

Chapter 1

Lagrange Multiplier

The mean-field Hamiltonian is

$$H_{\text{MF}} = \frac{i}{4} \sum_{ij} c_i^T A_{ij} c_j + \frac{i}{4} \sum_i c_i^T B_i c_i - C, \quad (1.1)$$

$$C = \frac{1}{8} \sum_{ij} \mathbf{J}_{ij}^{\alpha\beta} \left[\langle W_i, N^\alpha \rangle \langle W_j, N^\beta \rangle + 2 \langle U_{ij}, N^\alpha U_{ij} N^\beta \rangle \right], \quad (1.2)$$

$$B_i = \sum_j \mathbf{J}_{ij}^{\alpha\beta} N^\alpha \langle W_j, N^\beta \rangle - \sum_\gamma \left(2h_i^\gamma N^\gamma - \lambda_i^\gamma G^\gamma \right), \quad (1.3)$$

$$A_{ij} = 2\mathbf{J}_{ij}^{\alpha\beta} N^\alpha U_{ij} N^\beta. \quad (1.4)$$

Where $N^\gamma = (M^\gamma - G^\gamma)/2$ and $\gamma = 1, 2, 3$ for $\{M^\gamma\}$ and $\{G^\gamma\}$ being two linear independent real representations of the $\mathfrak{su}(2)$ algebra inside the fundamental representation of $\mathfrak{so}(4) = \mathfrak{su}(2) \times \mathfrak{su}(2)$. Thus, the span of $\{M^\gamma, G^\gamma\}$ is the 6-dimensional set of 4×4 antissymmetric matrices. The matrix representation of M^γ, G^γ is given in the appendix of the draft. These matrices are orthogonal with respect to the innerproduct $\langle A, B \rangle := \text{tr}(A^T B)$:

$$\langle M^\alpha, M^\beta \rangle = 4\delta^{\alpha\beta}, \quad \langle G^\alpha, G^\beta \rangle = 4\delta^{\alpha\beta}, \quad \langle M^\alpha, G^\beta \rangle = 0, \quad (1.5)$$

$$\langle N^\alpha, M^\beta \rangle = 2\delta^{\alpha\beta}, \quad \langle N^\alpha, G^\beta \rangle = -2\delta^{\alpha\beta}. \quad (1.6)$$

Since the matrices W are $2i\mathbb{1}_4$ plus an antissymmetric matrix they can be expanded as

$$W_j(\lambda) = 2i\mathbb{1}_4 + \sum_\gamma w_j^\gamma(\lambda) M^\gamma + \sum_\gamma \bar{w}_j^\gamma(\lambda) G^\gamma, \quad (1.7)$$

then

$$\langle W_j, M^\gamma \rangle = 4w_j^\gamma, \quad \langle W_j, N^\gamma \rangle = 2(w_j^\gamma - \bar{w}_j^\gamma), \quad \langle W_j, G^\gamma \rangle = 4\bar{w}_j^\gamma. \quad (1.8)$$

The energy functional is

$$\begin{aligned} \langle H_{\text{MF}} \rangle &= \frac{1}{8} \sum_{ij} \mathbf{J}_{ij}^{\alpha\beta} \left[\langle W_i, N^\alpha \rangle \langle W_j, N^\beta \rangle + 2 \langle U_{ij}, N^\alpha U_{ij} N^\beta \rangle \right] \\ &+ \frac{1}{4} \sum_{i,\gamma} \left[-2h_i^\gamma \langle W_i, N^\gamma \rangle + \lambda_i^\gamma \langle W_i, G^\gamma \rangle \right], \end{aligned} \quad (1.9)$$

