the spin dynamics evolve smoothly from the results of the pure Kitaev model using numerical [50–52] and parton mean-field methods [53] close to the QSL regime. In the following, we limit ourselves to the pure Kitaev model to calculate the polarization dynamical response of the fractionalized excitations, expecting that our results are meaningful physically in the putative proximate Kitaev spin liquids. We show that in this limit, the polarization dynamics is remarkably similar to the spin dynamics. The pure Kitaev limit is obtained from the electronic Hamiltonian by keeping only the 90° metal-ligand-metal hoppings ($t_1 = t_3 = t_4 = 0$). Then, for small trigonal CF, the spin Hamiltonian becomes $\mathcal{H}_K + O(\Delta)$. $\mathbf{P}_{\rm eff}$ is itself linear in Δ, therefore we do not need the $O(\Delta)$ correction in the Hamiltonian to calculate the response at first order in Δ.

The optical conductivity along the arbitrary in-plane direction $\hat{\boldsymbol{e}}_\alpha$ at $T = 0$ for $\omega > 0$ is

$$\sigma^\alpha(\omega) = \frac{\omega}{V}\mathrm{Re}\left\{\int_0^\infty dt\, e^{i\omega t}\langle P^\alpha(t)P^\alpha(0)\rangle\right\}, \tag{9}$$

where $\mathbf{P}(t) = e^{i\mathcal{H}t}\mathbf{P}e^{-i\mathcal{H}t}$, $P^\alpha = \mathbf{P}\cdot\hat{\boldsymbol{e}}_\alpha$, and V is the volume of the system. In the effective Kitaev model, we can substitute $\langle P^\alpha(t)P^\alpha(0)\rangle_{\rm Hubbard} \to \langle P^\alpha_{\rm eff}(t)P^\alpha_{\rm eff}(0)\rangle_{\rm Kitaev}$ with $\mathbf{P}_{\rm eff}(t) = e^{i\mathcal{H}_K t}\mathbf{P}_{\rm eff}e^{-i\mathcal{H}_K t}$ where the expectation value is taken with respect to the ground state of the Kitaev Hamiltonian.

For the calculation of $\langle P^\alpha_{\rm eff}(t)P^\alpha_{\rm eff}(0)\rangle$, we need to evaluate spin correlations of the form $\langle S^a_i(t)S^b_j(t)S^c_kS^d_l\rangle$ for pairs of bonds $\langle ij\rangle$ and $\langle kl\rangle$ and for $a,b,c,d \in \{x,y,z\}$. This is reminiscent of Raman scattering in the Kitaev Hamiltonian [19]. However, unlike Raman scattering for which only terms with $a = b$ and $c = d$ are relevant, only terms with $a \neq b$ and $c \neq d$ terms appear in $\langle P^\alpha_{\rm eff}(t)P^\alpha_{\rm eff}(0)\rangle$ due to the antisymmetric nature of $\mathbf{P}_{\rm eff}$. Moreover, we must have either $a = \gamma_{ij}$ or $b = \gamma_{ij}$ and similarly either $c = \gamma_{kl}$ or $d = \gamma_{kl}$.

On each hexagon, a specific product of Pauli matrices $W_p = 2^6 S^y_a S^z_b S^x_c S^y_d S^z_e S^x_f$ (refer to Fig. 1 for site labels) commutes with the Hamiltonian and has eigenvalues ± 1 so that there is a conserved $\mathbb{Z}_2$ flux in each hexagon. Kitaev introduced an enlarged Hilbert space of Majorana fermions [12] in which the spin operators read $2\hat{S}^a_i = i\hat{c}_i\hat{b}^a_i$. We use a hat symbol to indicate that the operators act on the enlarged Hilbert space. The Majorana fermions $\hat{c}_i$ and $\hat{b}^a_i$ are the matter and gauge fermions, respectively. The Kitaev Hamiltonian in terms of Majorana fermions is given by

$$\hat{\mathcal{H}}_K = iJ_K\sum_{\langle ij\rangle_a}\hat{u}_{\langle ij\rangle_a}\hat{c}_i\hat{c}_j, \quad \hat{u}_{\langle ij\rangle_a} = i\hat{b}^a_i\hat{b}^a_j, \tag{10}$$

