

Realization of Kitaev Model

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In this paper I will review the derivation of Kitaev coupling in honeycomb irridates.

谦谦君子，用涉大川。

—— 「易·谦卦」

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I. EFFECTIVE HILBERT SPACE AND PSEUDOSPIN

A. Preliminaries: Crystal Field Splitting — Representation of Octahedral Group O_h

In the crystal field of octahedral oxygen environment, the five d orbitals of Ir atom (each of which can accommodate two electrons) will split into two-fold e_g orbitals¹

$$|d_{x^2-y^2}\rangle \equiv r^2(Y_2^2 + Y_2^{-2}),$$
$$|d_{3z^2-r^2}\rangle \equiv r^2Y_2^0,$$

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¹ Fundamentally, this result comes from the decomposition of the reducible five-dimensional representation of octahedral group (expressing in the basis of d orbital wave function)

$$\Gamma_d \equiv E \oplus T_2.$$

See [1] and [2] for more details.

and three-fold t_{2g} orbitals

$$\begin{aligned} |d_{xz}\rangle &\equiv r^2(Y_2^1 - Y_2^{-1}), \\ |d_{yz}\rangle &\equiv r^2(Y_2^1 + Y_2^{-1}), \\ |d_{xy}\rangle &\equiv r^2(Y_2^2 - Y_2^{-2}), \end{aligned}$$

as is illustrated in FIG. 1. Further distortion, for example, Jahn-Teller effect, will split t_{2g} orbitals into a_{1g} and e'_g as well, but we only truncate our discussion at the level of t_{2g} orbitals.

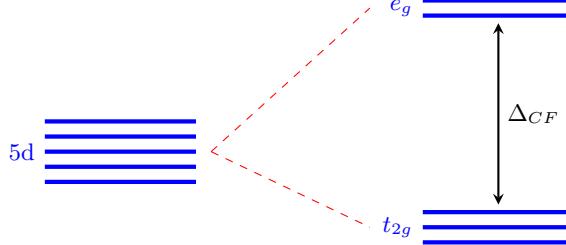


FIG. 1: Multi-orbital Effect: Octahedral Crystal Field Splitting.

B. Quenching of Orbital Angular Momentum — t_{2g} - p Equivalence

It can be seen that **the confining action of angular momentum operator L within the subspace of t_{2g} has exactly the same matrix element of the action within the standard p -orbital or $l_{\text{eff}} = 1$ basis [2]**. This phenomenon is called t_{2g} - p equivalence. More precisely, one can construct the $l_{\text{eff}} = 1$ eigen states $|m\rangle$ ($m \equiv +1, 0, -1$) with t_{2g} orbitals $|\alpha\rangle$ ($\alpha \equiv d_{xz}, d_{xy}, d_{yz}$) by

$$|+1\rangle \equiv \frac{1}{\sqrt{2}}(|d_{xz}\rangle + i|d_{yz}\rangle), \quad |0\rangle \equiv |d_{xy}\rangle, \quad |-1\rangle \equiv \frac{1}{\sqrt{2}}(|d_{xz}\rangle - i|d_{yz}\rangle), \quad (1)$$

and vice versa. We will work elaborately about the physical hopping of d -orbital electrons, both direct exchange of the Ir-Ir bond and indirect exchange of $\pi/2$ Ir-O-Ir bonds, so the unitary transformation between *orbital representation* and *angular momentum representation* is demanded

$$U_{\alpha m} \equiv \langle \alpha | m \rangle = \begin{pmatrix} 1/\sqrt{2} & 0 & 1/\sqrt{2} \\ 0 & 1 & 0 \\ i/\sqrt{2} & 0 & -i/\sqrt{2} \end{pmatrix}. \quad (2)$$

