Chapter 1

Lagrange Multiplier

The mean-field Hamiltonian is

$$H_{\mathrm{MF}} = \frac{\mathrm{i}}{4}\sum_{ij} c_i^{\mathrm{T}}\mathrm{A}_{ij}c_j + \frac{\mathrm{i}}{4}\sum_{i} c_i^{\mathrm{T}}\mathrm{B}_i c_i - \mathrm{C}\ , \tag{1.1}$$

$$\mathrm{C} = \frac{1}{8}\sum_{ij}\mathbf{J}_{ij}^{\alpha\beta}\Big[\,\langle \mathrm{W}_i\,,\,\mathrm{N}^\alpha\rangle\,\langle \mathrm{W}_j\,,\,\mathrm{N}^\beta\rangle + 2\,\langle \mathrm{U}_{ij}\,,\,\mathrm{N}^\alpha\mathrm{U}_{ij}\mathrm{N}^\beta\rangle\,\Big]\ , \tag{1.2}$$

$$\mathrm{B}_i = \sum_j \mathbf{J}_{ij}^{\alpha\beta}\mathrm{N}^\alpha\,\langle \mathrm{W}_j\,,\,\mathrm{N}^\beta\rangle - \sum_\gamma\Big(2h_i^\gamma\mathrm{N}^\gamma - \lambda_i^\gamma\mathrm{G}^\gamma\Big)\ , \tag{1.3}$$

$$\mathrm{A}_{ij} = 2\mathbf{J}_{ij}^{\alpha\beta}\mathrm{N}^\alpha\mathrm{U}_{ij}\mathrm{N}^\beta\ . \tag{1.4}$$

Where $\mathrm{N}^\gamma = (\mathrm{M}^\gamma - \mathrm{G}^\gamma)/2$ and $\gamma = 1,2,3$ for $\{\mathrm{M}^\gamma\}$ and $\{\mathrm{G}^\gamma\}$ 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 $\{\mathrm{M}^\gamma, \mathrm{G}^\gamma\}$ is the 6-dimensional set of 4×4 antissimetric matrices. The matrix representation of $\mathrm{M}^\gamma, \mathrm{G}^\gamma$ is given in the appendix of the draft. These matrices are orthogonal with respect to the innerproduct $\langle \mathrm{A}\,,\,\mathrm{B}\rangle := \mathrm{tr}\big(\mathrm{A}^{\mathrm{T}}\mathrm{B}\big)$:

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

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

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

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

then

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

The energy functional is

$$\begin{aligned}\langle H_{\mathrm{MF}}\rangle =& \frac{1}{8}\sum_{ij}\mathbf{J}_{ij}^{\alpha\beta}\Big[\langle \mathrm{W}_i\,,\,\mathrm{N}^\alpha\rangle\langle \mathrm{W}_j\,,\,\mathrm{N}^\beta\rangle + 2\langle \mathrm{U}_{ij}\,,\,\mathrm{N}^\alpha\mathrm{U}_{ij}\mathrm{N}^\beta\rangle\Big]\\ &+\frac{1}{4}\sum_{i,\gamma}\Big[-2h_i^\gamma\langle \mathrm{W}_i\,,\,\mathrm{N}^\gamma\rangle + \lambda_i^\gamma\langle \mathrm{W}_i\,,\,\mathrm{G}^\gamma\rangle\Big],\end{aligned}\tag{1.9}$$

$$\begin{aligned}=& \frac{1}{8}\sum_{ij}\mathbf{J}_{ij}^{\alpha\beta}\Big[4\big(\mathrm{w}_i^\alpha-\bar{\mathrm{w}}_i^\alpha\big)\big(\mathrm{w}_j^\beta-\bar{\mathrm{w}}_j^\beta\big) + 2\langle \mathrm{U}_{ij}\,,\,\mathrm{N}^\alpha\mathrm{U}_{ij}\mathrm{N}^\beta\rangle\Big]\\ &+\sum_{i,\alpha}\Big[-h_i^\alpha\big(\mathrm{w}_i^\alpha-\bar{\mathrm{w}}_i^\alpha\big)+\lambda_i^\alpha\bar{\mathrm{w}}_i^\alpha\Big],\end{aligned}\tag{1.10}$$