where $\hat{u}_{\langle ij\rangle_a} = \pm 1$ are constants of motions which fix the $\mathbb{Z}_2$ flux W_p in each hexagon. For a fixed flux pattern, the remaining matter Hamiltonian is quadratic and thus solvable. We further introduce the bond fermions, which are complex fermions defined by ($i \in A$, $j \in B$)

$$\hat{\chi}_{\langle ij\rangle_a} = \tfrac{1}{2}\left(\hat{b}^a_i + i\hat{b}^a_j\right), \quad a = x,y,z. \tag{11}$$

In terms of the bond fermions, the spin operators become

$$\begin{aligned}\hat{S}^a_i &= i\hat{c}_i\left(\hat{\chi}_{\langle ij\rangle_a} + \hat{\chi}^\dagger_{\langle ij\rangle_a}\right)/2 \quad (i \in A)\\ &= \hat{c}_i\left(\hat{\chi}_{\langle ij\rangle_a} - \hat{\chi}^\dagger_{\langle ij\rangle_a}\right)/2 \quad (i \in B).\end{aligned} \tag{12}$$

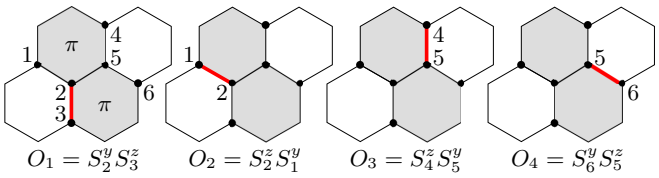

FIG. 2. Four different operators O_i appearing in $\mathbf{P}_{\rm eff}$ which create the same given pair of adjacent π fluxes. The shaded hexagons represent the π fluxes (plaquettes with $W_p = -1$).

In addition to adding a Majorana matter fermion at site i, $\hat{S}^a_i$ changes the bond fermion number of the bond $\langle ij\rangle_a$, which corresponds to the change $\hat{u}_{\langle ij\rangle_a} \to -\hat{u}_{\langle ij\rangle_a}$. Therefore, $\hat{S}^a_i$ adds one π flux to the two plaquettes sharing the bond $\langle ij\rangle_a$, which is also true for $\hat{S}^a_i(t)$, at all times t since all $\hat{u}_{\langle ij\rangle_a}$'s are constants of motion.

We now have a good criterion to identify which $\langle S^a_i(t)S^b_j(t)S^c_kS^d_l\rangle$ terms contribute to the optical conductivity. The expectation value of a product of any operator can be finite only if it does not change the flux in any hexagons. This is a direct consequence of the orthogonality of the subspaces with different flux patterns. Therefore, only combinations of four spin operators which overall leave the flux in each hexagon unchanged are relevant.

For a fixed bond $\langle ij\rangle$, each pair $S^a_iS^b_j$ in $\mathbf{P}_{\rm eff}$ changes the fluxes in two adjacent hexagons. There are four different possible pairs of hexagons, located around the bond $\langle ij\rangle$. Inversely, a fixed pair of adjacent hexagons is affected by four different operators $S^a_iS^b_j$. Let us consider the situation depicted in Fig. 2. The four aforementioned operators are labeled $O_{1\text{–}4}$. The different symmetries of the Kitaev model (mirror symmetries, C_3 symmetry, and inversion symmetry) leave us with only four independent correlation functions $\Gamma_i(t) = 2^4\langle O_1(t)O_i(0)\rangle$ for $i = 1$–4 from which we can calculate the full response,

$$\sigma(\omega) = \frac{e^2\mathbb{A}^2}{\hbar a_\perp}\frac{\omega}{8\sqrt{3}}(2\tilde{\Gamma}_1 - 2\tilde{\Gamma}_3 - \tilde{\Gamma}_2 + \tilde{\Gamma}_4), \tag{13}$$

where $\tilde{\Gamma}_i(\omega) = \mathrm{Re}\{\int_0^\infty dt\, e^{i\omega t}\Gamma_i(t)\}$. The calculated optical conductivity is independent of the direction of $\hat{\boldsymbol{e}}_\alpha$ and therefore isotropic on the plane.

