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Spin dynamics in the quantum spin system $KCu_5V_3O_{13}$

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Abstract

The complex Oxo-Cu-vanadate $KCu_5V_3O_{13}$ with 5 independent Cu-sites ($s = \frac{1}{2}$) per unit cell has an exchange topology described as a ladder of spin tetrahedra or triangles. Magnetic susceptibility measurements on single crystals show a steplike anomaly at 213 K and an antiferromagnetic transition at 7.5 K. ZF μSR reveals only nuclear relaxation above 10 K and two spontaneous precession signals in the long range ordered regime. Below 3.7 K, a spin reorientation leads to a broad frequency distribution typical for an incommensurate spin structure.

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Keywords: Low-dimensional quantum spin system; Spin ladder

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1. Introduction

The magnetic properties of low dimensional cuprates and vanadates have been studied intensively in recent years as they represent quantum spin systems with interesting properties. For some exchange topologies that are realized in these systems, peculiar phase diagrams are postulated that include short-range and long-range ordered phases as functions of exchange coupling parameters or doping. These phases are divided by quantum critical points. Of particular importance are systems that realize competing interactions, e.g. spin frustration due to antiferromagnetic exchange on triangular or tetrahedral lattices. Systems with ladder topologies are also interesting, as they may have spin liquid ground states that are predisposed to unconventional electronic and magnetic instabilities [1,2].

In the course of investigating the complex Oxo-Cu-vanadates $Cu_xV_2O_{5+x}$ with a linkage of differently coordinated chains of CuO_5 pyramids and VO_4 tetrahedra, we studied the magnetic properties of the related compound $KCu_5V_3O_{13}$. While the former systems form effectively one-dimensional chain or more complicated dimerized plane structures, the latter compound consists of ladders of Cu^{2+} $s = \frac{1}{2}$ triangles or tetrahedra [3]. It

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was the purpose of this study to investigate the magnetic properties of $KCu_5V_3O_{13}$ using nuclear probes and other experimental techniques to gain a better understanding of the effect and interplay of frustration with anisotropies in a quantum spin system.

2. Magnetic susceptibility and specific heat

Single crystals of $KCu_5V_3O_{13}$ show a Curie–Weiss-like magnetic susceptibility for $T > 213$ K with a $\theta_{CW} = -145$ K pointing to antiferromagnetic correlations. At $T^* = 213$ K a jump in the susceptibility is observed (Fig. 1). The magnitude and sign of this jump depend on the field orientation with respect to the crystallographic axes. In measurements on polycrystalline samples this effect is totally nullified. Therefore it must be related to anisotropies or orbital degrees of freedom. The respective anomaly in the specific heat is shown in the inset of Fig. 1.

Low temperature magnetic susceptibility and specific heat measurements give evidence for magnetic instabilities at 7.5 and 3.7 K. The related entropies at these temperatures are evident, but smaller than expected. Susceptibility and high field magnetization of polycrystalline $KCu_5V_3O_{13}$ have been reported and are in rough agreement with our data [4]. The transitions at 3.7 and 213 K have not been found.

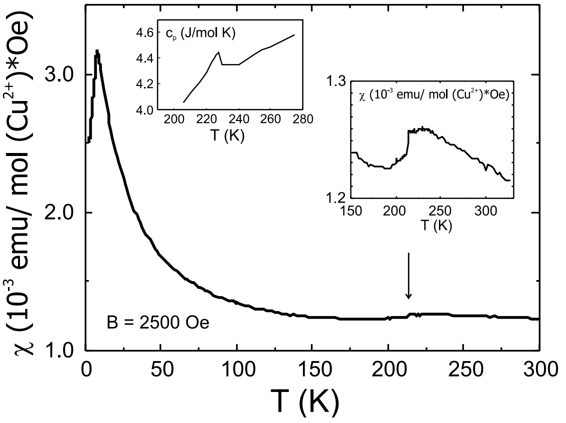

Fig. 1. Magnetic susceptibility of $KCu_5V_3O_{13}$ in $B = 2500$ Oe perpendicular to the crystallographic a-axis. Right inset: high temperature anomaly at 213 K. Left inset: high temperature specific heat of $KCu_5V_3O_{13}$ ($B = 0$).