$$\begin{aligned}=& \frac{1}{4}\sum_{ij}\mathbf{J}_{ij}^{\alpha\beta}\langle \mathrm{U}_{ij}\,,\,\mathrm{N}^\alpha\mathrm{U}_{ij}\mathrm{N}^\beta\rangle+\sum_{i,\alpha}\Big[-h_i^\alpha\big(\mathrm{w}_i^\alpha-\bar{\mathrm{w}}_i^\alpha\big)+\lambda_i^\alpha\bar{\mathrm{w}}_i^\alpha\Big],\\ &+\frac{1}{2}\sum_i\big(\mathrm{w}_i^\alpha-\bar{\mathrm{w}}_i^\alpha\big)\sum_j\mathbf{J}_{ij}^{\alpha\beta}\big(\mathrm{w}_j^\beta-\bar{\mathrm{w}}_j^\beta\big).\end{aligned}\tag{1.11}$$

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

$$\langle H_{\mathrm{MF}}\rangle = \frac{1}{4}\sum_{ij}\mathbf{J}_{ij}^{\alpha\beta}\langle \mathrm{U}_{ij}\,,\,\mathrm{N}^\alpha\mathrm{U}_{ij}\mathrm{N}^\beta\rangle+\sum_{i,\alpha}\Big[-\tilde{h}_i^\alpha\big(\mathrm{w}_i^\alpha-\bar{\mathrm{w}}_i^\alpha\big)+\lambda_i^\alpha\bar{\mathrm{w}}_i^\alpha\Big],\tag{1.12}$$

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

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

$$\mathrm{w}_i^\gamma=\mathrm{w}^\gamma\ ,\quad \bar{\mathrm{w}}_i^\gamma=\bar{\mathrm{w}}^\gamma\ .\tag{1.14}$$

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

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

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 $\bar{\mathrm{w}}=0$, let us vary F w.r.t. $\bar{\mathrm{w}}$ and find the critical value for the Lagrange multiplier

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

where

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

Note that $\det\left(\mathbb{J}^{\alpha\beta}\right) = (-\Gamma + 3J + K)^2(2\Gamma + 3J + K)$ 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

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

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

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

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

and the classical polarized energy is $F = -\frac{1}{2}\left(\mathbb{J}^{-1}\right)^{\alpha\beta} h^\alpha h^\beta$. Note however that this solution breaks in the region where $\det\left(\mathbb{J}^{\alpha\beta}\right) > 0$, 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

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

which gives the free energy

$$2N\, F = -\sum_i h_i^\gamma \mathrm{w}_i^\gamma + \frac{1}{2}\sum_{ij} J_{\langle ij\rangle}^{\alpha\beta}\left(\mathrm{w}_i^\alpha \mathrm{w}_j^\beta - \bar{\mathrm{w}}_i^\alpha \bar{\mathrm{w}}_j^\beta\right) . \tag{1.22}$$

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

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

Where δh_i is such that the solution of the self-consistent equation satisfies the constraint $\langle \mathrm{W}_i\, , \mathrm{G}^\gamma\rangle = 0$. 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 $\langle \mathrm{W}_i\,,\, \mathrm{G}^\gamma\rangle$, 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 $\alpha_1, \alpha_2, \alpha_3$

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

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

$$\delta\boldsymbol{\lambda} = \alpha\mathbf{h} \tag{1.25}$$

implies in the constraint fulfillment

$$\Delta\mathrm{V}_i(\alpha) := \frac{1}{\sqrt{3}} ||\ \bar{\mathrm{w}}_i\ || = \frac{1}{4}\sqrt{\frac{1}{3}\sum_{\gamma=1}^{3} \langle \mathrm{W}_i\,,\, \mathrm{G}^\gamma\rangle^2}\ \to 0 \tag{1.26}$$

