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PHYSICA B

Magnetic freezing and fluctuations in the Kagomé compound $SrCr_{8-x}Ga_{4+x}O_{19}$

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Abstract

We review recent neutron scattering experiments to determine the magnetic fluctuations and order in the geometrically frustrated magnet $SrCr_{8-x}Ga_{4+x}O_{19}$. The instruments used are the direct- and indirect-geometry instruments HET and IRIS at the ISIS pulsed spallation source. Results are compared with earlier data obtained using the triple-axis spectrometer TASI at the Risø DR3 reactor.

1. Introduction

Frustration is one of the most important concepts in modern many-body physics. The spin triangle, shown in Fig. 1, is one of the simplest illustrations of this concept. For antiferromagnetic interactions $JS_i \cdot S_j$ acting via the bonds between the spins, the minimum energy configuration of the entire triangle does not minimize the energy of each individual bond. Indeed, for Ising spins, the minimum energy E_{min} is simply $-2J + J = -J$, while for Heisenberg and xy spins, $E_{min} = -1.5\,J$, corresponding to the coplanar configuration illustrated in Fig. 1(c). Although it is simple to understand the isolated frustrated triangle, there is – even today – no complete knowledge of the ground state for infinite lattices formed when such triangles are linked together [1].

In view of the theoretical intractability of the problem of geometric frustrated antiferromagnets, it is important to perform experiments on compounds which contain geometrically frustrated spins. One such compound is $SrCr_{8-x}Gr_{4+x}O_{19}$ [2–9] (SCGO(x)), whose fundamental constituents are planes containing antiferromagnetically coupled $Cr^{3+}(S = \frac{3}{2})$ ions at the vertices of Kagomé lattices [10]. Fig. 2 shows a single Kagomé net, while Fig. 3 shows how these nets are assembled to form the compound.

Fig. 4(b) and (c) shows some of the fundamental physical properties [3] of the material for $x \approx 0$. In the magnetic-susceptibility data, there is a spin-glass like transition at $T_g \approx 3$ K, marked both by a cusp in the field-cooled data and a divergence of zero-field and field-cooled results. The ratio C/T of the specific heat C to the temperature T has a maximum at 5 K followed by linear, rather than T-independent (as is common for conventional spin glasses [11]) behavior as $T \to 0$.

2. Experimental details

We have performed neutron-scattering measurements on members of the SCGO(x) family both at the Risø DR3

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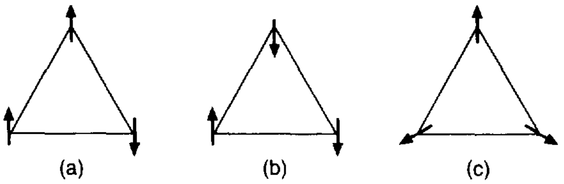

Fig. 1. Antiferromagnetic Ising ((a) and (b)) and *xy* spin triangles (c). The two configurations in (a) and (b) have the same energy.

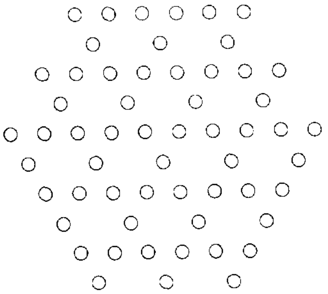

Fig. 2. Kagomé lattice [11].

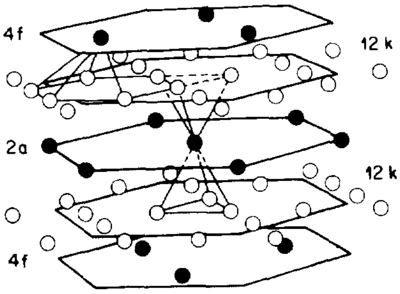

Fig. 3. Structure [2] of SCGO(*x*). Filled and open circles are Cr atoms in dilute triangular and dense Kagomé planes, respectively.

