Supplemental Material of "Mechanism for sub-gap optical conductivity in honeycomb Kitaev materials"

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PERTURBATION THEORY

First we derive the expression for the effective polarization Eq. (7) using second order perturbation in $\mathcal{H}_{\text{hop}}$ and treating all other Hamiltonian exactly. We also show explicitly the different hopping processes involved.

In the unperturbed Hilbert space of an N-site system, with $\mathcal{H}_{\text{hop}} = 0$, the 2^N degenerate eigenstates are the magnetic states $|\phi_n\rangle$, with exactly one hole per site. The perturbation lift the degeneracy and the new 2^N low-energy eigenstates $|\psi_n\rangle$ are adiabatically connected to the magnetic states, such that there are connected by a unitary transformation: $|\psi_i\rangle = e^{-S}|\phi_i\rangle$, where S is antihermitian.

For any observable $\mathcal{O}$ defined in the full Hilbert space, an effective low-energy operator $\mathcal{O}_{\text{eff}}$ can be defined by projecting in the subspace spanned by $\{|\psi_n\rangle\}$: $\mathcal{O}_{\text{eff}} = \mathbb{P}_\phi e^S \mathcal{O} e^{-S} \mathbb{P}_\phi$, where $\mathbb{P}_\phi$ is the projection operator onto the unperturbed magnetic Hilbert space. An equivalent definition in terms of individual matrix element is

$$\langle \phi_m | \mathcal{O}_{\text{eff}} | \phi_n \rangle \equiv \langle \psi_m | \mathcal{O} | \psi_n \rangle , \tag{S1}$$

which we use to calculate the effective polarization operator.

Without the trigonal distortion Δ, the magnetic states are split into effective $J = 1/2$ and $3/2$ states [S1–S4] that we denote $|J, M_J\rangle$. They are given by ($X = yz$, $Y = xz$, $Z = xy$)

$$\begin{aligned}
\left|\frac{1}{2}, \frac{1}{2}\right\rangle &= \frac{1}{\sqrt{3}}(-|d_X,\downarrow\rangle - i|d_Y,\downarrow\rangle - |d_Z,\uparrow\rangle) \\
\left|\frac{1}{2}, -\frac{1}{2}\right\rangle &= \frac{1}{\sqrt{3}}(-|d_X,\uparrow\rangle + i|d_Y,\uparrow\rangle + |d_Z,\downarrow\rangle) \\
\left|\frac{3}{2}, \frac{3}{2}\right\rangle &= \frac{1}{\sqrt{2}}(-|d_X,\uparrow\rangle - i|d_Y,\uparrow\rangle) \\
\left|\frac{3}{2}, \frac{1}{2}\right\rangle &= \frac{1}{\sqrt{6}}(-|d_X,\downarrow\rangle - i|d_Y,\downarrow\rangle + 2|d_Z,\uparrow\rangle) \\
\left|\frac{3}{2}, -\frac{1}{2}\right\rangle &= \frac{1}{\sqrt{6}}(|d_X,\uparrow\rangle - i|d_Y,\uparrow\rangle + 2|d_Z,\downarrow\rangle) \\
\left|\frac{3}{2}, -\frac{3}{2}\right\rangle &= \frac{1}{\sqrt{2}}(|d_X,\downarrow\rangle - i|d_Y,\downarrow\rangle).
\end{aligned} \tag{S2}$$

Note that the relative sign between the states is not always consistent in the literature. It is not relevant when only interested in the Hamiltonian, but it is for the polarization operator. We consistently choose the states such that $J^-|1/2, 1/2\rangle = |1/2, -1/2\rangle$, and similarly for the $J = 3/2$ states. The different hopping processes appearing in the perturbation theory at second order in $\mathcal{H}_{\text{hop}}$ can then be written schematically using those states. However, we want to track the same processes when $\Delta \neq 0$. As the e_g orbitals have been cast away, the quantization axis in Eq. (S2) is arbitrary. It is usually chosen to be the z axis of the octahedral environment but it does not need to. If the CF distortion direction coincides with the quantization axis of M_J, the structure of Eq. (S2) is left unchanged, even though J and M_J are no longer good quantum numbers. The eigenstates can still be labelled ϕ^J_M such that

$\phi^J_M \to |J,M\rangle$ when $\Delta \to 0$. They are given by (see [S1, S5])

$$\begin{aligned}
\phi^{1/2}_{1/2} &= \frac{1}{\sqrt{2}}\cos\theta(-|d_X,\downarrow\rangle - i\,|d_Y,\downarrow\rangle) - \sin\theta\,|d_Z,\uparrow\rangle \\
\phi^{1/2}_{-1/2} &= \frac{1}{\sqrt{2}}\cos\theta(-|d_X,\uparrow\rangle + i\,|d_Y,\uparrow\rangle) + \sin\theta\,|d_Z,\downarrow\rangle \\
\phi^{3/2}_{3/2} &= \frac{1}{\sqrt{2}}(-|d_X,\uparrow\rangle - i\,|d_Y,\uparrow\rangle) \\
\phi^{3/2}_{1/2} &= \frac{1}{\sqrt{2}}\sin\theta(-|d_X,\downarrow\rangle - i\,|d_Y,\downarrow\rangle) + \cos\theta\,|d_Z,\uparrow\rangle \\
\phi^{3/2}_{-1/2} &= \frac{1}{\sqrt{2}}\sin\theta(|d_X,\uparrow\rangle - i\,|d_Y,\uparrow\rangle) + \cos\theta\,|d_Z,\downarrow\rangle \\
\phi^{3/2}_{-3/2} &= \frac{1}{\sqrt{2}}(|d_X,\downarrow\rangle - i\,|d_Y,\downarrow\rangle),
\end{aligned} \tag{S3}$$