C. t_{2g} Splitting and Pseudospins from Spin-Orbital Coupling

It is believed that the d -orbital electron on each Ir atom experiences a *strong* spin-orbital² coupling

$$H_{\text{SOC}} = \lambda \mathbf{L} \cdot \mathbf{S} + \Delta L_z^2 \simeq \lambda \mathbf{L} \cdot \mathbf{S} \equiv \frac{\lambda}{2} ((\mathbf{L} + \mathbf{S})^2 - \mathbf{L}^2 - \mathbf{S}^2). \quad (3)$$

We will show in this subsection that such SOC Hamiltonian split t_{2g} orbitals further into $j_{\text{eff}} = 1/2$ doublet subspace and $j_{\text{eff}} = 3/2$ quadruplet subspace. So the low-energy effective spin (actually for the hole) is $1/2$.

² Clearly the “orbital momentum” here is the effective one with $l_{\text{eff}} = 1$.

In the (effective) $l\text{-}s$ representation $|ms\rangle \equiv (+1\uparrow, +1\downarrow, 0\uparrow, 0\downarrow, -1\uparrow, -1\downarrow)^T$, Hamiltonian (3) reads

$$\varepsilon_{ls} \equiv \langle ms | H_{\text{SOC}} | ms \rangle = \begin{pmatrix} \lambda/2 & 0 & 0 & 0 & 0 & 0 \\ 0 & -\lambda/2 & \lambda/\sqrt{2} & 0 & 0 & 0 \\ 0 & \lambda/\sqrt{2} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & \lambda/\sqrt{2} & 0 \\ 0 & 0 & 0 & \lambda/\sqrt{2} & -\lambda/2 & 0 \\ 0 & 0 & 0 & 0 & 0 & \lambda/2 \end{pmatrix}. \quad (4)$$

So under the basis of orbital representation $|\alpha s\rangle \equiv (d_{xz}\uparrow, d_{xz}\downarrow, d_{xy}\uparrow, d_{xy}\downarrow, d_{yz}\uparrow, d_{yz}\downarrow)^T$ we have

$$\varepsilon_{\alpha s} \equiv \langle \alpha s | H_{\text{SOC}} | \alpha s \rangle = \sum_m \langle \alpha s | ms \rangle \langle ms | H_{\text{SOC}} | ms \rangle \langle ms | \alpha s \rangle.$$

Now that spin degree of freedoms are included, the unitary transformation (2) should also be enlarged to

$$U \equiv U_{\alpha m} \otimes \sigma_0 = \begin{pmatrix} 1/\sqrt{2} & 0 & 0 & 0 & 1/\sqrt{2} & 0 \\ 0 & 1/\sqrt{2} & 0 & 0 & 0 & 1/\sqrt{2} \\ 0 & 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 & 0 \\ i/\sqrt{2} & 0 & 0 & 0 & -i/\sqrt{2} & 0 \\ 0 & i/\sqrt{2} & 0 & 0 & 0 & -i/\sqrt{2} \end{pmatrix} \quad (5)$$

Then

$$\varepsilon_{\alpha s} = U \varepsilon_{ls} U^\dagger = \frac{\lambda}{2} \begin{pmatrix} 0 & 0 & 0 & 1 & -i & 0 \\ 0 & 0 & 1 & 0 & 0 & i \\ 0 & 1 & 0 & 0 & 0 & -i \\ 1 & 0 & 0 & 0 & i & 0 \\ i & 0 & 0 & -i & 0 & 0 \\ 0 & -i & i & 0 & 0 & 0 \end{pmatrix}, \quad (6)$$

which have two eigenvalues $-\lambda$ and $\lambda/2$, indicating the further splitting of t_{2g} orbitals to low energy doublet subspace and higher energy of quadruplet subspace.

