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Gapless magnetic excitations in the kagome antiferromagnet Ca-kapellasite probed by ^{35}Cl NMR spectroscopy

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The low-energy magnetic excitations of the spin-1/2 kagome antiferromagnet $\text{CaCu}_3(\text{OH})_6\text{Cl}_2 \cdot 0.6\text{H}_2\text{O}$ (Ca-kapellasite) have been investigated by a ^{35}Cl NMR experiment in fields up to 18.9 T. Recently, Ca-kapellasite was found to be one of the most promising candidates to explore the intrinsic magnetic properties in kagome antiferromagnets, because magnetic defects, which deteriorate the intrinsic magnetic properties, are minimized by choosing Ca ions with a large ionic radius as the counterions. From our nuclear spin-lattice relaxation rate $1/T_1$ measurement, we found a power-law temperature dependence below the magnetic ordering temperature $T_M = 7.2$ K, which leads us to suggest that gapless magnetic excitations survive even in the ordered state. This power-law behavior is suppressed in high magnetic fields. We discuss a possible magnetic state that can generate gapless excitations at low fields.

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In quantum magnets with 1/2 spins, when the canonical magnetic ordering is disturbed by geometrical frustration and/or low-dimensionality effects, strong quantum fluctuations at low temperatures stabilize exotic spin states. The most apparent quantum fluctuation effect is expected for antiferromagnetically interacting spins on a kagome network, because the corner-sharing triangle network reduces the number of neighboring spins ($Z = 4$). In these kagome antiferromagnets (KAFMs), macroscopic numbers of spin states are degenerate or pseudodegenerate at very low energy, and therefore the exotic spin states, which never appear in canonical antiferromagnets, can be stabilized by a small energy perturbation, such as magnetic fields. Although interesting, an unperturbed ground state is hard to be picked out from the huge numbers of possible candidates [1]. In fact, theoretical studies have proposed various quantum spin-liquid states with gaps or without gaps in the spin excitation spectrum. A gapped topological Z_2 state was suggested from the density matrix renormalization group theory [2], while a gapless $U(1)$ -Dirac state was suggested from the Gutzwiller projection technique [3] and also from the recently developed tensor-network method [4]. To reveal the intrinsic ground state, which appears in real materials, an experimental search for a perfect KAFM is compulsory. At this time, the most promising candidate material for the perfect KAFM is the mineral herbertsmithite $[\text{ZnCu}_3(\text{OH})_6\text{Cl}_2]$ [5], for which many experimental studies have been performed [6–11], and a gapped spin-liquid ground state has been suggested from the latest ^{17}O NMR spectroscopy study for the single crystal [12]. In herbertsmithite, however, the site exchange between magnetic Cu and nonmagnetic Zn creates isolated spins [13], which can deteriorate the proposed intrinsic ground state for a clean system. Searches for the perfect KAFM have been carried out for long time [14–17].

The recently reported candidate for a KAFM is Ca-kapellasite $[\text{CaCu}_3(\text{OH})_6\text{Cl}_2 \cdot 0.6\text{H}_2\text{O}]$, for which an x-ray diffraction study evidences no mixture of Cu/Ca ions because of the large ionic radius of the Ca ions [18]. The structurally

perfect kagome network of Cu ions without magnetic defects allows us to study the intrinsic magnetic properties of KAFM. Ca-kapellasite shows a small peak in the temperature (T) dependence of heat capacity divided by temperature C/T at $T_M = 7.2$ K, at which the in-plane susceptibility shows a cusp anomaly [18]. These results suggest that a weak magnetic order occurs at T_M . Even in the magnetic phase, however, Ca-kapellasite has a finite T -linear term of C , which is attributed to unusual quasiparticle excitations. In magnetic fields, the T -linear term of C is suppressed by approximately 50% at 8 T, and the in-plane magnetization curve shows a convex anomaly above 20 T, at which the magnetization reaches to approximately $0.1\mu_B/\text{Cu}$. These results of bulk measurements evidence that an unconventional magnetic state is realized in Ca-kapellasite at low T and low fields. To understand this exotic magnetic state in the KAFM, magnetic properties should be investigated from a microscopic point of view.

To reveal microscopically the magnetic properties of Ca-kapellasite, we performed a ^{35}Cl NMR study. NMR spectroscopy is a powerful probe to study both the static and dynamic properties of interacting spins. We measured the ^{35}Cl NMR of a randomly oriented powder sample. The nuclear spin-lattice relaxation rate $1/T_1$ was measured at the central peak ($m = 1/2 \leftrightarrow -1/2$ transition) by the conventional saturation-recovery method. The high-field experiment at 18.9 T was carried out in the 20T-CSM magnet at the High Field Laboratory for Superconducting Materials, Tohoku University.