where $\tan(2\theta) = 2\sqrt{2}\frac{\lambda}{\lambda-2\Delta}$. When $\Delta = 0$, $\sin\theta = 1/\sqrt{3}$. The block-diagonal structure of the Kanamori Hamiltonian (4) is also left invariant [S5], so that the different hopping processes are the same with and without Δ. For Kitaev materials, the CF distortion direction is $\hat{\mathbf{n}}_{\rm CF} = [111]$. We thus rotate the orbital and spin angular momentum so that $[111] \to [001]$, corresponding to the SU(2) unitary transformation U and the SO(3) rotation R. The pay-off is that the hopping matrices now become $\mathbf{T}^\gamma \to R\mathbf{T}^\gamma R^{-1}$. Note that the final expression with respect to the effective spin operators has to be rotated back to the original frame to be consistent with Eq. (7).

The exact eigenstates of the full Hamiltonian can always be decomposed in a magnetic state and a polar state (with one or more doubly occupied site): $|\psi_m\rangle = \alpha\,|\phi_m\rangle + \beta\,|p_m\rangle$, where $\alpha \simeq 1$. Only the polar states are relevant for the polarization operator, $\langle\psi_m|\mathbf{P}|\psi_n\rangle \propto \langle p_m|\mathbf{P}|p_n\rangle$. To calculate $\mathbf{P}_{\rm eff}$ at second order in $\mathcal{H}_{\rm hop}$, all we need are the polar states at first order in $\mathcal{H}_{\rm hop}$,

$$|p_m\rangle_1 = \sum_{p\in\text{polar}} \frac{\langle p|\mathcal{H}_{\rm hop}|\phi_m\rangle}{E_0 - E_p}\,|p\rangle\,, \tag{S4}$$

from which we deduce Eq. (6) of the main text and

$$P_{ij} = \mathbb{P}\mathcal{H}^{ij}_{\rm hop}\mathbb{Q}\frac{1}{(E_0-\mathcal{H}_0)^2}\mathbb{Q}\mathcal{H}^{ji}_{\rm hop}\mathbb{P}. \tag{S5}$$

where $\mathcal{H}_0$ is the unperturbed Hamiltonian with ground state energy E_0, and $\mathbb{P}$ and $\mathbb{Q}$ denote the projection operators onto the low-energy subspace made of effective 1/2-spins, and the polar states (with doubly occupied sites), respectively. The analytical calculation is too heavy, so that we only calculated the $\mathbb{A}$ coefficient numerically, shown as a function of Δ in Fig. S1.

In order to obtain an analytical expression, we treat both $\mathcal{H}_{\rm hop}$ and $\mathcal{H}_{\rm CF}$ as perturbations. The ground state is thus constituted of the pure $J = 1/2$ states (S2) (the rotation $\hat{\mathbf{n}}_{\rm CF} \to [001]$ is unnecessary).

Up to third order, only terms scaling as $O(t^2\Delta)$ are relevant and we need to calculate polar states at first order in both $\mathcal{H}_{\rm hop}$ and $\mathcal{H}_{\rm CF}$. We obtain

$$P_{ij} = \left[-\frac{2}{3\lambda}\mathbb{P}_{\frac12}\mathcal{H}^{ij}_{\rm hop}\frac{\mathbb{Q}}{(E_0-\mathcal{H}_0)^2}\mathcal{H}^{ji}_{\rm hop}\mathbb{P}_{\frac32}\mathcal{H}_{\rm CF}\mathbb{P}_{\frac12} + \mathbb{P}_{\frac12}\mathcal{H}^{ij}_{\rm hop}\frac{\mathbb{Q}}{(E_0-\mathcal{H}_0)^2}\mathcal{H}_{\rm CF}\frac{\mathbb{Q}}{E_0-\mathcal{H}_0}\mathcal{H}^{ji}_{\rm hop}\mathbb{P}_{\frac12}\right] + \text{H.c.}, \tag{S6}$$

where $\mathbb{P}_{\frac12}$, $\mathbb{P}_{\frac32}$ and $\mathbb{Q}$ are the projection operators on the $J = 1/2$ states, the $J = 3/2$ states, and the polar states, respectively. The full expression of Eq. (8) of the main text is $\mathbb{A} = \frac{128}{81}\Delta t^2 J_H\frac{P}{Q}$ with

$$\begin{aligned}
P =& -16(J_H-U)(2J_H-U)(3J_H-U)^3 + 2\lambda(U-3J_H)^2\left(379J_H^2 - 404J_HU + 105U^2\right) \\
&- 3\lambda^2(3J_H-U)\left(1847J_H^2 - 1650J_HU + 363U^2\right) + 3\lambda^3\left(6004J_H^2 - 4743J_HU + 927U^2\right) \\
&+ 27\lambda^4(129U - 334J_H) + 1701\lambda^5 \\
Q =& \lambda(-6J_H + 3\lambda + 2U)^2\left(6J_H^2 - J_H(17\lambda + 8U) + (3\lambda+U)(3\lambda+2U)\right)^3.
\end{aligned} \tag{S7}$$