For the low-energy doublet subspace whose eigenvalue is $-\lambda$, the two-fold degenerate normalized eigen states, denoting as $|\tilde{\uparrow}\rangle$ and $|\tilde{\downarrow}\rangle$, are (note that we are now in the basis of $|\alpha s\rangle$)

$$|\tilde{\uparrow}\rangle = \frac{1}{\sqrt{3}} \left(|d_{xz}\downarrow\rangle - |d_{xy}\uparrow\rangle + |d_{yz}\downarrow\rangle \right), \quad |\tilde{\downarrow}\rangle = \frac{1}{\sqrt{3}} \left(-|d_{xz}\uparrow\rangle - i|d_{xy}\downarrow\rangle + |d_{yz}\uparrow\rangle \right). \quad (7)$$

That's why we say spin-orbital coupling leads to the emergence of pseudospin $j_{\text{eff}} = 1/2$. And **the low-energy doublet subspace can be extracted from arbitrary form of Hamiltonian in use of the projection operator constructed from (7)**

$$P = \frac{1}{\sqrt{3}} \begin{pmatrix} 0 & i \\ -i & 0 \\ i & 0 \\ 0 & -i \\ 0 & 1 \\ 1 & 0 \end{pmatrix}. \quad (8)$$

Particularly, one can check that

$$P^\dagger \varepsilon_{\alpha s} P = \begin{pmatrix} -\lambda & \\ & -\lambda \end{pmatrix}$$

II. HAMILTONIAN

A. On-site Hamiltonian

1. Free Part

Introducing a six-component electron (hole) field³ $d_i \equiv (d_{i,xz,\uparrow}, d_{i,xz,\downarrow}, d_{i,yz,\uparrow}, d_{i,yz,\downarrow}, d_{i,xy,\uparrow}, d_{i,xy,\downarrow})^T$ on the i -th Ir atom, for a nearest neighbor Ir-Ir link (which is enough in our further description of hopping and interaction) the energy is simply the summation of on-site Hamiltonian

$$H_0 = d_1^\dagger \varepsilon_0 d_1 + d_2^\dagger \varepsilon_0 d_2. \quad (9)$$

But since energy scale can be set to arbitrary value, we will ignore this trivial constant in the following discussion.

2. Coulomb Interaction Part

Except for the free part Hamiltonian, every electron (here is actually a hole) on the Ir site experiences strong (screened) Coulomb repulsive interaction (and we ignore the inter-site interaction for tight-binding approximation). So our next task is to express the second quantized two-body interaction on the i -th Ir atom

$$\begin{aligned} H_I &\equiv \frac{1}{2} \sum_{i \neq j} V(\mathbf{r}_i - \mathbf{r}_j) \equiv \frac{1}{2} \sum_{\substack{\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4 \\ \sigma_1, \sigma_2, \sigma_3, \sigma_4}} \langle \mathbf{r}_1, \sigma_1; \mathbf{r}_2, \sigma_2 | V(\mathbf{r}_i - \mathbf{r}_j) \otimes \mathbb{1} | \mathbf{r}_3, \sigma_3; \mathbf{r}_4, \sigma_4 \rangle \psi_{\sigma_1}^\dagger(\mathbf{r}_1) \psi_{\sigma_2}^\dagger(\mathbf{r}_2) \psi_{\sigma_3}(\mathbf{r}_3) \psi_{\sigma_4}(\mathbf{r}_4) \\ &= \frac{1}{2} \sum_{\sigma, \sigma'} \int d\mathbf{r} d\mathbf{r}' \psi_\sigma^\dagger(\mathbf{r}) \psi_{\sigma'}^\dagger(\mathbf{r}') V(\mathbf{r} - \mathbf{r}') \psi_{\sigma'}(\mathbf{r}') \psi_\sigma(\mathbf{r}) \end{aligned} \quad (10)$$

in terms of the orbital momentum representation.

