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Spin dynamics in the two-dimensional spin system $SrCu_2(BO_3)_2$

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Abstract

Muon spin relaxation measurements have been performed in the two dimensional spin system $SrCu_2(BO_3)_2$. As the temperature decreases, the relaxation rate increases below $T = 10$ K, and shows a saturation below $T = 3$ K. The line shape at low temperatures is nearly Gaussian, yet it cannot be easily decoupled by an application of longitudinal field. These results indicate the existence of magnetically active spins that show slowing down of fluctuations with decreasing temperatures, yet remain dynamic even at 0.1 K. We have also examined a sample doped with small amount of non-magnetic Zn^{2+} impurity, $Sr(Cu_{0.99}Zn_{0.01})_2(BO_3)_2$. The result was quite similar to the pure sample. Thus, anomalous behavior of the pure sample observed in low temperatures is not due to magnetic moments induced by impurities or a lattice defect.

Keywords: Spin singlet ground state; Two dimensional system; Dimer; $SrCu_2(BO_3)_2$

1. Introduction

Two dimensional (2D) quantum spin systems are quite interesting, not only from the viewpoint of magnetism but also of superconductivity. In 2D Heisenberg spin systems, long range order is established only at zero temperature [1,2]. In most real materials with layered magnetic structure, three dimensional (3D) long range order is formed at low temperatures due to small interactions between layers. Only a few 2D materials with a singlet ground state have been found. $SrCu_2(BO_3)_2$ provides a new type of 2D spin-gap system and show quite unique behavior [3].

$SrCu_2(BO_3)_2$ is a layered material with a 2D $CuBO_3$ plane. The arrangement of Cu^{2+} spins in that plane is topologically equivalent to the Shastry–Sutherland model [4,5], which is a 2D Heisenberg square lattice with alternating diagonal

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interactions. The susceptibility of $SrCu_2(BO_3)_2$ shows a maximum at ~ 15 K and decreases toward zero below ~ 15 K, which suggests a non-magnetic singlet ground state [3]. The spin gap is evaluated to be ~ 34 K [3,6]. Theoretical and experimental studies have been extensively performed, and it is considered that the ground state is an exact singlet dimer state though the spins are strongly connected in the 2D plane.

In order to study spin dynamics of $SrCu_2(BO_3)_2$, we performed muon spin relaxation (μSR) measurements. We also examined a sample doped with small amount of non-magnetic Zn^{2+} impurity, $Sr(Cu_{0.99}Zn_{0.01})_2(BO_3)_2$.

2. Experimental

We used powder samples of pure $SrCu_2(BO_3)_2$ and a sample doped with Zn, $Sr(Cu_{0.99}Zn_{0.01})_2(BO_3)_2$, for μSR measurements. The measurements were carried out using a surface muon beam at TRIUMF in Canada. We used a He-flow type cryostat and a dilution refrigerator. Time spectra were measured down to 100 mK.

3. Results and discussion

We show the temperature dependence of the time spectrum in zero field (ZF) in Fig. 1. As the temperature decreases below ~ 10 K, the relaxation becomes fast. Below ~ 3 K, the time spectrum is almost independent of temperature.

The shape of the time spectra and the decoupling behavior are quite anomalous. The details are described later. At present, we have not found a reasonable function explaining the relaxation in both of ZF and longitudinal fields. Thus, we define a half-width of the half-maximum (HWHM) of the time spectra, $T_{1/2}$, whose inverse reflects the relaxation rate. We plotted $1/T_{1/2}$ as a function of temperature in Fig. 2. As the temperature decreases, $1/T_{1/2}$ starts to decrease at ~ 10 K. It shows an anomaly at ~ 3 K and then remains nearly constant down to 0.1 K.

Thus, we see the existence of some magnetically active spins in a nominally pure $SrCu_2(BO_3)_2$ at low temperatures. This enhancement of relaxation rate shows the slowing down of fluctuations of electronic spins. Since the ground state is considered to be non-magnetic, the muon spin should not relax, at least, at low temperatures. Since the μSR method is quite sensitive to small magnetic moments, μSR might detect intrinsic magnetic properties of samples which cannot be detected by other methods. Impurity and probe effects are also suggested as a possibility of origin of anomalous enhancement [7,8]. The origin of the anomalous spin freezing has not yet been clarified.

