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Spin Glass State in S=3/2 Kagomé Antiferromagnet $Co(N_3)_2(bpg)[(CH_3)_2(NCOH)]_{4/3}$

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Specific heat, magnetic susceptibility and magnetization of $Co(N_3)_2(bpg)[(CH_3)_2(NCOH)]_{4/3}$, representing $S = 3/2$ kagomé system are reported. Long-range ordering at 16 K was revealed, however, at lower temperatures slow spin dynamics is still found. The analysis of alternating susceptibility suggests the onset of glassy state. The study of the time dependence of magnetization revealed the existence of more relaxation channels with pronounced different relaxation times. The observed behaviour is consistent with the formation of topological spin glass in which relaxation is governed by both spin and chiral degrees of freedom.

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1. Introduction

Geometrically frustrated magnets are of great interest due to novel phases, appearing because of large degeneracy of low energy states. In particular, for systems in kagomé lattice, the formation of disorder-free spin glass state, termed *topological spin glass*, has been predicted [1]. This state is believed to be a consequence of weak additional interactions, e.g. next nearest exchange, which using "order by disorder" mechanism select groups of states, from which $q = 0$ and $\sqrt{3} \times \sqrt{3}$ states are favoured [2]. These states are characterized by uniform and staggered chiralities, respectively, and different relaxation rate was predicted for chiral and spin degrees of freedom [3]. The purpose of this work is to investigate the potential formation of topological spin glass in $Co(N_3)_2(bpg)[(CH_3)_2(NCOH)]_{4/3}$ (I).

2. Experimental details

The studied compound has been synthesized following well established procedure [4]. The structure of the studied material consists of kagomé layers in which Co(II) magnetic ion is coordinated with six N atoms, two from *trans-bpg* ligands and four from azido ligands. The azido ligands link the Co(II) ions creating a kagomé layer featuring vertex-sharing triangles along the *bc* plane. The wavy kagomé layers are bridged by *bpg* ligands and generate a three – dimensional framework whose hexagonal channels are filled by $(CH_3)_2(NCOH)$ molecules. Specific heat of powder sample was studied in PPMS device from 2 K to 30 K, whereas SQUID magnetometer was used for ac susceptibility and magnetization measurements.

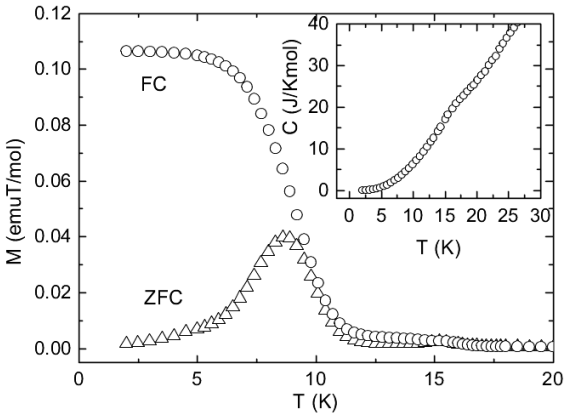

Fig. 1. Temperature dependence of magnetization of I in FC and ZFC regimes (open symbols). Inset: temperature dependence of specific heat of I in zero magnetic field.

3. Results and discussion

The magnetization in FC and ZFC regimes was studied from 2 K to 25 K in magnetic field 0.1 T, see Fig. 1. The data are characterized by small bifurcation at 16 K, followed by a pronounced difference between FC and ZFC data appearing at 9 K. The feature observed at 16 K may be associated with long-range ordering, as confirmed by specific heat measurements in zero magnetic field, where a small spike was found at 16 K. Large bifurcation of magnetization curves below 16 K, together with low entropy content of the spike in specific heat, suggest large fluctuations of spins even below ordering temperature. Such a situation is not uncommon for geometrically frustrated

