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Spin flop in one-dimensional Heisenberg antiferromagnet $KCuF_3$*

Shi Qing-Fan(史庆藩)[a)†], Li Liang-Sheng(李良生)[a)], and Wang Qi(王 琪)[b)]

[a)] *Department of Physics, School of Science, Beijing Institute of Technology, Beijing* 100081, *China*

[b)] *Department of Physics, Yancheng Teacher's Collegy, Yancheng* 224002, *China*

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Following Yamada and Kato [J. Phys. Soc. Japan 63 (1994) 289], we have calculated the frequency–field diagram of antiferromagnetic resonance of $KCuF_3$ using an eight-sublattice model and mean field approximation. A spin flop is found to take place from the $[110]_p$ to a direction perpendicular to $[110]_p$ in the c-plane at $H \simeq 42172$A/m, which is in good agreement with the data obtained by measuring magnetic susceptibility.

Keywords: $KCuF_3$, antiferromagnetic resonance, sublattice
PACC: 7650, 7550E

1. Introduction

The research for the ferro- and antiferromagnetic materials has attracted much attention due to its industrial and fundamental importance.[1−6] Generally, in order to clarify the magnetic structure of crystals, it is important to analyse the frequency–field diagram by solving the equations of motion, especially when the crystal has a multi-sublattice magnetic structure: e.g., non-collinear spin ordering which was discovered in $KMnF_3$[7] and $CuCl_2{\cdot}2D_2O$,[8] etc. Moreover, two or four or eight sublattices were found based on the presence of Dzyaloshinsky–Moriya (DM) antisymmetric exchange interaction, $\sum_{i>j} d_{ij} \cdot (S_i \times S_j)$ by comparing experimental results of antiferromagnetic resonance (AFMR) with the calculated frequency–field diagram.[9−11] AFMR measurement performed on the high-quality single crystal $KCuF_3$ and analyses of the AFMR field $H_{\rm res}$ have revealed that the compound $KCuF_3$ is a representative one-dimensional Heisenberg antiferromagnet at temperatures below T_N=39K; it has an eight-sublattice magnetic structure.[9,12−15] Linear antiferromagnetic chains are known to be along the c-axis with the intrachain exchange interaction J_c=−190K, and the very weak interchain exchange interaction in the c-plane estimated to be $J_a \simeq 0.01|J_c|$. The substantial easy direction is along $[110]_p$ and its equivalent axes, where $[\ ]_p$ represents an axis in a unit cell of a perovskite structure. However, an important phenomenon called spin flop observed in the experiments of neutron diffraction [16] and measurement of magnetic susceptibility [13] has never been reported by the experiment of AFMR. Furthermore, the external field $g_aH/2 \simeq 62860$ A/m (where $g_a = 2.27$), where the spin flop occurs from the $[110]_p$ axis to a direction perpendicular to $[110]_p$ in the c-plane predicted by numerical calculation for the frequency–field diagram only taken into account the simplified models of two- or four-sublattice systems,[9] is not in accordance with the value measured by magnetic susceptibility.[14] The purpose of the present study is to calculate the frequency–field diagram of AFMR of $KCuF_3$ using the eight-sublattices model based on the results of Yamada and Kato [13] with the classical method,[17,18] and determine the condition for spin flop occurrence.

2. Frequency–field relationship of AFMR for eight-sublattices

The Hamiltonian relevant to AFMR of $KCuF_3$ is

$$\begin{aligned}\hat{H} =& -2\sum\nolimits_{i>j}(J_c S_i \cdot S_j + D_c S_i^z \cdot S_j^z) \\ & -2\sum\nolimits_{i>m} J_a S_l \cdot S_m + \sum\nolimits_{i>j} d_{ij} \cdot (S_i \times S_j) \\ & + \mu_B \sum\nolimits_k S_k \cdot g_A \cdot H + \mu_B \sum\nolimits_{k'} S_{k'} \cdot g_B \cdot H. \quad (1)\end{aligned}$$