$$\begin{aligned} &= \frac{1}{8} \sum_{ij} \mathbf{J}_{ij}^{\alpha\beta} \left[4(w_i^\alpha - \bar{w}_i^\alpha)(w_j^\beta - \bar{w}_j^\beta) + 2 \langle U_{ij}, N^\alpha U_{ij} N^\beta \rangle \right] \\ &+ \sum_{i,\alpha} \left[-h_i^\alpha (w_i^\alpha - \bar{w}_i^\alpha) + \lambda_i^\alpha \bar{w}_i^\alpha \right], \end{aligned} \quad (1.10)$$

$$\begin{aligned} &= \frac{1}{4} \sum_{ij} \mathbf{J}_{ij}^{\alpha\beta} \langle U_{ij}, N^\alpha U_{ij} N^\beta \rangle + \sum_{i,\alpha} \left[-h_i^\alpha (w_i^\alpha - \bar{w}_i^\alpha) + \lambda_i^\alpha \bar{w}_i^\alpha \right], \\ &+ \frac{1}{2} \sum_i (w_i^\alpha - \bar{w}_i^\alpha) \sum_j \mathbf{J}_{ij}^{\alpha\beta} (w_j^\beta - \bar{w}_j^\beta). \end{aligned} \quad (1.11)$$

The matrix element $\mathbf{J}_{ij}^{\alpha\beta}$ is only non-zero for j in a bond with i , so it restricts to the three terms in the sum over γ in $\langle ij \rangle_\gamma$, as i can be in either sublattice here (to not confuse with the notation where $\langle ij \rangle$ forces $i/j \in A/B$ I will write this sum as a sum of nearest neighbors $\sum_{j \in V_i}$. From this, we can identify the effective magnetic field

$$\langle H_{\text{MF}} \rangle = \frac{1}{4} \sum_{ij} \mathbf{J}_{ij}^{\alpha\beta} \langle U_{ij}, N^\alpha U_{ij} N^\beta \rangle + \sum_{i,\alpha} \left[-\tilde{h}_i^\alpha (w_i^\alpha - \bar{w}_i^\alpha) + \lambda_i^\alpha \bar{w}_i^\alpha \right], \quad (1.12)$$

$$\tilde{h}_i^\alpha = h_i^\alpha - \frac{1}{2} \sum_{j \in V_i} J_{ij}^{\alpha\beta} (w_j^\beta - \bar{w}_j^\beta). \quad (1.13)$$

1.1 Partially polarized phase

We can solve the mean-field equation in the phase where all generalized Kitaev type parameters are zero $U_{ij}^{\alpha\beta} = 0$. Let us consider first the uniform case

$$w_i^\gamma = w^\gamma, \quad \bar{w}_i^\gamma = \bar{w}^\gamma. \quad (1.14)$$

The energy density $F = \langle H_{\text{MF}} \rangle / 2N$ is

$$F = \sum_\gamma \left[\mathbf{J}_\gamma^{\alpha\beta} \frac{1}{2} (w^\alpha - \bar{w}^\alpha)(w^\beta - \bar{w}^\beta) - h^\gamma (w^\gamma - \bar{w}^\gamma) + \lambda^\gamma \bar{w}^\gamma \right], \quad (1.15)$$

Where $\mathbf{J}_\gamma^{\alpha\beta} \equiv \mathbf{J}_{\langle ij \rangle_\gamma}^{\alpha\beta}$ and $\sum_\gamma \equiv \sum_{j \in V_i}$ for the set $V_i = \{i + \delta_x, i + \delta_y, i + \delta_z\}$ of N.N. of i . Instead of directly imposing the minimization of F w.r.t to λ that enforces $\bar{w} = 0$, let us vary F w.r.t. \bar{w} and find the critical value for the Lagrange multiplier

$$\frac{\partial F}{\partial \bar{w}^\beta} = - \left(\sum_\gamma \mathbf{J}_\gamma^{\alpha\beta} \right) (w^\alpha - \bar{w}^\alpha) + h^\beta + \lambda^\beta \Rightarrow \boxed{\lambda^\alpha = -h^\alpha + \mathbb{J}^{\alpha\beta} (w^\beta - \bar{w}^\beta)}, \quad (1.16)$$

where

$$\mathbb{J}^{\alpha\beta} = \frac{\partial^2 F}{\partial \bar{w}^\alpha \partial \bar{w}^\beta} = \sum_\gamma \mathbf{J}_\gamma^{\alpha\beta} = \begin{pmatrix} 3J+K & \Gamma & \Gamma \\ \Gamma & 3J+K & \Gamma \\ \Gamma & \Gamma & 3J+K \end{pmatrix}^{\alpha\beta} \quad (1.17)$$

