QUANTUM MAGNETISM
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20.10.2023

PROBLEM SHEET 2
Winter Term 2023/24

Submission deadline for the homework questions: 3.11.2023 (hand in via moodle).
Please prepare the remaining questions for the tutorials in the following week and be ready to present the questions marked with a star (*).

Exercise 2.1*: Van Vleck paramagnetism

Let's consider an ion whose ground state (electronic configuration) is not degenerate with respect to total orbital angular momentum which is given in Eq.1 or Eq.(33) of the lecture notes.

$$\boldsymbol{L} = \sum_i \frac{\hbar}{i} \boldsymbol{r}_i \times \nabla_i \tag{1}$$

In this case, $\boldsymbol{L}$ of an ion should be quenched, $\langle g|\boldsymbol{L}|g\rangle = 0$ where $|g\rangle$ is the unique ground state of an ion.

(a) Prove that $\langle g|\boldsymbol{L}|g\rangle = 0$ using the fact that physical quantities should be real in the real-space (Schrodinger) representation.

Note: If the ground state is degenerate, the proof fails since the ground state wavefunction can be a superposition of the degenerate states with complex coefficients.

A small orbital contribution of magnetic moment can be restored if we include spin-orbit coupling as below in a perturbative manner.

$$H_1 = \lambda \boldsymbol{L} \cdot \boldsymbol{S} + \mu_B \boldsymbol{B} \cdot (2\boldsymbol{S} + \boldsymbol{L}) \tag{2}$$

(b) Derive the first order, ΔE_1, and second-order, ΔE_2, energy correction of the ground state. Assume that $\boldsymbol{S}$ is a good quantum number, i.e.

the bare Hamiltonian H_0 commute with $\boldsymbol{S}$. You should find

$$\begin{aligned} \Delta E_1 &= 2\mu_B \boldsymbol{B}\cdot\boldsymbol{S} \\ \Delta E_2 &= \sum_{\mu,\nu}(\lambda^2\Lambda_{\mu,\nu}S_\mu S_\nu + 2\lambda\mu_B\Lambda_{\mu,\nu}B_\mu S_\nu + \mu_B^2\Lambda_{\mu,\nu}B_\mu B_\nu) \\ \text{where } \Lambda_{\mu,\nu} &= \sum_{|\psi\rangle\neq|g\rangle}\frac{\langle g|L_\mu|\psi\rangle\langle\psi|L_\nu|g\rangle}{E_\psi - E_g} \text{ with excited states } |\psi\rangle. \end{aligned}$$

(c) Using Eq.3 or Eq.(41) of the lecture notes, derive the magnetic moment of a single ion.

$$M_\mu = -\frac{\partial E}{\partial B_\mu} \tag{3}$$

where $E = \Delta E_1 + \Delta E_2$.

Exercise 2.2*: Low-spin v.s. High-spin crossover

We have so far assumed that the crystal field splitting Δ, is weaker than the first (and second) Hund couplings so that a high-spin configuration is always stabilized within an ion as in Fig.1. However, the crystal field effect can be the strongest in some materials and we now have to treat Hund rules as a weaker perturbation. If the crystal field splitting is strong, all 6 electrons go into the lower-energy t_{2g} orbitals as in Fig.2.

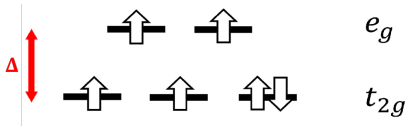

Figure 1: High-spin configuration

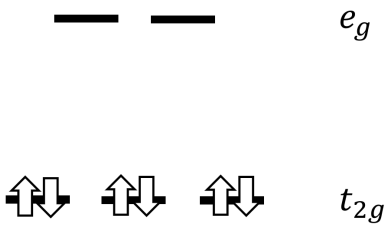

Figure 2: Low-spin configuration

The average spin of an ion should be

$$S = \frac{S_{LS}G_{LS} + S_{HS}G_{HS}e^{-\Delta/k_BT}}{G_{LS} + G_{HS}e^{-\Delta/k_BT}} \tag{4}$$

where G_{LS} and G_{HS} are the degeneracies of the low-spin and high-spin state, respectively.

(a) Assume that the degeneracies only depend on the spin and plot the average spin as a function of k_BT/Δ for the case of Fig.1 and Fig.2.