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†E-mail: qfshi@163.com

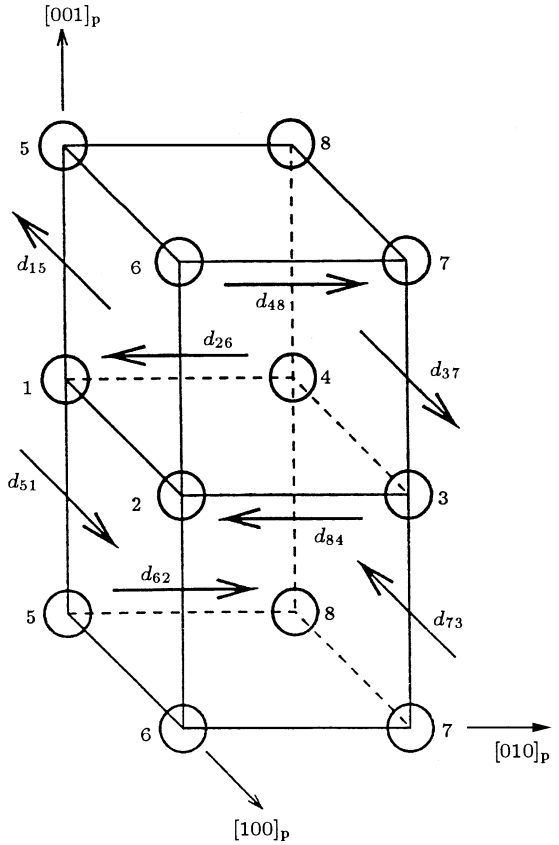


Fig.1. The directions of the DM vectors and the positions of eight sublattices 1–8.[11]

In Eq.(1), J_c represents the intrachain exchange interaction among nearest-neighbour (nn) ions in the c-axis, J_a means the ferromagnetic interchain exchange interaction among nn ions in the c-plane, and D_c=0.04K produces an XY-like anisotropy field which forces the spins to lie in the c-plane.[19] The third term represents DM interaction between nn spins in the c-axis. The last two terms are Zeeman interactions for the spins having inequivalent g-tensors, g_A and g_B, where the two summations $\sum_k$ and $\sum_{k'}$ represent spins having g_A and g_B, respectively. Because of these g factors, the spins are canted and divided into eight sublattices as shown in Fig.1.

Applying the mean field approximation to the Hamiltonian given by Eq.(1), the free energy F is formulated as follows:

$$\begin{aligned} F =& A(M_1 \cdot M_5 + M_2 \cdot M_6 + M_3 \cdot M_7 + M_4 \cdot M_8) \\ & - A'(M_1 \cdot M_2 + M_1 \cdot M_4 + M_2 \cdot M_3 \\ & + M_3 \cdot M_4 + M_5 \cdot M_6 + M_6 \cdot M_7 \\ & + M_7 \cdot M_8 + M_5 \cdot M_8) \\ & + B[d_{15} \cdot (M_1 \times M_5) + d_{26} \cdot (M_2 \times M_6) \\ & + d_{37} \cdot (M_3 \times M_7) + d_{48} \cdot (M_4 \times M_8)] \\ & + \frac{K}{2M^2} \sum_{i=1}^{8} M_{iz^2} - H \cdot \frac{g_A}{g} \cdot (M_1 + M_3 + M_6 + M_8) \\ & - H \cdot \frac{g_B}{g} \cdot (M_2 + M_4 + M_5 + M_7), \end{aligned} \tag{2}$$

where each $M_i = -Ng\mu_B\langle S_i\rangle$ (i=1,2,..., 8) is the magnetization of the ith sublattice, in which N is the number of magnetic ions in the ith sublattice, g the g-factor of a free electron, and $\langle\ \ \rangle$ a thermal average. The coordinate axes (x, y and z) are taken parallel to the $[100]_p$, $[010]_p$ and $[001]_p$ axes, respectively. The parameters in Eq.(2) are determined by

$$\begin{aligned} A &= \frac{4|J_c|}{N\gamma^2\hbar^2}, \quad A' = \frac{4J_a}{N\gamma^2\hbar^2}, \\ B &= \frac{4}{N\gamma^2\hbar^2}, \quad K = \frac{4D_c}{N\gamma^2\hbar^2}, \end{aligned} \tag{3}$$