Note that $\det(\mathbb{J}^{\alpha\beta}) = (-\Gamma + 3J + K)^2(2\Gamma + 3J + K)$ is negative close to the pure FM Kitaev limit $K < 0$ so the energy is a minimum.

This minimization procedure implies that the Lagrange multiplier is equal to $\lambda = h - 2\tilde{h}$. Plugging this value for the Lagrange multiplier in the energy density

$$F = \frac{1}{2} \mathbb{J}^{\alpha\beta} (w^\alpha - \bar{w}^\alpha)(w^\beta - \bar{w}^\beta) - h^\alpha w^\alpha + \mathbb{J}^{\alpha\beta} \bar{w}^\alpha (w^\beta - \bar{w}^\beta), \quad (1.18)$$

$$= \frac{1}{2} \mathbb{J}^{\alpha\beta} (w^\alpha w^\beta - \bar{w}^\alpha \bar{w}^\beta) - h^\alpha w^\alpha. \quad (1.19)$$

From that, we immediately see that if $\det(\mathbb{J}^{\alpha\beta}) < 0$ the energy is minimized for $\bar{w} = 0$. Minimizing the energy w.r.t. w^α we find

$$\frac{\partial F}{\partial w^\alpha} = \mathbb{J}^{\alpha\beta} w^\beta - h^\alpha \Rightarrow \boxed{w^\alpha = (\mathbb{J}^{-1})^{\alpha\beta} h^\beta}, \quad (1.20)$$

and the classical polarized energy is $F = -\frac{1}{2}(\mathbb{J}^{-1})^{\alpha\beta} h^\alpha h^\beta$. Note however that this solution breaks in the region where $\det(\mathbb{J}^{\alpha\beta}) > 0$, i.e. $\Gamma > -\frac{K}{2} - \frac{3J}{2}$, which gives a explanation for the transition at $\Gamma/(-K) = 0.5$ in the $h\Gamma$ phase diagram for $J = 0$.

In the inhomogeneous case, the Lagrange multiplier is

$$\lambda_i^\alpha = -h_i^\alpha + \frac{1}{2} \sum_{j \in V_i} J_{ij}^{\alpha\beta} (w_j^\beta - \bar{w}_j^\beta), \quad (1.21)$$

which gives the free energy

$$2N F = - \sum_i h_i^\gamma w_i^\gamma + \frac{1}{2} \sum_{ij} J_{ij}^{\alpha\beta} (w_i^\alpha w_j^\beta - \bar{w}_i^\alpha \bar{w}_j^\beta). \quad (1.22)$$

1.2 Kitaev spin liquid phase

Treating the mean-field parameters U, W as independent variables, the minimization procedure for W gives the same expression for the Lagrange multiplier. Allowing a correction, I will write the Lagrange multiplier as

$$\lambda_i^\alpha = -h_i^\alpha + \frac{1}{2} \sum_{j \in V_i} J_{ij}^{\alpha\beta} (w_j^\beta - \bar{w}_j^\beta) + \delta\lambda_i. \quad (1.23)$$

Where δh_i is such that the solution of the self-consistent equation satisfies the constraint $\langle W_i, G^\gamma \rangle = 0$. As there is no way to solve the self-consistent equations in the general case,

then one cannot compute $\delta\lambda_i$. A not elegant solution for this is: for each fixed value of $\{\delta\lambda_i\}$ one test if it solves the self-consistent equations and computes the value of $\langle W_i, G^\gamma \rangle$, iterating this process one can find the solution by sampling a set of values one.

