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Impurity-Robust Bulk Gapless Excitation in the Yb-Based Zigzag Chain Compound YbCuS₂

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Abstract. We have performed ⁶³Cu-nuclear quadrupole resonance (NQR) measurements using a lump sample of the Yb zigzag-chain compound YbCuS₂ with a small surface area to investigate the sample dependence of low-temperature magnetic properties in YbCuS₂ by comparing with the previous study with different powdered sample. The line width of NQR signals in the present lump sample is larger than that in the previous powdered sample. In addition, the transition temperature $T_N \sim 0.92$ K in the present lump sample is lower than that in the previous powdered sample (~ 0.95 K). These results suggest that the quality of the present lump sample is worse than that of the previous powdered sample. However, the T -linear behavior of the nuclear spin-lattice relaxation rate $1/T_1$ was observed below 0.5 K and the value of $1/T_1 T$ in both samples is almost the same even though the sample quality and sample geometry are different. This suggests that T -linear behavior in $1/T_1$ arises from the impurity-robust bulk gapless excitation inherent in YbCuS₂ rather than from sample issues such as the sample quality or geometry.

1. Introduction

Rare-earth-based frustrated spin systems have been attracted much attention [1–5]. This is because the strong spin-orbit coupling and crystalline electric field associated with the rare-earth ion lead to anisotropic exchange interactions, which may induce strong quantum fluctuations and host novel quantum spin states.

YbCuS₂ is a magnetic semiconductor with Yb zigzag chains. This compound has an orthorhombic structure with the space group $P2_12_12_1$, and Yb ions form the zigzag chains along the a -axis [6]. At zero magnetic field $H = 0$, the magnetic specific heat shows a peak at $T_N \sim 0.95$ K [7, 8]. In the previous study, we performed ^{63/65}Cu nuclear magnetic resonance and nuclear quadrupole resonance (NQR) measurements on a powdered sample of YbCuS₂ [9] and revealed the occurrence of the antiferromagnetic (AFM) phase transition at T_N . In addition, the nuclear spin-lattice relaxation rate $1/T_1$ was proportional to T below 0.5 K, which is generally observed in metals. The value of $1/T_1 T$ below 0.5 K in YbCuS₂ is by more than one order of magnitude larger than that in Cu metals [10]. This indicates the presence of the novel gapless spin excitation at low temperatures in YbCuS₂ with the magnetic frustration based on the Yb zigzag chain.



In this study, to investigate the origin and characteristics of the peculiar T -linear behavior of $1/T_1$ in YbCuS_2 , we performed the ^{63}Cu -NQR measurements by changing the following two conditions from the previous study [9]. First, by using different quality of a sample as the previous study, we investigated the sample quality dependence of the low-temperature magnetic excitations. Second, by using a lump sample with a small surface area, which is different sample geometry from the previous powdered sample, we checked whether the low-temperature magnetic excitation comes from the surface or the bulk. We observed the same T -linear behavior of $1/T_1$ below 0.5 K and we confirmed the value of $1/T_1 T$ in both samples is almost the same even though the sample quality and geometry were different. This suggests that T -linear behavior of $1/T_1$ arises from the impurity-robust bulk gapless excitation inherent in YbCuS_2 rather than from extrinsic sample issues such as the sample quality or sample geometry.

2. Experiments

Polycrystalline samples of YbCuS_2 were synthesized by the melt-growth method [8]. ^{63}Cu -NQR measurements were performed on a cuboid lump sample of $6 \times 6 \times 5 \text{ mm}^3$ size. A conventional spin-echo technique was used for the ^{63}Cu -NQR measurements (^{63}Cu nucleus have nuclear spin $I = 3/2$ with nuclear gyromagnetic ratios of $^{63}\gamma/2\pi = 11.289 \text{ MHz/T}$, and there are nuclear quadrupolar interactions). A ^3He - ^4He dilution refrigerator was used down to 0.075 K. The NQR spectra were obtained by the frequency-swept method without an external field. $1/T_1$ was evaluated by fitting the relaxation curve of the nuclear magnetization after the saturation to a theoretical function for the nuclear spin $I = 3/2$. While $1/T_1$ was determined by a single relaxation component down to T_N , the whole of the relaxation curve cannot be fitted to the single relaxation component below T_N . Thus, we picked up the slowest components below T_N in the this study as well as the previous study [9].

