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Experimental observation of the antiferromagnetic resonance linewidth in $KCuF_3$

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Abstract
We report the results of antiferromagnetic resonance (AFMR) measurements conducted on $KCuF_3$ at various frequencies from 3.8 to 10.6 GHz at 4.2 K. The resonance linewidth is first found to depend on the frequency, i.e., the lower the frequency the greater the resonance linewidth, no matter whether the AFMR field is forced on the easy axis or uneasy axis. Moreover, a linewidth peak seems to exist for $H \parallel [100]_p$ at about 4 GHz. Based on the model of Yamada and Kato (1994 *J. Phys. Soc. Japan* **63** 289) and considering the Laudau–Lifshitz damping term, the result of numerical calculation for the resonance linewidth is in good agreement with the data of AFMR experiments.

The linewidth of magnetic resonance is characterized by various relaxation mechanisms: for example, the specimen shape, the anisotropy, etc [1–6]. Because the anisotropy is related to the temperature, the relaxation of ferro- and antiferro-magnetic systems, especially for the antiferromagnetic material, is often investigated by a temperature-dependent effect [1, 7–9]. However, measurements of the antiferromagnet linewidths in the past were carried out only at a few frequencies, and the values of the linewidths are not the same [8, 10]. For example, the linewidths of MnF_2, which is a two-sublattice antiferromagnet without Dzyaloshinsky–Moriya interaction, are 39 Oe at 70 GHz, 5 Oe at 23 GHz and 4–9 Oe at low frequencies, respectively [8]. Therefore, the linewidth seems to reduce with decreasing frequency.

In this paper, we report the results of AFMR of $KCuF_3$ at frequencies varying from 3.8 to 10.6 GHz. The resonance linewidth is found to depend on the frequency, i.e., the lower the frequency, the greater the resonance linewidth, no matter whether the AFMR is conducted on the easy axis $[110]_p$ and its equivalent axes, or the uneasy axis $[100]_p$, where $[\,]_p$ represents an axis in a unit cell of a perovskite structure. Moreover, it seems that a linewidth peak exists for $H \parallel [100]_p$ at about 4 GHz, In addition, two kinds of linewidth are first observed to appear in the substantial uneasy direction of the spin $[100]_p$ axis at the lower-frequency branch of the C-band. Based on the model of Yamada and Kato [11] and considering the Laudau–Lifshitz damping term, the results of numerical calculation for the frequency–field

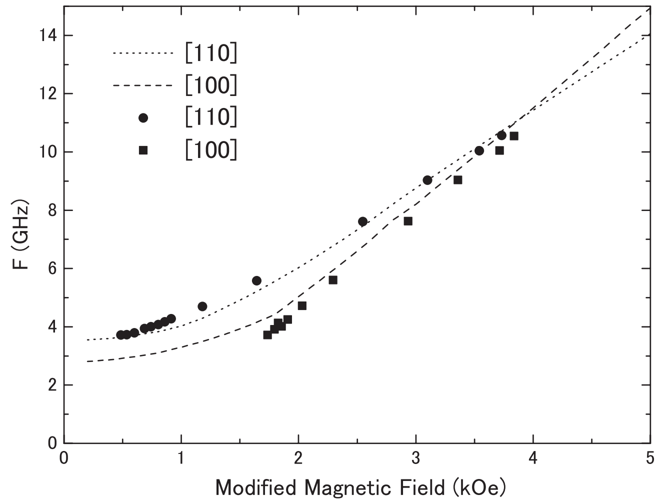

Figure 1. Experimental data of the frequency–field (ω–**H**) relation. Calculated results are shown by a dotted line for $\mathbf{H} \parallel [110]_p$ and a dashed line for $\mathbf{H} \parallel [100]_p$.

diagram and resonance linewidth are in good agreement with the AFMR measurements, which indicates that the nature of the unusual frequency dependence of the linewidth might come from the DM interaction.