To study whether a small amount of impurity affects the behavior, we examined a sample doped

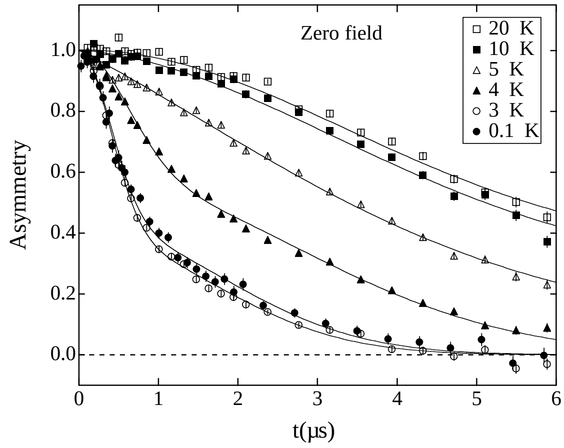

Fig. 1. Temperature dependence of time spectra in zero field. The solid lines are guides for the eyes.

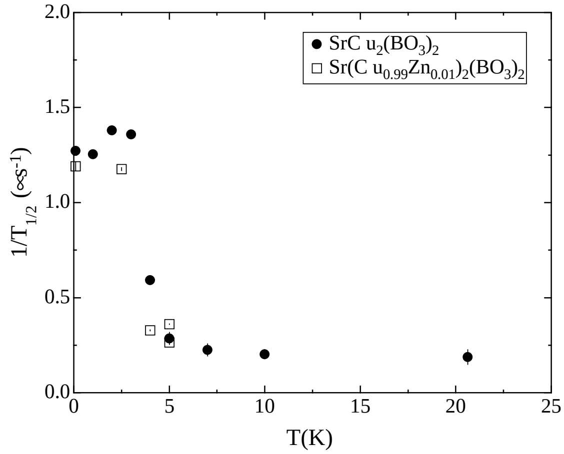

Fig. 2. Temperature dependence of $1/T_{1/2}$ in zero field.

with 1% Zn^{2+}. The susceptibility is almost identical to that in the pure sample, though the increase of the susceptibility at low temperatures is slightly larger than that in the pure sample. This suggests that the ground state is still a spin singlet state, but there are quasi-free spins induced near the doped Zn^{2+} ions. The results of the μSR measurements were almost the same as for the pure sample. We have plotted $1/T_{1/2}$ of the time spectra for the doped sample in Fig. 2. This result suggests that the origin of the enhancement of the relaxation rate observed in the pure sample is not due to Cu^{2+} moments induced by impurities or lattice defects.

The temperature dependence of $1/T_{1/2}$ in Fig. 2 shows an anomaly at $T \sim 3$ K. It seems to suggest some phase transition or spin freezing. However, the behavior is quite different from generally observed phase transition or spin freezing. In general phase transitions or spin freezing, the time spectrum relaxes exponentially above transition or freezing temperatures. In $SrCu_2(BO_3)_2$, the shape of the time spectrum at early times in ZF is Gaussian-like over the whole temperature region examined. Gaussian-like line shapes generally result from quasi-static local fields.

The most noticeable point is the decoupling behavior by a longitudinal field (LF). We show the LF dependence of time spectra measured at 0.1 K in Fig. 3. The long time tail shows only slight relaxation and shifts upward (decouples) with increasing LF. The time spectrum at early time is independent of LF. All of these facts are characteristic of decoupling behavior of quasi-static local fields. Assuming that the local field is static and the distribution of the magnitude is Gaussian (not Maxwellian), the value estimated from the time spectrum at $t < 1$ μs is ~ 30 G. This value of the static field should be decoupled by a small LF. However, even in a LF of 2 kG, the time spectrum shows relaxation. Thus, the value of the local field estimated from the LF dependence is much larger than the one estimated from the shape of the time spectra at early times, it is an 'undecouplable Gaussian'. This tendency is observed in the time spectra even at $T \sim 4$ K.

Such anomalous 'undecouplable Gaussian' relaxation is observed in several systems with