Expanding the position representation eigen states with t_{2g} orbital states that

$$|\mathbf{r}, \sigma\rangle \equiv \left(\sum_{\alpha \in \{xz, yz, xy\}} |\alpha\rangle \langle \alpha| \right) \otimes |\sigma\rangle \equiv \sum_{\alpha} \varphi_{\alpha}(\mathbf{r}) |\alpha\rangle \otimes |\sigma\rangle \equiv \sum_{\alpha} \varphi_{\alpha}(\mathbf{r}) |\alpha, \sigma\rangle,$$

then one immediately gets⁴

$$H_I = \frac{1}{2} \sum_{\sigma, \sigma'} \sum_{\alpha_1, \alpha_2, \alpha_3, \alpha_4} d_{\alpha_1, \sigma}^\dagger d_{\alpha_2, \sigma'}^\dagger V_{\alpha_1, \alpha_2, \alpha_3, \alpha_4} d_{\alpha_3, \sigma'} d_{\alpha_4, \sigma}, \quad (11)$$

where

$$V_{\alpha_1, \alpha_2, \alpha_3, \alpha_4} \equiv \int d\mathbf{r} d\mathbf{r}' \varphi_{\alpha_1}^*(\mathbf{r}) \varphi_{\alpha_2}^*(\mathbf{r}') V(\mathbf{r} - \mathbf{r}') \varphi_{\alpha_3}(\mathbf{r}') \varphi_{\alpha_4}(\mathbf{r}).$$

Symmetry argument [3, 4] shows that

$$V_{\alpha, \beta, \mu, \nu} \equiv U \delta_{\alpha\beta} \delta_{\mu\nu} \delta_{\alpha\mu} + V \delta_{\alpha\nu} \delta_{\beta\mu} (1 - \delta_{\alpha\beta}) + J \left(\delta_{\alpha\beta} \delta_{\mu\nu} (1 - \delta_{\alpha\mu}) + \delta_{\alpha\mu} \delta_{\beta\nu} (1 - \delta_{\alpha\beta}) \right), \quad (12)$$

with $U \equiv V + 2J$, where U is the intra-orbital Hubbard interaction and V is the inter-orbital direct interaction. The first term of J is the Hund's rule exchange interaction, while the second term is the pair hopping interaction [5].

³ Note that the spin degree of freedoms on d -orbital electron field operators here are the pseudospin we introduced after consideration of spin-orbital-coupling induced splitting.

⁴ In our convention of notation, $|\mathbf{r}, \sigma\rangle \equiv \psi_\sigma(\mathbf{r})^\dagger |\Omega\rangle$ and $|\alpha, \sigma\rangle \equiv d_{\alpha, \sigma}^\dagger |\Omega\rangle$.

Finally we have

$$H_I = \frac{U}{2} \sum_{\sigma, \sigma'} \sum_{\alpha} d_{\alpha, \sigma}^\dagger d_{\alpha, \sigma'}^\dagger d_{\alpha, \sigma'} d_{\alpha, \sigma} + \frac{V}{2} \sum_{\sigma, \sigma' \neq \beta} \sum_{\alpha} d_{\alpha, \sigma}^\dagger d_{\beta, \sigma'}^\dagger d_{\beta, \sigma'} d_{\alpha, \sigma} \\ + \frac{J}{2} \sum_{\sigma, \sigma' \neq \beta} \left(d_{\alpha, \sigma}^\dagger d_{\beta, \sigma'}^\dagger d_{\alpha, \sigma'} d_{\beta, \sigma} + d_{\alpha, \sigma}^\dagger d_{\alpha, \sigma'}^\dagger d_{\beta, \sigma'} d_{\beta, \sigma} \right). \quad (13)$$

3. On-Site Two-Particle Hilbert Space

On each i -th site of Ir atom Coulomb interaction H_I involves in two electrons (holes), so to combine all terms in Hamiltonian for further projection onto the correct low-energy sector, we have to enlarge the previous 6-dimensional single-particle basis $\{d_{\alpha, \sigma}^\dagger | \Omega \rangle\}$ to two-particle one

$$\mathcal{H}_{\text{intra-site}} \equiv \{d_{i, \alpha, \sigma}^\dagger d_{i, \alpha', \sigma'}^\dagger | \Omega \rangle\}. \quad (14)$$

