

Dispersion Curves of the Magnetic Excitation of KCuCl_3

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The magnetic excitation of the spin gap system KCuCl_3 was studied using the neutron inelastic scattering technique. The dispersion curves along the diagonal directions in the a^*-c^* plane such as $(h, 0, \pm 2h+1)$ were experimentally determined in detail. This enabled to choose the function for the best fitting uniquely and to obtain highly reliable fitting parameters, *i.e.*, the inter-dimer interactions in KCuCl_3 in the random phase approximation. The exchange network on the $(10\bar{2})$ plane was found to be more important than the coupling along the double-chain.

KEYWORDS: spin gap system, inter-dimer interactions, dispersion, neutron inelastic scattering

§1. Introduction

Quantum spin system on the low-dimensional lattice¹⁾ is one of the most interesting topics for researchers of magnetism. Recent technological progress of high-field experiments²⁾ and theoretical analysis³⁾ enables to make more precise evaluation of the exchange parameters at low temperatures for the singlet ground-state system. Even though an antiferromagnetic dimer is known as a classical spin gap system, the dimer materials are possible to behave as a quantum system if the inter-dimer interactions are comparable to the intra-dimer interactions and as a consequence a low-dimensional lattice is constructed.

About a quarter of a century ago, Willet and his co-workers⁴⁾ studied magneto-structural properties of KCuCl_3 and related compounds and concluded that the temperature dependence of the susceptibility of KCuCl_3 is attributed to the antiferromagnetic dimerization at low temperatures in the planar unit of Cu_2Cl_6 . The crystal structure of KCuCl_3 has monoclinic $P2_1/c$ symmetry with the lattice parameters $a = 4.029 \text{ \AA}$, $b = 13.785 \text{ \AA}$, $c = 8.735 \text{ \AA}$ and $\beta = 97^\circ 20'$.⁴⁾ An important character of this structure is the one-dimensional Cu^{2+} zigzag chains, running parallel to the a -axis, which consist of stacked Cu_2Cl_6 planes. The spin gap of KCuCl_3 can be of quantum origin if the intra-chain inter-dimer interaction is relevant.

Recent studies of magnetic susceptibility⁵⁾ and high-field magnetization process²⁾ of KCuCl_3 using single crystals indicate that this crystal has the singlet ground state with the spin gap of 2.74 meV. Previous inelastic neutron scattering studies^{6,7)} revealed that the dispersion along the b^* - and c^* -axes is comparable to that along the a^* -axis. Accordingly, the dimers are connected not only within the double-chains but also along the *inter*-double-chain directions. Moreover, the analysis of the dispersion data seemed to be impossible because there is an arbitrariness of choice of the fitting functions for the dispersion curves if the excitations were measured only along the a^* -, b^* - and c^* -axes. In the previous pub-

lication,⁸⁾ we proposed a quasi-two-dimensional dimer network along the $(10\bar{2})$ plane of KCuCl_3 with some preliminary data. More detail of the experimental data of the dispersion along the diagonal directions are needed to check the validity of this model. The present study was aimed to fix the picture of the inter-dimer interactions of KCuCl_3 through the dispersion determination by the measurements of the inelastic neutron scattering along the diagonal directions.

§2. Experimental Procedures and Results

The experimental details were the same as in the previous reports.^{6,8)} The spectrometer ISSP-PONTA installed at JRR-3M in JAERI was used with fixed incident neutron energy ($E_i = 14.7 \text{ meV}$) and the collimation condition $40'-40'-40'-80'$. The constant- Q energy scans were performed at 1.5 K ($=0.13 \text{ meV}$) along some reciprocal lines in the a^*-c^* plane, which are shown in Fig. 1(A) with obtained profiles in Fig. 1(a)-(e). The excitation energy at (001) which corresponds to the spin gap was obtained as 2.87 meV. In some profiles, an additional peak arises in the same energy range. The higher-energy peak in the double-peak profile of (101) in Fig. 1(b) is found as a phonon mode, because its intensity in the profile of the equivalent point (001) is not large and the existence of the phonon mode was confirmed by the measurements of some profiles at 60 K and 100 K. The results of the Gaussian fitting to evaluate the center of the magnetic peak are shown as curves in Fig. 1.

