Hybridized Criticality and Elementary Excitations in LiHoF4

Haifu Ma
Graduate Center, City University of New York
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by

Haifu Ma

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Date

Professor Vadim Oganesyan
Chair of Examining Committee

Date

Professor Igor Kuskovsky
Executive Officer

Professor Eugene Chudnovsky

Professor Dmitry Garanin

Professor Pouyan Ghaemi

Professor Steven Girvin

Professor Tobias Schäfer

Supervisory Committee

THE CITY UNIVERSITY OF NEW YORK
Abstract

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Advisor: Vadim Oganesyan, Professor of Physics

In this dissertation, I study the magnetic properties of LiHoF$_4$. Quantum criticality in rare earth ferromagnet LiHoF$_4$ is complicated by the presence of strong crystal field and hyperfine interactions resulting, e.g., in incomplete mode softening reported by Rønnnow et al. We construct a systematic framework for treating elementary excitations in this material across the phase diagram. These excitations interpolate between purely electronic, nuclear and lattice modes and exhibit two-types of quantum critical softening, both complete (as anticipated by elementary treatments, see e.g. Sachdev) but also incomplete, in close correspondence with nuclear scattering results.
To My Dear Parents.
Acknowledgements

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Introduction

1.1 Background

Quantum criticality is a continuous phase change between ground states of matter that differ macroscopically and therefore cannot be reached by adiabatic evolution of the ground state. While manifestations of quantum criticality in the real world are complicated and oftentimes controversial, model theoretical studies firmly established this phenomenon. Perhaps the simplest such model is the quantum (transverse-field) Ising model, which also appears to be realized in a number of materials. One such material is a rare-earth compound LiHoF$_4$. Here the Ising degree of freedom is the crystal-field doublet ground state separated by about 10 K from the first excited state. While early thermodynamic measurements clearly established the existence of quantum criticality near 5 T, subsequent studies and careful theoretical work\[1\] highlighted puzzles in the shape of the phase diagram but also in the nature of collective dynamics. In particular, neutron scattering\[2\] appears to suggest absence of quantum critical mode softening, an observation interpreted by its discoverers as evidence of entanglement with "nuclear bath".

1.2 Summary of Ising criticality in LiHoF$_4$

Uniaxial ferromagnets (and ferroelectrics) have been studied since early days of classical phase transitions research. Dipolar systems in particular have been of interest in part due to enhanced universality of phase transitions\[3\] and mesoscale pattern formation\[4\] induced by frustrating nature
of the interaction. Much of the attention lavished on LiHoF$_4$ in recent years is due to the possibility of realizing a transverse field driven quantum critical point in this material. Theoretical and experimental results confirm this scenario. In particular, high quality thermodynamic susceptibility measurements clearly display critical scaling over 4 decades, while nuclear scattering spectroscopy exhibits field induced softening, albeit incomplete of the spin wave spectrum. Notwithstanding this progress several puzzles remain, which have motivated this work and which we now outline. First, the shape of the experimentally observed phase boundary in the limit of vanishing transverse field appears inconsistent with existing theoretical calculations. Second, incomplete mode softening is inconsistent with observed thermodynamic signatures of the transition. Proposed explanation for this phenomenon in terms of the so-called "1/z" theory essentially dismisses the observed critical mode as not critical and appears to leave open the question of the nature of the true critical mode, as it must involve nuclear dynamics.

1.3 Summary of this work

We build on prior work by deriving transverse field dependent interactions of the Ising doublet with the nuclear spin and the ionic lattice. We analyse collective physics induced by these couplings using standard "single mode" approximation for hybrid spin-nuclear-phonon excitations of this system. We find that in addition to the "electron spin" mode already documented in neutron scattering this model supports a sharp low energy "hyperfine" mode, with typical dispersion on scales of 1GHz, which becomes strongly hybridized with the electron mode near the quantum critical point. We also reconsider the equilibrium phase diagram in the presence of both hyperfine and magnetoelastic couplings.
Review of basic theory – the transverse field Ising model (TFIM)

2.1 Mean field theory for 3D Ising model (T=0 K)

Consider the nearest neighbor coupled Ising model on the cubic lattice, described by Hamiltonian

\[ H = -J \sum_j \sigma^z_j \sigma^z_{j+1} - h \sum_j \sigma^x_j \]  

(2.1)

J is exchange energy, h is transverse field.