In the homogeneous case, one can solve it by brute force. Define functions $\alpha_1, \alpha_2, \alpha_3$

$$\delta\boldsymbol{\lambda} = \alpha_1 \mathbf{a} + \alpha_2 \mathbf{b} + \alpha_3 \mathbf{c}, \quad (1.24)$$

These are functions of the field and the couplings. From trial and error, I found that $\delta\boldsymbol{\lambda}$ is small and scales linearly with \mathbf{h} . The problem reduces to find α such that

$$\delta\boldsymbol{\lambda} = \alpha \mathbf{h} \quad (1.25)$$

implies in the constraint fulfillment

$$\Delta V_i(\alpha) := \frac{1}{\sqrt{3}} \|\bar{\mathbf{w}}_i\| = \frac{1}{4} \sqrt{\frac{1}{3} \sum_{\gamma=1}^3 \langle W_i, G^\gamma \rangle^2} \rightarrow 0 \quad (1.26)$$

The constraint violation for $W^{\mu\nu} = \langle ic^\mu c^\nu \rangle$ is

$$(\Delta V)^2 = \frac{\left(\frac{\langle ic^0 c^x \rangle + \langle ic^y c^z \rangle}{2} \right)^2 + \left(\frac{\langle ic^0 c^y \rangle + \langle ic^z c^x \rangle}{2} \right)^2 + \left(\frac{\langle ic^0 c^z \rangle + \langle ic^x c^y \rangle}{2} \right)^2}{3}. \quad (1.27)$$

The normalization is chosen so that the maximal value is one (the factor $1/3$ is due to three terms in the γ sum, and the $1/4$ is because of the normalization of $G : \langle W, G^\gamma \rangle = 4\bar{w}^\gamma$). (**remark** to change $W_i \rightarrow V_i$ in all text). In the isotropic case, this quantity reduces to the expected expression $\Delta V = \frac{1}{2} |\langle ic^0 c^\gamma \rangle + \langle ic^\alpha c^\beta \rangle|$ with normalization $\frac{1}{2}$. The bound for the constraint violation is

$$\begin{aligned} 0 \leq \|\bar{\mathbf{w}}_i\|^2 &\leq \|\mathbf{w}_i\|^2 + \|\bar{\mathbf{w}}_i\|^2 = \frac{1}{4^2} \sum_{\gamma} (\langle W_i, M^\gamma \rangle^2 + \langle W_i, G^\gamma \rangle^2) \\ &= \frac{1}{2} \left(\langle ic^0 c^1 \rangle^2 + \langle ic^0 c^2 \rangle^2 + \langle ic^0 c^3 \rangle^2 + \langle ic^1 c^2 \rangle^2 + \langle ic^2 c^3 \rangle^2 + \langle ic^3 c^1 \rangle^2 \right) \\ &\leq \frac{1}{2} \left(\langle (ic^0 c^1)^2 \rangle + \langle (ic^0 c^2)^2 \rangle + \langle (ic^0 c^3)^2 \rangle + \langle (ic^1 c^2)^2 \rangle + \langle (ic^2 c^3)^2 \rangle + \langle (ic^3 c^1)^2 \rangle \right) \\ &= 3 \\ &\Rightarrow 0 \leq \Delta V \leq 1. \end{aligned}$$

Now let us turn to analyze how much the constraint is violated for some values of $\delta\lambda$ or α . Numerically $\alpha = 0$ already gives a small constraint violation in the KSL region

$$\alpha = 0 \quad \Rightarrow \quad \Delta V_i(\alpha) \lesssim 10^{-2}. \quad (1.28)$$

If one wants a more precise solution, one can try different values for α such that ΔV is minimized. Here I will consider the homogeneous case with the same α in both sublattices. For example, for $\Gamma = 0.1$ one can take $\alpha \approx -0.11$ to get $\Delta V < 10^{-5}$ for any value of h as shown in Fig. 1.1.