The dispersion data was analyzed as follows. The spin Hamiltonian of the inter-dimer interactions was rewritten with the base of dimer vectors $\mathbf{L}_i^\pm = \mathbf{S}_{i1} \pm \mathbf{S}_{i2}$:

$$\mathcal{H} = \sum_{\langle ij \rangle} \sum_{\mu, \nu}^{1,2} J_{ij\mu\nu} \mathbf{S}_{i\mu} \cdot \mathbf{S}_{j\nu} \sim \frac{1}{2} \sum_{\langle ij \rangle} \mathcal{J}_{ij} \mathbf{L}_i^- \cdot \mathbf{L}_j^-, \quad (2.1)$$

where $\langle ij \rangle$ indicates the dimer pairs,

$$\mathcal{J}_{ij} = (J_{ij11} + J_{ij22})/2 - (J_{ij12} + J_{ij21})/2, \quad (2.2)$$

is the interaction between the i -th and j -th dimers, and

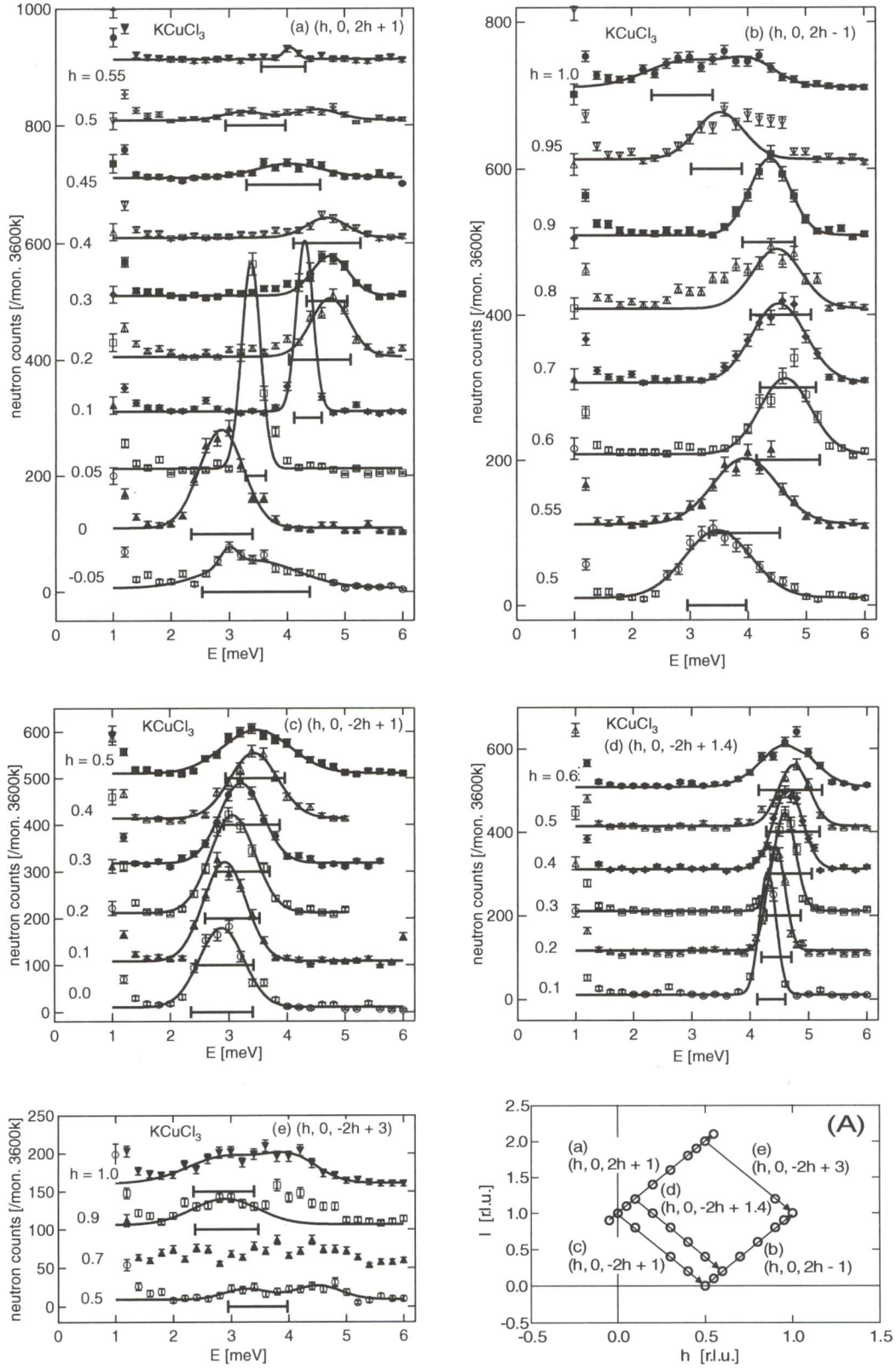


Fig. 1. Profiles of the constant- Q energy scans of KCuCl_3 for the reciprocal lines of the diagonal directions (a)~(e), which are lying on the a^*-c^* plane (A). In some profiles, *e.g.* at (101) in (b) or (e), a phonon excitation is also observed. The results of the Gaussian fitting are shown as curves. The horizontal error bars on the baselines indicate the resolution widths calculated using the Cooper-Nathans formalism⁹⁾ and the best-fit dispersion curves shown in Fig. 2. One should remark that the a^* - and c^* - axes are not orthogonal. An exact drawing for this reciprocal plane can be found in Fig.1(c) of ref. 8.