3. Results and Discussion

Figure 1(a) shows the frequency-swept ^{63}Cu -NQR spectra measured at 4.2 K (upper panel) and 0.3 K (lower panel). Similarly to the previous study [9], a single ^{63}Cu -NQR signal was observed above T_N , and the NQR peak splits into multi peaks due to the internal magnetic fields arising from the AFM ordered moments below T_N . The NQR-peak positions in the present lump sample are almost the same with those in the previous powdered samples. On the other hand, the line width of NQR signals in the present lump sample is larger than that of the previous powdered sample, suggesting that the quality of the present lump sample is worse than that of the previous powdered sample.

The difference of the sample quality in the present sample and previous one was also confirmed from the comparison of the transition temperatures determined by the decrease in the NQR peak intensity. Figure 1(b) shows the temperature dependence of the products of the NQR peak intensity and the temperature $IT(T)$ at $f = 9.3 \text{ MHz}$. We define T_N as the temperature when $IT(T)$ decreases rapidly with decreasing temperature. In the present lump sample, $IT(T)$ decreases rapidly at $T_N \sim 0.92 \text{ K}$, which is slightly lower than that in the previous powdered sample ($\sim 0.95 \text{ K}$).

In contrast to the NQR spectra and T_N , the behaviors of the nuclear spin-lattice relaxation rate $1/T_1$ were almost the same in the previous powdered sample and the present lump sample as shown in Fig. 2. Due to the difference of T_N , the value of $1/T_1$ in the present lump sample is slightly larger than that in the previous powdered sample for $0.5 \text{ K} \leq T \leq T_N$. On the other hand, T -linear behavior of $1/T_1$ was observed below T_N and the value of $1/T_1 T$ at lowest temperatures is almost the same in both samples, suggesting that T -linear behavior of $1/T_1$ is independent of the sample quality and geometry.