Let \( m = \langle \sigma^z_j \rangle \), \( n = \langle \sigma^x_j \rangle \)

\[ H_{MF} = - \sum_j (qJm \sigma^z_j + h \sigma^x_j) \]  

(2.2)

Single site Hamiltonian is:

\[ H_{MF}' = -(qJm \sigma^z + h \sigma^x) = -h_n \sigma_n \]  

(2.3)

\[ h_n = \sqrt{h^2 + (qJm)^2} \]  

(2.4)

\[ \sigma^z = \sin \theta \sigma_n \]  

(2.5)

\[ \sigma^x = \cos \theta \sigma_n \]  

(2.6)
And if we let $\langle \sigma^n \rangle = 1$, then we can get,

\[
m = \langle \sigma^z \rangle = \sin \theta = \frac{qJm}{\sqrt{h^2 + (qJm)^2}} \quad (2.7)
\]

\[
n = \langle \sigma^z_j \rangle = \cos \theta = \frac{h}{\sqrt{h^2 + (qJm)^2}} \quad (2.8)
\]

### 2.2 Mean field theory for 3D Ising model ($T > 0K$)

\[
H_{MF} = -\sum_j (qJm\sigma^z_j + h\sigma^x_j) \quad (2.9)
\]

The partition function is:

\[
Z = (2 \cosh [\beta \sqrt{(qJm)^2 + h^2}])^N \quad (2.10)
\]

$\beta = \frac{1}{T}, K_B = 1$

\[
m = \frac{1}{N\beta} \frac{\partial \ln Z}{\partial m} = \frac{qJm}{\sqrt{h^2 + (qJm)^2}} \tanh[\beta \sqrt{h^2 + (qJm)^2}] \quad (2.11)
\]

Cancel $m$, then let $m=0$, we can get the critical relation:

\[
h = qJ \tanh(\beta h) \quad (2.12)
\]

We can calculate $T = T(h_c)$.

### 2.3 Dynamics:

semi-classical spin-wave (single particle) approximation

\[
H = -J \sum_j \sigma^z_j \sigma^z_{j+1} - h \sum_j \sigma^x_j \quad (2.14)
\]
Use Holstein Primakoff transformation, we define:

\[ \sigma_j^n = m \sigma_j^z + n \sigma_j^x = \sigma - b_j^\dagger b_j \]  
(2.15)

\[ \sigma_j^m = -n \sigma_j^z + m \sigma_j^x = \frac{\sqrt{2}\sigma}{2}(b_j^\dagger + b_j) \]  
(2.16)

\[ (\sigma_j^n)^+ = \sigma_j^m + i \sigma_j^y = \sqrt{2}\sigma b_j^\dagger \]  
(2.17)

\[ (\sigma_j^n)^- = \sigma_j^m - i \sigma_j^y = \sqrt{2}\sigma b_j \]  
(2.18)

Then, we can get:

\[ \sigma_j^z = m \sigma_j^n - n \sigma_j^m \]  
(2.19)

\[ \sigma_j^x = n \sigma_j^n + m \sigma_j^m \]  
(2.20)

To the momentum space:

\[ \sum_k \sum_j b_j e^{ikj} = \sum_k b_k \]  
(2.21)

