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Physica B 374–375 (2006) 138–141
PHYSICA B
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Low-T dynamics in the highly frustrated $S=\frac{3}{2}$ kagomé bilayers: A phenomenological function for a spin liquid state?

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Abstract

A μSR study of the spin dynamics of the archetypes of the highly frustrated magnets, $SrCr_{9p}Ga_{12-9p}O_{19}$ and $Ba_2Sn_2ZnGa_{10-7p}Cr_{7p}O_{22}$, is summarized. Especially, low dilutions of the magnetic network could be achieved and we take advantage of the close similarity between these kagome bilayers to single out their typical properties. The phenomenological model for the μ^+ relaxation, based on sporadic dynamics due to spin excitations in a singlet sea, proposed by Uemura et al., is extended to all fields, temperature and defects range. Its connection to a RVB picture is discussed.

PACS: 75.50.Lk; 75.40.Gb; 76.75.+i

Keywords: Kagomé; Spin liquid; Dynamics; Spinons

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1. Introduction

Geometric frustration in magnetism (GFM) has proven to yield various original ground states in the past decade, from RVB "spin liquids", to exotic freezings as spin ice [1]. In particular, the ground state of the kagomé lattice with $S=\frac{1}{2}$ Heisenberg spins and near neighbor (*nn*) antiferromagnetic interactions is expected to be the long sought resonating valence bond (RVB) state [2] also advocated in many fashionable fields of condensed matter beyond GFM, such as high-T_c cuprates or recently discovered cobaltates [3]. In this context, longitudinal field (LF) μSR has proven to be quite adequate and unique to probe spin dynamics. Only recently, the first spin echo experiments could bring some complementary information.

Among all the "highly frustrated magnets," the $S=\frac{3}{2}$ kagomé bilayers $Ba_2Sn_2ZnGa_{10-7p}Cr_{7p}O_{22}$ [BSZCGO(p)], and the isostructural $SrCr_{9p}Ga_{12-9p}O_{19}$ [SCGO(p)] [4], are ones of the few good candidates as they feature an ideal Heisenberg lattice with *nn* couplings. Although the RVB ground state has only been discussed theoretically for $S=\frac{1}{2}$, the physics of these systems retains the essential signatures of the RVB state: beyond the absence of ordering well below the Curie–Weiss temperature [5], the unusual large value of the specific heat and its field independence are consistent with the predicted continuum of low lying excitations [6]. The ground state is found essentially fluctuating as proven by neutron experiments, and μSR for SCGO(p) [7,8], although an intrinsic spin glass (SG) signature is observed at $T_g \approx 1.5$ K for BSZCGO and 3.5 K in SCGO in macroscopic susceptibility measurements. In itself this SG freezing remains a puzzle and T_g is found to weakly decrease upon dilution of the magnetic network [4,9].

We present here recently published LF μSR experiments [9] in BSZCGO(p) and also SCGO(p) for recently achieved

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doi:10.1016/j.physb.2005.11.035

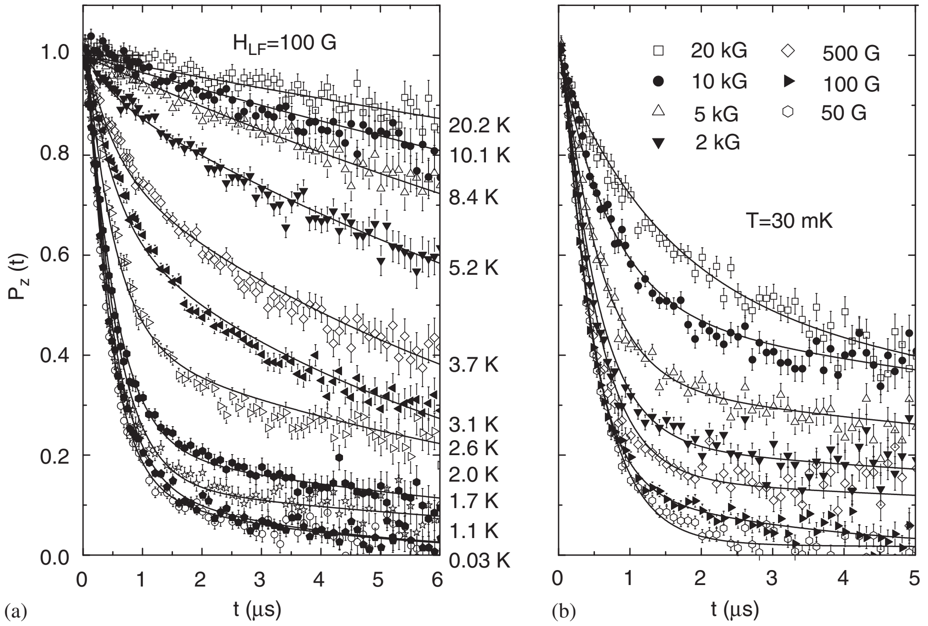

Fig. 1. (a) H_{LF}- and T-dependence of the μ^+ polarization $P_z(t)$ in BSZCGO(0.97). The lines are fits with Eq. (1).