The ^{35}Cl NMR spectrum obtained at 100 K is shown in Fig. 1, together with the result of a simulation for the powder NMR spectrum. The experimental spectrum has a double-horn peak at 5.57 T (inset of Fig. 1), and broad satellites at 5.4 and 5.75 T. These features are consistently explained by the powder NMR spectrum for the nuclear spin $I = 3/2$. From the fitting, the quadrupole frequency and the asymmetric parameter are determined to be $\nu_Q = 2.25(9)$ MHz and $\eta = 0.4(1)$. Comparable values of ν_Q were reported for Zn-kapellasite, for which the local environment around the Cl site is similar to that for Ca-kapellasite [19]. In Zn-kapellasite, however,

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FIG. 1. ^{35}Cl NMR spectrum of the powder sample obtained at 100 K. The insets show the close view near (a) the central peak and (b) the satellite peak. The double-horn structure for the central peak and the broad satellites are consistently explained by the simulation, which is shown as the thin red line. From the fitting we found that the anisotropy of the Knight shift is small.

$\eta = 0$ for the Cl site with three nearest-neighbor Cu sites, which is natural for the Cl nuclei located on the trigonal axis. A finite η was found for Cl near the Zn-substituted sites. In Ca-kapellasite, since no Ca/Cu site exchange has been observed from the x-ray diffraction study, the finite η cannot be ascribed to the local distortion caused by the Ca substitution. Instead, we suggest that the hydrogen bonds formed between Cl and the partially occupied crystal water are responsible for the finite η . However, the asymmetry of the electric interactions is irrelevant in this study because we focus on the magnetic properties originating from the Cu kagome network. The powder NMR spectrum can be fit without introducing the anisotropy of the Knight shift, meaning an isotropic coupling between the electronic spins and the nuclear spins. With this isotropic hyperfine coupling constant, we can measure the intrinsic nuclear spin-lattice relaxation rate at the central peak of the powder spectrum.

To quantitatively discuss the magnetism of Cu 3d spins, we need to characterize the hyperfine coupling constant A_{hf} by comparing the T dependence of K and the bulk susceptibility χ . As shown in Fig. 2, the T dependence of K is scaled to that of χ in the paramagnetic state above 15 K, confirming a linear relationship of $K(T) = A_{\text{hf}}\chi(T) + K_0$. A linear fit to the K - χ plot (inset of Fig. 2) results in $A_{\text{hf}} = -375(10)$ mT/ μ_B . The absolute value of A_{hf} is smaller than that obtained in Zn-kapellasite, in which $A_{\text{hf}} = -498$ mT/ μ_B for the Cl site with three nearest-neighbor Cu sites [19]. This is because the distance between the Cl and Cu sites is shorter by 2.6%

FIG. 2. The T dependence of the Knight shift determined by the peak positions of the center line. The T dependence of K (triangles) scales to that of χ (solid line) with a negative coefficient. The inset shows the K - χ plot. From the linear relationship we determined the hyperfine coupling constant A_{hf} .

in Zn-kapellasite (2.76 Å) than in Ca-kapellasite (2.835 Å). Although an almost identical value of $A_{\text{hf}} = -380$ mT/ μ_B was reported for herbertsmithite [9], we point out that this is accidental, that is, a small transfer coupling due to the large distance (3.42 Å) is compensated by twice as many numbers of nearest-neighbor Cu sites.

In the magnetic state at $T = 3$ K, we observed a spectrum broadening at the central peak as shown in Fig. 3. The double-horn structure is smeared by the distribution of the local magnetic fields generated by the ordered moments. The spectrum width at 3 K is much narrower than that expected when the full moment is ordered. The internal magnetic field induced at the Cl site by the ordered moment of $0.1\mu_B$ was estimated using A_{hf} , and is represented in Fig. 3 by the horizontal arrow. The narrow spectrum width evidences that the ordered moment is strongly reduced. In fact, the spectrum width at 3 K is comparable to that for herbertsmithite, in which no long-range magnetic order has been reported [6,7]. We can exclude the possibility that a part of the spectrum weight is lost by the effects of the large ordered moments, because the spectrum intensity multiplied by T (inset of Fig. 3) in the ordered state is almost the same as that in the normal state.

To discuss quantitatively the static internal fields, we calculated the second moment $\sqrt{M_2} = \mu_0\sqrt{\sum(H - H_0)^2 I(H)}$, where H_0 is the gravity center of the spectrum and $I(H)$ is the normalized spectrum intensity. The T dependence of $\sqrt{M_2}$ is shown in the inset of Fig. 3. From the paramagnetic state at 20 K to the lowest T , $\sqrt{M_2}$ increases by approximately 20 mT, which indicates that the typical size of the ordered moment is $0.05\mu_B$. This reduced ordered moment leads us to suggest that the magnetic fluctuations survive even below the weak magnetic ordering at T_M . The dynamic properties of the spins will be investigated later by the $1/T_1$ measurement.