Then,

\[ H = \sum_{k>0} [\epsilon_k (b_k^\dagger b_k + b_{-k}^\dagger b_{-k}) + \rho_k (b_k^\dagger b_{-k} + b_{k} b_{-k})] \]  
(2.22)

\[ \epsilon_k = qJm^2\sigma - nh - n^2J\sigma \cos k \]  
(2.23)

\[ \rho_k = -n^2J\sigma \cos k \]  
(2.24)

use quantization

\[ b_k = x_1 + ip_1 \]
\[ b_k^\dagger = x_1 - ip_1 \]
\[ b_{-k} = x_2 + ip_2 \]
\[ b_{-k}^\dagger = x_2 - ip_2 \]
get

\[
H = \epsilon_k (x_1^2 + x_2^2 + p_1^2 + p_2^2) + \rho_k (2x_1 x_2 - 2p_1 p_2)
\]
(2.25)

\[
H = \frac{1}{2} (PM^{-1}P^T + XKKX^T)
\]
(2.26)

\[
P = (p_1, p_2)
\]

\[
X = (x_1, x_2)
\]

\[
M^{-1} = \begin{pmatrix}
\epsilon_k & -\rho_k \\
-\rho_k & \epsilon_k
\end{pmatrix}
\]

\[
K = \begin{pmatrix}
\epsilon_k & \rho_k \\
\rho_k & \epsilon_k
\end{pmatrix}
\]

the frequency is

\[
\omega^2 X = M^{-1} K X
\]
(2.27)

\[
\omega^2 = \frac{1}{2} \left\{ \frac{\epsilon_k^2 - \rho_k^2}{2} \pm \sqrt{\left(\frac{\epsilon_k^2 - \rho_k^2}{2}\right)^2} \right\}
\]
(2.28)
Moments and models for LiHoF$_4$

3.1 Degrees of freedom and the Hamiltonian

Relevant degrees of freedom in insulating LiHoF$_4$ are electron and nuclear spins of Ho$^{3+}$ ions (denoted, respectively, by total spin vector $\mathbf{J}$ and $\mathbf{I}$) and lattice displacement field, $\mathbf{U}$.

We parse the complete Hamiltonian of the system into four parts

$$H = H_J + H_{JJ} + H_{JU} + H_U, \tag{3.1}$$

capturing electron spin interactions (dipolar, exchange and with external and crystal fields, respectively)

$$H_J = \sum_i V_C(\vec{J}_i) - g_L\mu_B \sum_i B_x J_i^x + \frac{1}{2}(g_L\mu_B)^2 \sum_{i\neq j} L_{ij}^{\mu\nu} J_i^\mu J_j^\nu + \frac{1}{2}(g_L\mu_B)^2 J_{ex}^a \sum_{i,nn} \vec{J}_i \cdot \vec{J}_{nn} \tag{3.2}$$

$V_C(\vec{J}_i)$is crystal field, $B_x$is transverse field, $L_{ij}$is dipolar interaction, $J_{ex}$is exchange energy. And Lande g factor $g_L = \frac{5}{2}$, Bohr magneton $\mu_B = 0.6717K/T$.

---

1Spin-orbit, the largest of these couplings, is straightforwardly dealt with by switching to $J = L + S$ basis, with $J = 8$. 
hyperfine coupling

\[ H_{II} = A \sum_i \vec{I}_i \cdot \vec{J}_i \]  

(3.3)

This first two terms are well characterized in LiHoF\(_4\) and fully captured (we believe) by the terms above, the latter two a more complicated generally, so our treatment is necessarily a simplified caricature, e.g. we ignore nonlinearities and intricate details of full phonon bandstructure. Neither of which we believe to be important here. Importantly, as we describe below, these terms lead to direct, bilinear, coupling between Ising pseudospins and lattice deformations, leading to a possibility of strong hybridization among elementary excitations of the two subsystems.

Local physics of Ho\(^{3+}\) and other rare-earth moments is complicated by the interplay of spin-orbit, crystal field and hyperfine interactions. In the next subsection we focus on summarizing some salient features, including important matrix elements etc, in the absence of lattice displacements, i.e. \(U = 0\). Impatient reader should skip these details and return to them later for reference.