low dilutions in comparison to the seminal work of Uemura, Keren et al. [7,8]. Only dilution rates ($0.7<p\leqslant 0.97$) above the percolation threshold are discussed in this paper and we put the emphasis on the phenomenological depolarization function we proposed in Ref. [9]. We relate it to the essence of the RVB ground state picture. A careful control of the Cr content could be achieved through X-ray checks of the absence of impurity phases after preparation (accuracy ~3%) and NMR control of Ga/Cr substitution in SCGO. In addition to dilution defects, magnetic bond disorder was found in BSZCGO [5,10].

2. Experimental results

We used a conventional LF setup, with z the axis of the initial μ^+ spin polarization, noted P_z. $P_z(t)$ is presented in Fig. 1(a) for BSZCGO(0.97) at various T. It is found to be qualitatively the same in SCGO(p) and BSCGO(p) for $p>0.7$. A weak LF $H_{\mathrm{LF}}=100\,\mathrm{G}$ was used to decouple the static nuclear Kubo–Toyabe contribution. For $T>5\,\mathrm{K}$, an exponential decrease of the polarization, typical of dense paramagnetic systems, is evidenced. Below 5 K, the relaxation rate increases by more than two orders of magnitude to reach a T-independent value for $T<T_{\mathrm{g}}$. For $T=0.03\,\mathrm{K}$, $P_z(t)$ displays a shape in between exponential and Gaussian at early times and reaches zero value at long times. It also displays a much weaker H_{LF}-dependence (Fig. 1(b)) than for a frozen magnetic state ($H_{\mathrm{LF}}\sim 500\,\mathrm{G}$ would completely decouple $P_z(t)$). This undecoupled Gaussian is the typical signature of an unconventional dynamical magnetic state at low T, following the arguments initially developed for SCGO(0.89) [7]. On the contrary, we find more "conventional" dynamics, i.e. paramagnetic-like, for $p<0.6$ in both SCGO(p) and BSZCGO(p) [9].

3. Sporadic relaxation function

One of the challenges in the analysis of a μSR experiment is to derive an appropriate relaxation fitting function *further* supported by a relevant physical picture. Here, and more generally for the kagomé frustrated antiferromagnets, one has to find a theoretical model reproducing $P_z(t)$ for *all* fields, *all* temperatures, and *all* dilution rates $(1-p)$ of the magnetic network. We derive it from the $S=\frac{1}{2}$ RVB framework and conjecture that such picture could extend to the $S=\frac{3}{2}$ case. To induce some relaxation of the muon, one needs to create unpaired spins from the resonating RVB singlets. Theoretical works demonstrate that such excitations can be ascribed to unconfined spinons, for which the location of spin $\frac{1}{2}$ vary in time with no loss of coherence of the excited state [2]. A "step by step" sketch for these spinon excitations is given in Fig. 2. Within the RVB ground state, only resonating singlets are present (a). If the energy provided by the environment is larger than the singlet–triplet gap, calculated to be $\sim J/20$ for $S=\frac{1}{2}$ and even possibly smaller [2], $S=\frac{1}{2}$ spins may be created, by first "breaking a singlet" (b). Then, the resonance of the singlets, defining the RVB state, authorizes each unpaired spin to move independently from each other in the background of a "singlet sea" (c and d). After a given coherence time the process is re-initialized (a).

Two different situations can therefore occur for the muon. (i) When a μ^+ is surrounded by singlets, its spin senses H_{LF} only, and therefore $P_z(t)$ remains constant. (ii) When an unpaired spin comes on the nearby sites, it induces a paramagnetic-like depolarization, given by the dynamical Kubo–Toyabe function $P_z^{\mathrm{DKT}}(t, \Delta, H_{\mathrm{LF}}, \nu)$. Δ/γ is the average field created on a μ^+ site by one *nn* unpaired spin, and ν is the on-site fluctuation frequency of the paramagnetic-like unpaired spin. Finally a variable *f*, related to the hopping frequency of an unpaired spin from one magnetic site to another, has to be introduced, and accounts for the effective average time *ft* a μ^+ spin suffers a fluctuating internal field.