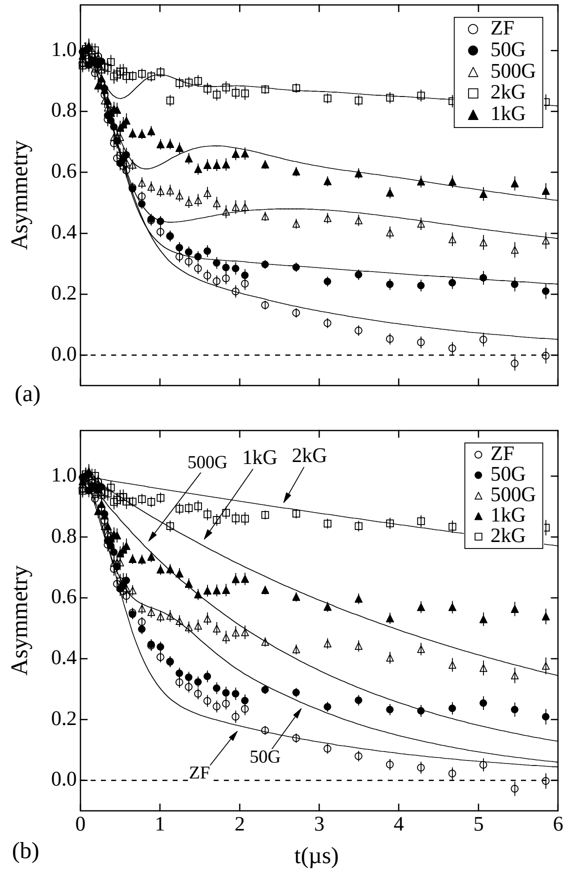

Fig. 3. Longitudinal field dependence of time spectra measured at $T = 0.1$ K. (a) The solid lines are the result of fitting by Eq. (3) with $f_{LF} = 0.034$. (b) The solid lines are the results of fitting by Eq. (3) without multiplying H_{LF} by f_{LF} ($f_{LF} = 1$).

geometrically-frustrated exchange interactions, such as *kagomé*-lattice $SrCr_8Ga_4O_{19}$ [9], the heavy fermion compound $CePt_2Sn_2$ [10], and the 1D spin system Y_2BaNiO_5 with charge doping [11]. The details of this line shape have not yet been understood. In these materials, the decoupling behaves as if the efficiency of the applied field were multiplied by a numerical factor $f_{LF}(0 < f_{LF} < 1)$, which is independent of the applied field [10].

We analyzed the data by a similar method to Ref. [10]. In $SrCu_2(BO_3)_2$, we assumed that the distribution of the magnitudes of the static local fields H_s is Gaussian,

$$P(H_s) = \frac{\sqrt{2}}{\sqrt{\pi}\sigma} \exp\left(-\frac{H_s^2}{2\sigma^2}\right), \qquad (1)$$

where σ is the width of the Gaussian distribution. This distribution is different from $P(H_{x,y,z}) = (1/\sqrt{2\pi}\sigma)\exp(-H_{x,y,z}^2/2\sigma^2)$, which is assumed in the Kubo-Toyabe function [12]. The time spectrum of powder samples with static local fields is represented by

$$G_{\mathrm{stat}}(t, H_{\mathrm{LF}}) = \frac{1}{2}\int_0^{\pi} \mathrm{d}\theta \int_0^{\infty} \mathrm{d}HP(H_{\mathrm{s}}) \times \frac{(H_s\cos\theta + H_{\mathrm{LF}})^2 + H_s^2\sin^2\theta\cos(\gamma_\mu\sqrt{H_s^2 + 2H_s\cos\theta H_{\mathrm{LF}} + H_{\mathrm{LF}}^2 t})}{H_s^2 + H_{\mathrm{LF}}^2 + 2H_sH_{\mathrm{LF}}\cos\theta} \quad (2)$$

We approximate the relaxation function by a product of a static function $G_{\mathrm{stat}}(t, H_{\mathrm{LF}})$ and a dynamical function $\exp(-\lambda(H_{\mathrm{LF}})t)$. Since decoupling behaves as if $f_{\mathrm{LF}}H_{\mathrm{LF}}$ is applied instead of H_{LF}, the fitting function is

$$G_z(t, f_{\mathrm{LF}}H_{\mathrm{LF}}) = \exp(-\lambda(H_{\mathrm{LF}})t) \times G_{\mathrm{stat}}(t, f_{\mathrm{LF}}H_{\mathrm{LF}}). \quad (3)$$

The solid lines in Fig. 3(a) are the results of fitting. The value of σ was common in all LF. From the fitting, we obtained $\sigma = 28$ G and $f_{\mathrm{LF}} = 0.034$, and the LF dependence is successfully reproduced. For comparison, we also fit the time spectra by Eq. (3) without multiplying H_{LF} by f_{LF} ($f_{\mathrm{LF}} = 1$). The decoupling behavior cannot be reproduced at all as shown in Fig. 3(b).

At present, we consider that the 'undecouplable Gaussian' results from dynamically fluctuating fields. If the fluctuations of $SrCu_2(BO_3)_2$ are general thermal fluctuations that result in spins fluctuating randomly in time, the shape of the time spectrum should be exponential-like. Even if some conventional distribution of the magnitude of the fluctuating field or fluctuating time is assumed, the Gaussian-like line shape cannot be reproduced. One of the anomalous points of the experimental results in $SrCu_2(BO_3)_2$ is that the relaxation rate remains constant below ~ 3 K down to 0.1 K. These results may suggest that the fluctuations are not general thermal fluctuations but rather have a quantum origin.

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