Antiferromagnetic resonance modes for the $S = 1/2$ kagome antiferromagnet $\text{Cs}_2\text{Cu}_3\text{SnF}_{12}$

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Abstract. We have investigated the antiferromagnetic resonance modes of the kagome antiferromagnet $\text{Cs}_2\text{Cu}_3\text{SnF}_{12}$ both theoretically and experimentally. High-field electron spin resonance experiments on single crystals were conducted in the ordered phase at different frequencies and temperatures with the external magnetic field parallel to the $c$ axis. Considering three sublattices, we calculated the resonance modes for the motions of the sublattice magnetizations within the framework of the mean field approximation. It was found that the frequency-field diagram is in good agreement with the experimental results.

1. Introduction
A two-dimensional (2-D) spin frustrated system is an interesting subject in condensed matter physics, because of the competition between the frustration of spins and the anisotropy due to the low dimensionality. The basic building block of most geometrically frustrated systems is a set of three spins on a triangle interacting antiferromagnetically. An example of such a system is the kagome lattice which may be seen as a diluted triangular lattice with larger geometrical frustration and lower coordination number than the triangular lattice [1].

Calculating the antiferromagnetic resonance modes (AFMR) provides a microscopic view of the magnetic structure, leading to measurements of magnetic exchange, magnetic anisotropy, spin-flop and critical fields, and spin canting. Also, there is no inversion center at the middle point of two neighboring magnetic ions in the kagome lattice which differs from the situation in the triangular lattice. Thus, in general, the Dzyaloshinskii-Moriya (DM) interaction is allowed in the kagome lattice [3]. AFMR modes should also be able to provide information on the DM interaction. In this work, we present a calculation of the resonance modes of a stoichiometrically pure $S = 1/2$ ideal kagome lattice antiferromagnet $\text{Cs}_2\text{Cu}_3\text{SnF}_{12}$ with full occupancy of spin-$1/2$ at the Cu sites in the easy-plane anisotropy case and compare the theoretical results with experimental data.
2. Kagome antiferromagnet Cs$_2$Cu$_3$SnF$_{12}$

Cs$_2$Cu$_3$SnF$_{12}$ is a relatively new antiferromagnetic (AF) kagome structure which possesses quite a number of properties that are not well understood. The spins are 120° apart from each other as expected in the so-called $q = 0$ or $\sqrt{3} \times \sqrt{3}$ spin structures proposed as the possible ground states of a kagome antiferromagnet. The magnetic Cu$^{2+}$ ions are surrounded by F$^-$ ions octahedrally and CuF$_6$ octahedra are linked in the $ab$ plane with sharing corners. Since CuF$_6$ octahedra are elongated along the principal axes approximately parallel to the $c$ axes, the hole orbitals $d(x^2 - y^2)$ of Cu$^{2+}$-ions spread within the kagome layer. The value of the bond angle $\alpha$ for Cs$_2$Cu$_3$SnF$_{12}$ is 138.9° at room temperature [4]. Furthermore, the kagome layers are well separated by non-magnetic Cs$^+$, Sn$^{4+}$ and F$^+$ layers. We observe the presence of antiferromagnetic long-range ordering at around 20K and a structural transition $T_1 \sim 185K$. The $g$ factors determined self-consistently in the temperature range higher than the structural phase-transition temperature are $g_\parallel = 2.48$ and $g_\perp = 2.10$ where $g_\parallel$ and $g_\perp$ are the $g$ factors for the magnetic field parallel and perpendicular to the $c$ axis. Exchange constant $J$ for $T > T_1$ is 240 K. The spins are aligned parallel to an easy $c$ axis and a weak ferromagnetic moment along the $ab$-plane has been established in the magnetization measurements [5].

There exists a mirror plane that passes the middle points of neighboring Cu$^{2+}$ ions and is perpendicular to the line connecting these two ions. Therefore the D vector should be parallel to the mirror plane. Because there are twofold screw axes along the [1,0,0], [0,1,0] and [1,1,0] directions, the D vectors change their directions alternately along these directions [5] [see Fig. 1].

![Figure 1](image-url)

**Figure 1.** The configuration of D vectors and $q = 0$ spin structure with plus chirality and staggered field directions. Arrangement of the D vectors for $T > T_1$ in Cs$_2$Cu$_3$SnF$_{12}$ systems, (a) the $c$ axis component $D_\parallel$ and (b) the $c$ plane component $D_\perp$. Symbols $\circ$ and $\otimes$ in (a) and short thick arrows in (b) denote the local positive directions of parallel and perpendicular components $D_\parallel$ and $D_\perp$, respectively. Large arrows in (a) denote the $q = 0$ spin structure stabilized when $D_\parallel > 0$. Long thin arrows in (b) denote the local direction of staggered field $h_i$.