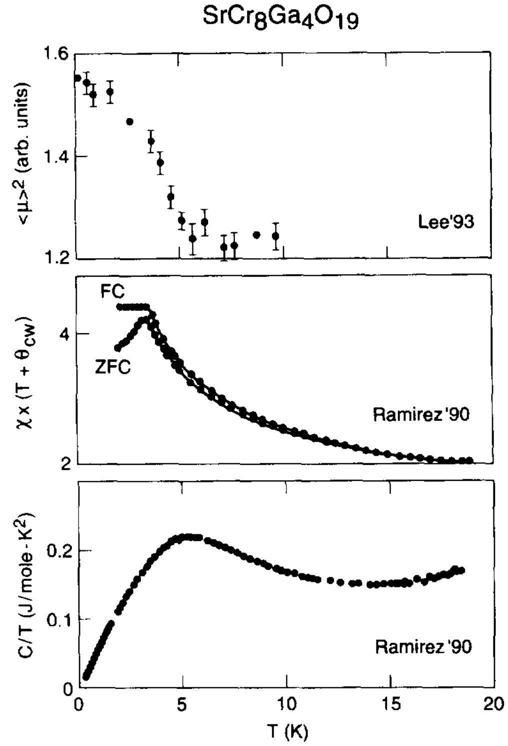

Fig. 4. Temperature dependence of (a) elastic signal measured [15] at IRIS (energy resolution = 5 μeV FWHM) using mica 004 analyzer (b) zero-field and field-cooled magnetic susceptibilities [3] and (c) specific heat [7] divided by T. All data are for $x \approx 0$ samples.

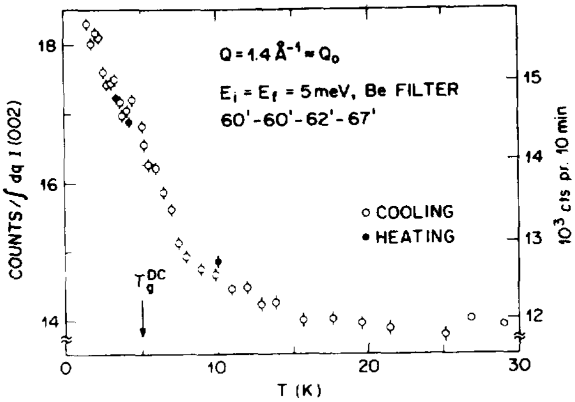

Fig. 5. Temperature dependence of elastic signal measured using TASI instrument (energy resolution = 0.2 meV FWHM) at Risø DR3 reactor for SCGO(x) with $x = 0.87$.

reactor and at the ISIS facility. At Risø, we used the TASI 3-axis instrument which views the cold source from a position in the main reactor hall. At ISIS, we have used the direct-geometry time-of-flight chopper spectrometer HET as well as the indirect-geometry, high-resolution backscattering spectrometer IRIS [12, 13].

3. Low frequency measurements: IRIS and TASI

Fig. 5 shows how the nominally elastic scattering, proportional to the mean-square frozen moment in the limit of perfect energy resolution [14–16] depends on temperature [4]. The sample has $x = 0.87$, and the corresponding bulk glass-transition temperature is 5 K. It is clear

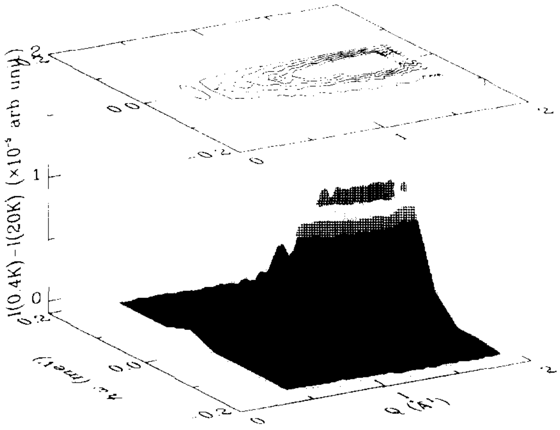

Fig. 6. Difference between 1.4 and 20 K data taken using IRIS with pg(002) analyzer for which the energy resolution is 20 μeV FWHM [9].

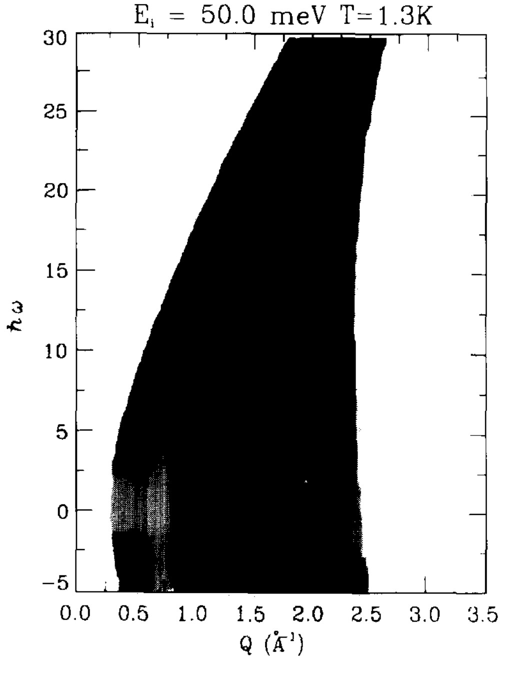

Fig. 7. Survey in momentum transfer – energy space of scattering measured [17] at 1.4 K using HET.