The dimensionality of this *on-site* two-particle Hilbert space becomes $5 + 4 + \dots + 1 = 15$ due to Pauli exclusion principle. More precisely, under the 15-dimentional basis (for convenience labeled from 1 to 15)

$$\underbrace{(d_{xz\uparrow}^\dagger d_{xz\downarrow}^\dagger, \dots, d_{xz\uparrow}^\dagger d_{xy\downarrow}^\dagger)}_5; \underbrace{(d_{xz\downarrow}^\dagger d_{yz\uparrow}^\dagger, \dots, d_{xz\downarrow}^\dagger d_{xy\downarrow}^\dagger)}_4; \underbrace{(d_{yz\uparrow}^\dagger d_{yz\downarrow}^\dagger, \dots, d_{yz\uparrow}^\dagger d_{xy\downarrow}^\dagger)}_3; \underbrace{(d_{yz\downarrow}^\dagger d_{xy\uparrow}^\dagger, d_{yz\downarrow}^\dagger d_{xy\downarrow}^\dagger)}_2; \underbrace{(d_{xy\uparrow}^\dagger d_{xy\downarrow}^\dagger)}_1)^T | \Omega \rangle,$$

the matrix of H_I in (13) takes the form of (after re-arrangement)

$$H_I = \begin{pmatrix} -U & 0 & 0 & 0 & 0 & 0 & 0 & 0 & -J & 0 & 0 & 0 & 0 & -J \\ 0 & J - V & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & -V & 0 & 0 & J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & J - V & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -V & 0 & 0 & J & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & J & 0 & 0 & -V & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & J - V & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & J & 0 & 0 & -V & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & J - V & 0 & 0 & 0 & 0 & 0 & 0 \\ -J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & -U & 0 & 0 & 0 & 0 & -J \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & J - V & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & -V & J & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & J & -V & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & J - V & 0 \\ -J & 0 & 0 & 0 & 0 & 0 & 0 & 0 & -J & 0 & 0 & 0 & 0 & 0 & 0 & -U \end{pmatrix}. \quad (15)$$

B. Inter-Site Hamiltonian

1. Hopping Part

For a general hopping (keeping the spin orientaion) on Ir-Ir link we have

$$H_t = d_1^\dagger T_{12} d_2 + h.c. \quad (16)$$

Following the supplementary material of [6], since such Ir-Ir bond (as well as the surrounding octahedral oxygen atoms) possesses *time-reversal symmetry* and *inversion symmetry* about the bond center, we must have

$$\rho(T)^\dagger H_t \rho(T) = d_1^\dagger T_{12}^* d_2 + h.c. \equiv H_t \implies T_{12}^* \equiv T_{12}$$

since representaiton of time-reversal symmetry is anti-unitary and *anti-linear*, and

$$\rho(P)^\dagger H_t \rho(P) = d_2^\dagger T_{12} d_1 + h.c. \equiv H_t \implies T_{12} \equiv T_{21}.$$

Therefore the hopping matrix T_{12} must be *real* and *symmetric*

$$T_{12} = \begin{pmatrix} T_{12}^{xz,xz} & T_{12}^{xz,yz} & T_{12}^{xz,xy} \\ T_{12}^{xz,yz} & T_{12}^{yz,yz} & T_{12}^{yz,xy} \\ T_{12}^{xz,xy} & T_{12}^{yz,xy} & T_{12}^{xy,xy} \end{pmatrix} \otimes \sigma_0.$$