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

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

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 $\mathrm{G} : \langle \mathrm{W}, \mathrm{G}^\gamma\rangle = 4\bar{\mathrm{w}}^\gamma$). (**remark** to change $\mathrm{W}_i \to \mathrm{V}_i$ in all text). In the isotropic case, this quantity reduces to the expected expression $\Delta\mathrm{V} = \frac{1}{2}\left|\langle ic^0c^\gamma\rangle + \langle ic^\alpha c^\beta\rangle\right|$ with normalization $\frac{1}{2}$. The bound for the constraint violation is

$$\begin{aligned}
0 \le ||\bar{\mathrm{w}}_i||^2 \le ||\mathrm{w}_i||^2 + ||\bar{\mathrm{w}}_i||^2 &= \frac{1}{4^2}\sum_\gamma \left(\langle \mathrm{W}_i\,,\, \mathrm{M}^\gamma\rangle^2 + \langle \mathrm{W}_i\,,\, \mathrm{G}^\gamma\rangle^2 \right) \\
&= \frac{1}{2}\Big(\langle \mathrm{i}c^0c^1\rangle^2 + \langle \mathrm{i}c^0c^2\rangle^2 + \langle \mathrm{i}c^0c^3\rangle^2 + \langle \mathrm{i}c^1c^2\rangle^2 + \langle \mathrm{i}c^2c^3\rangle^2 + \langle \mathrm{i}c^3c^1\rangle^2 \Big) \\
&\le \frac{1}{2}\Big(\langle(\mathrm{i}c^0c^1)^2\rangle + \langle(\mathrm{i}c^0c^2)^2\rangle + \langle(\mathrm{i}c^0c^3)^2\rangle + \langle(\mathrm{i}c^1c^2)^2\rangle + \langle(\mathrm{i}c^2c^3)^2\rangle + \langle(\mathrm{i}c^3c^1)^2\rangle \Big) \\
&= 3 \\
&\Rightarrow \quad 0 \le \Delta\mathrm{V} \le 1\ .
\end{aligned}$$

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

$$\alpha = 0 \quad \Rightarrow \quad \Delta\mathrm{V}_i(\alpha) \lesssim 10^{-2}\ . \tag{1.28}$$

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 $\Delta\mathrm{V} < 10^{-5}$ for any value of h as shown in Fig. 1.1.

