19F-NMR Study on Antiferromagnetic Heisenberg Chain KCuGaF₆

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Abstract. $S = \frac{1}{2}$ one-dimensional antiferromagnet KCuGaF₆, which has the alternation of both the $g$-tensor and Dzyaloshinsky-Moriya (DM) vector along the chain, is theoretically expected to show the field-induced spin gap. We have measured the longitudinal nuclear spin relaxation rate $T_{1-1}$ of 19F-NMR and found that it shows the thermal activation type temperature dependence, indicating the existing of the spin gap. The field-dependence of the gap is found to be proportional to $H^{2/3}$. These results are consistent with the theory of Affleck and Oshikawa.

1. Introduction

The magnetic ground state of the $S = \frac{1}{2}$ antiferromagnetic Heisenberg chain (AFHC) is in general gapless [1]. However, the field-induced gap has recently been observed in some $S = \frac{1}{2}$ AFHC systems such as Cu (C₆H₅COO)₂·3H₂O [2], PM-Cu (NO₃)₂·(H₂O)₂ (PM = pyrimidine) [3]. The gap $\Delta(H)$ was found to be

$$\Delta(H) = AH^\alpha,$$

where the coefficient $A$ depends on the field direction, and $\alpha$ is approximately equal to $2/3$. Affleck and Oshikawa investigated theoretically $S = \frac{1}{2}$ AFHC, in which both $g$ tensor and Dzyaloshinsky-Moriya (DM) vector alternate along the chain, described as the following model [4]

$$\mathcal{H} = \sum_i [J \vec{S}_i \cdot \vec{S}_{i+1} - g\mu_B H S_i^z + (-1)^i g\mu_B h S_i^z],$$

where $h = c_sH$ is the effective staggered field induced by applied magnetic field, and $J$ is the exchange interaction. The constant $c_s$ depending on the field direction is determined experimentally from the magnetic susceptibility as [6]

$$\chi \equiv \chi_u + c_s^2 \chi_s,$$

where $\chi_u$ is the uniform susceptibility of the intrinsic part [5] and $\chi_s$ is the staggered susceptibility.
induced by the magnetic field. The former per a spin is written as

$$\chi_u = \frac{g^2 \mu_B^2}{k_B T} F \left( \frac{J}{k_B T} \right),$$

(4)

and $F(x)$ is an empirical function proposed by Feyerherm et al. [3]

$$F(x) = \frac{1 + 0.08516x + 0.2335x^2}{1 + 0.73382x + 0.1369x^2 + 0.53568x^3}.$$  

(5)

For $T > \Delta$, $\chi$ per a spin is given [6] by

$$\chi_s = 0.2779 \cdot \frac{g^2 \mu_B^2}{k_B T} \sqrt{\ln \left( \frac{J}{k_B T} \right)}.$$  

(6)

So far, Umegaki and Morisaki et al. have intensively studied the $S = 1/2$ AFHC system KCuGaF$_6$, which is described by the model (2), to find that the specific heat shows the thermal-activation-type temperature dependence and that the magnetic susceptibility obeys the Curie law below 20 K. These results suggest that the gap and staggered magnetization are induced by the external magnetic field, indicating that their results are consistent with the theory of Oshikawa and Affleck [4], [6], and also with [8], [9] lately developed.

In this paper, we show microscopically by utilizing the NMR technique that KCuGaF$_6$ exhibits both the field-induced gap and the field induced staggered field.

2. Experimental

KCuGaF$_6$ has a monoclinic crystal structure with the space group $P2_1/c$, in which Cu$^{2+}$ and Ga$^{3+}$ ions form a pyrochlore lattice [10]. The single crystal of KCuGaF$_6$ with the size of about 500 mm$^3$ grown by I. U. was used for present NMR measurements. The temperature dependence of the longitudinal nuclear spin relaxation rate of $^{19}$F-NMR under various fields was measured by the saturation-recovery method with a pulse train. The temperature and the field regions of measurements were around 1.8 – 20 K and 2 – 5 T, respectively. The temperature dependence of the uniform magnetization is measured under the field approximately the same as NMR measurements.

3. Results and Discussion

Figure 1 shows typical profiles of $^{19}$F-NMR spectra at various temperatures. We observed six NMR resonance lines that correspond to the crystallographically-inequivalent six fluorine sites. The hyperfine coupling constant for each site was obtained by comparing the NMR shift and the uniform magnetization as shown in Fig. 2. The result is listed in Table 1. The variety of both the magnitude and the sign of $A$ comes from the geometrical location of each F site around Cu.

