Temperature and magnetic-field dependencies of magnetic excitations of spin-pair system $KCuCl_3$

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Abstract

Temperature and magnetic-field dependencies of magnons in $KCuCl_3$ are investigated in detail on the basis of the pair mean-field approximation. From the condition that the gap energy vanishes a region of long range order in the plane of temperature *vs* field is determined.

Key words: spin pair system; magnetic excitation; field-induced long range order; $KCuCl_3$

1. Introduction

The quantum double spin chain systems of $KCuCl_3$-type structure such as $KCuCl_3$, $TlCuCl_3$ and NH_4CuCl_3 are typical examples of the so-called singlet-ground-state or spin-gap systems,[1,2] and much attention have been paid to their quite interesting magnetic properties in connection with other singlet-ground-state systems such as spin Peierls systems, Haldane systems and so on.

In this paper, first the temperature and magnetic-field dependencies of magnetic excitations (magnons) of $KCuCl_3$ are investigated on the basis of the pair mean-field approximation (Pair-MFA).[3] Secondly, the magnetic phase diagram of $KCuCl_3$ in the plane of temperature *vs* field is constructed.

2. Magnetic excitations

$KCuCl_3$ crystallizes in a monoclinic structure and its unit cell contains four Cu ions with spin S=1/2. We adopt the following Heisenberg Hamiltonian for $KCuCl_3$ under magnetic field along the z-axis:

$$\begin{aligned}\mathcal{H}_0 = &-J\sum_i (\boldsymbol{S}_{i1}\cdot\boldsymbol{S}_{i2} + \boldsymbol{S}_{i3}\cdot\boldsymbol{S}_{i4}) \\ &-\frac{1}{2}\sum_{i,j}\sum_{\mu\nu} J_{i\mu,j\nu}\boldsymbol{S}_{i\mu}\cdot\boldsymbol{S}_{j\nu} \\ &-g\mu_B H\sum_i (S^z_{i1}+S^z_{i2}+S^z_{i3}+S^z_{i4}),\end{aligned} \qquad (1)$$

where $\boldsymbol{S}_{i\mu}$ ($\boldsymbol{S}_{j\nu}$) denotes the spin μ (spin ν) in the unit cell i (unit cell j) with μ, $\nu = 1 \sim 4$.

The effective Hamiltonian for the spin pair $\boldsymbol{S}_{i1}$ and $\boldsymbol{S}_{i2}$ in the mean-field approximation is written as

$$\mathcal{H}^{\rm eff}_{12} = -J\boldsymbol{S}_{i1}\cdot\boldsymbol{S}_{i2} - H'(S^z_{i1}+S^z_{i2}) \qquad (2)$$

with $H' = g\mu_{\rm B}H + J_0\langle S^z\rangle$ and $J_0 = \sum_{j\nu} J_{i\mu,j\nu}$. Here $\langle S^z\rangle \equiv \langle S^z_{i1}\rangle = \langle S^z_{i2}\rangle = \langle S^z_{i3}\rangle = \langle S^z_{i4}\rangle$ denotes the spin moment induced by the external magnetic field. We have assumed no spontaneous long range order. The effective Hamiltonian for the spin pair $\boldsymbol{S}_{i3}$ and $\boldsymbol{S}_{i4}$ has the same form as that of $\mathcal{H}^{\rm eff}_{12}$. The value of $\langle S^z\rangle$ is determined from usual self-consistency condition.

Now the dynamical susceptibilities for the whole system can be calculated within the Pair-MFA[3], and the magnon energies are determined from the poles of the dynamical susceptibilities $\chi^{+-}_{\mu\nu}(\boldsymbol{q},\omega : T)$ where $\boldsymbol{q}$, ω and T denote wave vector, frequency and temperature, respectively. Under no magnetic field we obtain two

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kinds of triply-degenerate magnon branch whose dispersion are expressed as follows:

$$\omega^{\pm}(\boldsymbol{q}:T) = \sqrt{J^2 + J\rho(A(\boldsymbol{q}) \pm B(\boldsymbol{q}))} \qquad (3)$$

where $\rho = (1-e^{-\beta|J|})(1+3e^{-\beta|J|})$ with $\beta = 1/(k_B T)$, and $A(\boldsymbol{q})$ and $B(\boldsymbol{q})$ are expressed in terms of the Fourier transforms of exchange couplings $J_{\mu\nu}(\boldsymbol{q})$ (μ, $\nu = 1 \sim 4$). $\omega^-(\boldsymbol{q}:T)$ denotes the mode with lower energy. In Ref. 4 we have analyzed the magnetic excitations of $KCuCl_3$ observed at low temperatures under no magnetic field,[5,6] and determined the inter-dimer exchange couplings as well as the intra-dimer one. In $KCuCl_3$ the lowest magnon energy is realized at $\boldsymbol{q}$=0.