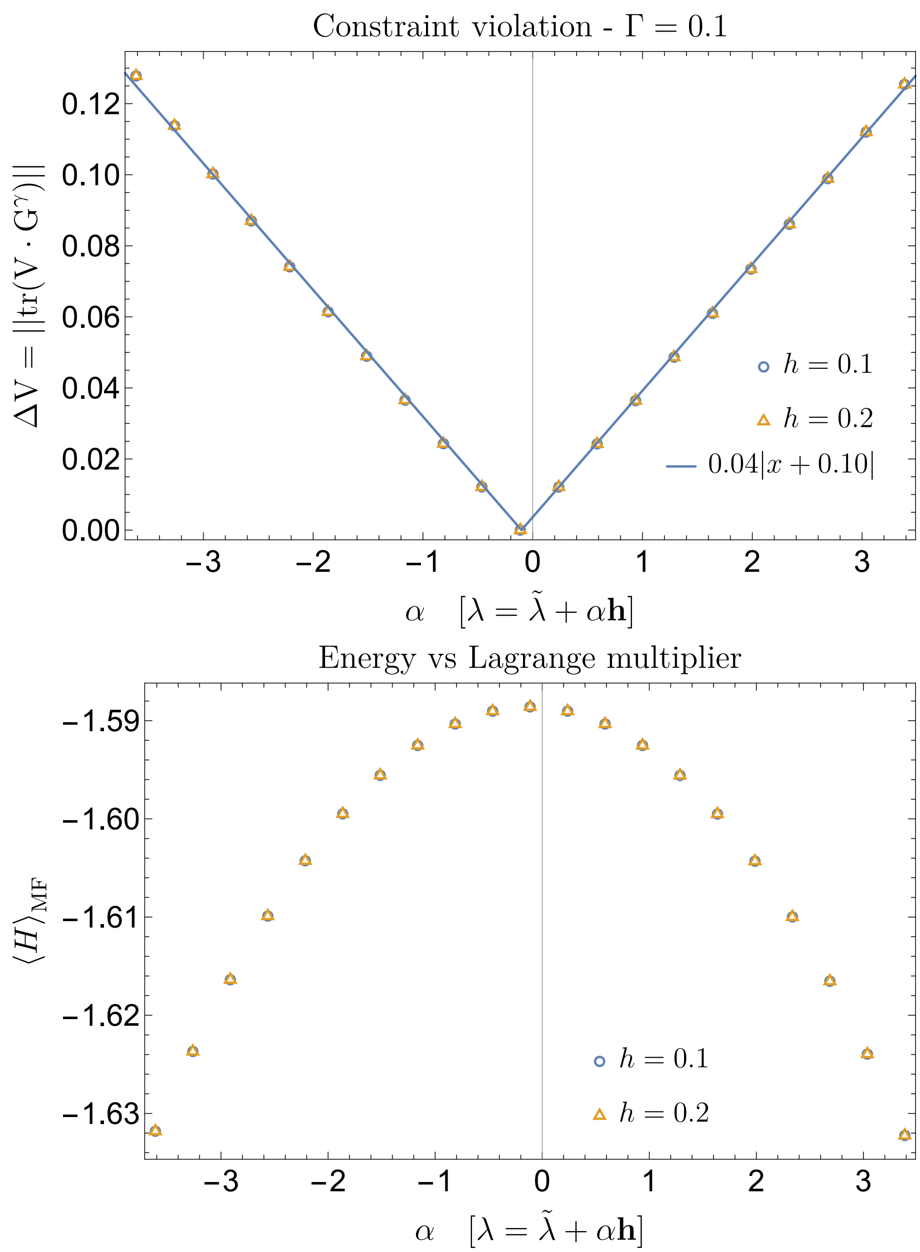


Figure 1.1: The value for the Lagrange multiplier is $\delta\lambda = -0.11\mathbf{h}$.

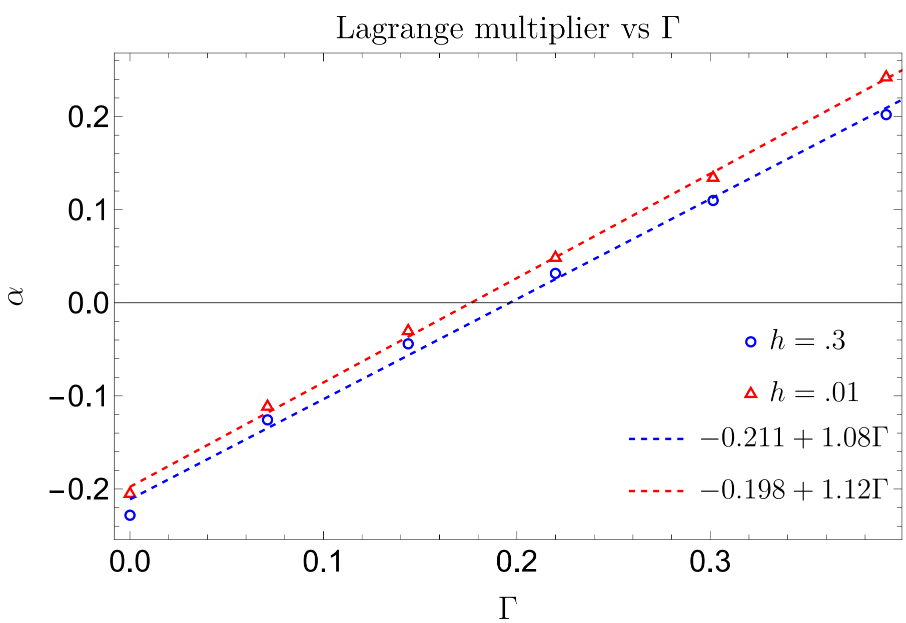


Figure 1.2: $\alpha(\Gamma)$ for two values of the magnetic field. The dependence in the magnetic field is very weak and in the KSL phase a good approximation is $\alpha(\Gamma) = -0.2 + 1.1\,\Gamma$.

