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2007 J. Phys.: Condens. Matter 19 145254

(http://iopscience.iop.org/0953-8984/19/14/145254)

J. Phys.: Condens. Matter **19** (2007) 145254 (6pp) doi:10.1088/0953-8984/19/14/145254

Low-temperature relaxation in kagome bilayer antiferromagnets

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Received 16 September 2006
Published 23 March 2007
Online at stacks.iop.org/JPhysCM/19/145254

Abstract
The pyrochlore slab (kagome bilayer) compounds $SrCr_{9x}Ga_{12-9x}O_{19}$ (SCGO; $x < 1$) and $Ba_2Sn_2ZnCr_{7x}Ga_{10-7x}O_{22}$ (BSZCGO; $x < 1$), are frustrated systems with quite similar magnetic properties of the spin $S = 3/2$ Cr^{3+} ions. Neutron scattering studies have shown that the two compounds have a completely dynamic magnetic response in a broad temperature range. In both systems the development of short-ranged dynamic correlations leads to a low-T state that can be understood as local clusters with antiferromagnetic character. At liquid He temperatures a partial freezing of the magnetic fluctuations is observed as an increase of the elastic resolved response. A large majority of the magnetic moments remain fluctuating and one also observes a low-energy (long-time) relaxation in the vicinity of the macroscopic freezing. Time and temperature dependence of this relaxation appear system dependent without critical behaviour, and we conclude that the freezing is a consequence of the establishment of a coherent quantum state.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The pyrochlore slab (or kagome bilayer) compounds $SrCr_{9x}Ga_{12-9x}O_{19}$ (SCGO (x)) and $Ba_2Sn_2ZnCr_{7x}Ga_{10-7x}O_{22}$ (BSZCGO (x)) have been extensively studied in the context of an expected manifestation of classical or quantum ground states relevant to theoretical predictions concerning spin systems on the kagome lattice [1]. Caution must be exercised, however, since the kagome bilayer magnetic sub-lattice is more complicated than the ideal planar kagome lattice. An additional complication is the substitution by non-magnetic Ga atoms on the magnetic Cr sites. This dilution is inevitable, but to some level controllable, and thus it provides a handle for the study of defect properties. It is rather clear that these systems should not be regarded as ideal models of generic kagome behaviour, rather as remarkable examples of strongly fluctuating magnets without any sign of conventional long-range order. The two compounds have in common intriguing low-T phenomena, such as a partial freezing of the magnetic fluctuations. In spite of the overall similarity, certain details of the low-temperature properties are clearly dominated by system-dependent interactions as would be expected for any highly frustrated magnets [2]. Due to the very broad dynamic range of the magnetic response it has become evident that many different experimental techniques are necessary for completing the picture of their magnetic properties. Features such as resonating valence bonds, spinon excitations or quantum spin-glass—theoretical concepts worked out for more ideal cases—-have been applied to explain the comprehensive experimental studies. In the present paper we shall review the experimental evidence on the low temperature phenomena, including recent work, and present our actual ideas about the physics involved.

2. Spin-glass ground state? Kagome physics?

The observation of a spin-glass like freezing in SCGO [3] triggered a continuing interest in the physics of kagome bilayers. It was realized that the ground state of this strongly fluctuating antiferromagnet could not be easily described within the standard framework of spin-glass physics. One essential feature difficult to match with such a picture is, for example, the relative insensitivity of the freezing to disorder; moreover, with a counter-intuitive dependence the freezing temperature decreases with increasing magnetic dilution [4]. The scaling laws usually applicable to insulating spin-glasses with nearest neighbour interactions did not give a satisfactory account of the bulk ac susceptibility [5]. Another point of concern was found when the short-range correlations evident in the neutron diffraction were quantified with the help of energy-resolved neutron scattering: it was shown that only a fraction (of the order of 30%) of the total magnetic moment involved was frozen in the form of antiferromagnetic correlations with very short correlation length, essentially limited to nearest neighbours only [6].

A particular non-ideal feature in SCGO is the presence of magnetic ions on the $4f_{vi}$ sites that do not belong to the bilayer [7]. However, the moments on those sites couple in singlet ground state dimers with a first excited triplet level at about $\hbar\omega/k_B \approx 215$ K, indicating that the kagome bilayer (also called the pyrochlore slab) is the relevant magnetic lattice at low temperature [8]. The subsequent discovery of the BSZCGO compound [9] in which the similar kagome bilayer carries all the magnetic ions brought fresh motivation to the study of these spin systems. Indeed, static bulk properties of the two compounds show a lot of qualitatively similar features, for example a T^2-dependence of the specific heat at low temperatures [3, 9]. It is tempting to treat the two compounds on the same footing.

The local spin dynamics of the two compounds have been extensively studied on using muon spin relaxation (μSR) (for a review see [10]). The first μSR studies of SCGO pointed out the particularities of the observed relaxation, especially the dynamic nature of the system persisting down to mK temperatures. The μSR studies were later extended to a wide range of external (T, H) and internal (x) parameters on both of the compounds. The outcome was qualitatively very similar with regard to the low-T behaviour of the spin system both in SCGO and in BSZCGO. It also became evident that the magnetic dilution had a rather similar effect in the two compounds. With respect to the dilution, the earlier observation of the tendency to complete disappearance of the freezing (T_g extrapolating to $T = 0$) at the percolation threshold of the kagome bilayer was confirmed in these studies [10]. Such a dependence suggests that the freezing phenomenon has an intrinsic origin and is therefore perturbed by the dilution of the lattice.