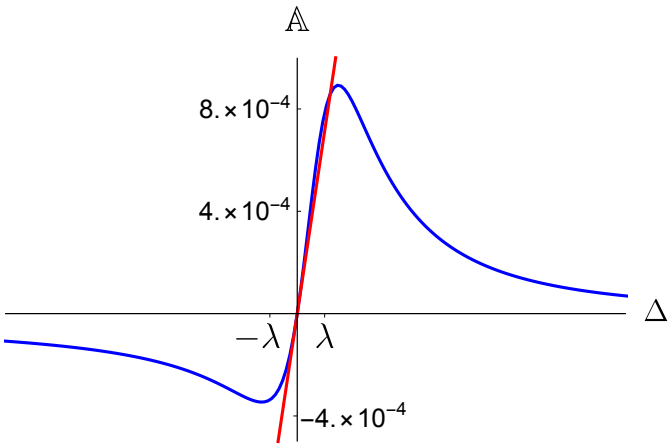

FIG. S1. Coefficient $\mathbb{A}$ as a function of Δ. Red line: analytical result linear in Δ. Blue line: calculation exact in Δ. $U = 3000$, $J_H = 600$, $\lambda = 150$, $t = 100$.

For the more general hopping matrices which preserve the C_3 symmetry,

$$\mathbf{T}^x = \begin{pmatrix} t_3 & t_4 & t_4 \\ t_4 & t_1 & t_2 \\ t_4 & t_2 & t_1 \end{pmatrix}, \mathbf{T}^y = \begin{pmatrix} t_1 & t_4 & t_2 \\ t_4 & t_3 & t_4 \\ t_2 & t_4 & t_1 \end{pmatrix}, \mathbf{T}^z = \begin{pmatrix} t_1 & t_2 & t_4 \\ t_2 & t_1 & t_4 \\ t_4 & t_4 & t_3 \end{pmatrix}, \tag{S8}$$

we find

$$\mathbb{A} = \Delta(t_1+t_2-t_3-t_4)\left[\frac{128}{81}\frac{8U^3(t_1+t_2+t_3)+3\lambda U^2(11t_1+7t_2+9t_3)+24\lambda^2U(2t_1+t_3)+9\lambda^3(2t_1+t_3)}{\lambda U^2(3\lambda+2U)^4}J_H+O(J_H^2)\right]. \tag{S9}$$

The full expression can also be written as a fraction of polynomials but we do not write it explicitly. We see that even with more general hopping matrices, the effective polarization vanishes when either $J_H = 0$ or $\Delta = 0$. (This is still true with hopping matrices breaking the C_3 symmetry.) Numerical calculations show that when $t_1 + t_2 - t_3 - t_4 = 0$ the polarization vanishes, even when treating Δ exactly.

Let us now consider explicitly the hopping processes. Without the CF distortion Δ, the processes leading to the Kitaev Hamiltonian are handily visualized by choosing the usual quantization axis of M_J (along [001]) and considering the hopping matrices with only $t_2 \neq 0$. The only four possible processes for a z-bond are shown in Fig. S2, up to the exchange of the two sites. The mechanism is Ising-like as no spin flips are possible [S6]. The Hund's coupling J_H is responsible for the Ising ferromagnetism ($J_K > 0$). When $J_H = 0$, the effective Hamiltonian is proportional to the identity ($J_K = 0$).

Due to spatial inversion symmetry, only transitions between the singlet state and a triplet state are relevant to the effective polarization. With $\Delta = 0$, the processes (c_0) and (d_0) actually individually allow such transitions, between the singlet and the $M_J = 0$ triplet. However, they interfere destructively, resulting in a vanishing $\mathbf{P}_{\text{eff}}$.

We now consider the more delicate case $\Delta \neq 0$. Therefore, we switch to the states (S3), for which M_J is quantized along [111]. We will only consider transitions between the triplet state $|\uparrow,\uparrow\rangle$ and the singlet state (if the polarization is considered) or the $M_J = 0$ triplet state (if the Hamiltonian is considered).

The different processes are represented graphically in Fig. S3. Note that as the hopping matrices are rotated, no additional processes appear when considering the more general hopping matrices of Eq. (S8). As before, when $\Delta = 0$, the processes (a)-(g) of Fig. S3 interfere destructively. However, when both $\Delta \neq 0$ and $J_H \neq 0$, the processes (a)-(g) do not completely cancel and $\mathbf{P}_{\text{eff}}$ is finite. Te process (h) is only possible when $\Delta \neq 0$ and $J_H \neq 0$ and also contribute to $\mathbf{P}_{\text{eff}}$ (though its relative amplitude is small compared to the other processes).