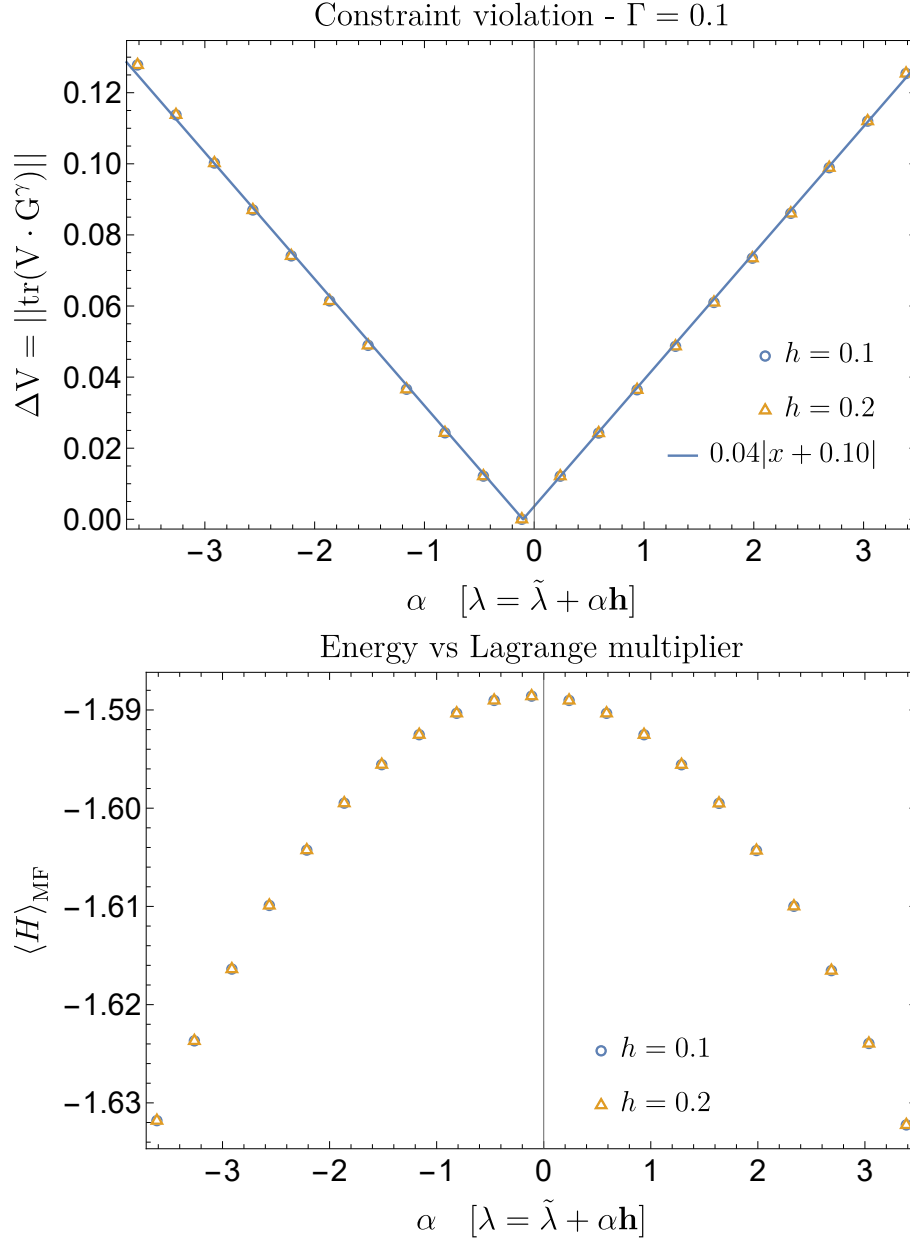


Figure 1.1: The value for the Lagrange multiplier is $\delta\lambda = -0.11\mathbf{h}$.