Two different matter Hamiltonians are needed to calculate $\Gamma_{1\text{–}4}$ (see the Supplemental Material [49]): the flux-free Hamiltonian $\hat{\mathcal{H}}_0$ (all $\hat{u}_{\langle ij\rangle} = 1$'s) and the two-flux Hamiltonian $\hat{\mathcal{H}}'$, whose π fluxes correspond to those depicted in Fig. 2 ($\hat{u}_{\langle ij\rangle} = 1$ expected for $\hat{u}_{\langle 25\rangle} = -1$). The ground state of the full Kitaev Hamiltonian is in the flux-free sector [54] so that it is given by the ground-state $|M_0\rangle$ of $\hat{\mathcal{H}}_0$. Interestingly, the calculation of the simpler spin-spin dynamical correlation functions requires the same Hamiltonian $\hat{\mathcal{H}}'$ [14,15], implying that the magnetic dipole and electric dipole transitions take place between the same flux sectors. Using the Lehmann spectral representation, we generally define the operator,

$$\Pi_{ab}(\omega) = \pi\sum_\lambda\langle M_0|c_a|\lambda\rangle\langle\lambda|c_b|M_0\rangle\delta(\omega - \Delta_\lambda), \tag{14}$$

where $|\lambda\rangle$'s are the eigenvectors of $\hat{\mathcal{H}}'$ with energy E_λ and $\Delta_\lambda = E_\lambda - E_0$. We find

$$\tilde{\Gamma}_1 = \Pi_{33}, \quad \tilde{\Gamma}_2 = -\Pi_{31}, \quad \tilde{\Gamma}_3 = i\Pi_{34}, \quad \tilde{\Gamma}_4 = -i\Pi_{36}, \tag{15}$$

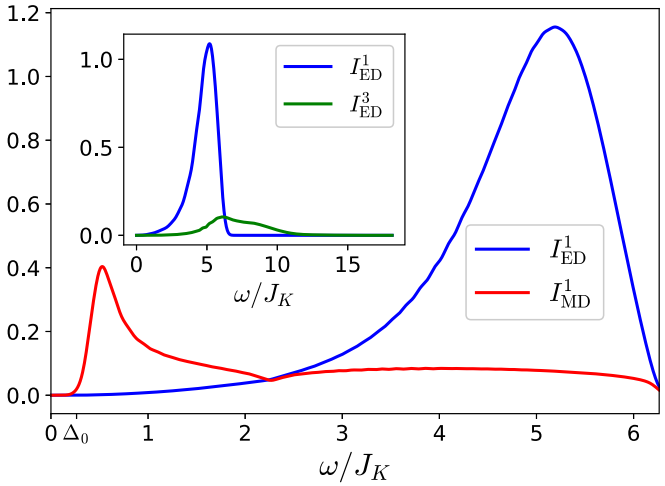

FIG. 3. Single-particle responses $I^1_{\rm ED} = \sigma(\omega)$ in units of $e^2\mathbb{A}^2/(\hbar a_\perp)$ and $I^1_{\rm MD}$ where we set $R = (\mathbb{A}/g)^2 \times (a/\lambda\!\!\!^{-})^2$ to 0.5. The inset: single- and three-particle responses $I^1_{\rm ED}$ and $I^3_{\rm ED}$.

where the matter fermions are labeled according to Fig. 2. The imaginary part of the spin susceptibility can be written as $\chi''(\omega) \propto (\Pi_{22} + \Pi_{55} - i\Pi_{25} + i\Pi_{52})$ [15]. Using complex matter fermions and the appropriate Bogoliubov transformations, the Hamiltonians become

$$\hat{\mathcal{H}}_0 = \sum_{n>0} \omega_n a_n^\dagger a_n + E_0, \quad \hat{\mathcal{H}}' = \sum_{n>0} \omega'_n b_n^\dagger b_n + E'_0, \qquad (16)$$

where $E_0 = -\frac{1}{2}\sum_{n>0}\omega_n$ and $E'_0 = -\frac{1}{2}\sum_{n>0}\omega'_n$. Even though all the $|\lambda\rangle$ states are defined in the same flux sector, we find that the states that contribute to $\sigma(\omega)$ and $\chi''(\omega)$ are mutually exclusive. This can be explained using symmetries of the Hamiltonians $\hat{\mathcal{H}}_0$ and $\hat{\mathcal{H}}'$ (see the Supplemental Material [49] and Ref. [55]).