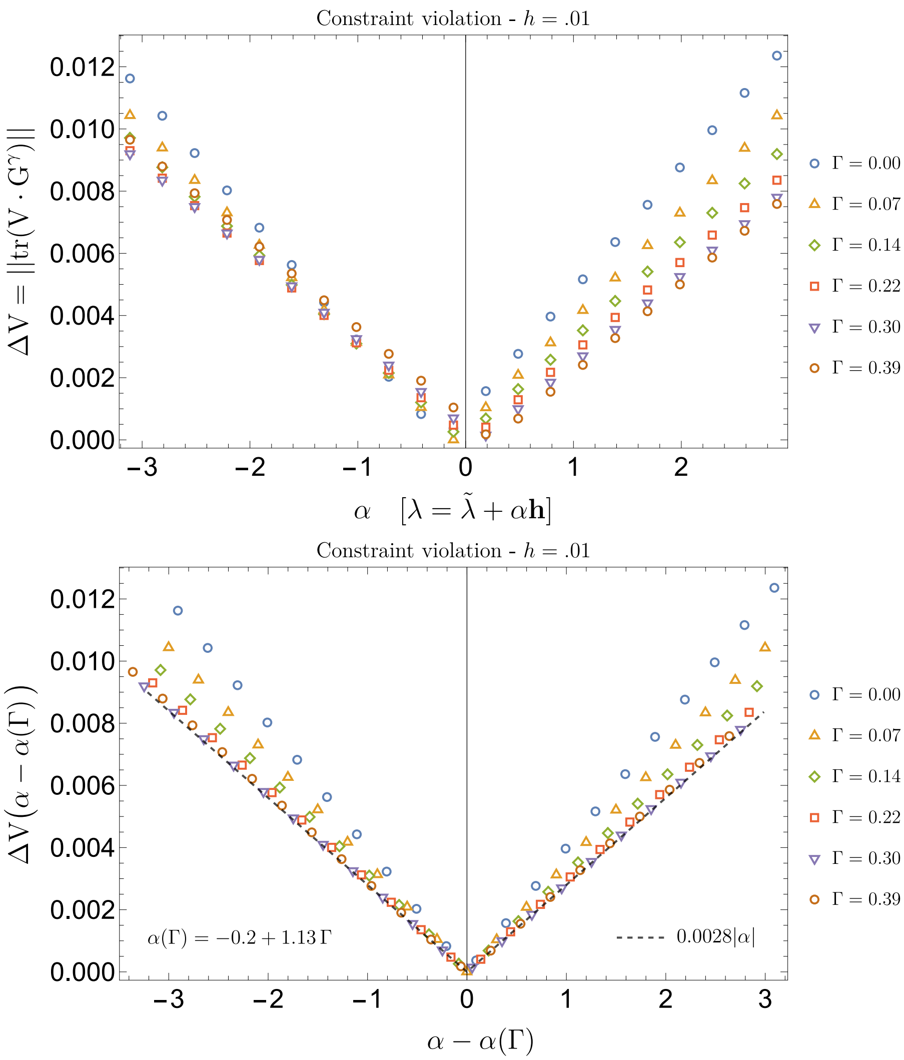