where $\gamma = -g\mu_B/\hbar$. Using the mean field theory, H_i that acts on the sublattice magnetization M_i is given by

$$H_i = -\frac{\partial F}{\partial M_i} \quad (i = 1, 2, \ldots, 8), \tag{4}$$

and every H_i has a similar structure as the following:

$$\begin{aligned} H_1 =& \frac{g_A}{g} \cdot H - AM_5 + A'(M_2 + M_4) - B(M_5 \times d_{15}) + H_{A1}, \\ H_2 =& \frac{g_B}{g} \cdot H - AM_6 + A'(M_1 + M_3) - B(M_6 \times d_{26}) + H_{A2}, \\ H_3 =& \frac{g_A}{g} \cdot H - AM_7 + A'(M_2 + M_4) - B(M_7 \times d_{37}) + H_{A3}, \\ H_4 =& \frac{g_B}{g} \cdot H - AM_8 + A'(M_1 + M_3) - B(M_8 \times d_{48}) + H_{A4}, \\ H_5 =& \frac{g_A}{g} \cdot H - AM_1 + A'(M_6 + M_8) - B(M_1 \times d_{15}) + H_{A5}, \\ H_6 =& \frac{g_B}{g} \cdot H - AM_2 + A'(M_5 + M_7) - B(M_2 \times d_{26}) + H_{A6}, \\ H_7 =& \frac{g_A}{g} \cdot H - AM_3 + A'(M_6 + M_8) - B(M_3 \times d_{37}) + H_{A7}, \\ H_8 =& \frac{g_B}{g} \cdot H - AM_4 + A'(M_5 + M_7) - B(M_4 \times d_{48}) + H_{A8}, \end{aligned} \tag{5}$$

in which $H_{Ai}=\left(0,0,-\frac{K}{M^2}M_{iz}\right)$ $(i=1,2,\ldots,8)$ are the XY-like anisotropy fields. The equations of motion for the eight-sublattices can be written by

$$\frac{\mathrm{d}M_i}{\mathrm{d}t}=-\gamma[M_i\times H_i],\qquad (i=1,2,\ldots,8). \tag{6}$$

In order to obtain the resonance frequency ω we change Eq.(6) into the following expressions:

$$\begin{aligned}
\frac{\mathrm{d}^2M_i}{\mathrm{d}t^2}=&-\gamma\frac{\mathrm{d}M_i}{\mathrm{d}t}\times\frac{\mathrm{d}H_i}{\mathrm{d}t}=\gamma^2\left(M_i\times\frac{\partial F}{\partial M_i}\right)\times\frac{\mathrm{d}\dfrac{\partial F}{\partial M_i}}{\mathrm{d}t},\\
\frac{\mathrm{d}\dfrac{\partial F}{\partial M_i}}{\mathrm{d}t}=&F\left(M_1,\ldots,M_8,\frac{\mathrm{d}M_1}{\mathrm{d}t},\ldots,\frac{\mathrm{d}M_8}{\mathrm{d}t}\right)\\
=&F(M_1,\ldots,M_8,-\gamma[M_1\times H_1],\ldots,\\
&-\gamma[M_8\times H_8])\\
=&F\left(M_1,\ldots,M_8,-\gamma\left(M_1\times\frac{\partial F}{\partial M_{1i}}\right),\ldots,\right.\\
&\left.-\gamma\left(M_8\times\frac{\partial F}{\partial M_8}\right)\right)\\
=&F(M_1,\ldots,M_8),\quad (i=1,2,\ldots,8),
\end{aligned}\tag{7}$$

then, we have the nonlinear autonomous vibration equations

$$\frac{\mathrm{d}^2M_i}{\mathrm{d}t^2}+g_i(M_i)=f(M_1,\ldots,M_{i-1},M_{i+1},\ldots,M_8),$$

$$(i=1,2,\ldots,8). \tag{8}$$

In order to solve Eq.(8), we first consider the homogeneous equations, and let $f(M_1,\ldots,M_{i-1},M_{i+1},\ldots,M_8)=0$, then, we have