3.2 Non-Kramers doublet+nuclear spin of Ho

3.2.1 splitting of non-Kramers doublets

Now we follow Chakraborty’s[7] way to construct the model, single site \(H\) can be truncated as:

\[ H_T = V_C(\vec{J}) - g_L \mu_B B_x J_x \]  

(3.4)

\(V_C(\vec{J})\) is fixed crystal field of LiHoF\(_4\), for \(J=8\), the details can be found in the appendix. The only variable is \(B_x\), from 0 T to 10 T.

The ground state of \(V_C(\vec{J})\) is a doublet, which can be split by transverse field \(B_x\). Let the two lowest state be \(|\alpha(B_x)\rangle\) and \(|\beta(B_x)\rangle\), then the \(H_T\) in 17-dimension Hilbert space can be shown in 2-dimension subspace:

\[ H_T = E_C(B_x) - \frac{1}{2} \Delta(B_x) \sigma^x \]  

(3.5)

Where

\[ E_C = \frac{1}{2}(E_{\alpha} + E_{\beta}), \Delta(B_x) = (E_{\beta} - E_{\alpha}) \]  

(3.6)
3.2.2 projecting to Ising Model

To generate the full $H$, we need to project $\vec{J}$ to this 2-dim subspace:

$$J^\mu = C_\mu + \sum_\nu C_{\mu\nu} \sigma^\nu$$  \hspace{1cm} (3.7)

$\mu, \nu = x, y, z$. The basis of $\sigma^\nu$ is a combination of $|\alpha\rangle$ and $|\beta\rangle$, and we chose the phase so that $J_z$ is real:

$$J_z = C_{zz} \sigma^z$$  \hspace{1cm} (3.8)

Each C-number can be calculated for every $B_x$. After dropping all the small Cs, we can get the Ising model of LiHoF4:

$$H_{J, Ising} = \sum_i E_{C,i}(B_x) - \frac{1}{2} \Delta(B_x) \sum_i \sigma_i^x$$

$$+ \frac{1}{2} (g_L \mu_B C_{zz})^2 \sum_{i\neq j} L_{ij}^z \sigma_i^z \sigma_j^z$$

$$+ \frac{1}{2} \frac{g_L \mu_B C_{zz}}{a^3} J_{ex} \sum_{i, nn} \sigma_i^z \sigma_{nn}^z$$  \hspace{1cm} (3.9)
Let’s only consider nearest neighbour and drop the long range interaction, so this two terms can be combined as:

\[-J \sum_j \sigma^z_j \sigma^z_{j+1} = \frac{1}{2} \left( g_L \mu_B C_{zz} \right)^2 \sum_{i \neq j} L_{ij}^{zz} \sigma^z_i \sigma^z_j + \frac{1}{2} \left( g_L \mu_B C_{zz} \right)^2 \frac{J_{ex}}{a^3} \sum_{i,nn} \sigma^z_i \sigma^z_{nn} \]

(3.10)

We let $\Delta = \frac{1}{2} \Delta(B_x)$, and neglect the constant term, then get:

\[H_J = -J \sum_j \sigma^z_j \sigma^z_{j+1} - \Delta \sum_j \sigma^x_j \]

(3.11)

We know the transition temperature is $T_c = 1.53K$ for $B_x = 0$, so here we use this value to fit in the equation, to obtain $J$’s value:

\[qJ = T_c - \frac{A_z^2}{T_c} = 1.41K \]

(3.12)

$q$ is the coordination number, and we will get $A_z$’s value later.
3.2.3 hyperfine interactions

Hyperfine interactions in LiHoF$_4$ are simply given by fully symmetric antiferromagnetic coupling on each Ho$^{3+}$

$$H_{IJ} = AJ \cdot I, \quad (3.13)$$

with $A \approx 0.039 K$. Despite the smallness of this coupling constant, e.g. compared to ordering temperature in zero field, the effects of hyperfine interactions are known to be very strong, primarily due to very large magnitudes of both sets of spins involved.