A $KCuF_3$ single crystal was grown by the Bridgman method; it had high quality and had been used in previous studies [10–12]. AFMR measurements were performed at 4.2 K, far below $T_N = 39$ K. A wide-band spectrometer operated from 2 to 20 GHz was employed to investigate the existence of the eight-sublattice. The sample was placed at the bottom centre of a rectangular cavity and a water-cooled magnet was rotated around a vertical axis of the c-plan by a rotation mechanism. In order to lower the resonance frequency, Teflon is used to fill the cavity.

Figure 1 shows the experimental data of the resonance frequency–field (ω–**H**) relation. The circles are for $\mathbf{H} \parallel [110]_p$, the squares are for $\mathbf{H} \parallel [100]_p$. Figures 2(a) and (b) indicate changes in the derivative peak-to-peak linewidth ΔH_{pp} as a function of frequency at 4.2 K for $\mathbf{H} \parallel [100]_p$ and $\mathbf{H} \parallel [110]_p$, respectively. The stars in figure 2 represent the second observed absorption mode. The linewidth ΔH_{pp} can be found to depend apparently upon the frequency and increases with decreasing frequency in the low-frequency branch for both directions. Moreover, the distinct change of linewidth with frequency at $\mathbf{H} \parallel [110]_p$ is significantly larger than that at $\mathbf{H} \parallel [100]_p$, and the linewidth of the second mode occurring at $\mathbf{H} \parallel [100]_p$ is wider than that of the first mode. Obviously, the trend of the linewidth of $KCuF_3$ is different from the linewidth of MnF_2 in which the magnetic structure is two-sublattice and DM interaction is not exist.

Let us now calculate the frequency–field (ω–**H**) relation and the frequency dependence of the linewidth in light of the eight-sublattice model developed by Yamada and Kato [11] but taking into account the Laudau–Lifshitz damping term. According to the conventional AFMR theory, the Hamiltonian can be written as

$$\hat{H} = -2\sum_{i>j}(J_c\mathbf{S}_i \cdot \mathbf{S}_j + D_c S_i^z \cdot S_j^z) - 2\sum_{l>m} J_a \mathbf{S}_l \cdot \mathbf{S}_m + \sum_{i>j} d_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j) + \mu_{\mathrm{B}} \sum_k \mathbf{S}_k \cdot \mathbf{g}_A \cdot \mathbf{H} + \mu_{\mathrm{B}} \sum_{k'} \mathbf{S}_{k'} \cdot \mathbf{g}_B \cdot \mathbf{H}, \quad (1)$$