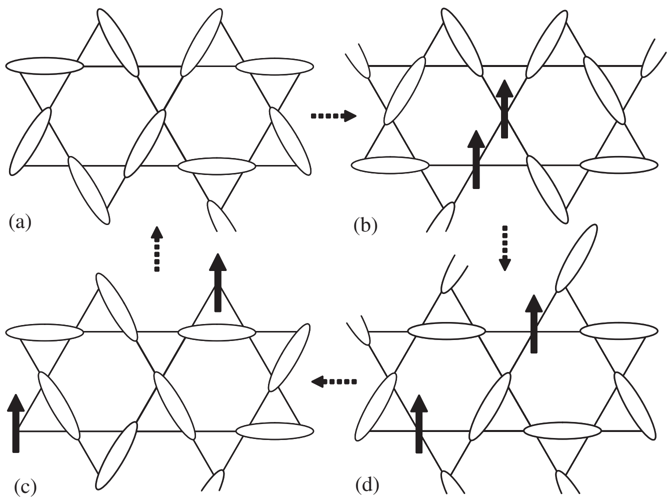

Fig. 2. Sketch of unconfined spinon excitations for the RVB state on the kagomé lattice.

As initially proposed, after a time *t*, the average muon polarization is finally $P_z^{\mathrm{DKT}}(ft, \Delta, H_{\mathrm{LF}}, \nu) = P_z^{\mathrm{DKT}}(t, f\Delta, fH_{\mathrm{LF}}, f\nu)$ [7]. The LF is effectively reduced to a value fH_{LF}. This function accounts for the weakness of the field dependence, the Gaussian initial decrease and the absence of the $\frac{1}{3}$ tail in $P_z(t)$ at low-*T* in SCGO(0.89) [7]. Yet, this sporadic relaxation function cannot fit the long times tail ($t > 3\,\mu$s) for all fields and all *T* and our major input is that a more conventional Markovian relaxation needs to be introduced to fit the long times tail ($t > 3\,\mu$s) for all fields and all *T* [9]. We therefore propose the phenomenological fitting function

$$P_z(t) = xP_z^{\mathrm{DKT}}(ft, \Delta, H_{\mathrm{LF}}, \nu) + (1 - x)\mathrm{e}^{-\lambda t}. \tag{1}$$

Since we use a *sum* of two different functions, this model implies that two different muon sites are distinguishable. The first ones, which proportion is *x*, were discussed above and are associated with the spinons dynamics. The second ones have *always* a *nn* paramagnetic-like spin.

In order to limit the number of free parameters, we make the minimal assumption that the external field does not influence the dynamics (ν, *f*) of the coherent spinon term, also taken identical in the very similar lattices SCGO(*p*) and BSZCGO(*p*) for equivalent *p*. Δ, related to the μ^+ location in the unit cell, and ν are shared for all *p* and H_{LF} and are set by our low field data. Δ is expected to vary from BSZCGO to SCGO. *f* is adjusted for each *p* and its variation accounts for the evolution of the initial decrease