We may use $J \to \sigma$ mapping to approximate hyperfine interactions (assuming no longitudinal fields)

$$H_{JI} = A \sum_i \vec{I}_i \cdot \vec{J}_i = A(C_{\mu} + \sum_{\nu} C_{\mu\nu} \sigma^\nu) I^\nu \quad (3.14)$$

The only significant large terms are: $\Delta_I = AC_x, A_z = AC_z$. This reduction is valid for static transverse fields that induce static expectation value of transverse electron polarization responsible for local effective nuclear field. In fact, at low temperatures one expects this form to hold even for time dependent external fields, provided it is sufficiently slow as not to excite across the 10 K gap in the electronic system.

Now, the Hyperfine interactions is:

$$H_{JI} = \Delta_I \sum_j I^x_j + A_z \sum_j \sigma^z_j I^z_j \quad (3.15)$$

3.3 The effective Ising model with nuclei

After projecting onto the Ising pseudospin we are left with the following reduced/effective Hamiltonian of the system

$$H_{\text{Ising}} = -J \sum_j \sigma^z_j \sigma^z_{j+1} - \Delta \sum_j \sigma^x_j + \Delta_I \sum_j I^x_j + A_z \sum_j \sigma^z_j I^z_j \quad (3.16)$$

We plot characteristic dependence of these on the external transverse field $H_\perp(B_x)$. 
Figure 3.3: The hyperfine couplings
4

Application of mean field theory to LiHoF$_4$

4.1 Mean field theory

Phase boundary separating paramagnet and ferromagnet is complicated by presence of hyperfine interactions, which have been previously accounted for self-consistently numerically, see e.g. chakraborty’s [7]. We begin by deriving the phase diagram analytically as we are interested in attributing clear physical interpretation to known features of $T_C(B_x)$ curve. Starting with the Hamiltonian describing electron and nuclear spins only

$$H = -J \sum_{(ij)} \sigma_i^z \sigma_j^z - \sum_j (\Delta \sigma_j^x - \Delta I_j^x - A_z \sigma_j^z I_j^z)$$  \hspace{1cm} (4.1)$$

where all parameters depend on the externally applied transverse field $B_x$ (see Fig. 3.2, Fig. 3.3), let

$$m = \langle \sigma_j^z \rangle, \hspace{1cm} (4.2)$$

$$u = \langle I_j^z \rangle, \hspace{1cm} (4.3)$$
we can apply the standard Hubbard-Stratonovich transformation to the partition function to obtain uniform mean-field approximation from the saddle point condition for the action

\[
Z = \text{tr} \int \mathcal{D}m \mathcal{D}ue^{-\beta(J \sum_{(ij)} m_im_j - A_z \sum_j m_ju_j) - \beta H} \tag{4.4}
\]

\[
\approx e^{-\beta V(Jdm^2 - A_zmu)} Z^V_\sigma (\beta|h_\sigma|) Z^V_I (\beta|h_I|), \tag{4.5}
\]

where “tr” denotes traces over spin variables, \(d = 3\) is the dimension of space, \(V = L^d\) is the total number of lattice sites and single electron/nuclear spin partition function and effective field, respectively,

\[
Z_\sigma = \text{tr} \left[ e^{-\beta (\vec{\sigma} \cdot \vec{h}_\sigma)} \right], \quad h_\sigma = (\Delta, 0, A_zu - 2dJm); \tag{4.6}
\]

\[
Z_I = \text{tr} \left[ e^{-\beta (\vec{I} \cdot \vec{h}_I)} \right], \quad h_I = (-\Delta_I, 0, A_zm). \tag{4.7}
\]

Finally, the two mean field equations are

\[
2dJm - A_zu = T \frac{\partial}{\partial m} \log(Z_\sigma Z_I) \tag{4.8}
\]

\[
-A_zm = T \frac{\partial}{\partial u} \log(Z_\sigma) \tag{4.9}
\]