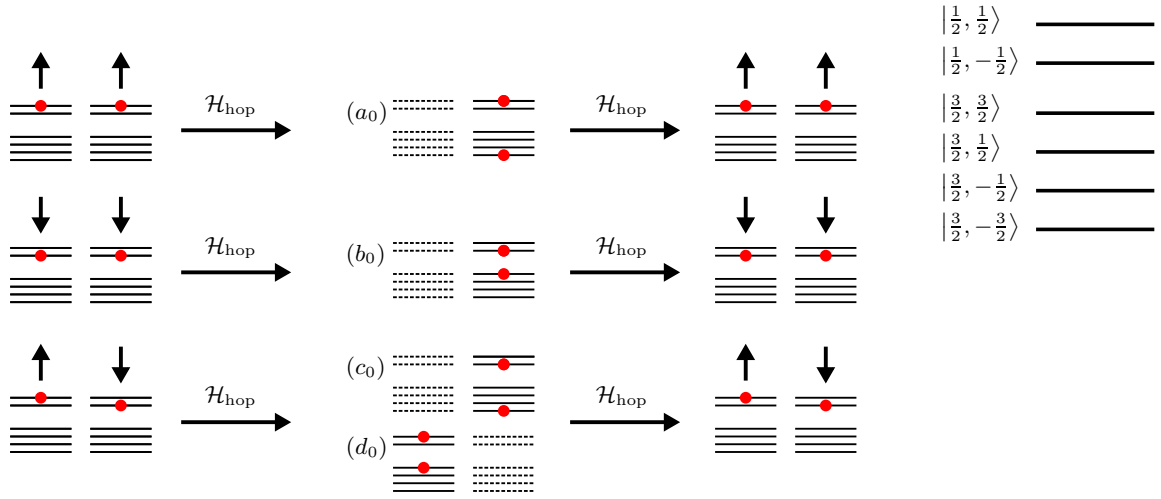

FIG. S2. Different allowed hopping processes for a z bond without the trigonal CF distortion, leading to the Kitaev Hamiltonian. Here, M_J is quantized along the usual z axis [001] of the octahedral environment (in which the Kitaev Hamiltonian is written) as $\Delta = 0$.