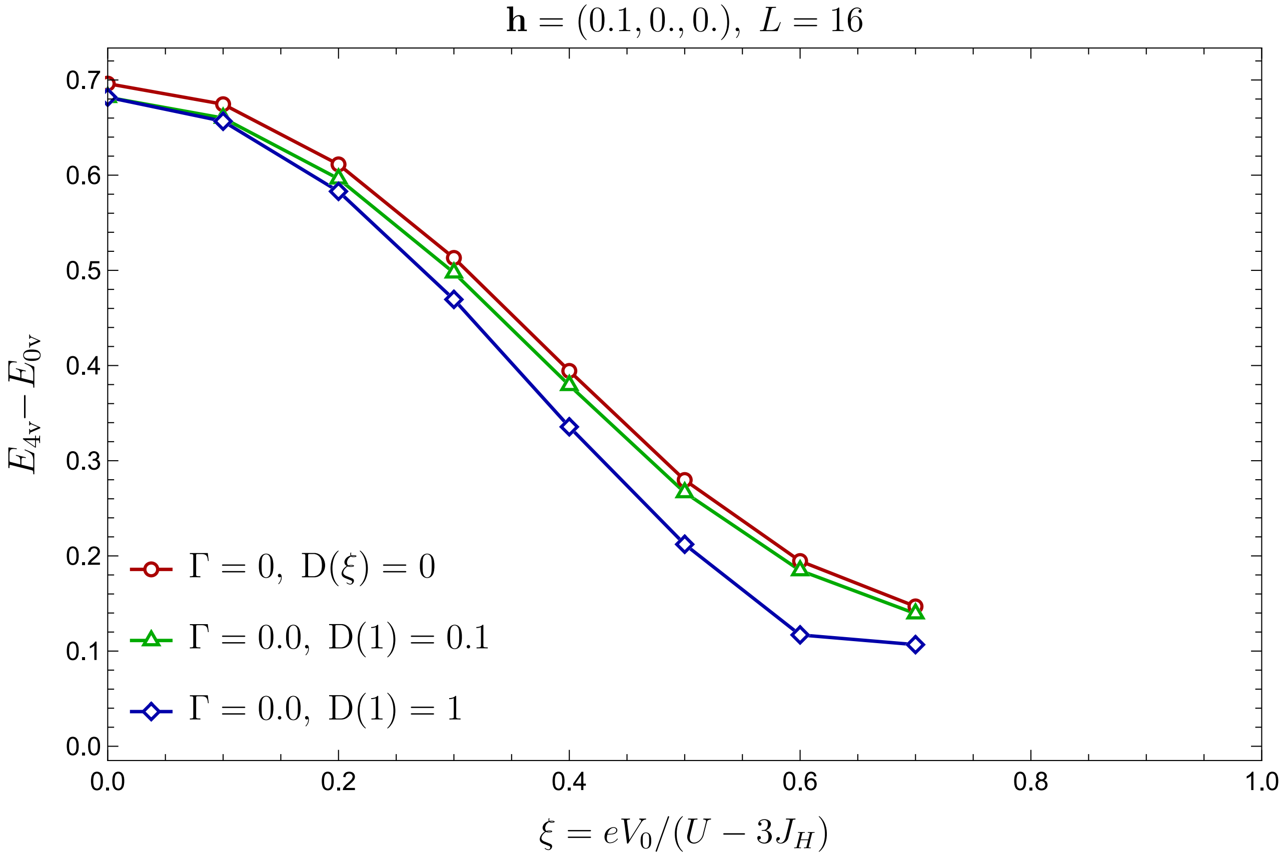