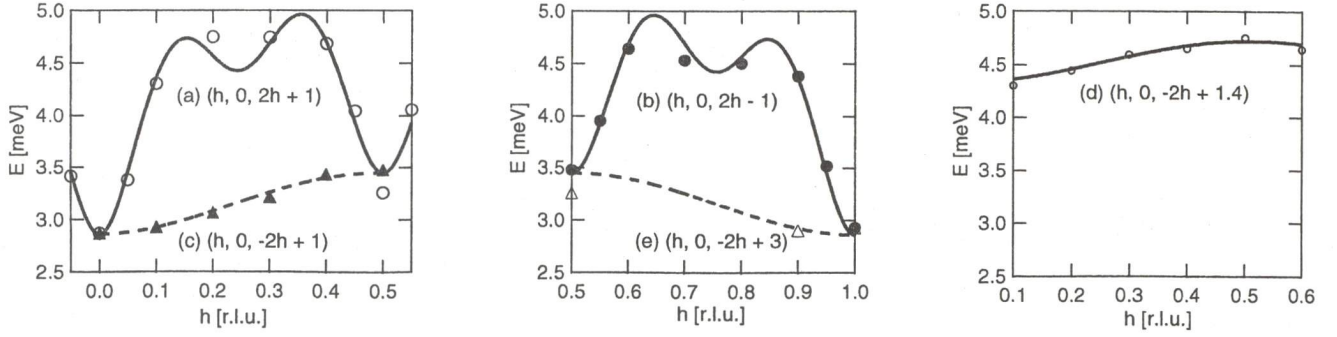


Fig. 2. Diagonal-direction dispersion curves of the magnetic excitation of KCuCl_3 . The excitation energy at (001) which corresponds to the spin gap was obtained as 2.87 meV.

the terms related with L_i^+ were omitted because only the lowest temperatures ($k_B T \ll J_{\text{dimer}}$) were concerned. Here we defined the intra-dimer part of the Hamiltonian as $\mathcal{H}_0 = J_{\text{dimer}} \sum_i \mathbf{S}_{i1} \cdot \mathbf{S}_{i2}$. Applying the random phase approximation⁸⁾ to our idea of the quasi-two-dimensional dimer network,⁶⁾ the fitting function for the dispersion curves becomes

$$E = \sqrt{J_{\text{dimer}}^2 - J_{\text{dimer}} \mathcal{J}(\mathbf{Q}_{hkl})}, \quad (2.3)$$

$$\begin{aligned} \mathcal{J}(\mathbf{Q}_{hkl}) = & 2\mathcal{J}_a \cos(2\pi h) + 2\mathcal{J}_{2a} \cos(4\pi h) \\ & + 2\mathcal{J}_{2ac} \cos(4\pi[h + l/2]) + 4\mathcal{J}_{bc} \cos(\pi l) \cos(\pi k) \\ & + 4\mathcal{J}_{abc} \cos(2\pi[h + l/2]) \cos(\pi k) + 2\mathcal{J}_c \cos(2\pi l). \end{aligned}$$