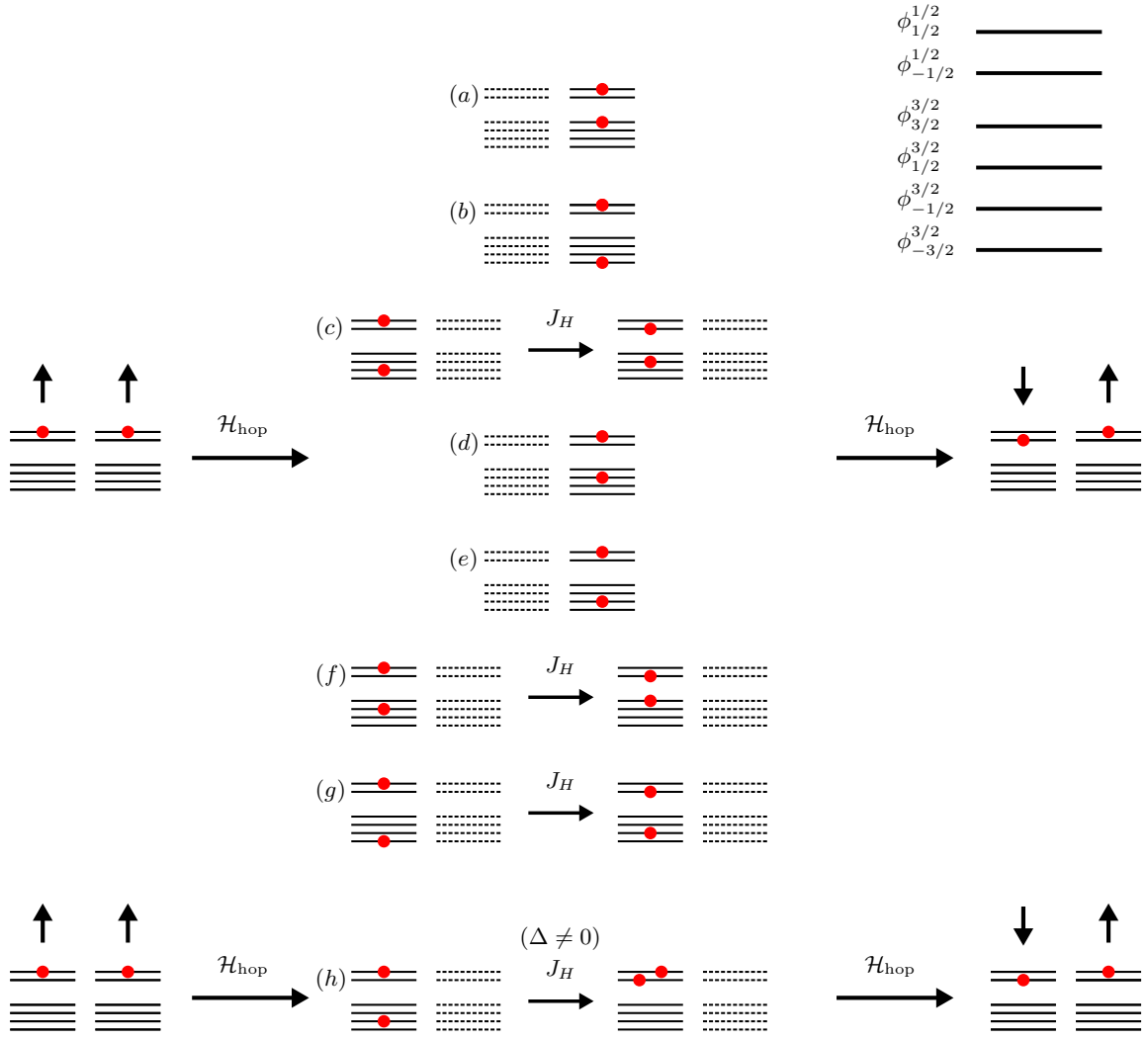

FIG. S3. Different allowed hopping processes. The process (h) is only possible when $\Delta \neq 0$. Here M_J is quantized along $\hat{\mathbf{n}}_{\rm CF} = [111]$ and not the z axis [001] of the octahedral environment.

4-SPIN DYNAMICAL CORRELATION FUNCTION OF THE KITAEV HAMILTONIAN

Here we derive the expressions for the different correlation functions Γ_i given in Eqs. (14) and (15). In the Majorana representation,

$$\begin{aligned}
\Gamma_1(t) &= \langle M_0| \langle F_0| \sigma_2^y(t)\sigma_3^z(t)\sigma_2^y(0)\sigma_3^z(0) |F_0\rangle |M_0\rangle \\
\Gamma_2(t) &= \langle M_0| \langle F_0| \sigma_2^y(t)\sigma_3^z(t)\sigma_2^z(0)\sigma_1^y(0) |F_0\rangle |M_0\rangle \\
\Gamma_3(t) &= \langle M_0| \langle F_0| \sigma_2^y(t)\sigma_3^z(t)\sigma_4^z(0)\sigma_5^y(0)P |F_0\rangle |M_0\rangle \\
\Gamma_4(t) &= \langle M_0| \langle F_0| \sigma_2^y(t)\sigma_3^z(t)\sigma_6^y(0)\sigma_5^z(0)P |F_0\rangle |M_0\rangle ,
\end{aligned} \tag{S10}$$

where $|F_0\rangle |M_0\rangle$ is the ground state of the Kitaev Hamiltonian $\mathcal{H}_K$ decomposed into the gauge and matter sectors, such that $\hat{u}_{\langle ij\rangle} |F_0\rangle = |F_0\rangle$. P is the projector onto the physical Hilbert space, defined by $D_j |\Psi\rangle_{\text{phys}} = |\Psi\rangle_{\text{phys}}$ where $D_j = c_j b_j^x b_j^y b_j^z$. It can be shown that P is only needed when some of the bond fermion number operators $\chi^\dagger_{\langle ij\rangle_a}\chi_{\langle ij\rangle_a} = (\hat{u}_{\langle ij\rangle_a} + 1)/2$ are not conserved (even though the $\mathbb{Z}_2$ fluxes are). This is the case for Γ_3 and Γ_4. Note that P commutes with the spin operators.

The general strategy is to calculate separately the expectation value in the gauge sector and the matter sector. In terms of Majorana fermions,

$$\Gamma_1 = \langle M_0| \langle F_0| e^{i\mathcal{H}_0 t}(ic_2\chi^\dagger_{21})(-c_3\chi^\dagger_{23})e^{-i\mathcal{H}t}(ic_2\chi_{21})(c_3\chi_{23}) |F_0\rangle |M_0\rangle , \tag{S11}$$

where $\mathcal{H}_0$ refers to the matter fermion Hamiltonian with all $u_{\langle ij\rangle_a} = 1$. We now use the important relation

$$\chi^\dagger_{\langle ij\rangle_\gamma} e^{-i\mathcal{H}t} = e^{-i(\mathcal{H}+V_{ij})t}\chi^\dagger_{\langle ij\rangle_\gamma}, \tag{S12}$$

where $V_{ij} = -2iJ_K c_i c_j$. This implies that $\mathcal{H}_0 + V_{ij}$ is the Hamiltonian with all bond operators $\hat{u} = 1$ except on the bond $\langle ij\rangle_\gamma$ where $u_{\langle ij\rangle_\gamma} = -1$. Therefore,

$$\begin{aligned}
\Gamma_1 &= \langle M_0| e^{i\mathcal{H}_0 t}c_2c_3e^{-i(\mathcal{H}_0+V_{21}+V_{23})t}c_2c_3 |M_0\rangle \langle F_0| \chi^\dagger_{21}\chi^\dagger_{23}\chi_{21}\chi_{23} |F_0\rangle \\
&= -\langle M_0| e^{i\mathcal{H}_0 t}c_2c_3e^{-i(\mathcal{H}_0+V_{21}+V_{23})t}c_2c_3 |M_0\rangle
\end{aligned} \tag{S13}$$