$$\frac{\mathrm{d}^2M_i}{\mathrm{d}t^2}+g_i(M_i)=0,(i=1,2,\ldots,8). \tag{9}$$

Supposing the results being in the following form:

$$M_i=a_i\cos(\varphi_i)+b_i,\quad (i=1,2,\ldots,8), \tag{10}$$

$$\frac{\mathrm{d}\varphi_i}{\mathrm{d}t}=\phi_{i0}(\varphi_i),\quad (i=1,2,\ldots,8), \tag{11}$$

and putting them in Eq.(9), then we integrate the equations with respect to φ_i, and we obtain

$$\frac{1}{2}[a_i\phi_{i0}(\varphi_i)\sin(\varphi_i)]^2=v_i(a_i+b_i)-v_i(a_i\cos(\varphi_i)+b_i),$$

$$(i=1,2,\ldots,8). \tag{12}$$

$$v_i(M_i)=\int_0^{M_i}g_i(x)\mathrm{d}x,\quad (i=1,2,\ldots,8). \tag{13}$$

In addition, we assume the solutions of Eq.(8) are

$$\begin{aligned}
M_i=&a_i\cos(\varphi_i)+b_i+x_{i1}(a_1\cos(\varphi_1)\\
&+b_1,\ldots,a_8\cos(\varphi_8)+b_8),\\
&(i=1,2,\ldots,8).
\end{aligned}\tag{14}$$

In fact, we can use a complementary function to describe the general solution. Using the asymptotic method, we finally have

$$\begin{aligned}
\varphi_i=&\varphi_j+\sum_{n=1}^{N}\frac{1}{nc_{i0}}(c_{jn}\sin n\varphi_j-c_{in}\sin n\varphi_i)\\
&+c_{i0}^{-1}(r_i-r_j),
\end{aligned}\tag{15}$$

where the Fourier coefficient is

$$c_{in}=\frac{1}{2\pi}\int_0^{2\pi}\frac{1}{\phi_{i0}}\cos n\varphi_i\mathrm{d}\varphi_i,\quad (n=0,1,2,\ldots).$$

The nonhomogeneous term f in Eq.(8) is described by a periodic solution

$$\begin{aligned}
&f_{i0}(a_1,\ldots,a_8,\varphi_i)\\
=&f_i(a_1\cos(\varphi_1)+b_1,\ldots,a_8\cos(\varphi_8)+b_8);
\end{aligned}\tag{16}$$

then we have the function of first-order approximation

$$\phi_{i1}=\frac{-\displaystyle\int_0^{\varphi_i}f_{i0}(a_1,\ldots,a_8,\varphi_i)\sin\varphi_i\mathrm{d}\varphi_i-A_{i1}\int_0^{\varphi_i}\left(2\phi_{i0}+a_i\frac{\partial\phi_{i0}}{\partial a_i}\right)\sin^2\varphi_i\mathrm{d}\varphi_i}{a_i\phi_{i0}\sin^2\varphi_i}, \tag{17}$$

where

$$A_{i1}=-\int_0^{2\pi}f_{i0}(a_1,\ldots,a_8,\varphi_i)\sin\varphi_i\mathrm{d}\varphi_i\Big/\int_0^{2\pi}\left(2\phi_{i0}+a_i\frac{\partial\phi_{i0}}{\partial a_i}\right)\sin^2\varphi_i\mathrm{d}\varphi_i,$$

so the period of the system is given by

$$T_i=\int_0^{2\pi}\frac{\mathrm{d}\varphi_i}{\phi_{i0}+\phi_{i1}},\qquad \omega_i=2\pi/T_i. \tag{18}$$

3. Calculated results and discussion

Using the experimental data $J_c = -190$K, J_a=0.01$|J_c|$ and D_c=0.04K,[19,20] and adjusting the value of d, the angular dependence of resonance field $H_{\rm res}$ is firstly calculated using Eq.(18). It can be seen from Fig.2 that $H_{\rm res}$ has a fourfold symmetry and reaches its higher peak for $H//[100]_{\rm p}$, and its lower peak for $H//[110]_{\rm p}$ directions. The result is in good agreement with the experimental data [13] if d=0.027$|J_c|$ is used.