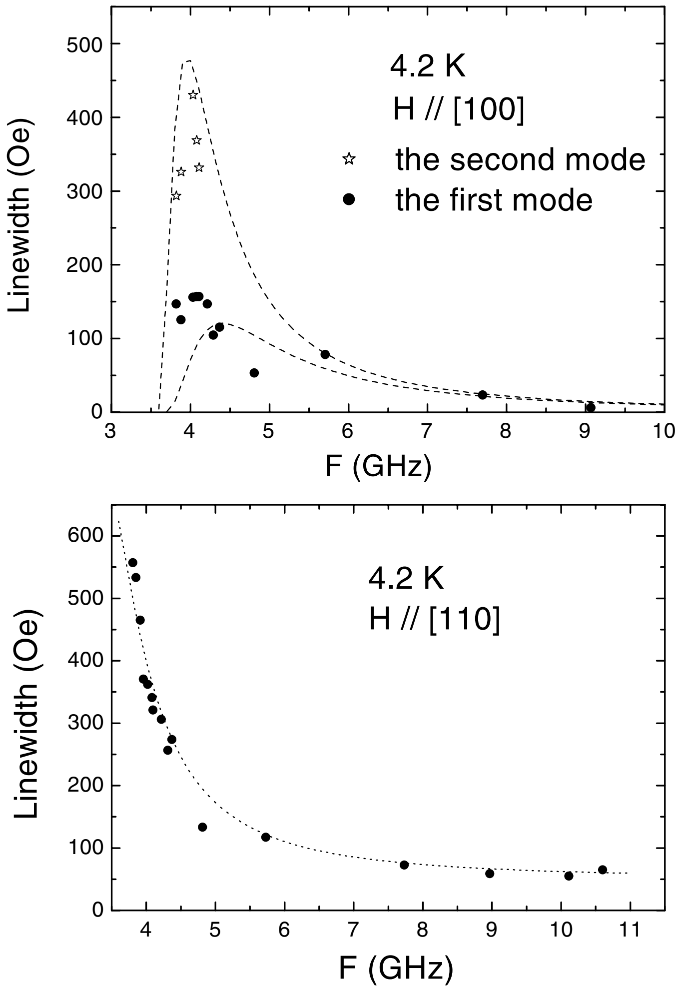

Figure 2. Linewidth ΔH_{pp} as a function of frequency at 4.2 K. (a) $\mathbf{H} \parallel [100]_p$, (b) $\mathbf{H} \parallel [110]_p$. The dashed line and the dotted line represent the calculated results for $\mathbf{H} \parallel [100]_p$ and $\mathbf{H} \parallel [110]_p$, respectively.

where $J_c = -190$ K represents the intrachain exchange interactions between nearest-neighbour (nn) ions in the c-axis [13], $J_a \simeq 0.01|J_c|$ means the ferromagnetic interchain exchange interaction among nn ions in the c-plane [14], and $D_c = 0.04$ K produces an XY-like anisotropy field which forces the spins to lie in the c-plane [14]. The third term represents the Dzyaloshinsky–Moriya (DM) antisymmetric exchange interaction $\sum_{i>j} \mathbf{d}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j)$ occurring between the nearest-neighbour (nn) spins along the c-axis [13, 15]. The last two terms are Zeeman interactions for the spins having inequivalent g-tensors, $\mathbf{g}_A$ and $\mathbf{g}_B$, respectively. $\mathbf{g}_A$ and $\mathbf{g}_B$ defined along the three principal axes of the F^- octahedron have been theoretically and experimentally confirmed as [2, 3, 6]

$$\mathbf{g}_A = \begin{pmatrix} 2.49 & & \\ & 2.05 & \\ & & 2.14 \end{pmatrix}, \qquad \mathbf{g}_B = \begin{pmatrix} 2.05 & & \\ & 2.49 & \\ & & 2.14 \end{pmatrix}.$$

Because of these g factors, the spins are canted and divided into eight sublattices.

In the next step, for simplicity of calculating the 24 secular equations, we rewrite the equations of motion for the eight-sublattice as follows:

$$\frac{\mathrm{d}\mathbf{M}_{i\alpha}}{\mathrm{d}t} = [-\gamma[\mathbf{M}_i \times \mathbf{H}_i] - \mathbf{T}_i]_\alpha, \qquad (i = 1\text{–}8,\ \text{and}\ \alpha = x, y, z), \tag{2}$$

with

$$\begin{aligned} \mathbf{M}_i &= \mathbf{M}_{i0} + \mathbf{m}_i, \\ \mathbf{H}_i &= \mathbf{H}_{i0} + \mathbf{h}_i, \\ \mathbf{T}_i &= -\frac{\alpha\gamma}{|\mathbf{M}_i|}\mathbf{M}_i \times (\mathbf{M}_i \times \mathbf{H}_i), \end{aligned}$$

where $\gamma = -g\mu_\mathrm{B}/\hbar$; $\mathbf{M}_{i0}$ and $\mathbf{H}_{i0}$ are the steady parts of the sublattice magnetization and mean field, respectively; $\mathbf{m}_i$ and $\mathbf{h}_i$ represent the parts containing $\exp(\mathrm{i}\omega t)$ in $\mathbf{M}_i$ and $\mathbf{H}_i$, and $\mathbf{T}_i$ is the Laudau–Lifshitz damping term, respectively. Thus, the resonance frequencies should be the eigenvalues of the matrix with 24×24 elements. From the expressions of the Hamiltonian and the mean field $\mathbf{H}_i$ acting on the sublattice magnetization $\mathbf{M}_i$, we have