Under the external magnetic field the triply degenerate modes split, but it is impossible to obtain general analytical expressions of magnon energies under the external filed. Exceptions are the cases of $\boldsymbol{q}$=0 and $T = 0$ K. In the following section we confine ourselves to $\boldsymbol{q}$=0 and determine the magnetic phase diagram by investigate the field and temperature dependencies of the lowest magnon energy, *i.e.* gap energy.

3. Magnetic phase diagram

First we consider the case of T =0 K. For small magnetic fields we obtain $\langle S^z \rangle$=0 and the triply degenerate $E_G^0 \equiv \omega^-(\boldsymbol{q}=0:T=0)$ splits into $E_G^0 + g\mu_B H$, E_G^0 and $E_G^0 - g\mu_B H$. With increasing field the lowest magnon energy $E_G^0 - g\mu_B H$ becomes zero for $H = H_{c1} = E_G^0/(g\mu_B)$. At this critical field H_{c1} the static susceptibility χ^{+-} diverges and it indicates a occurence of a long range order in which a spontaneous spin moment $S^{\perp}$ perpendicular to the static field along the z-axis appears. For $KCuCl_3$ the value of H_{c1} is evaluated to be 20.5 T with use of g=2.91 and the spin moments $S^{\perp}$ of four sublattices satisfy the relation $S_1^{\perp} = -S_2^{\perp} = -S_3^{\perp} = S_4^{\perp}$.

For large magnetic field the induced spin moments are saturated, that is, $\langle S^z \rangle = \frac{1}{2}$. In this case the lowest magnetic excitation originates from the S^z=1 state of S=1 to the S^z=0 state of S=0 of the spin pair states, and its energy $E_G(H)$ is expressed as

$$E_G(H) = g\mu_B H - |J| + J_{12}(0) + J_{13}(0). \qquad (4)$$

With decreasing field the value of $E_G(H)$ vanishes for $H = H_{c2} = \frac{1}{g\mu_B}(|J| - J_{12}(0) - J_{13}(0))$. At this critical field H_{c2} the static susceptibility χ^{+-} diverges and again it indicates occurrence of long range magnetic order. The value of H_{c2} of $KCuCl_3$ is evaluated as $H_{c2} = |J| - J_1 - J_4 - J_5 - J_6 = 52$ T. For $H_{c1} < H < H_{c2}$ both the uniform moment $\langle S^z \rangle$ and the staggered moments $S_i^{\perp}$ (i=1 $\sim$ 4) exist, namely a spin canted long range ordered state is realized.

We can determine also for finite temperatures the phase boundary between the ordered and disordered states from the condition that the lowest magnon energy vanishes. By performing numerical calculations we have determined the region of the ordered state of $KCuCl_3$. The results are shown in Fig. 1. The lower and upper critical fields, H_{c1}=20.5 T and H_{c2}=52 T, determined theoretically show excellent agreements with the observed values, 20 T and 54 T.[2,7]

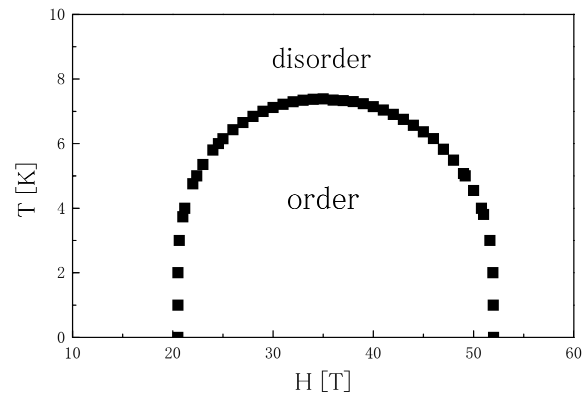

Fig. 1. Magnetic phase diagram of $KCuCl_3$. The ordered state is a spin-canted state.

For the fixed value of H the gap energy decreases with decreasing temperature. For $H < H_{c1}$ and $H > H_{c2}$ it remains finite down to 0 K, but for $H_{c1} < H < H_{c2}$ it vanishes at the boundary of the spin canted phase.

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