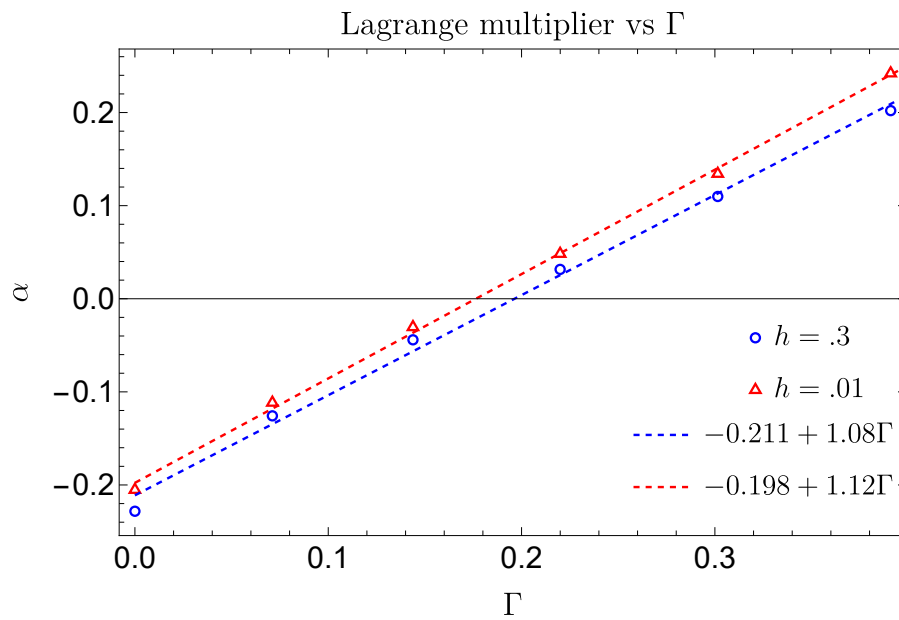


Figure 1.2: $\alpha(\Gamma)$ for two values of the magnetic field. The dependence in the magnetic field is very weak and in the KSL phase a good approximation is $\alpha(\Gamma) = -0.2 + 1.1 \Gamma$.