For the Kitaev Hamiltonian in a general flux sectors characterized by the set $\{u_{\langle ij\rangle_\gamma}\}$, noted $\mathcal{H}(\{u\})$, we have the relation

$$c_i\mathcal{H}(\{u\})c_i = \mathcal{H}(\{\tilde{u}\}), \qquad \text{where} \quad \tilde{u}_{\langle mn\rangle} = \begin{cases} -u_{\langle mn\rangle} & \text{if } i = m \text{ or } n \\ u_{\langle mn\rangle} & \text{else} \end{cases} . \tag{S14}$$

Together with the relation $c_i e^O c_i = e^{c_i O c_i}$, we have

$$\Gamma_1 = \langle M_0| e^{i\mathcal{H}_0 t}c_3e^{-i\mathcal{H}'t}c_3 |M_0\rangle . \tag{S15}$$

with $\mathcal{H}' = \mathcal{H}_0 + V_{25}$. We could further simplify $c_3e^{-i\mathcal{H}'t}c_3 = e^{-i\mathcal{H}''t}$, but we do not for the following reason. $\mathcal{H}_0$, $\mathcal{H}'$ and $\mathcal{H}''$ are all Hamiltonians in the matter Hilbert space with a fixed bond fermion parity. However, the matter parities of their respecting ground states do not necessarily match. For example, in the general Kitaev Hamiltonian with different parameters for the three bonds (J_K^X, J_K^Y and J_K^Z), the parity of the ground state in a fixed gauge sector depends on the values of the three parameters (see Ref. [S7]). Numerically, we find that the ground state of $\mathcal{H}_0$ ($|M_0\rangle$) has the same parity as that of $\mathcal{H}'$ ($|M'\rangle$) and that the ground state of $\mathcal{H}''$ ($|M''\rangle$) has the opposite parity so that $\langle M_0|M''\rangle = 0$. For this reason, we work with $\mathcal{H}'$ so that we can find a relation between $|M_0\rangle$ and $|M'\rangle$ explicitly, which will then be used in a Bogoliubov transformation.

For Γ_2 we similarly find

$$\Gamma_2 = -\langle M_0| e^{i\mathcal{H}_0 t}c_3e^{-i\mathcal{H}'t}c_1 |M_0\rangle . \tag{S16}$$

For Γ_3 and Γ_4 we need to add the projection operator P. Here, it is enough to replace P with D_2D_5 [S8, S9] , which reads

$$D_2D_5 = -iu_{\langle 25\rangle_x}c_2c_5(\chi^\dagger_{21} + \chi_{21})(\chi^\dagger_{23} + \chi_{23})(\chi^\dagger_{65} - \chi_{65})(\chi^\dagger_{45} - \chi_{45}). \tag{S17}$$

Therefore,

$$\begin{aligned}\Gamma_3 =& i \langle M_0| e^{i\mathcal{H}_0 t} c_2 c_3 e^{-i(\mathcal{H}_0+V_{21}+V_{23})t} c_4 c_5 c_2 c_5 |M_0\rangle \langle F_0| \chi_{21}^\dagger \chi_{23}^\dagger \chi_{45}^\dagger \chi_{65}^\dagger \chi_{21}\chi_{23}\chi_{65}\chi_{45} |F_0\rangle \\ =& - i \langle M_0| e^{i\mathcal{H}_0 t} c_2 c_3 e^{-i(\mathcal{H}_0+V_{21}+V_{23})t} c_2 c_4 |M_0\rangle \\ =& i \langle M_0| e^{i\mathcal{H}_0 t} c_3 e^{-i\mathcal{H}' t} c_4 |M_0\rangle . \end{aligned} \tag{S18}$$

Similarly,

$$\Gamma_4 = -i \langle M_0| e^{i\mathcal{H}_0 t} c_3 e^{-i\mathcal{H}' t} c_6 |M_0\rangle , \tag{S19}$$

from which we finally obtain Eq. (15) after a time integration using the Lehmann spectral representation.

BOGOLIUBOV TRANSFORMATIONS

We introduce complex matter fermions on each z bond $\langle ij\rangle_z$ ($i \in A, j \in B$) at position r, and we relabel the Majorana fermions as $c_i = c_{Ar}$, $c_j = c_{Br}$, such that

$$f_r = \frac{c_{Ar} + i c_{Br}}{2}, \qquad f_r^\dagger = \frac{c_{Ar} - i c_{Br}}{2}. \tag{S20}$$