FIG. 3. Temperature variation of the central peak near T_M . The double-horn spectrum at high T becomes a single broad peak at 3 K. The horizontal arrow represents the internal field sensed by the ^{35}Cl nuclei when $0.1\mu_B$ of Cu spins are ordered. The inset shows the T dependence of $\sqrt{M_2}$ (left scale) and the intensity multiplied by T (right scale). At high T , $\sqrt{M_2}$ originates from the electric quadrupole effect on the powder spectrum. The onset T for the spectrum broadening is higher than T_M .

From the T dependence of $\sqrt{M_2}$, we note that the spectrum broadening by the short-range order increases below approximately 10 K. At the same T , the in-plane susceptibility shows an abrupt increase, which cannot be explained by the antiferromagnetic short-range order [18]. We suggest that the ferromagnetic next-nearest interaction is responsible for the spectrum broadening very close to T_M . The spectrum broadened by the weak ferromagnetic spin configuration was also observed in a ^{51}V NMR study for vesignieite [20]. In vesignieite, the onset T of spectrum broadening coincides with a weak ferromagnetic increase in the magnetic susceptibility below 13 K, which is then followed by a peak at the long-range magnetic transition T of 9 K.

Now, to study the dynamics of the correlated quantum spins, we measured $1/T_1$ at three different fields, and the results are shown in Fig. 4. At high T , $1/T_1$ converges to the same value of $1/T_1 = 15 \text{ s}^{-1}$ at all fields. We can estimate

FIG. 4. Temperature dependence of $1/T_1$ measured at 2.0, 5.6, and 18.9 T. At high T , $1/T_1$ is independent of T and fields. We observed two peaks at $T^* = 20 \text{ K}$, and $T_M = 7.2 \text{ K}$. The peak at T^* is ascribed to the short-range spin dynamics. Below T_M , a power-law T dependence was observed. This behavior is fitted to a two-term power function $A(BT + T^3)$ (see text), and the results for each field are shown as dashed lines. The inset is the field dependence of the coefficient A . The extrapolation to high fields (dotted line) suggests that the low-energy spin excitations are completely suppressed above 22 T.

the effective superexchange interaction J_{eff} using the following equations [19],

$$\frac{1}{T_1} = \frac{\sqrt{2\pi}}{3} \frac{g^2 S(S+1)}{z' \omega_e \hbar^2} (\gamma_n \hbar A_{\text{hf}})^2, \quad (1)$$

$$\omega_e^2 = \frac{2ZS(S+1)}{3\hbar^2} J_{\text{eff}}^2. \quad (2)$$

Here, $z' = 3$ is the number of Cu spins that couple to the Cl nuclear spins, and $Z = 4$ is the number of nearest-neighbor Cu sites. We averaged the anisotropic g -factor [18], and used $g = 2.06$ for this analysis. As a result, we obtained $|J_{\text{eff}}| = 30 \text{ K}$, which is comparable to the Weiss temperature $\Theta_W^{ab} = -60 \text{ K}$ and $\Theta_W^c = -55 \text{ K}$ determined from χ measured in the fields parallel to the ab and c directions, respectively.

The T dependence of $1/T_1$ shows two peaks at $T^* = 20 \text{ K}$ and $T_M = 7.2 \text{ K}$. The peak at T^* is ascribed to the development

of the short-range spin dynamics, for which the characteristic time scale progressively slows down at temperatures lower than the energy scale of J_{eff} . The peak in $1/T_1$ is observed when the time scale for the slow spin dynamics becomes comparable to the NMR frequency. The gradual development of the short-range spin dynamics is also observed from the bulk measurements as the broad hump in C/T at 12 K and the abrupt increase in χ below 10 K [18]. The peak of $1/T_1$ is suppressed in high magnetic fields, although the dominant J_{eff} is antiferromagnetic. We think that the ferromagnetic component of the next-nearest interaction is suppressed in the fields. A similar peak behavior was observed in herbertsmithite at approximately 50 K. This peak was first interpreted as being caused by the slow dynamics of the OH bonds [9], and in the later works by the defect Cu spins at the Zn intersites, which couple to the intrinsic kagome plane [12,21]. We note that the ratio of the peak T between herbertsmithite and Ca-kapellasite is comparable to the ratio of J_{eff} ($J_{\text{eff}} \simeq 170$ K for herbertsmithite [9]). This result indicates that the high- T peak behavior originates from the intrinsic magnetic interactions.