While \(Z_\sigma = 2 \cosh \beta|h_\sigma| \equiv Z_2(2\beta|h_\sigma|)\) may be evaluated exactly and manipulated straightforwardly (see below), \(Z_I = 2 \sum_{j=1/2}^{7/2} \cosh j\beta|h_I|\) of the eight level nuclear levels is cumbersome. There are two alternative simplification strategies, one to replace \(I = 7/2\) quantum spin with an effective two level system via \(\vec{I} \rightarrow I\vec{\sigma}\) and another by taking the continuum (classical) limit, replacing spin with a classical three component vector \(\vec{I} \rightarrow I\hat{n}\), where \(|\hat{n}| = 1\). These two approximations have complimentary domains of validity, with “classical” one being particularly accurate at finite temperatures of interest, so we will work out the rest of the mean-field theory identifying \(Z_I(\beta h_I) \approx Z_\infty(\beta I h_I)\) and

\[
Z_\infty = \int dcos(\theta) e^{-\beta h_I \cos(\theta)} = \frac{2 \sinh \beta I|h_I|}{\beta I|h_I|} \tag{4.10}
\]
Mean field Equations. 4.8 and 4.9 become

\[ m = \frac{(2dJm - A_zu)}{|h_\sigma|} \tanh \beta|h_\sigma| \]  
(4.11)

\[ u = \frac{-A_zmI}{|h_I|} (\coth \beta I|h_I| - \frac{1}{\beta I|h_I|}) \]  
(4.12)

In addition to trivial (paramagnetic) solution \( u = m = 0 \), ordered solution(s) may be found by explicitly dividing out \( m \) and/or \( u \), e.g. by substituting Eq. 4.9 into Eq. 4.8 and then working in the limit of vanishing (but finite) \( m \) and \( u \):

\[ \Delta = \left[ 2dJ + \frac{A_z^2 I}{\Delta I} (\coth \beta I\Delta I - \frac{1}{\beta I\Delta I}) \right] \tanh \beta \Delta \]  
(4.13)

We solve this equation for \( T = T(B_x) \) using parameters shown in Fig. 3.2 Fig. 3.3 get \( \Delta_c = 2dJ + \frac{A_z^2 I}{\Delta I}, B_{x,c} = 4.97(T) \), and plot the solution in Fig. 4.1 .

The results fit very well with Bitko etal’s[5] experiment values, see Fig. 4.2.

For \( A=0 \) this reduces to the commonly studied TFIM case, which we shall use to quantify hyperfine effects. Due to separation of scales between hyperfine and dipolar interactions, we may think of three distinct regimes/approximations demarcated with help of a temperature scale, \( \Delta T_C \approx 0.12K \): near the classical \( T_C(B_x \to 0) \) both nuclear and electron spins are thermally distributed; both
Figure 4.2: Compare with experiment (filled circles), $B_{x,c} \sim 4.9T$.

nuclear and electron spins are nearly frozen near their respective groundstates near the quantum critical point, $T_C \to 0$, and up to $\Delta T_C$;

only nuclear spins thermally excited, just above $\Delta T_C$.

Now compare with $A=0$ (Fig 4.3):

$$\Delta T_C = A_z^2 I / 2dJ, \Delta B_{x,c} = A_z^2 I / \Delta I, \Delta T_A = 3\Delta I$$

(4.14)
Figure 4.3: $T$ vs $B_x$ with $A=0$ (dash line)
5

Elementary excitations

5.1 Elementary excitations

Elementary excitation dispersion may be computed using any one of standard "semiclassical" tools, e.g. Holstein-Primakoff or Schwinger bosons, also various large N schemes, and more simply equation-of-motion approach. We shall use Holstein-Primakoff bosons as this also affords us direct access to wave functions of these excited states.

\[ H_{Ising} = -J \sum_j \sigma_j^z \sigma_{j+1}^z - \Delta \sum_j \sigma_j^x + \Delta_I \sum_j I_j^x + A_z \sum_j \sigma_j^z I_j^z \]  \hspace{1cm} (5.1)