The Hamiltonians $\mathcal{H}_0$ and $\mathcal{H}'$ can then be diagonalized on a finite system. The resulting complex fermions $a^\dagger$ and $b^\dagger$ of Eq. (16) and the $f^\dagger$ fermions are related by a Bogoliubov transformation

$$\begin{pmatrix} a \\ a^\dagger \end{pmatrix} = \begin{pmatrix} X_a^* & Y_a^* \\ Y_a & X_a \end{pmatrix} \begin{pmatrix} f \\ f^\dagger \end{pmatrix}, \qquad \begin{pmatrix} b \\ b^\dagger \end{pmatrix} = \begin{pmatrix} X_b^* & Y_b^* \\ Y_b & X_b \end{pmatrix} \begin{pmatrix} f \\ f^\dagger \end{pmatrix}, \qquad \begin{pmatrix} b \\ b^\dagger \end{pmatrix} = \begin{pmatrix} \mathcal{X}^* & \mathcal{Y}^* \\ \mathcal{Y} & \mathcal{X} \end{pmatrix} \begin{pmatrix} a \\ a^\dagger \end{pmatrix}, \tag{S21}$$

where $\mathcal{X}$ and $\mathcal{Y}$ are related to $X_{a,b}$ and $Y_{a,b}$ [S9, S10]. We use the notation $f^\dagger = (f_1^\dagger \dots f_N^\dagger)$, and similarly for column vectors. As the ground states of $\mathcal{H}_0$ and $\mathcal{H}'$, $|M_0\rangle$ and $|M'\rangle$ respectively, have the same parity, $\langle M_0|M'\rangle \neq 0$ and

$$|M'\rangle = [\det(\mathcal{X}^\dagger\mathcal{X})]^{1/4} e^{-\frac{1}{2}\sum_{nm} \mathcal{F}_{nm} a_n^\dagger a_m^\dagger} |M_0\rangle , \tag{S22}$$

where $\mathcal{F} = \mathcal{X}^{*-1}\mathcal{Y}$ [S10]. For Hamiltonians with different ground state parities, $\mathcal{X}$ is singular and such expression does not exist. For single-particle eigenstates $|\lambda\rangle = b_\lambda^\dagger |M'\rangle$ of $\mathcal{H}'$ we find

$$\begin{aligned}\langle M_0|c_{Ar} b_\lambda^\dagger|M'\rangle =& \ [\det(\mathcal{X}^\dagger\mathcal{X})]^{1/4} \left[(\mathcal{X}^\dagger)^{-1}(X_a + Y_a)\right]_{\lambda r} \\ \langle M_0|c_{Br} b_\lambda^\dagger|M'\rangle =& \ i[\det(\mathcal{X}^\dagger\mathcal{X})]^{1/4} \left[(\mathcal{X}^\dagger)^{-1}(Y_a - X_a)\right]_{\lambda r} , \end{aligned} \tag{S23}$$

and for three-particle eigenstates $|\lambda\rangle = b_{\lambda_1}^\dagger b_{\lambda_2}^\dagger b_{\lambda_3}^\dagger |M'\rangle$, we find

$$\begin{aligned}\langle M_0|c_{Ar} b_{\lambda_1}^\dagger b_{\lambda_2}^\dagger b_{\lambda_3}^\dagger|M'\rangle =& \ [\det(\mathcal{X}^\dagger\mathcal{X})]^{1/4} \Big\{ \left[(\mathcal{X}^\dagger)^{-1}(X_a + Y_a)\right]_{\lambda_1 r} \left[(\mathcal{Y}(\mathcal{X}^*)^{-1}\right]_{\lambda_2\lambda_3} \\ &+ \left[(\mathcal{X}^\dagger)^{-1}(X_a + Y_a)\right]_{\lambda_2 r} \left[(\mathcal{Y}(\mathcal{X}^*)^{-1}\right]_{\lambda_3\lambda_1} \\ &+ \left[(\mathcal{X}^\dagger)^{-1}(X_a + Y_a)\right]_{\lambda_3 r} \left[(\mathcal{Y}(\mathcal{X}^*)^{-1}\right]_{\lambda_1\lambda_2} \Big\} \\ \langle M_0|c_{Br} b_{\lambda_1}^\dagger b_{\lambda_2}^\dagger b_{\lambda_3}^\dagger|M'\rangle =& \ i[\det(\mathcal{X}^\dagger\mathcal{X})]^{1/4} \Big\{ \left[(\mathcal{X}^\dagger)^{-1}(Y_a - X_a)\right]_{\lambda_1 r} \left[(\mathcal{Y}(\mathcal{X}^*)^{-1}\right]_{\lambda_2\lambda_3} \\ &+ \left[(\mathcal{X}^\dagger)^{-1}(Y_a - X_a)\right]_{\lambda_2 r} \left[(\mathcal{Y}(\mathcal{X}^*)^{-1}\right]_{\lambda_3\lambda_1} \\ &+ \left[(\mathcal{X}^\dagger)^{-1}(Y_a - X_a)\right]_{\lambda_3 r} \left[(\mathcal{Y}(\mathcal{X}^*)^{-1}\right]_{\lambda_1\lambda_2} \Big\} \end{aligned} \tag{S24}$$

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SYMMETRIES OF $\mathcal{H}_0$ AND $\mathcal{H}'$