The peak at 7.2 K is attributed to the long-range magnetic order. However, the peak height is much smaller than that typically observed in canonical antiferromagnets, for which $1/T_1$ diverges at the transition T because of the critical slowing down of the magnetic fluctuations. The peak is small because the size of the ordering moments is strongly reduced by quantum fluctuations. The peak height at T_M is further suppressed in high magnetic fields, while keeping the peak T almost unchanged. The field independent T_M is consistent with the results of previous bulk measurements [18], which can also be explained by the small size of the ordered moments.

Below T_M , $1/T_1$ shows a power-law T dependence with an exponent $d = 2.3$, when the result is fitted to a single component power function. The power-law T dependence was observed in all the external fields, although the absolute values decrease in accordance with the suppression of the peak at T_M . The exponent found in Ca-kapellasite is larger than that in herbertsmithite, in which d was determined as 0.47 at 8.3 T in the T range between 2 and 30 K [9]. A steeper T dependence was observed from the ^{17}O NMR experiment below 1 K [22]. This low- T behavior resembles what we observed below T_M in Ca-kapellasite.

To understand the power-law T dependence of $1/T_1$, we assume that the nuclear spin relaxation is caused by magnetic excitations, for which the density of states at an energy ϵ is expressed by $\rho(\epsilon)$. The power-law T dependence of $1/T_1$ originates from the power-law energy dependence of $\rho(\epsilon)$ at the low-energy limit. Since the heat capacity measurement revealed the T -linear and T^2 (spin-wave) terms [18], we introduce two terms $\rho_1(\epsilon) = \text{const}$ and $\rho_2(\epsilon) \propto \epsilon$. As NMR spectroscopy can mainly detect magnetic fluctuations at low temperatures, the lattice contributions observed in the heat capacity measurement (T^3 and T^5 terms) are irrelevant. Thus the T dependence of $1/T_1$ derived from these $\rho(\epsilon)$ has two terms, which are $1/T_1 = A(BT + T^3)$. The relative weight B was determined by fitting the result at 18.9 T as $B = 5.84$. The good fit to the $1/T_1$ data validates the observation of the T -linear term by the heat capacity measurement, and suggests that gapless excitations survive below T_M .

In the inset of Fig. 4, we show the field dependence of the coefficient A , which was determined by fitting the results to the above function. The coefficient is suppressed in high magnetic fields, and extrapolates to zero at approximately 22 T. This means that the low-energy excitations are completely suppressed above 22 T. Around this field, the in-plane magnetization shows a convex anomaly, which is indicative of a modification in the magnetic structure [18]. From the coincidence between the suppression of the low-energy excitations and the modification in the magnetic structure, we suggest that the magnetic structure at low fields is crucial for low-energy gapless excitations.

On the basis of these observations, we speculate that the possible magnetic structures are coplanar or fan at low fields and collinear at high fields. With this model, the convex anomaly in the magnetization curve can be explained, because the ferromagnetic response becomes smaller in the collinear spin state than that in the coplanar spin state. This mechanism is similar to that for the $1/3$ magnetization plateau theoretically predicted for quantum spin systems [23], although in Ca-kapellasite, the anomaly in the magnetization curve appears at approximately $0.1\mu_B/\text{Cu}$.

For the KAFM with next- and third-nearest-neighbor interactions, which is the case for kapellasites [24], theoretical studies have suggested several types of ground states including the long-range ordered states [25–29]. Such farther nearest-neighbor interactions are mediated by the Cu-metal ion-Cu path, as in kapellasites the metal ions are located at the center of the Cu hexagon. In contrast, in herbertsmithite the farther nearest-neighbor interaction is small because Cl is located at the center of the Cu hexagon. The intriguing character of Ca-kapellasite is the antiferromagnetic nearest-neighbor interaction. In this case, a theoretical study predicts that several classical ground states appear in a narrow parameter region in the phase diagram [30]. We expect that unusual magnetic excitations will be generated in such a situation by the effect of strong quantum fluctuations.

To conclude, we have performed a ^{35}Cl NMR experiment for the kagome antiferromagnet Ca-kapellasite, and revealed two anomalies in the T dependence of $1/T_1$. We interpreted that the short-range spin dynamics develops below $T^* \simeq 20$ K, and the long-range magnetic transition occurs at $T_M = 7.2$ K. However, the ordered moment, which was estimated from the spectrum width, is strongly reduced. Below T_M we observed a power-law T dependence of $1/T_1$, from which we conclude that gapless magnetic excitations survive below T_M . The unusual magnetic excitations are suppressed by high magnetic fields. We suggest that a spin configuration, which is stabilized at low magnetic field, is crucial to generate gapless magnetic excitations.

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