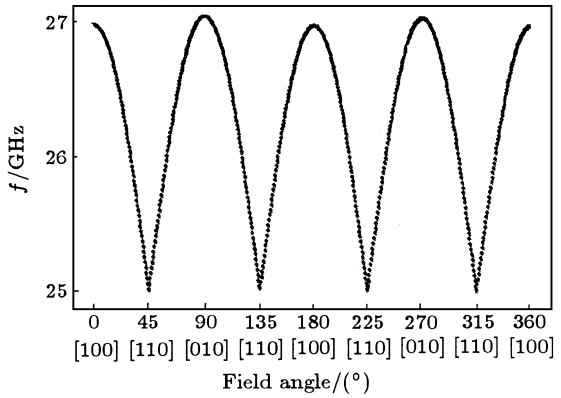


Fig.2. The calculated angular dependence of resonance field $H_{\rm res}$ at the lowest-frequency mode, in which the external field H is fixed at 795700A/m and rotated in the c-plane.

The frequency–field diagram (ω–H) of AFMR calculated for the lowest-frequency mode and $H//[110]_{\rm p}$ is shown in Fig.3. When the ω–H diagram is scaled up for the lower $H_{\rm res}$ as shown in Fig.4, a change of $H_{\rm res}$ ends at $H_{\rm sf}$=$|g_m \cdot H/g|$=42172A/m for $H//[110]_{\rm p}$. According to the conventional AFMR theory, such a change of $H_{\rm res}$ indicates the spin flop taking place from $[110]_{\rm p}$ to the direction perpendicular to $[110]_{\rm p}$ in the c-plane. Our calculated results about $H_{\rm res}$ is in good agreement with $H_{\rm res}$=43764A/m, which is measured for magnetic susceptibility.[14] We also propose to perform the AFMR experiment to parallel and validate our calculated result about $H_{\rm sf}$.

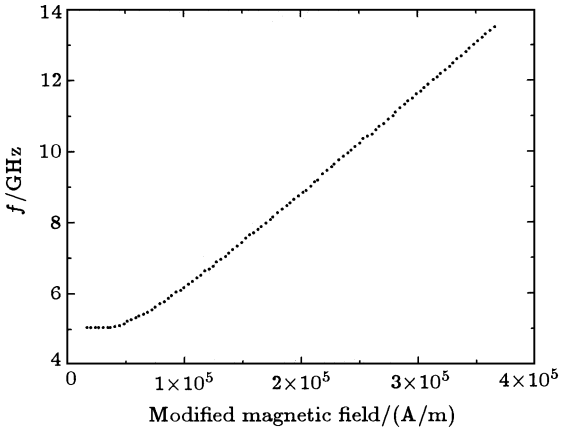


Fig.3. Frequency–field diagram of the lowest-frequency mode.

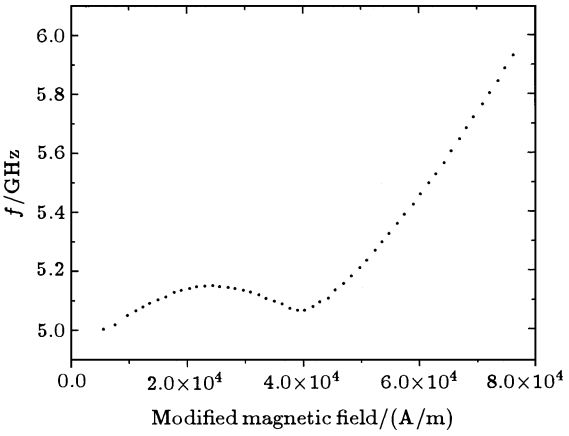


Fig.4. Scaled-up frequency–field diagram of the lowest-frequency mode. Spin flop is predicted to occur at $H_{\rm sf}$=$|g_m \cdot H/g|$=42172A/m.

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