Exercise 2.3: Antiferromagnetic Exchange from Hubbard Model with 2nd quantization (Homework Question)

Here, we introduce how to derive antiferromagnetic exchange acting within a restricted Hilbert space when the Hubbard interaction U is much bigger than the hopping parameter t. Being able to systematically carry out the transformation to the restricted space is of practical value and the techniques are presented in this problem using the Hubbard model as the starting point.

$$H = -t\sum_{ij\sigma}\left(c^\dagger_{i\sigma}c_{j\sigma} + h.c\right) + U\sum_i n_{i\uparrow}n_{i\downarrow} \tag{5}$$

Here, $c^\dagger_{i\sigma}$ creates an electron at site i with spin σ. We have used t to describe the nearest-neighbor hopping only. The following derivation is largely taken from "Phys. Rev. B 37, 9753" or "Lecture Notes on Electron Correlation and Magnetism, Fazekas, section 5.1". Using the fact that the electron occupation plus the hole occupation at any site for spin σ adds to one, $n_{i\sigma} + h_{i\sigma} = 1$, one can split the kinetic term into three pieces, $T = T_0 + T_1 + T_{-1}$ where

$$\begin{aligned} T_1 &= -t\sum_{ij\sigma} n_{j\bar{\sigma}}c^\dagger_{j\sigma}c_{i\sigma}h_{i\bar{\sigma}} \\ T_{-1} &= -t\sum_{ij\sigma} h_{j\bar{\sigma}}c^\dagger_{j\sigma}c_{i\sigma}n_{i\bar{\sigma}} \\ T_0 &= -t\sum_{ij\sigma} h_{j\bar{\sigma}}c^\dagger_{j\sigma}c_{i\sigma}h_{i\bar{\sigma}} - t\sum_{ij\sigma} n_{j\bar{\sigma}}c^\dagger_{j\sigma}c_{i\sigma}n_{i\bar{\sigma}}. \end{aligned} \tag{6}$$

In Eq.6 $\bar{\sigma}$ is $|\uparrow\rangle$ for $\sigma\,|\downarrow\rangle$ and $|\downarrow\rangle$ for σ being $|\uparrow\rangle$. T_1 increases the number of doubly occupied site by one because it has a non-zero matrix element if and only if the initial state has the configuration $|\uparrow_i\downarrow_j\rangle$ or $|\downarrow_i\uparrow_j\rangle$. Then T_1 acting on the initial state produces $|0_i d_j\rangle$ or $|d_i 0_j\rangle$, d implying the doubly

occupied site. Similarly, T_{-1} decrease the number of doubly occupied sites by one by acting on the initial state $|0_i d_j\rangle$ or $|d_i 0_j\rangle$. T_0 does not change the number of double occupancy because it acts only on the initial state $|0_i \sigma_j\rangle$, $|\sigma_i 0_j\rangle$, or $|\sigma_i d_j\rangle$, $|d_i \sigma_j\rangle$.

Now consider a canonical transformation (same as in perturbation theory)

$$H_{eff} = e^{iS} H e^{-iS} \tag{7}$$

using $iS = (T_1 - T_{-1})/U$. $T_{\pm 1}$ satisfies the commutation $[V, T_{\pm 1}] = \pm U T_{\pm 1}$ where $V = U \sum_i n_{i\uparrow} n_{i\downarrow}$.

(a) Expand Eq.7 using the Baker-Hausdorff formula up to second order in S and keep the leading terms in $O(1/U)$ to obtain H_{eff}.

(b) Now, let H_{eff} only act between states with no doubly occupied sites. Simplify H_{eff} from (a) under this restriction and express the result in terms of T operators from Eq.6.

A short consideration gives that the operator $T_{-1}T_1$ have the following action on the possible spin states on sites i and j:

$$T_{-1}T_1 = \sum_{ij} t^2(|\uparrow_i\downarrow_j\rangle - |\downarrow_i\uparrow_j\rangle)(\langle\uparrow_i\downarrow_j| - \langle\downarrow_i\uparrow_j|) = 2t^2 |s_{ij}\rangle\langle s_{ij}| \tag{8}$$

where the symbol s_{ij} refers to the singlet state in the ij bond.

(c) Write $|s_{ij}\rangle\langle s_{ij}|$ in terms of operators n_i and $\boldsymbol{s}_i$, spin 1/2 operators. Using Eq.8 and H_{eff} from (b) obtain

$$H_{\text{eff}} = T_0 + \frac{4}{U} \sum_{ij} t^2 (\boldsymbol{s}_i \cdot \boldsymbol{s}_j - \frac{1}{4} n_i n_j). \tag{9}$$

Exercise 2.4: Spin-dependent hopping and anisotropic interactions (Homework Question)

In this problem, we introduce a way to derive spin-anisotropic interactions, DM interactions and Kitaev interactions. Under time reversal operation the spin-up and spin-down operators transform as $c_{i\uparrow} \to c_{i\downarrow}$ and $c_{i\downarrow} \to -c_{i\uparrow}$. The most general fermion hopping between a pair of sites ij obeying the time-reversal symmetry can be written down using the spinor notation $\psi = (c_{i\uparrow}, c_{i\downarrow})$.