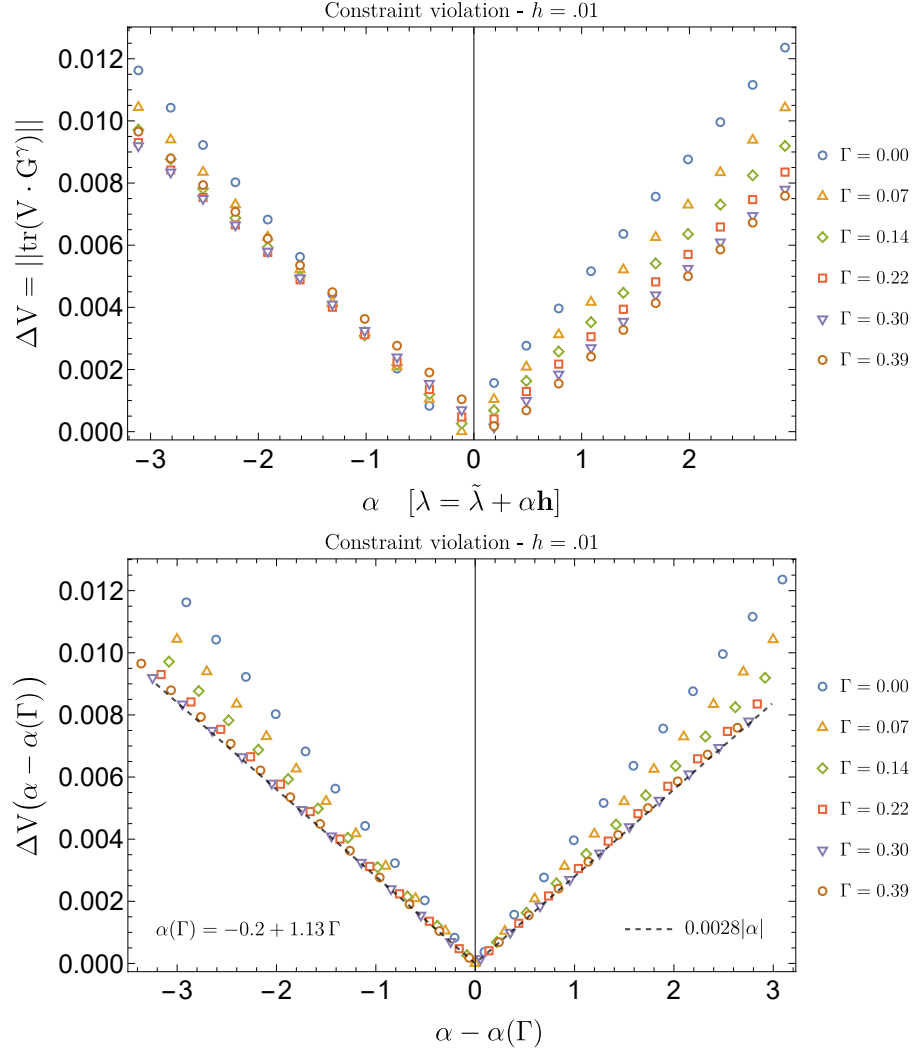


Figure 1.3: Constraint violation vs Lagrange multiplier for some values of Γ . Numerical fit: $\alpha(\Gamma) = -0.2 + 1.13\Gamma$.

1.3 Kitaev + Dzyaloshinskii Moriya

Manually inserting a Dzyaloshinskii Moriya interaction $D(\sigma_i^\alpha \sigma_j^\beta - \sigma_i^\beta \sigma_j^\alpha)$ proportionally to the applied ultralocal electric field,

$$D(\xi) = D(1)\xi, \quad \xi = eV_0/(U - 3J_H), \quad (1.29)$$

The minimal Hamiltonian to describe the physics of $4d^5$ orbitals in an octahedral environment, RuCl_6 of a spin-orbit enhanced Mott insulator is the sum of the contribution for the crystal field $10Dq \sim 2 - 2.2\text{eV}$ [?], spin-orbit coupling $\lambda_{\text{Ru}} \sim 100 - 150\text{meV}$, coulomb exchange $U \sim 2.4 - 2.6\text{eV}$, $J_H \sim 0.3 - 0.4\text{eV}$ and hopping integrals $t_2 \sim 160\text{meV}$, $t_1 \sim 60\text{meV}$, $t_3 \sim -100\text{meV}$.

This minimal model generates an effective spin $j_{\text{eff}} = 1/2$ model with dominant Kitaev interaction, symmetric Γ coupling, and Heisenberg, but not Γ' and antisymmetric Dzyaloshinskii Moriya interaction. These interactions are generically created in the materials due to trigonal distortion and lowering the C_3 symmetry to the $C2/m$ space group (e.g. eqs. 15 and 16 of [?]). In general, the anisotropy is small, e.g. there is a small trigonal splitting $\Delta_{\text{tri}} \sim 20\text{meV}$, but for t_3 the anisotropy is rather large, about 50meV .

To correctly deal with the anisotropy in the hopping integral and in the crystal field one would need to introduce many additional parameters. Here we will stay with only t_1, t_2, t_3 and add an external electric field. The electric field has many effects: (1) to change the chemical potential $\sum_i eV_0 N_i$, (2) enhance the trigonal distortion $\Delta_{\text{tri}}(V_0)$, and (3) to change the orbital wave-functions. Previously we only had to take into account (1), which only renormalizes the couplings.