that the scattering captured within the 0.2 meV full-width-at-half-maximum (FWHM) resolution of the Risø TASI instrument used here begins to grow above the non-magnetic elastic background at $T = 15$ K $= 3\ T_g$. It also does not saturate above 1.2 K, the base temperature of the cryostat. These data are to be contrasted with the IRIS results [17] shown in Fig. 4(a), obtained for a more Cr-rich sample ($x \approx 0$) . The energy resolution here is 5 μeV FWHM. The intensity begins to grow in the fashion one would associate with an ordinary magnetic order parameter as T passes below 5 K, a temperature which is less than twice the bulk spin-glass transition temperature. We conclude that improving the energy resolution (as, e.g., in going from TASI to IRIS) results in better-defined spin freezing, as it does in traditional spin glasses such as CuMn [14] and amorphous MnSi [16].

While the neutron data provide an excellent demonstration of conventional T-dependent spin freezing in SCGO(x), they also suggest that SCGO(x), is not a conventional spin glass. In particular, Fig. 6 [9] which shows the difference between spectra taken at $T = 1.4$ and 20 K for pg(002) analyzers on IRIS (resolution = 20 μeV FWHM), clearly indicates a strongly Q-dependent elastic structure factor. Indeed, $S(Q)$ appears to become small as $Q \to 0$. A vanishing forward-scattering amplitude is certainly not a property of conventional spin glasses [14] and suggests a hidden order among the spins in the Kagomé lattice. This hidden order is most likely [5] associated with each triangle possessing net zero moment by virtue of spin configurations such as that illustrated in Fig. 1(c).

4. High and medium frequencies: HET

In addition to providing detailed information on the lower limit of $S(Q, \omega)$, pulsed neutron spectroscopy is also an excellent method for probing the medium and high frequency response. Fig. 7 shows a map of the inelastic scattering [17] for SCGO($x = 0$) obtained using HET. One important feature in this map is the magnetic scattering ridge emanating parallel to the energy axis from $E = 0$ and $Q_0 = 1.4$ A^{-1}, the wavevector associated with the triangular spin order of Fig. 1(c). This feature is due to the Kagomé planes, and its extent in energy yields a bandwidth of ~20 meV for the magnetic excitations in SCGO($x \approx 0$). A second feature, almost certainly due [17] to pairs of spins in the pairs of dilute triangular planes located between every other Kagomé plane, is the sharp peak at $E = 21$ meV.

5. Conclusions

Direct and indirect geometry time-of-flight instruments are very useful for measuring the magnetic structure function in powders of $SrCr_{8-x}Ga_{4+x}O_{19}$, a compound with strong geometric frustration, over frequencies ranging from several μeV to tens of meV. Important new physical information includes a measure of the bandwidth (~ 20 meV) of the magnetic excitations in this compound, and identification of the feature, namely the nearly vanishing forward-scattering amplitude, which distinguishes the frozen state of SCGO($x \approx 0$) from that of ordinary spin glasses.

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References

[1] A recent review of the theoretical issues is by P. Chandra, P. Coleman and I. Ritchey, J. Phys. I (France) 3 (1993) 591.
[2] X. Obradors et al., Solid State Comm. 65 (1988) 189.
[3] A.P. Ramirez et al., Phys. Rev. Lett. 64 (1990) 2070.
[4] C. Broholm et al., Phys. Rev. Lett. 65 (1990) 3173.
[5] G. Aeppli et al., J. Magn. Magn. Mat. 90-91 (1990) 255.
[6] Y.J. Uemura et al., Hyperfine Interactions 85 (1994) 133.
[7] M. Sumikawa et al., submitted to Physica B (1994).
[8] C. Broholm et al., J. Appl. Phys. 69 (1991) 4968.
[9] G. Aeppli et al., ISIS Annual Report - 1993 (RAL, Didcot) p. 83.
[10] I. Syozi, Prog. Theor. Phys. 6 (1951) 306.
[11] See, e.g., D. Meschede et al., Phys. Rev. Lett. 44 (1980) 102.
[12] C.J. Carlile et al., Nucl. Instr. and Meth. 338 (1994) 78.
[13] C.J. Carlile and M.A. Adams, Physica B 182 (1992) 431.
[14] A.P. Murani and A. Heidemann, Phys. Rev. Lett. 41 (1978) 1402.
[15] G. Aeppli et al., Phys. Rev. B 29 (1984) 2589.
[16] G. Aeppli et al., Phys. Rev. Lett. 54 (1985) 843.
[17] S. Lee et al., to be published.