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magnets. In order to clarify the origin of the magnetization behavior at low temperatures, systematic study of alternating (ac) susceptibility was performed in the frequency range 1 Hz – 1 kHz. The sensitivity of ac susceptibility to the used excitation frequencies suggests slow spin dynamics below 16 K. The data were evaluated using Cole-Cole formalism based on the equation:

$$\chi^* = \chi_S + \frac{\chi_0 - \chi_S}{1 + (i\omega\tau_C)^{1-\alpha}},$$

in which χ_0 and χ_S denote isothermal and adiabatic susceptibilities, respectively, α characterizes the width of the distribution of relaxation times, τ_C represents mean relaxation time and ω denotes the excitation frequency. The results are presented in Fig. 2. Below 10 K the parameter α from Cole-Cole formula was found to be bigger than 0.85, indicating a wide distribution of relaxation times characteristic for spin glass state. It should be stressed, that the temperature dependence of the isothermal susceptibility χ_0 obtained from the fit, is in very good agreement with ac susceptibilities found at lowest frequencies. For simplicity, adiabatic susceptibility χ_S was set to zero. Notably, the observed spin glass state is not induced by structural and/or bond disorder as is the case of canonical spin glasses.

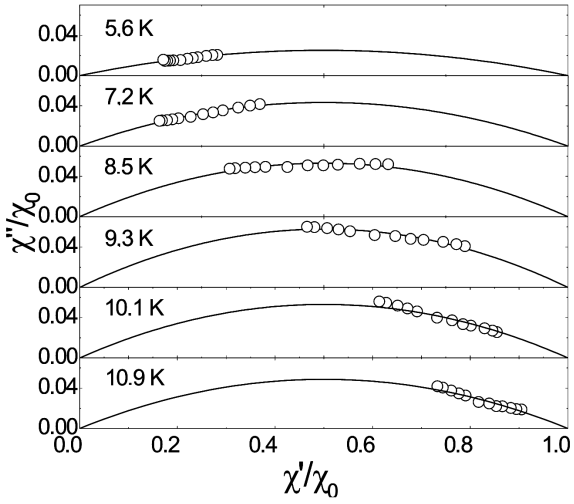

Fig. 2. Cole-Cole diagrams of I, normalized to isothermal susceptibility χ_0 analyzed at various temperatures.

In order to get insight in slow dynamics revealed by ac susceptibility studies, time dependence of magnetization was investigated at various temperatures. To this end, the sample was cooled in magnetic field from above 16 K to the desired temperature between 2 K and 10 K. Then magnetic field was removed and the relaxation of magnetization was monitored. Very long relaxation of the magnetization was observed with only little effect of the temperature. The obtained results are illustrated in Fig. 3. The time dependence of magnetization could be

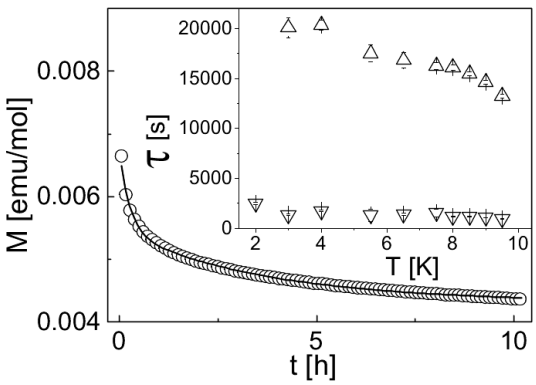

Fig. 3. Time dependence of magnetization of I at 9 K (open circles), fitting using two exponentials is denoted by a solid line. Inset: Temperature dependence of the two relaxation times.

analyzed using sum of two exponentials. The obtained weak temperature dependence of relaxation times suggest an important role of quantum tunnelling in the relaxation process. The existence of more relaxation processes with significantly different relaxation times is consistent with relaxation of spin and chiral degrees of freedom in topological spin glass.

4. Conclusions

The investigation of relaxation behaviour of $Co(N_3)_2(bpg)[(CH_3)_2(NCOH)]_{4/3}$ revealed the formation of spin glass state below ordering temperature. The spin glass state does not seem to be induced by structural disorder and has properties anticipated in topological spin glass. However, further investigation at lower temperatures and frequencies using single crystals is necessary to elucidate the origin of the observed behaviour.

Acknowledgments

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