Figure 1.3: Constraint violation vs Lagrange multiplier for some values of Γ. Numerical fit: $\alpha(\Gamma) = -0.2 + 1.13\Gamma$.



1.3 Kitaev + Dzyaloshinskii Moriya

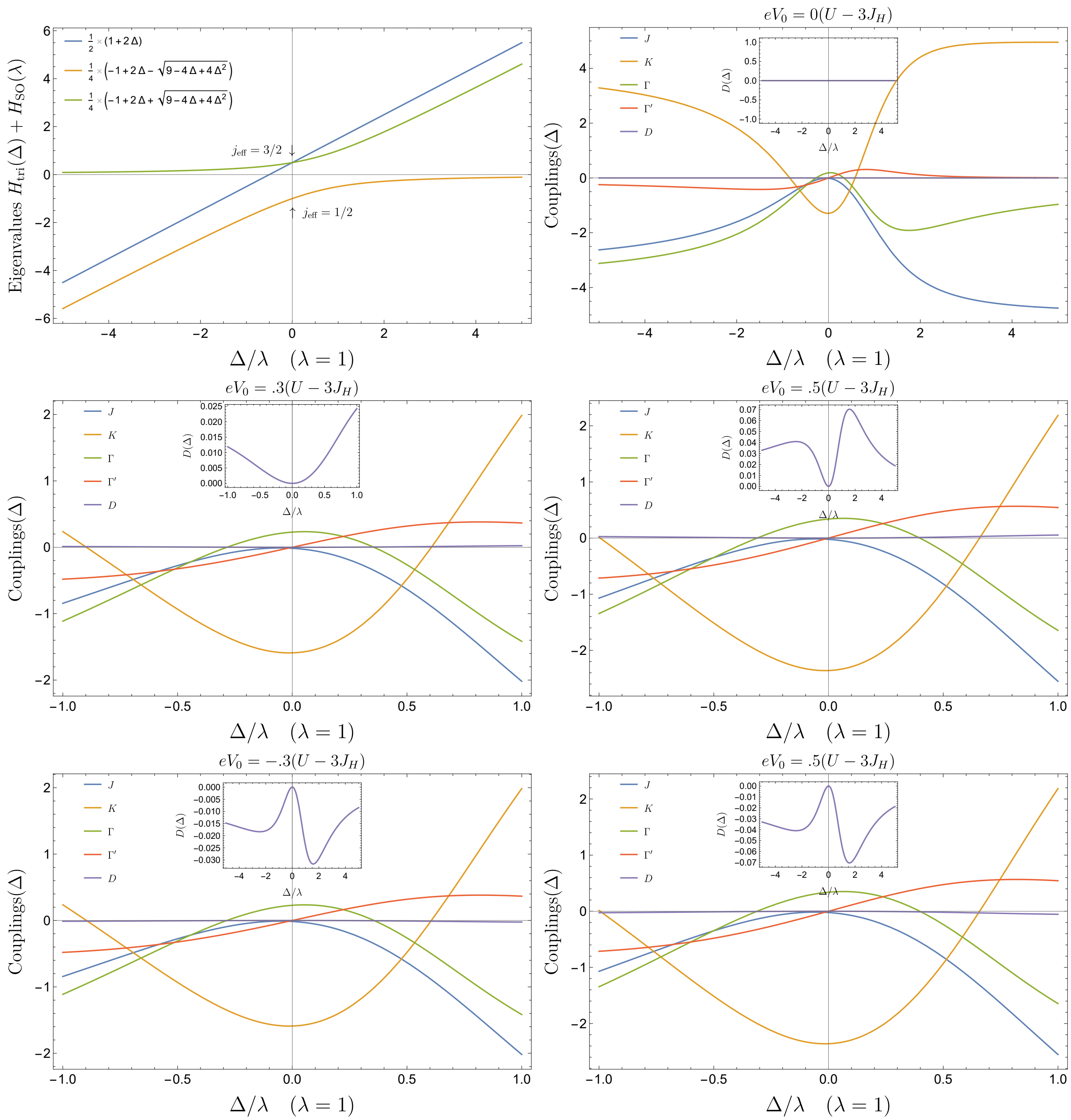
Manually inserting a Dzyaloshinskii Moriya interaction $D(\sigma_i^\alpha \sigma_j^\beta - \sigma_i^\beta \sigma_j^\alpha)$ proportionally to the applyed ultralocal electric field,

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

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 $10Dq \sim 2 - 2.2$eV [**?**], spin-orbit coupling $\lambda_{\mathrm{Ru}} \sim 100 - 150$meV, coulomb exchange $U \sim 2.4 - 2.6$eV, $J_H \sim 0.3 - 0.4$eV and hopping integrals $t_2 \sim 160$meV, $t_1 \sim 60$meV, $t_3 \sim -100$meV.

This minimal model generates an effective spin $j_{\mathrm{eff}} = 1/2$ 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 $\Delta_{\mathrm{tri}} \sim 20$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 $\sum_i eV_0N_i$, (2) enhance the trigonal distortion $\Delta_{\mathrm{tri}}(V_0)$, and (3) to change the orbital wave-functions. Previously we only had to take into account (1), which only renormalizes the couplings.



$\frac{1}{2} \times (1+2\Delta)$
$\frac{1}{4} \times \left(-1+2\Delta-\sqrt{9-4\Delta+4\Delta^2}\right)$
$\frac{1}{4} \times \left(-1+2\Delta+\sqrt{9-4\Delta+4\Delta^2}\right)$
$j_{\mathrm{eff}} = 3/2$
$j_{\mathrm{eff}} = 1/2$
Eigenvalues $H_{\mathrm{tri}}(\Delta) + H_{\mathrm{SO}}(\lambda)$
$\Delta/\lambda \quad (\lambda = 1)$
$eV_0 = 0(U - 3J_H)$
$eV_0 = .3(U - 3J_H)$
$eV_0 = .5(U - 3J_H)$
$eV_0 = -.3(U - 3J_H)$
$eV_0 = .5(U - 3J_H)$
J
K
Γ
Γ'
D
Couplings(Δ)
$D(\Delta)$
Δ/λ