We then analyzed the temperature dependence of the resonance line shift, using the following equation [4]:

$$\nu / 19\gamma - H = AH (\chi_u + c^2 \chi_s),$$  

(7)

where $\nu$ and $19\gamma$ are the resonance frequency and the gyromagnetic ratio of $^{19}$F. The temperature dependence of the Knight shift obeys the Curie-like behavior as shown in Fig. 3. With parameters reported by I. U., as $J/k_B = 103$ K, $g = 2.12$, and $c_s = 0.18$ [11], we can reproduce the observed temperature dependence only by modifying the value of $A$ to 40 % larger than the one determined...
above. We also note that it deviates from the Curie law and tends to saturate below 4 K. This is simply due to the magnetization saturation in high field, and hence does not affect the existence of the Curie term.

Figure 1. The temperature dependence of $^{19}$F-NMR spectra for $c // H$.

Figure 2. The plot of the resonance line shift vs. the macroscopic magnetic magnetization with the temperature as the implicit parameter. A typical temperature region is between 4 – 16 K.

Figure 3. The temperature dependence of the resonance line shift of each peak in $^{19}$F-NMR spectrum. The dashed curve shows the Curie-Weiss temperature dependence of eq. (7).

Figure 4. Typical longitudinal nuclear spin relaxation curves for $c // H$. Dashed curves are the fitting function.

Table 1. Hyper fine coupling constants $A$ for each site.

| Peak | P1   | P2   | P3   | P4   | P5   | P6   |
|------|------|------|------|------|------|------|
| $A$  | 1.12 | 0.88 | 0.53 | 0.23 | −0.098 | −0.37 |

Next, $T_1$ was measured at the peak of P3, that is, at $H = 4.91$ T. Figure 4 shows the typical relaxation curves; they are described with the two components short and long, $T_{1S}$ and $T_{1L}$, respectively. The fraction of these two components was found to be temperature independent and approximately unity. Figure 5 shows the temperature dependence of $\log T_1^{-1}$’s for the two components. The values of
gap $\Delta$ were estimated from gradients of $\log T_1^{-1}$ to be approximately 8.7 K, for both components measured at 4.91 T. We also investigated the field dependence of the gap in the region 2 and 5 T as shown in Fig. 6. The gap increased with increasing field $H$, and obeyed a power law $H^{2/3}$, which agrees with the ESR measurement reported by I. U. [11].

![Graph](image)

**Figure 5.** Temperature dependence of $T_1^{-1}$ measured at $H = 4.91$ T. The dashed lines show the thermal activation type function, from which the gap was estimated to be $\Delta = 8.7$ K.

**Figure 6.** Field dependence of the gap measured in the region 2 and 5 T for $c \parallel H$. The dashed line denotes the function $AH^{\alpha}$ with $\alpha = 2/3$ and $A = 3 (K/T^{2/3})$.

4. Conclusion

By utilizing $^{19}$F-NMR technique as a microscopic probe, the existence of the field-induced gap in the $S = 1/2$ AFHC KCuGaF$_6$ was confirmed. The field dependence of the gap was found to be proportional to $H^{2/3}$, which is consistent with the theory of Affleck and Oshikawa.

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References

[1] des Cloizeaux J and Pearson J J 1962 Phys. Rev. 128 2131.
[2] Dender D C, Hammar P R, Reich Daniel H, Briholm C, and Aeppli G 1997 Phys. Rev. Lett. 79 1750.
[3] Feyerherm R, Abens S, Günther D, Ishida T, Meißner M, Meschke M, Nomgami T, and Steiner M 2000 J. Phys. Condens. Matter 12 8495.
[4] Oshikawa M and Affleck I 1997 Phys. Rev. Lett. 79 2883.
[5] Eggert S, Affleck I, and Takahashi M 1994 Phys. Rev. Lett. 73 332.
[6] Affleck I and Oshikawa M 1999 Phys. Rev. B 60 1038.
[7] Morisaki R, Ono T, Tanaka T, and Uekusa H 2006 J. Phys. Conf. Ser. 51 17.
[8] Hikihara T, and Furusaki A. 2004 Phys. Rev. B 69 064427.
[9] Takayoshi S, and Sato M 2010 Phys. Rev. B 82 214420.
[10] Dahlke P, Fehler J, and Babel D 2005 Z. Anorg. Allg. Chem. 631 115.
[11] Umegaki I, Ono T, Uekusa H, and Nojiri H 2009 Phys. Rev. B 79 184401.