Let:

\[ m' = \langle \sigma_j^z \rangle / \sigma = \sin \theta, \]  \hspace{1cm} (5.2)

\[ n = \langle \sigma_j^x \rangle / \sigma = \cos \theta, \]  \hspace{1cm} (5.3)

\[ u' = \langle I_j^z \rangle / I = \sin \phi, \]  \hspace{1cm} (5.4)

\[ v = \langle I_j^x \rangle / I = \cos \phi, \]  \hspace{1cm} (5.5)
Again, we use Holstein Primakoff transformation,

\begin{align}
\sigma^n &= m'\sigma^z + n\sigma^x = \sigma - b^\dagger b \\
\sigma^m &= -n\sigma^z + m'\sigma^x = \frac{\sqrt{2}\sigma}{2}(b^\dagger + b) \\
I^n &= -u'I^z - vI^x = I - a^\dagger a \\
I^m &= vI^z - u'I^x = \frac{\sqrt{2I}}{2}(a^\dagger + a)
\end{align}

Then get,

\begin{align}
\sigma^z &= m'\sigma^n - n\sigma^m \\
\sigma^x &= n\sigma^n + m'\sigma^m \\
I^z &= -u'I^n + vI^m \\
I^x &= -vI^n - u'I^m
\end{align}

to the momentum space,

\[\sum_k \sum_j b_j e^{ikj} = \sum_k b_k, \sum_k \sum_j a_j e^{ikj} = \sum_k a_k\]

\[H_{\text{Ising}} = \sum_{k>0} \left[ \epsilon_k (b_k^\dagger b_k + b_{-k}^\dagger b_{-k}) + \rho_k (b_k^\dagger b_{-k}^\dagger + b_k b_{-k}) \right. \]

\[+ \gamma(a_k^\dagger a_k + a_{-k}^\dagger a_{-k}) \]

\[+ \xi(b_k^\dagger a_{-k}^\dagger + b_{-k}^\dagger a_k^\dagger + b_k a_{-k} + b_{-k} a_k + b_k^\dagger a_k^\dagger + b_{-k}^\dagger a_{-k}^\dagger) \]

\[= 2m'^2 J\sigma - n\Delta - n^2 J\sigma \cos k + m'u'A_z I \]

\[\epsilon_k = 2m'^2 J\sigma - n\Delta - n^2 J\sigma \cos k + m'u'A_z I \]

\[\rho_k = -n^2 J\sigma \cos k \]

\[\gamma = v\Delta I + m'u'A_z \sigma \]

\[\xi = -\frac{1}{2}(nvA_z)\sqrt{\sigma I} \]
Use quantization

\[ b_k = x_1 + ip_1 \]
\[ b_k^\dagger = x_1 - ip_1 \]
\[ b_{-k} = x_2 + ip_2 \]
\[ b_{-k}^\dagger = x_2 - ip_2 \]
\[ a_k = y_1 + iq_1 \]
\[ a_k^\dagger = y_1 - iq_1 \]
\[ a_{-k} = y_2 + iq_2 \]
\[ a_{-k}^\dagger = y_2 - iq_2 \]

get

\[ H_{\text{Ising}} = \frac{1}{2} (PM^{-1}P^T + KXX^T) \]  \hspace{0.5cm} (5.15)

\[ P = (p_1, p_2, q_1, q_2) \]
\[ X = (x_1, x_2, y_1, y_2) \]

\[
M^{-1} = \begin{pmatrix}
\epsilon_k & -\rho_k & \xi & -\xi \\
-\rho_k & \epsilon_k & -\xi & \xi \\
\xi & -\xi & \gamma & 0 \\
-\xi & \xi & 0 & \gamma
\end{pmatrix}
\]

\[
K = \begin{pmatrix}
\epsilon_k & \rho_k & \xi & \xi \\
\rho_k & \epsilon_k & \xi & \xi \\
\xi & \xi & \gamma & 0 \\
\xi & \xi & 0 & \gamma
\end{pmatrix}
\]
the frequency is

\[ \omega^2 X = M^{-1} K X \]  \hspace{1cm} (5.16)

\[ \omega^2 = \frac{1}{2} \left\{ \epsilon_k^2 - \rho_k^2 + \gamma^2 \pm \sqrt{(\epsilon_k^2 - \rho_k^2 - \gamma^2)^2 + 16 \xi^2 \gamma (\epsilon_k - \omega_k)} \right\} \]  \hspace{1cm} (5.17)