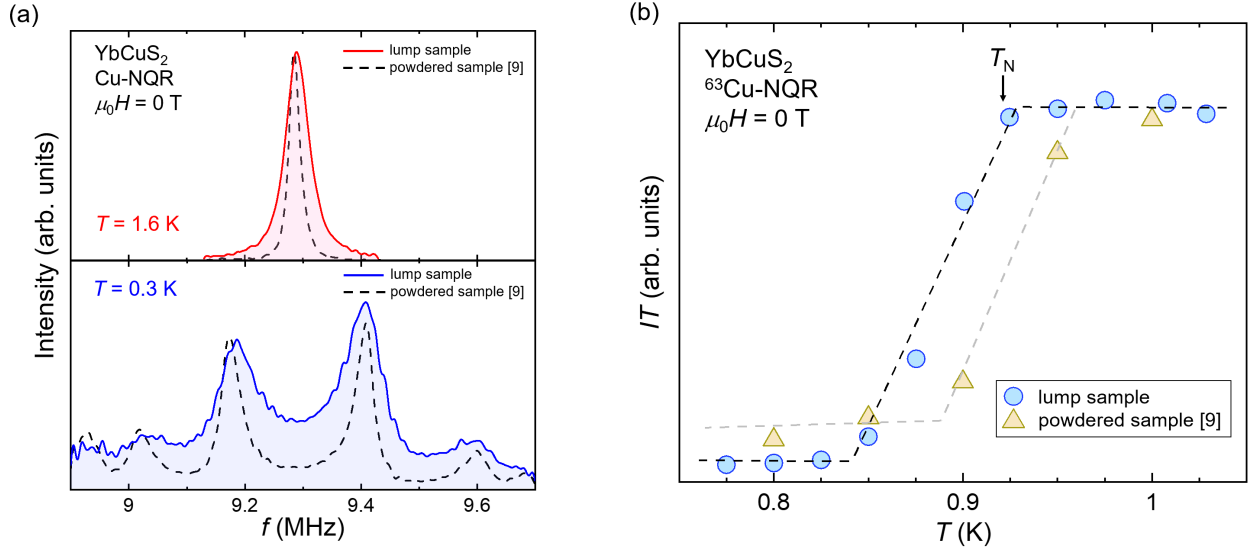


Figure 1. (Color online) (a) ^{63}Cu -NQR spectra obtained by the frequency-swept method at 4.2 K (upper panel) and 0.3 K (lower panel): the (red and blue) solid lines and (black) dashed line represents ^{63}Cu -NQR spectra in present lump sample and the previous powdered sample, respectively. (b) The temperature dependence of the products of the NQR peak intensity and the temperature $IT(T)$: the circles and triangles denote $IT(T)$ in the present lump sample and the previous powdered sample, respectively.

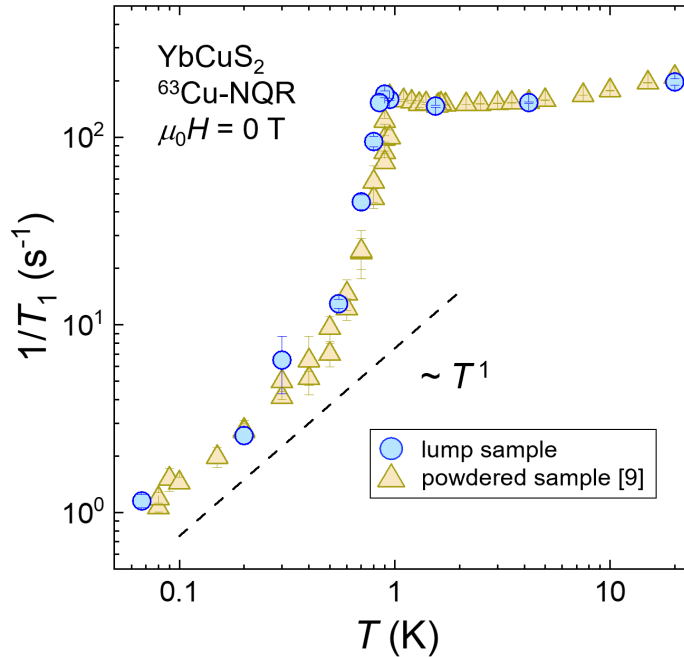


Figure 2. Temperature dependence of the ^{63}Cu -NQR nuclear spin-lattice relaxation rates $1/T_1$ in YbCuS_2 : the circles and triangles denote $1/T_1$ in the present lump sample and the previous powdered sample, respectively.

As mentioned in the previous study [9], the similar T -linear behavior of $1/T_1$ below transition temperature was reported in several kagomé systems, and spinon excitations were proposed in these systems [11–13]. Recently, the ground states of several Yb triangular lattices such as YbMgGaO₄ [1–3] and YbNaSe₂ [4, 5] were also reported to be quantum spin liquid states with a spinon Fermi surface. However, YbMgGaO₄ has an inhomogeneous charge environment arising from a site mixing of Mg²⁺ and Ga³⁺, which gives rise to the orientational spin disorder and mimic a spin-liquid-like state as different interpretation of the ground state. [14, 15]. In contrast, the novel gapless excitation in YbCuS₂ is not related to sample extrinsic effect such as the sample quality or sample geometry.

4. Conclusion

In conclusion, we have performed ⁶³Cu-NQR measurements using a lump sample of YbCuS₂ with a small surface area to investigate the sample dependence of low-temperature magnetic properties in YbCuS₂ by comparing with the previous study with different powdered sample. The line width of NQR signals in the present lump sample is larger than that of the previous powdered sample. Moreover, the transition temperature $T_N \sim 0.92$ K in the present lump sample is slightly smaller than that in previous one (~ 0.95 K). These results suggest that the quality of the present lump sample is worse than that of the previous powdered sample. In contrast, the novel T -linear behavior of $1/T_1$ was observed below 0.5 K and the value of $1/T_1 T$ in both samples is almost the same even though the sample quality and sample geometry are different. This suggests that the behavior of $1/T_1$ at low temperature arises from the impurity-robust bulk gapless excitation inherent in YbCuS₂ and is not related to sample issues such as the sample quality or sample geometry. The properties of this novel gapless magnetic excitation deserve further investigation.

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