The electric dipole and magnetic dipole response functions were numerically calculated in systems of sizes up to 82×82 unit cells. As mentioned in Refs. [15,18,19], the leading contribution comes from the single-particle states $|\lambda\rangle = b_\lambda^\dagger |M'\rangle$, where $|M'\rangle$ is the ground state of $\mathcal{H}'$ which satisfies $b_\lambda |M'\rangle = 0$. We verified this property by calculating the single- and three-particle responses of a 20×20 system (see the Supplemental Material [49]), shown in the inset of Fig. 3.

The electric and magnetic dipole absorption rates scale as $\sigma(\omega)$ and $\omega(g\mu_B/c)^2\chi''(\omega)$, respectively, where μ_B is the Bohr magneton and g is the effective Landé g factor so that we set $I_{\rm ED} = \sigma(\omega)$ and $I_{\rm MD} = \omega(g\mu_B/c)^2\chi''(\omega)$. The intensities are related by the ratio $R = (\mathbb{A}/g)^2 \times (a/\lambda\!\!\!^{-})^2$, where a is the spacing between the transition-metal atoms on the honeycombs and $\lambda\!\!\!^{-}$ is the reduced Compton wavelength. In the literature, a wide range of values for the different physical parameters has been reported, resulting in different values for $\mathbb{A}$. For α-$RuCl_3$, the ratio R roughly ranges between 0.01 and 10. Figure 3 shows $I_{\rm ED}$ in units of $e^2\mathbb{A}^2/(\hbar a_\perp)$ and the corresponding $I_{\rm MD}$ where the ratio R is arbitrarily set to 0.5. Around $\omega = \Delta_0$, $I_{\rm MD}$ seems to be dominant. However, $I_{\rm MD}$ is of course independent of $\mathbb{A}$, and its calculated value (with $g = 2$) can only account for about 7% of the measured signal just above the sharp gap in Ref. [10]. With a ratio of $R = 10$, the order of magnitude of $I_{\rm ED}$ is comparable to the measured quantity, but the sharp gap at Δ_0 disappears as $I_{\rm MD}$ becomes negligible.

Discussion. We showed that the complex interplay of the Hund's coupling, SOC, and a trigonal CF distortion results in a nontrivial polarization operator originating from nearest-neighbor hopping processes, shedding some light on an unexpected charge fluctuation mechanism in Kitaev materials. By calculating the effective polarization operator and its dynamical correlation function, we determined the electric dipole absorption spectrum originating from the pure Kitaev model. This shows that, like other spin liquids with a continuum of low-energy excitations [6–9], the fractionalized magnetic excitations respond to an external ac electric field. As measured in the terahertz absorption measurements of α-$RuCl_3$ [10], the electric dipole spectral weight is expected to dominate over the magnetic dipole one. Our results for the optical conductivity in Fig. 3 are valid for the pure Kitaev model. However, the derived polarization operator (7) is valid even in the effective KH$\Gamma\Gamma'$ model (only the coefficient $\mathbb{A}$ is affected). Therefore, we expect the optical response to be modified smoothly when introducing integrability breaking terms in the proximity of the QSL regime as for the spin structure factor [50–53]. Nonetheless, substantial changes in the spin Hamiltonian, such as a large Γ term (expected in real materials), most probably significantly alter the calculated response [17].

Additionally, other corrections can potentially affect the optical conductivity in real materials, such as longer-range hopping or breaking of the C_3 symmetry, which should explain the dependence on the direction of the probing ac field.

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