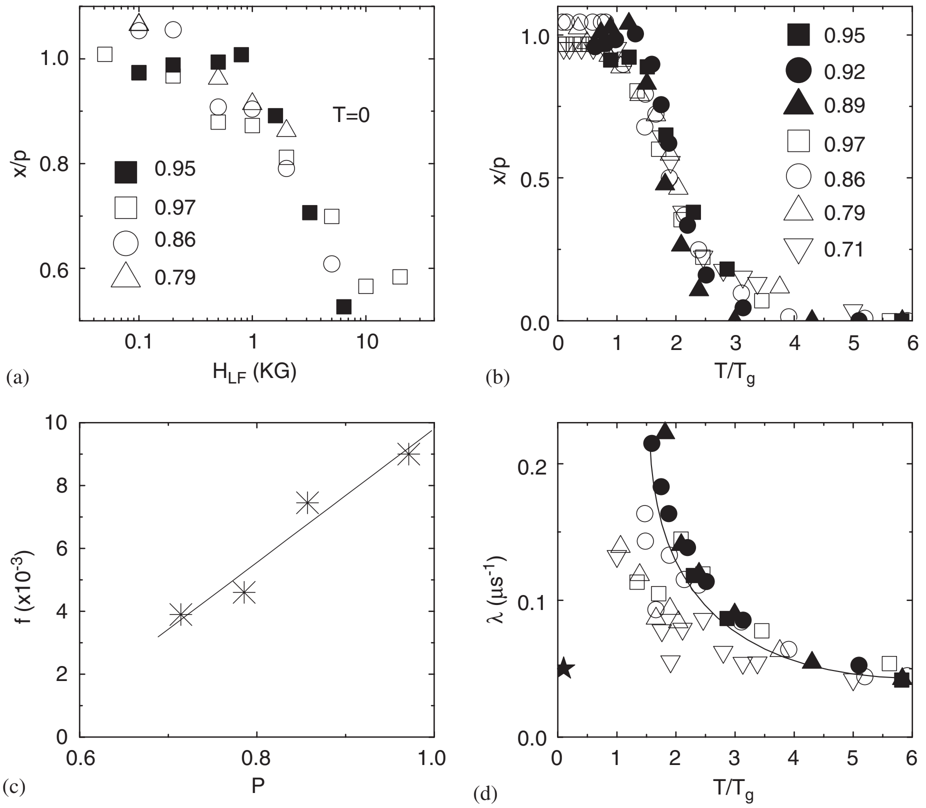

Fig. 3. Fitting parameters of Eq. (1) for BSZCGO and SCGO (open and full symbols). *f* is common for both systems, with $f(1) \approx 0.01$. For low fields, $x \sim 1$ makes λ a non-relevant fitting parameter. The black star in (d) was obtained while fitting the low-*T* H_{LF}-dependence. The lines are guide to the eye.

of $P_z(t)$, related to $f\Delta$. Then, x and λ are obtained for all H_{LF} and T [9].

We find perfect fits of our data (Fig. 1) with $\nu \sim 1000\,\mu\mathrm{s}^{-1}$ and $\Delta \sim 350\,\mu\mathrm{s}^{-1}$ for BSZCGO(p) ($\Delta \sim 1200\,\mu\mathrm{s}^{-1}$ for SCGO(p)) in agreement with previous work on SCGO(0.89) [7] and with high-T approximation [9]. The other parameters are displayed in Fig. 3.

Spin vacancies induced effects: At low T and low fields, x is of the order of p (Fig. 3(a,b)). Within our model, it means that we find the same fraction of μ^+ located near a conventional paramagnetic-like spin as the fraction of spin vacancies, which suggests that *one* spin vacancy disturbs the RVB state around it and releases *one spatially fixed* spin. This could be related qualitatively to the predicted enhancement of the correlations around a spin vacancy in the kagomé lattice [11], which would destroy locally the liquid state. Moreover, we observe a linear variation of f with p (Fig. 3(c)) and f tends to vanish around $p \sim 0.5$. This indicates that even far from the substituted sites, the coherent state is somehow affected, e.g. the density of spinons could be smaller.

Energy scale(s): The weight of the sporadic term decreases appreciably for $H_{\mathrm{LF}} \sim 10\,\mathrm{kG}$ whatever the value of p, corresponding to an energy scale $\sim 1\,\mathrm{K} \sim T_{\mathrm{g}}$. The same energy scale is found in the T-dependence, with an expected decrease of x between T_{g} and $3T_{\mathrm{g}}$. Above, it finally enters a high-T regime (Fig. 3(b)), with a conventional exponential μ^+ relaxation ($x = 0$), consistent with the "cooperative paramagnetic state" measured in susceptibility [5,9]. Then, both the field and T-dependence of x indicate that the sporadic regime is destroyed with an energy of the order of T_{g}.

Spin glass transition: As for the "conventional" relaxation channel in weight $(1 - x)$, λ seems to diverge at T_{g} when T decreases (Fig. 3(d)). Below, the weight of the exponential term at low fields ($1 - x \sim 1 - p$) could correspond to localized frozen spins, reflecting a glassy component. We could further confirm the existence of such a frozen component in SCGO($p = 0.95 - 0.89$), since a clear recovery of a small part (~4%) of the asymmetry is found at long times [9].

4. Conclusion

As a conclusion, a picture based on a $S = \frac{3}{2}$ analogue of a coherent RVB state, which magnetic excitations are mobile fluctuating spins on the kagomé lattice, explains well the data, provided that (i) these excitations can be generated for $T \geqslant 30\,\mathrm{mK}$, which underlines the smallness of the "magnetic" gap, if any; (ii) an energy scale related to T_{g}, which varies very little with p, is high enough to destroy the coherent RVB-like state; (iii) the substitution defects are accounted for by an additional classical relaxation process.

In this framework, spinons could mediate the interactions between magnetic defects localized around spin vacancies and induce the SG-like state. The transition would correspond then to the formation of the coherent singlet state rather than any interaction strength between defects. This would explain why T_{g} is little affected by dilutions and, at the opposite of canonical SG slightly decreases with $(1 - p)$, which could be viewed as a progressive breakdown of spinon excitations.

Acknowledgements

These experiments were performed in the SμS, PSI, Villigen, CH, and ISIS, RAL, Didcot, UK. Part of this work was financially supported by the EC IHP Programme for large scale facilities and by the EC Framework Programmes V and VI.

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