3. Nuclear probes

We performed zero field μSR experiments on a polycrystalline sample of $KCu_5V_3O_{13}$ in the temperature range from 1.7 to 250 K. Between room temperature and 10 K only a static gaussian relaxation due to nuclear moments and no electronic relaxation is observed (Fig. 2). Therefore no slow spin fluctuations occur in this temperature range.

Below 7.5 K a static internal field distribution of electronic origin is found (Fig. 3). Between 7.5 and 3.8 K an isotropic internal field distribution with two well defined muon precession frequencies with equal amplitude are observed. Both signals can be described as exponentially-relaxing precession,

$$P_\mu(t) = \tfrac{2}{3}\big(\tfrac{1}{2}\cos(2\pi\nu_1 t + \phi_1)\,e^{-\lambda_{T1}t} + \tfrac{1}{2}\cos(2\pi\nu_2 t + \phi_2)\,e^{-\lambda_{T2}t}\big) + \tfrac{1}{3}\,e^{-\lambda_L t}. \quad (1)$$

The narrow line shape (Fig. 4) proves a commensurate long range ordered spin system in this temperature range. For temperatures below 3.7 K, the internal field distribution is changed qualitatively. A strong asymmetric broadening of the two signals is observed. The signals are described by first-order Bessel functions j_0 which have been derived for an incommensurate modulation vector

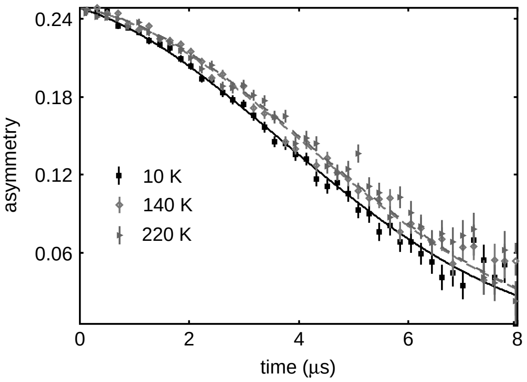

Fig. 2. Zero field muon spin relaxation spectra of $KCu_5V_3O_{13}$ at 10, 140 and 220 K.

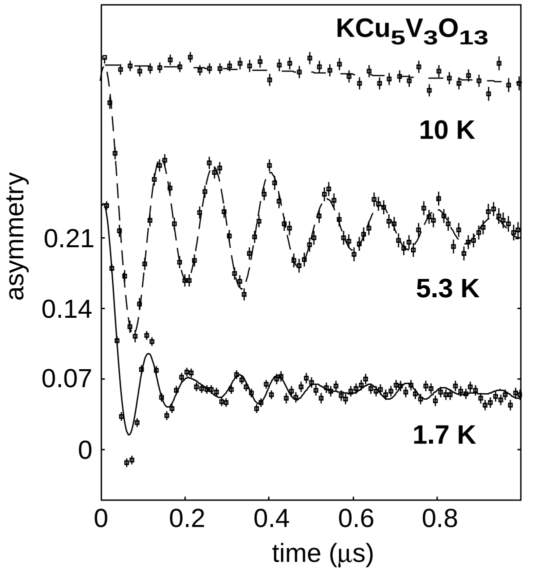

Fig. 3. Low temperature zero field muon spin relaxation spectra of $KCu_5V_3O_{13}$. Spectra at 5.3 and 10 K are shifted vertically for clarity. Lines are fits using the different models described in the text.