Let $\boldsymbol{r}_0$ be the center of the $\langle 25\rangle$ bond depicted in Fig. 2 of the main text. Then, to each site i corresponds a site $p(i)$ such that $(\boldsymbol{r}_i - \boldsymbol{r}_0) = -(\boldsymbol{r}_{p(i)} - \boldsymbol{r}_0)$ modulo the periodic boundary conditions. If $i \in A$ then $p(i) \in B$ and vice versa. The transformation p is bijective and $p^{-1} = p$. The Hamiltonian $\mathcal{H}_0$ and $\mathcal{H}'$ are such that for all bonds $\langle ij\rangle_\gamma$,

$$u_{\langle ij\rangle_\gamma} = u_{\langle p(j)p(i)\rangle_\gamma}. \tag{S25}$$

We then define the unitary transformation U_I such that

$$U_I c_i U_I^\dagger = c_{p(i)}, \tag{S26}$$

for all $i \in A, B$, and the particle-hole unitary transformation U_{PH},

$$\begin{aligned} U_{PH} c_i U_{PH}^\dagger &= c_i \quad \forall i \in A \\ U_{PH} c_j U_{PH}^\dagger &= -c_j \quad \forall j \in B. \end{aligned} \tag{S27}$$

It corresponds to a particle-hole symmetry in the sense that $U_{PH} f^\dagger U_{PH}^\dagger = f$ and $U_{PH} f U_{PH}^\dagger = f^\dagger$. Finally, we define the combined unitary transformation $V = U_{PH} U_I$, which satisfies

$$\begin{aligned} V c_i V^\dagger &= -c_{p(i)} \quad \forall i \in A \\ V c_j V^\dagger &= c_{p(j)} \quad \forall j \in B, \end{aligned} \tag{S28}$$

so that $V^2 c_i (V^\dagger)^2 = -c_i$, $\forall i$. Additionally, for any matter Hamiltonian $\mathcal{H}$ satisfying Eq. (S25),

$$\{\mathcal{H}, U_I\} = 0, \quad \{\mathcal{H}, U_{PH}\} = 0, \quad [\mathcal{H}, V] = 0. \tag{S29}$$

We can therefore choose a basis of eigenstates of $\mathcal{H}$ so that each element $|\lambda\rangle$ satisfies

$$\mathcal{H} |\lambda\rangle = E_\lambda |\lambda\rangle, \qquad V |\lambda\rangle = v_\lambda |\lambda\rangle. \tag{S30}$$

Using the properties of V, the following eigenstates of V can be constructed from any $|\lambda\rangle$,

$$\begin{aligned} |\lambda_+\rangle &= d_m^\dagger |\lambda\rangle, \\ |\lambda_-\rangle &= d_m |\lambda\rangle, \\ V |\lambda_\pm\rangle &= \mp i v_\lambda |\lambda_\pm\rangle, \end{aligned} \tag{S31}$$

where we have defined the complex fermions

$$d_m^\dagger = \frac{c_m + i c_{p(m)}}{2}, \quad d_m = \frac{c_m - i c_{p(m)}}{2}, \quad \text{for } m \in A. \tag{S32}$$

Then using a Bogoliubov transformation relating the $d^\dagger$ fermions to the $b^\dagger$ fermions, and a series of arguments, we argue that we can sort the fermions into two species: $b^\dagger = (b_+^\dagger \; b_-^\dagger)$, which satisfy

$$V b_+^\dagger V^\dagger = -i b_+^\dagger, \qquad V b_-^\dagger V^\dagger = i b_-^\dagger. \tag{S33}$$

Thanks to Eq. (S22), we can also show that $|M_0\rangle$ and $|M'\rangle$ have the same V eigenvalue: $V |M_0\rangle = v_0 |M_0\rangle$ and $V |M'\rangle = v_0 |M'\rangle$. Note that we have assumed that $\omega_n \neq 0$ and $\omega_n' \neq 0$ for all n.

Finally, for $j \in B$,

$$\begin{aligned} \langle M_0 | c_j | \lambda \rangle &= \langle M_0 | V^\dagger c_{p(j)} V | \lambda \rangle \\ &= \frac{1}{v_0} \langle M_0 | c_{p(j)} V b_{\lambda_1}^\dagger \dots b_{\lambda_m}^\dagger | M' \rangle, \quad (m \text{ odd}) \\ &= \pm i \langle M_0 | c_{p(j)} | M' \rangle, \end{aligned} \tag{S34}$$

where the sign depends on the composition of $|\lambda\rangle$ in terms of $b_+^\dagger$ and $b_-^\dagger$ fermions. Moreover, for single-particle states $|\lambda\rangle = b_\lambda^\dagger |M'\rangle$ we have

$$\langle M_0 | c_j b_\lambda^\dagger | M' \rangle = \begin{cases} -i \langle M_0 | c_{p(j)} b_\lambda^\dagger | M' \rangle, & \text{if } b_\lambda \in \{b_+\} \\ i \langle M_0 | c_{p(j)} b_\lambda^\dagger | M' \rangle, & \text{if } b_\lambda \in \{b_-\} \end{cases} \tag{S35}$$

For $j \in A$, the signs are reversed.

In the optical conductivity, only expressions of the form $\langle M_0 | c_j b_\lambda^\dagger | M' \rangle - i \langle M_0 | c_{p(j)} b_\lambda^\dagger | M' \rangle$ with $j \in B$ appears so that only $b_-^\dagger$ states contributes. In the magnetic susceptibility however, only $b_+^\dagger$ states contribute. This can be generalized to any odd-particle energy eigenstates.

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