$$\begin{aligned} \mathbf{H}_1 &= \frac{\mathbf{g}_A}{g} \cdot \mathbf{H} - A\mathbf{M}_5 + A'(\mathbf{M}_2 + \mathbf{M}_4) - B(\mathbf{M}_5 \times \mathbf{d}_{15}) + \mathbf{H}_{A1}, \\ \mathbf{H}_2 &= \frac{\mathbf{g}_B}{g} \cdot \mathbf{H} - A\mathbf{M}_6 + A'(\mathbf{M}_1 + \mathbf{M}_3) - B(\mathbf{M}_6 \times \mathbf{d}_{26}) + \mathbf{H}_{A2}, \\ \mathbf{H}_3 &= \frac{\mathbf{g}_A}{g} \cdot \mathbf{H} - A\mathbf{M}_7 + A'(\mathbf{M}_2 + \mathbf{M}_4) - B(\mathbf{M}_7 \times \mathbf{d}_{37}) + \mathbf{H}_{A3}, \\ \mathbf{H}_4 &= \frac{\mathbf{g}_B}{g} \cdot \mathbf{H} - A\mathbf{M}_8 + A'(\mathbf{M}_1 + \mathbf{M}_3) - B(\mathbf{M}_8 \times \mathbf{d}_{48}) + \mathbf{H}_{A4}, \\ \mathbf{H}_5 &= \frac{\mathbf{g}_A}{g} \cdot \mathbf{H} - A\mathbf{M}_1 + A'(\mathbf{M}_6 + \mathbf{M}_8) - B(\mathbf{M}_1 \times \mathbf{d}_{15}) + \mathbf{H}_{A5}, \\ \mathbf{H}_6 &= \frac{\mathbf{g}_B}{g} \cdot \mathbf{H} - A\mathbf{M}_2 + A'(\mathbf{M}_5 + \mathbf{M}_7) - B(\mathbf{M}_2 \times \mathbf{d}_{26}) + \mathbf{H}_{A6}, \\ \mathbf{H}_7 &= \frac{\mathbf{g}_A}{g} \cdot \mathbf{H} - A\mathbf{M}_3 + A'(\mathbf{M}_6 + \mathbf{M}_8) - B(\mathbf{M}_3 \times \mathbf{d}_{37}) + \mathbf{H}_{A7}, \\ \mathbf{H}_8 &= \frac{\mathbf{g}_B}{g} \cdot \mathbf{H} - A\mathbf{M}_4 + A'(\mathbf{M}_5 + \mathbf{M}_7) - B(\mathbf{M}_4 \times \mathbf{d}_{48}) + \mathbf{H}_{A8}, \end{aligned} \tag{3}$$

where the first terms on the right-hand side are the external fields respectively modified by $\mathbf{g}_A$ and $\mathbf{g}_B$; the second and the third terms respectively correspond to the exchange field from J_c and J_a; the fourth terms come from DM interaction characterized by the following vector $\mathbf{d}_{ij}$:

$$\begin{aligned} \mathbf{d}_{15} &= \mathbf{d}_{73} = [-d, 0, 0], \\ \mathbf{d}_{51} &= \mathbf{d}_{37} = [d, 0, 0], \\ \mathbf{d}_{26} &= \mathbf{d}_{84} = [0, -d, 0], \\ \mathbf{d}_{48} &= \mathbf{d}_{62} = [0, d, 0]; \end{aligned}$$

and the last terms $\mathbf{H}_{Ai} = (0, 0, -\frac{K}{M^2}M_{iz})$ $(i = 1\text{–}8)$ are the XY-like anisotropy fields. In (3) $M_i = -Ng\mu_\mathrm{B}\langle S_i\rangle$ $(i = 1\text{–}8)$ 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 system $[x, y, z]$ is taken as parallel to the $[100]_p$, $[010]_p$ and $[001]_p$ axes, respectively.