But this is not the end of the story. Along Ir-Ir bond direction, or [110] direction in a local coordinate system, we also have a C_2 rotational symmetry such that t_{2g} orbitals transform like $d_{xz,\sigma} \mapsto d_{yz,\sigma}$, $d_{yz,\sigma} \mapsto d_{xz,\sigma}$, and $d_{xy,\sigma} \mapsto d_{xy,\sigma}$. Therefore, hopping matrix T_{12} takes the form of

$$\rho(C_2)^\dagger H_t \rho(C_2) \equiv H_t \implies T_{12} = \begin{pmatrix} t_1 & t_2 & t_4 \\ t_2 & t_1 & t_4 \\ t_4 & t_4 & t_3 \end{pmatrix} \otimes \sigma_0. \quad (17)$$

Matrix (17) consists of both direct Ir-Ir exchange and indirect $\pi/2$ Ir-O-Ir exchange, as is shown in FIG. 2 and FIG. 3. Similar illustration can also be found in the recent analysis of d^7 cobalt compound in [7] and [8].

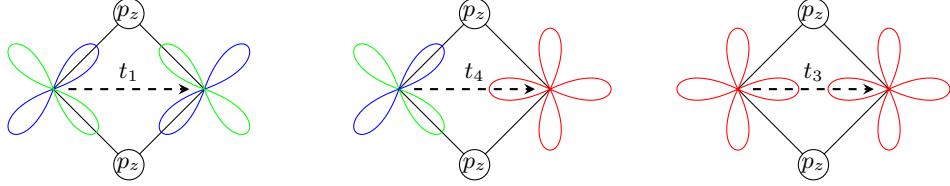


FIG. 2: **Direct Exchange between Ir-Ir Orbitals.** Blue one is the t_{2g} orbital of $d_{yz,\sigma}$, red one is $d_{xy,\sigma}$, and green one is $d_{yz,\sigma}$.

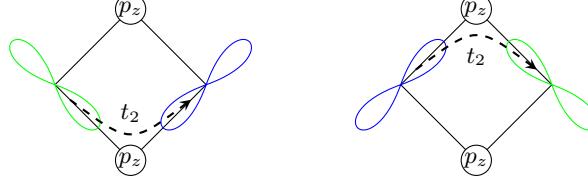


FIG. 3: **Hopping Mediated by Oxygen p -Orbitals.**

Detailed physical meanings of hopping matrix elements relating to so-called *Slater-Koster* parameter [9] are given in the supplementary material of [6].

2. Inter-Site Two-Particle Hilbert Space

Similar to the *quartic* form of Coulomb term we encountered before, where *on-site* two-particle Hilbert space is demanded for accommodation of the matrix elements, since the *quadratic* hopping Hamiltonian H_t involves two electrons (holes) on *distinct* sites of Ir atoms this time, we still have to consider the two-particle Hilbert space to accommodate the hopping matrix elements, while Pauli exclusion principle is quenched this time. So the dimensionality of our new *inter-site* two-particle basis, (taking the hopping processes from site 2 to site 1 as an example)

$$\mathcal{H}_{\text{inter-site}} = \{(d_1)_{\alpha,\sigma}^\dagger (d_2)_{\alpha',\sigma'} | \Omega \rangle\} \quad (18)$$

is $6 \times 6 = 36$. And hopping matrix under (18) takes the form of

$$T = \mathbb{1} \otimes T_{12} + T_{21} \otimes \mathbb{1}, \quad (19)$$

where $\mathbb{1}$ is a 6-dimensional identity matrix on the opposite transition sector.

Let us focus on the site-1 physics. Hopping matrix $d_1^\dagger T_{12} d_2$ will transfer electrons (holes) from site 2 to site 1, formally resulting in a new 36-dimensional *on-site* two-particle basis

$$\tilde{\mathcal{H}}_{\text{on-site}} \equiv d_1^\dagger T_{12} d_2 (\mathcal{H}_{\text{inter-site}}) = \{(d_1)_{\alpha,\sigma}^\dagger (d_1)_{\alpha',\sigma'} | \Omega \rangle\}.$$