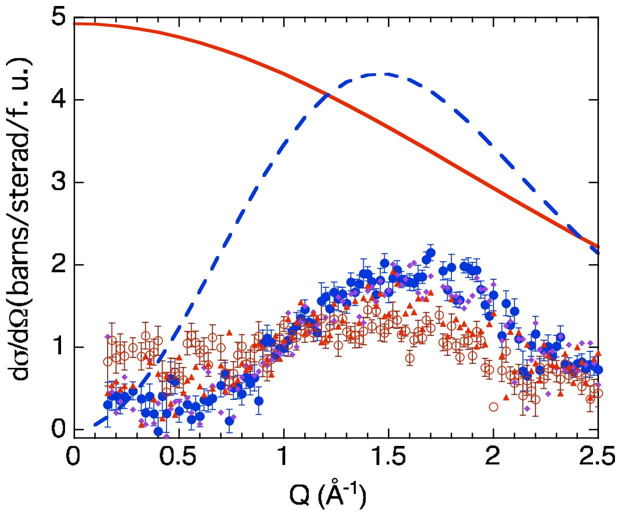

Figure 1. The temperature dependence of the magnetic cross-section of BSZCGO obtained at $T = 100$ K (open dots), 50 K (triangles), 20 K (diamonds) and 2.5 K (full dots) showing the development of the short-ranged dynamic correlations. Comparing with the ideal paramagnetic cross-section calculated for the $S = 3/2$ moment of 6.7 Cr^{3+}/f.u. (solid line) one can see that already at $T = 100$ K some correlation exist and also that the cross-section is less than expected for the full moment because the dynamic range of the instrument does not cover the full width of the response. At $T = 1.5$ K the Q dependence of the response is similar to the form expected for nearest neighbour dimers as discussed in the text (dashed line).

Indeed, the major outcome from detailed analysis of the recent μSR data is the proposition that the freezing is associated with the establishment of a coherent quantum state at low temperatures [11]. Such a state would be akin to the resonating valence bond (RVB) state proposed for the spin $S = 1/2$ kagome system by theoretical and numerical works, e.g. [12]. It is not straightforward to assign the properties of such an ideal model to the $S = 3/2$ kagome bilayer and there is surely no *a priori* justification for doing that, but these ideas have provided an interesting framework for discussing the recent experiments and therefore we keep to this line in the present paper.

3. Low-T, low-energy magnetic response

The magnetic response of short-range correlated systems with broad features in $(Q, \hbar\omega)$-space is best examined using neutrons with polarization analysis. As depicted in figure 1 obtained in diffraction mode (no energy analysis) on the instrument D7 at the ILL, the paramagnetic scattering from the BSZCGO ($x = 0.97$) spin system already shows some Q-dependent modulation at $T = 100$ K, and on cooling one can observe the growth of a characteristic hump associated with the antiferromagnetic short-range correlations. The maximum of the hump occurs at $Q \approx 1.5$ Å^{-1} and the width is such that one concludes that the important correlation has to do with a tendency for local singlet formation between nearest neighbour magnetic moments separated by a distance R_{nn}. In fact, as shown in the figure, the powder averaged dimer structure factor, $S(Q) \sim \sin(QR_{nn})/(QR_{nn})$, where R_{nn} is the average nearest neighbour Cr–Cr distance, is a reasonable first approximation for the form of the hump. A comparison with the ideal paramagnetic response also shows that in the present experimental conditions one cannot find the full fluctuating magnetic moment expected for the composition; this is due to the notable width of the response that exceeds the dynamic window of the experiment (i.e. failure of the quasi-static approximation). Indeed the magnetic response extends over a few tens of meV in both of these compounds [8, 13], with a energy dependence that appears rather independent of the wavevector transfer [13]. The overall features of the magnetic response are quite similar in the SCGO compound [14]. The NMR Knight shifts on samples of the same batch gave evidence of the appearance of a pseudo-gap and were interpreted as indicative of the intrinsic local response of the bilayer system [15].

Neutron scattering has not shown any indications of a gapped response in either of the two compounds. High-resolution energy resolved neutron scattering has been used to investigate the very low energy ($\hbar\omega < 10\ \mu$eV) range [6, 16]. These studies were used to quantify the strongly slowed-down frozen response that is seen as the growth of the elastic (resolution limited) response. That contribution is only about $1/4$ to $1/3$ of the possible full frozen moment, but anyhow it is much more than the some % level corresponding to the effective moment of the static bulk susceptibility obtained either by the two-population model [17] or simply as $\chi T/C$, where C is the Curie constant for free Cr moments. Further, it was demonstrated that the low-T static bulk susceptibility above the freezing follows very well a power-law dependence $\chi(T) \sim T^{-\alpha}$ with $\alpha \approx 0.6$ for BSZCGO [18] or $\alpha \approx 0.4$ for SCGO [19] (see also [20]). This can be taken as an indication that the defects are not quasi-free but are involved in an interacting assembly with random exchange [21]. Such a picture is consistent with the neutron response that shows no gap nor any distinct narrow response that one could associate with a weakly interacting defect moment population separable from the global response.