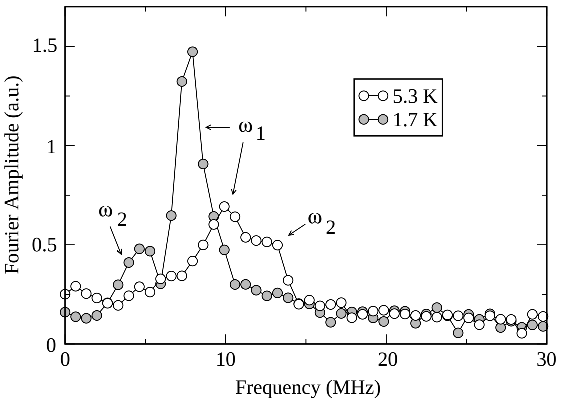

Fig. 4. Fourier spectra of $KCu_5V_3O_{13}$ below and above 3.7 K showing a significant line broadening at 1.7 K with respect to the data at 5.3 K.

of the local field at the muon site [5]:

$$P_\mu(t) = \tfrac{2}{3}\left(\tfrac{1}{2} j_0(2\pi\nu_1 t + \phi_1)\, e^{-\lambda_{T1} t} + \tfrac{1}{2} j_0(2\pi\nu_2 t + \phi_2)\, e^{-\lambda_{T2} t}\right) + \tfrac{1}{3}\, e^{-\lambda_L t}. \qquad (2)$$

The onset temperatures of both precession signals coincide with the ordering temperature derived from susceptibility and specific heat (Fig. 5). This proves a homogeneous sample with static magnetic order observed at two magnetically inequivalent muon sites. The change of the line shape and the step-like increase of the precession frequency ν_2 are interpreted with a spin reorientation to an incommensurate ordered phase. To draw more specific conclusions on the magnetic structures Knight shift experiments on a single crystal are needed to identify the magnetically inequivalent muon sites in the lattice.

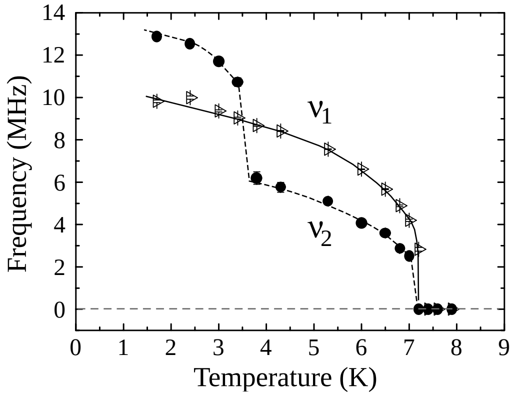

Fig. 5. Spontaneous muon spin precession frequencies in $KCu_5V_3O_{13}$.

^{51}V-NMR measurements in low (≈1 T) and high (≈8 T) external field reveal a temperature independent hyperfine coupling in the paramagnetic temperature regime above 10 K. This result supports the dynamic nature of the switching phenomenon observed in the susceptibility. The magnitude of the hyperfine coupling constant points to a spin singlet pairing of four Cu spins and one additional unpaired Cu spin per unit cell.

4. Conclusions

The Oxo-Cu-vanadate $KCu_5V_3O_{13}$ shows a coexistence of unpaired and dimerized Cu spin-$\frac{1}{2}$ spin species and a complex and low symmetry exchange topology. This is proposed to lead to the anisotropic switching phenomena in the magnetic susceptibility between 20 and 220 K. The zero field μSR experiments reveal no electronic relaxation above 7.5 K, confirming the dynamic nature of the magnetization switching. Below 7.5 K, two well-defined spontaneous muon spin precession signals confirm long range magnetic order. At 3.7 K a strong broadening of the spectrum is interpreted as a spin reorientation. The discrepancy between the large Curie–Weiss temperature and the small ordering temperatures are taken as evidence for a considerable competition of magnetic exchange processes. These are compatible with a local singlet formation of a part of the spins.

Acknowledgements

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