But as is mentioned above, because of the existence of Pauli exclusion principle, not all hopping channels are physically allowed. In fact, real physical hoppings occur only on the 15-dimensional *on-site* two-particle Hilbert space $\mathcal{H}_{\text{on-site}}$ we constructed in the previous section instead of $\tilde{\mathcal{H}}_{\text{on-site}}$. To achieve this, again we need a projection

$$\mathcal{P}_t \tilde{\mathcal{H}}_{\text{on-site}} = \mathcal{H}_{\text{on-site}}, \quad (20)$$

where \mathcal{P}_t is a 15×36 matrix

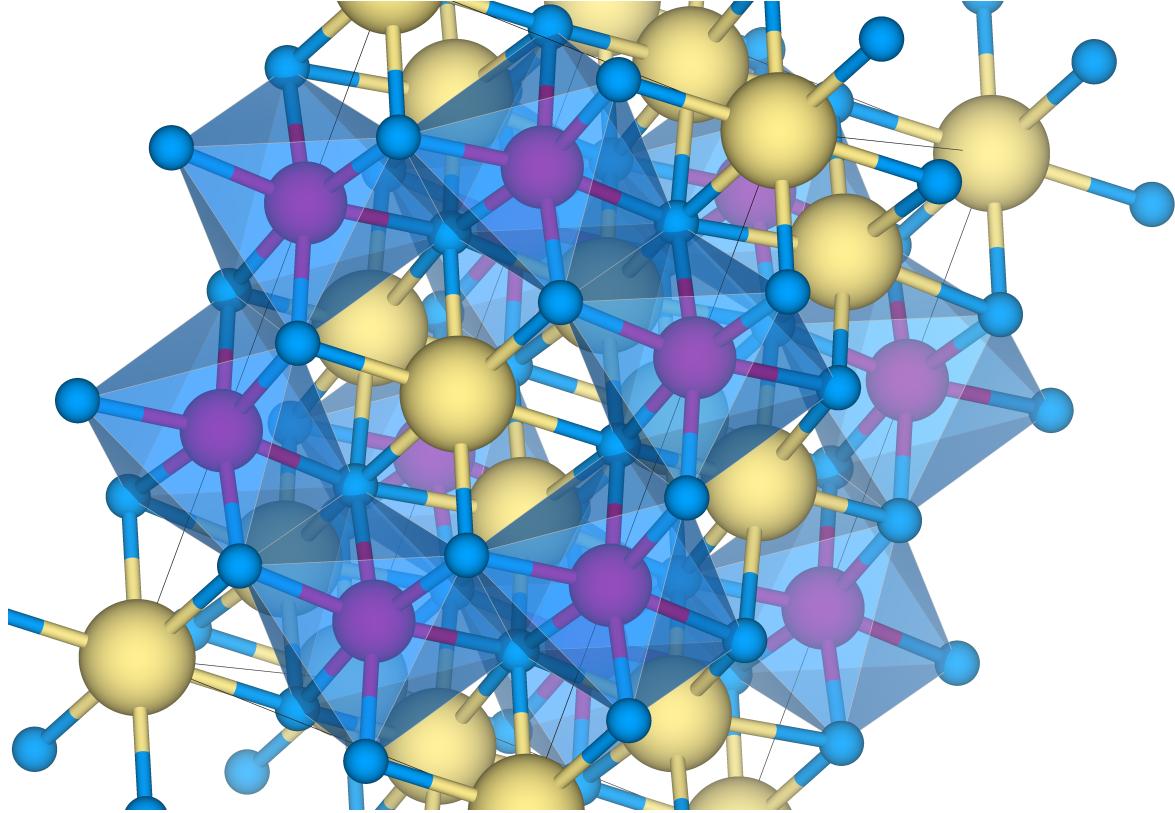
III. LARGE-U APPROXIMATION

A. Self-Energy Operator

B. Projection to Low-Energy Pseudospin Subspace

C. Kitaev Coupling

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FIG. 4: **caption:**

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