3. Antiferromagnetic resonance modes

We derive the resonance modes for $H_\parallel|c$ within the framework of the mean field approximation. In Cs$_2$Cu$_3$SnF$_{12}$, we may consider only three sublattices, which lie in the kagome plane with 120° structure. We assume the $q = 0$ structure with plus chirality as shown in Fig. 1(a).
We consider the Hamiltonian under an external magnetic field $H$ as follows:

$$H = \sum_{\langle i,j \rangle} J S_i \cdot S_j + \sum_{\langle i,j \rangle} D_{ij} \cdot [S_i \times S_j] - g \mu_B H \sum_i S_i^z - \sum_i g \mu_B h_i \cdot S_i$$

(1)

$D^\perp$ and $D^\parallel$ are respectively the real values of the parallel and perpendicular components of the Dzyloshinskii-Moriya (DM) interaction, $J$ is the exchange constant, $h_i$ the staggered field induced by inclination of the principal axes of the $g$ tensors associated with the low local symmetry of the Cu$^{2+}$ magnetic centers. In this paper, we do not consider exchange interactions between kagome layers.

For a simple three sublattice antiferromagnet, three resonance frequencies should be observed. The resonance conditions of these three observable modes are given by:

$$\omega_0 = \sqrt{3(\langle d^\perp \rangle^2 + HM_0 \left\{ \frac{h}{H} (3B + \sqrt{3}d^\parallel) + \sqrt{3}d^\perp \right\} } \, ,$$

(2)

$$\omega_{\pm} = \sqrt{\frac{3}{2} (B + \sqrt{3}d^\parallel) M_0 (\sqrt{3}d^\parallel M_0 + h) + \frac{\sqrt{3}}{4} d^\perp M_0 (\sqrt{3}d^\parallel M_0 + H) + \frac{3(B + \sqrt{3}d^\parallel)}{4(3B + \sqrt{3}d^\parallel)} H^2} \pm \frac{3B - \sqrt{3}d^\parallel}{2(3B + \sqrt{3}d^\parallel)} H ,$$

(3)

where $M_0$ is the magnitude of the sublattice magnetizations, given by $M_0 = (N/3)g \mu_B \langle S \rangle$, where $\langle S \rangle$ is the mean value of spin $S$, $h$ is the magnitude of the staggered field and

$$B = \frac{3}{N} \frac{2J}{\langle g \mu_B \rangle^2} , \quad d^\perp = \frac{3}{N} \frac{2D^\perp}{\langle g \mu_B \rangle^2} .$$

(4)

The $\omega_0$ mode corresponds to the global rotation of spins around the external field $H$. Thus, its frequency becomes zero in the case of $D^\perp = 0$ and $h = 0$. When the staggered field $h_i$ is parallel to the sublattice spin $S_i$, i.e., $h/H > 0$, the frequency of the $\omega_0$ mode increases with increasing $H$. On the other hand, when $h_i$ and $S_i$ are antiparallel, i.e., $h/H < 0$, the frequency of the $\omega_0$ mode decreases and becomes zero at

$$H_c = -\frac{3\langle d^\perp \rangle^2 M_0}{c_s (3B + \sqrt{3}d^\parallel) + \sqrt{3}d^\perp} ,$$

(5)

where $c_s$ is given by $c_s = h/H$. As shown below, $c_s < 0$ in Cs$_2$Cu$_3$SnF$_{12}$.

4. Experimental results

High-field ESR experiments were performed on single crystals of Cs$_2$Cu$_3$SnF$_{12}$ at different frequencies. Only one antiferromagnetic resonance mode $\omega_0$ was observable below 210 GHz, as shown in Fig. 2. This is to be expected as the frequencies $\omega_{\pm}$ calculated from Equation (3) should be in the terahertz region.

In Figure 2 the field dependence of the frequencies for $H||c$ measured at $T = 4.2$ and 2 K are shown. The resonance mode can be assigned as the $\omega_0$ mode. The frequency decreases with increasing magnetic field, which indicates that $h_i$ and $S_i$ are antiparallel, $c_s < 0$. The solid lines follow Equation (2) with $BM_0 = 178$ T. At both temperatures the experimental values fit well to the theoretical equation. The fit gives the values of other interaction parameters: $d^\perp M_0 = 4.93$ T and $c_s = -0.057$ for $T = 4.2$ K, and $d^\perp M_0 = 4.63$ T and $c_s = -0.058$ for $T = 2.0$ K. The values of $d^\perp M_0$ evaluated at $T = 2.0$ and 4.2 K are different. This seems not intrinsic but experimental error. For difference in critical field $H_c$, we have no reasonable explanation at present.
Figure 2. Frequency-field diagram for $H \parallel c$ obtained at $T = 2.0$ and 4.2 K. The solid lines show the fit of Eq. (2) to the experimental data with interaction parameters shown in the text.

5. Conclusion
High-field ESR experiments have been conducted on single crystals of the kagome antiferromagnet Cs$_2$Cu$_3$SnF$_{12}$ at different frequencies and at different temperatures. The experimental results were compared with the theoretical data derived from the model proposed using the mean field approximation to calculate the resonant modes in kagome antiferromagnets consisting of three sublattices with easy-plane anisotropy. It was found that the experimental and theoretical data are in good agreement. Values of the magnitudes of the Dzyaloshinskii-Moriya interaction and the staggered field were evaluated.

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6. References
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