Because of the difficulty of analytically solving such a large-scale system of equations, the equations of motion (2) are numerically calculated to obtain the eigenfrequencies. In the calculation, the experimental data $J_c = -190$ K, $J_a = 0.01|J_c|$, $D_c = 0.04$ K, and $d = 0.027|J_c|$ are employed. The parameters in equation (1) are determined by [11]

$$A = \frac{4|J_c|}{N\gamma^2 h^2}, \qquad A' = \frac{4J_a}{N\gamma^2 h^2}, \qquad B = \frac{4}{N\gamma^2 h^2}, \qquad K = \frac{4D_c}{N\gamma^2 h^2}, \tag{4}$$

where $\gamma = -g\mu_B/\hbar$. Moreover, the first approximation

$$\mathbf{M}_{i0} \times \mathbf{H}_{i0} = 0$$

is also used as the initial condition, and therefore we can obtain the linear equations of motion:

$$\frac{i\omega}{\gamma}\mathbf{m}_i + \frac{i\alpha_i\omega}{|\mathbf{M}_{i0}|}\mathbf{m}_i \times \mathbf{M}_{i0} + \mathbf{m}_i \times \mathbf{H}_{i0} + \mathbf{M}_{i0} \times \mathbf{h}_i = 0, \qquad (i = 1\text{–}8). \tag{5}$$

The calculated frequency–field diagram for the lowest-frequency mode and linewidth–frequency relation are shown in figures 1 and 2. The dotted line and the dashed line in the figures correspond to $\mathbf{H} \parallel [110]_p$ and $\mathbf{H} \parallel [100]_p$, respectively. In the calculated eight eigenmodes, we only show the two which are basically in accordance with the observed two modes, and the detailed discussion for the multi-sublattice modes in $KCuF_3$ will be published elsewhere. Spin flop is suggested to occur at $H_{sf} = |\frac{1}{2}(\mathbf{g}_A + \mathbf{g}_B) \cdot \frac{\mathbf{H}}{g}| = 500$ Oe for H $\parallel [110]_p$.

One can see from the figures that there are some slight differences between the experimental data and the calculated curves. Because the analytical method used here is based on the mean field approximation, exact agreement between experimental results and calculation should not be expected. Nevertheless, the calculated result based on the interdependent effect of the DM interaction and inequivalent g-tensors actually governing $\mathbf{H}_{res}$ in the c-plane has reproduced the essential features of the experimental data.

Furthermore, it is difficult to derive the explicit expression of frequency–field relation for each of the eight sublattices owing to the complexity of the 24 secular equations. However, by taking the values of $\mathbf{H}_{DM}$, $\mathbf{g}_A \cdot \mathbf{H}$, $\mathbf{g}_B \cdot \mathbf{H}$, $A\mathbf{M}_i$, $A'\mathbf{M}_i$, and $\mathbf{H}_{Ai}$ into the matrix with 24×24 elements, we can reasonably postulate the analytical expressions of both $\mathbf{H}_{res}$ and ΔH_{pp} for the eight resonance modes of $KCuF_3$ as follows:

$$\mathbf{H}_{res} \sim \frac{\omega}{\gamma}\left(f\frac{1}{\cos(4\theta)} + \cos(4\theta)\right), \tag{6}$$

$$\Delta H_{pp} \sim \gamma \frac{\frac{k}{\omega}}{\exp(\frac{m}{\omega}) + 1}, \tag{7}$$

where θ is the angle between the $[110]_p$ axis and $\mathbf{H}$; k, m and f are all adjustable parameters, and $k, m \propto 1/\mathbf{H}_{res}$.

In summary, the resonance linewidth of $KCuF_3$ is first observed to depend on the frequency, i.e., the lower the frequency the greater the resonance linewidth, no matter whether the AFMR field is forced on the easy axis or the uneasy axis. Moreover, a linewidth peak seems to exist for $H \parallel [100]_p$ at about 4 GHz. The linewidth behaviour in $KCuF_3$ is different from the behaviour of the linewidth in usual two-sublattice antiferromagnets without the Dzyaloshinsky–Moriya interaction in which the linewidth seem to reduce with decreasing frequency. In addition, two kinds of the absorption linewidth are first found to take place in the substantial uneasy direction of the spin $[100]_p$ axis at the lower-frequency (<5 GHz) branch of the C-band. A numerical calculation considering the Laudau–Lifshitz damping term for the resonance linewidth and frequency–field relation based on the model of Yamada and Kato [11] is in good agreement with the AFMR measurements, which indicates that the nature of the unusual frequency dependence of the linewidth might be suspected to come from the DM interaction.

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