Let momentum \( k=0, \sigma = 1, I = 7/2 \), and use mean field equations 4.11 and 4.12, we can calculate the \( \omega_\pm \) for a given \( T \). For \( T=0 \) K, \( B_{x,c} = 4.97(T) \) as shown in Fig 5.1, this confirm our former result. \( \omega \)'s unit is K.

![Graph](image)

Figure 5.1: \( \omega \) vs Bx

### 5.2 Discussion

#### 5.2.1 case 1

Add the \( A=0 \) curve, and compare with Rønnow et al.'s experiment values, we can find that for \( A > 0 \), there is an energy gap, and two curves will coincide after critical point. (Fig 5.2, 5.3)
Figure 5.2: $\omega_+/K$ vs $B_x/T$

Figure 5.3: Rønnow et al.’s experiment values
5.2.2 case 2

We can see the temperature dependence of the Energy Gap, the calculation shows the same energy peak movements as in the Conradin Kraemer’s work. (Fig 5.4, 5.5)

Figure 5.4: Temperature dependence of the Energy Gap, for $T=0, 0.06, 0.15, 0.3, 0.5, 0.75, 1, 1.2, 1.53\,\text{(K)}$
Figure 5.5: Conradin Kraemer’s work
5.2.3 case 3

In $\omega_-$, we find probably detectable lower mode in $0.015 \text{ K} \sim 0.045 \text{ K}$ range, that is $500 \text{ MHz} \sim 900 \text{ MHz}$ in microwave(Fig 5.6, 5.7), which doesn’t show in Rønnow etal’s[2] experiment that using neutron scattering (Fig.5.3).

![Figure 5.6: Probably detectable lower mode](image)

$\omega_- / \text{K}$

![Figure 5.7: detectable range of $\omega_-$](image)
5.2.4 case 4

Along critical line of "T vs Bx", plot $\omega_+ \text{ vs } B_x$. We can see it fits well with Fig.5.4 (Fig 5.8, 5.9)

$\omega_+/K$

![Graph 1](image1)

$B_x/T$

Figure 5.8: $\omega_+ \text{ vs } B_x$ along critical line

$\omega_+/K$

![Graph 2](image2)

$B_x/T$

Figure 5.9: dash line fits the Temperature dependence
Conclusion

We calculate the full hamiltonian of LiHoF$_4$ with mean field theory, and get the phase diagram. The figure fits the experiment values very well.

We also use semi-classical approximation, get the energy mode of LiHoF$_4$. Then we discuss different cases and compare with the experiment results.

At last, we suggest a possible mode to be detected in the experiment using microwave.
Appendix A

The crystal field hamiltonian

In the LiHoF$_4$, the crystal field hamiltonian could be written as:

\[ V_c = B_2^0 O_2^0 + B_4^0 O_4^0 + B_6^0 O_6^0 + B_4^1(C)O_4^1(C) + B_6^1(C)O_6^1(C) + B_6^4(S)O_6^4(S). \]  

(A.1)

The $O_m^l$ are the Stevens operators. And in this paper, we use parameters shown in the Rønnow et al.'s [6]. The values are:

- $B_2^0 = -0.696$  
- $B_4^0 = 4.06 \times 10^{-3}$  
- $B_6^0 = 4.64 \times 10^{-6}$  
- $B_4^1(C) = 0.0418$  
- $B_6^1(C) = 8.12 \times 10^{-4}$  
- $B_6^4(S) = 1.137 \times 10^{-4}$  

(A.2) \hspace{1cm} (A.3) \hspace{1cm} (A.4) \hspace{1cm} (A.5) \hspace{1cm} (A.6) \hspace{1cm} (A.7)
Bibliography


Publications