Here \mathcal{J}_a and \mathcal{J}_{2a} are the intra-double-chain interactions among the nearest and next nearest dimers, respectively, and \mathcal{J}_{2ac} and \mathcal{J}_{abc} are the couplings on the $(10\bar{2})$ plane. The relevant paths of the spin-spin interaction corresponding to \mathcal{J}_{2ac} and \mathcal{J}_{abc} had been shown in Fig. 1(a) of ref. 8. Additional two terms with the parameters \mathcal{J}_c and \mathcal{J}_{bc} are included in order to confirm the uniqueness of the choice of functions: Three terms with the coefficients \mathcal{J}_{2ac} , \mathcal{J}_{2a} and \mathcal{J}_c are independent functions for the present dispersion data along the diagonal directions, though only the two coefficients ($\mathcal{J}_{2ac} + \mathcal{J}_{2a}$ and $\mathcal{J}_c + \mathcal{J}_{2a}$) are possible to be determined by fitting to the dispersion along the principle axes.⁶⁾⁷⁾ Similar merit of the diagonal-direction dispersion is expected for another set of parameters \mathcal{J}_{abc} , \mathcal{J}_a and \mathcal{J}_{bc} .

The best fit result is easily obtained as

$$\begin{aligned} J_{\text{dimer}} &= 4.34 \pm 0.02 \text{ meV}, \\ \mathcal{J}_{2ac} &= 0.45 \pm 0.03 \text{ meV } (\mathbf{r} = \pm 2[\mathbf{a} + \mathbf{c}/2]), \\ \mathcal{J}_{abc} &= -0.28 \pm 0.02 \text{ meV } (\mathbf{r} = \pm[\mathbf{a} + \mathbf{c}/2] \pm \mathbf{b}/2), \\ \mathcal{J}_a &= 0.21 \pm 0.07 \text{ meV } (\mathbf{r} = \pm \mathbf{a}), \end{aligned} \quad (2.4)$$

and $\mathcal{J}_{2a}(\mathbf{r} = \pm 2\mathbf{a})$, $\mathcal{J}_{bc}(\mathbf{r} = \pm \mathbf{c}/2 \pm \mathbf{b}/2)$ and $\mathcal{J}_c(\mathbf{r} = \pm \mathbf{c})$ were smaller than the errors of ± 0.05 , ± 0.03 and ± 0.03 meV, respectively. Here the displacement vector \mathbf{r} indicates the positional difference between the interacting dimers, and \mathbf{a} , \mathbf{b} and \mathbf{c} are the primitive lattice vectors. Using the dispersion formula (2.3) with these best-fit parameters (2.4), the instrumental resolution widths

were calculated by the Cooper-Nathans formalism⁹⁾ and exhibited in Fig. 1 as the horizontal bars on the base-lines. Almost all the magnetic peaks were found to be resolution limited.

§3. Discussion

The present result that the interactions \mathcal{J}_{bc} and \mathcal{J}_c are negligible indicates that the dispersion along the c^* -axis originates from the \mathcal{J}_{2ac} and \mathcal{J}_{abc} on the $(10\bar{2})$ plane. The spin system of KCuCl_3 is well described as the model of the three-dimensionally coupled dimers, and the interactions in the $(10\bar{2})$ plane were found to be more important than the interaction along the double-chain (\mathcal{J}_a).

It must be remarked that the sign of the dimer-dimer interaction depends on the definition of the sign of each dimer vector $\pm \mathbf{L}_i^-$, even if one of four corresponding spin-spin interactions in Eq. (2.2) is relevant and it is antiferromagnetic. From the present results corresponding to \mathcal{J}_{2ac} and \mathcal{J}_{abc} , one can easily deduce that the spin-spin interactions on the $(10\bar{2})$ plane shown in Fig. 1(a) of Ref. 8 are antiferromagnetic. For the case of \mathcal{J}_a , the situation is not clear because two kinds of the spin-spin interactions contribute to \mathcal{J}_a with opposite signs.

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