$$H_{ij} = t\psi_i^\dagger (\sigma_0 \cos\theta_{ij} + i(\hat{d}_{ij} \cdot \boldsymbol{\sigma}) \sin\theta_{ij}) \psi_j = t\psi_i^\dagger (e^{i\theta_{ij}\hat{d}_{ij}\cdot\boldsymbol{\sigma}}) \psi_j \tag{10}$$

Here, $\hat{d}$ is an arbitrary 3-dimensional real unit vector and the second part of Eq.10 describes spin-dependent hopping which might originate from the presence of inter-atomic spin-orbit coupling. To ensure that the Hamiltonian remains Hermitian we will require $\hat{d}_{ji} = -\hat{d}_{ij}$. The superexchange calculation in the case of such a general hopping matrix can be worked out from the two-site model

$$t\psi_1^\dagger(e^{i\theta_{12}\hat{d}_{12}\cdot\boldsymbol{\sigma}})\psi_2 + Un_1(n_1-1) + Un_2(n_2-1)$$

Following "Phys. Rev. Lett. 69, 836" we can eliminate the 2 by 2 unitary matrix, $e^{i\theta_{12}\hat{d}_{12}\cdot\boldsymbol{\sigma}}$, by the transformation

$$\tilde{\psi}_1 \to e^{i(\theta_{12}/2)\hat{d}_{12}\cdot\boldsymbol{\sigma}}\psi_1,\ \ \tilde{\psi}_2 \to e^{i(-\theta_{12}/2)\hat{d}_{12}\cdot\boldsymbol{\sigma}}\psi_2. \tag{11}$$

(a) How does the Hubbard interaction $Un_1(n_1-1) + Un_2(n_2-1)$ change under the transformation Eq.11?

In the rotated basis we can use Exercise 2.3 and find the effective Hamiltonian

$$J_{12}\boldsymbol{s}_1\cdot\boldsymbol{s}_2 \text{ with } J_{12} \sim \frac{t^2}{U}$$

(b) Rotate the spins, $\boldsymbol{s}_1\cdot\boldsymbol{s}_2$, back to the original basis. You might need the relation $e^{i(-\theta/2)\hat{d}\cdot\boldsymbol{\sigma}}\boldsymbol{\sigma}e^{i(\theta/2)\hat{d}\cdot\boldsymbol{\sigma}} = \cos\theta\boldsymbol{\sigma} + (1-\cos\theta)\hat{d}(\hat{d}\cdot\boldsymbol{\sigma}) - \sin\theta(\hat{d}\times\boldsymbol{\sigma})$. Identify the three different terms you find with Superexchange, Kitaev and DM interaction.

Exercise 2.5: Spin wave dispersion via the classical Bloch spin wave (Homework Question)

Here, we introduce an alternative way to find low-energy excitations for the ferromagnetic spin-chain. First, we write down the equation of motion for the spin operator in the Heisenberg picture,

$$\frac{d\boldsymbol{s}_i}{dt} = i[H, \boldsymbol{s}_i] \tag{12}$$

where we take H to be the ferromagnetic spin-chain Hamiltonian

$$H = -\frac{J}{2}\sum_{ij}\boldsymbol{s}_i\cdot\boldsymbol{s}_j. \tag{13}$$

(a) Using Eq.12 and Eq.13, show that $\frac{ds_i^x}{dt} = -J\sum_j(s_j^y s_i^z - s_j^z s_i^y)$. Derive the similiar expression for $\frac{ds_i^y}{dt}$.

(b) Assuming the system to be close to the fully polarized ground state and the sites r_i to form Bravais lattice points of the spin chain, perform a Fourier transformation and derive $s_{\boldsymbol{k}}^-(t)$ where $s_{\boldsymbol{k}}^- = s_{\boldsymbol{k}}^x - is_{\boldsymbol{k}}^y$.

(c) Derive $s_i^x(t)$ and $s_i^y(t)$.

Exercise 2.6: Ferromagnetic magnon specific heat - Bloch law

The total energy of the thermally excited ferromagnetic magnons is $U(T) = \frac{1}{N}\sum_{\boldsymbol{k}} \epsilon_{\boldsymbol{k}} \langle b_{\boldsymbol{k}}^\dagger b_{\boldsymbol{k}} \rangle$ where $b_{\boldsymbol{k}}^\dagger$ creates a magnon with momentum $\boldsymbol{k}$ with the dispersion $\epsilon_{\boldsymbol{k}} = \Delta + \frac{k^2}{2m^*}$ as shown in Eq.(115) of the lecture notes.

(a) Show that the specific heat, $C = \frac{\partial U}{\partial T}$, of a ferromagnetism goes as $T^{3/2}$ for isotropic 3 dimensional systems at low T as shown in Eq.(122) of the lecture notes.