The Q dependence of the neutron response shows that both the dynamic response and the elastic resolution limited part originate from similar short-range correlated entities. The minimal realization for those entities is a nearest neighbour dimer. For the dynamic part the first excited triplet levels of such a collective assembly of dimers should then represent a continuum extending up to the full nearest neighbour exchange. According to theoretical work on the ideal $S = 1/2$ kagome lattice the dimers around a non-magnetic defect have a tendency for staggered magnetization [22], i.e. static short-range order. This is seen as the frozen short-range order in the high-resolution elastic neutron response Such a picture was also proposed to be consistent with the NMR results that show a broadening of the NMR line-width at low temperatures [23].

Detailed analysis of the μSR work led to the suggestion that the dynamics observed with that method can be described as quantum excitations in a coherent singlet matrix [10], proposed to be akin to the spinons expected in the case of the ideal $S = 1/2$ kagome system with the RVB ground state. Overall, the analysis was capable of extracting several contributions in the μSR signal, identified through their behaviour in applied field as well as their dependence on temperature and magnetic dilution x. Accordingly it was shown that thermally excited and defect induced spinons contribute to the low-T quantum dynamics, below the temperature of the coherence. The apparent freezing then is an indication of the establishment of a coherent state.

The results obtained with the neutron spin echo (NSE) method on SCGO ($x = 0.95$) and BSZCGO ($x = 0.97$) have given new insights into the properties of these compounds [24]. The NSE method appears to be the ideal tool for examination of the low-T, very-low-energy magnetic response [25]. The main advantages of NSE are the magnetic selectivity due to the use of polarized neutrons and especially the broad dynamic range not reachable by other high-resolution neutron techniques. As shown in figure 2 there is no divergence indicative of a critical regime of the relaxation time at the macroscopic freezing temperature. This is unlike the behaviour of the canonical spin-glasses, in which a critical regime occurs [26–28]. The other interesting finding was the thermally activated phonon assisted regime in the SCGO compound [24]. In BSZCGO such a regime does not appear and the relaxational dynamics

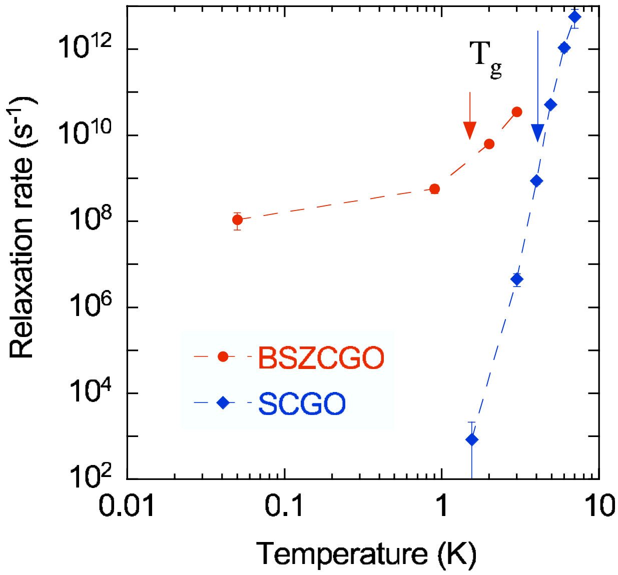

Figure 2. The temperature dependence of the magnetic relaxation rate in the low-T range where it is observable with NSE. Data for SCGO (diamonds) and BSZCGO (dots) show no critical behaviour, i.e. divergence of τ, at the macroscopic freezing indicated by the arrows. In SCGO a thermally activated regime is seen at $4\ K \approx T_g < 7\ K$ [24]. In BSZCGO no such regime is observed and there is a marked saturation at low temperature.

is faster by orders of magnitude. This is not consistent with the standard spin-glass transition that ideally should follow the universal behaviour. These quite dissimilar observations in apparently quite similar spin systems suggest further that the slowing down leading to the partial freezing of the magnetic response is a system-dependent process reflecting indirectly the onset of coherence in the ground state. In this context it is interesting to recall that at the very low dilution limit the frozen moment decreases notably and might extrapolate to zero for $x \rightarrow 0$, while the apparent freezing temperature tends to a finite value [29]. However, it has proved to be quite difficult, if not impossible, to produce a sample with no dilution at all and an experimental proof of the no freezing limit may well be unreachable.

4. Conclusions

We have presented a short overview of the main experimental results concerning the low-temperature, low-energy magnetic response in the kagome bilayer compounds, with special emphasis on the neutron work. The dimer correlations, and their partial, defect induced freezing, are the main ingredients of the observed phenomena, and following the earlier suggestions based on the μSR work we propose that the freezing is a consequence the establishment of a coherent quantum state within the phenomenology proposed for the resonating valence bond state.

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