Observation of the topological phase transition in the triangular Heisenberg antiferromagnet NiGa$_2$S$_4$

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Abstract. It has been shown in 2005 by neutron powder diffraction that the triangular layer Heisenberg compound NiGa$_2$S$_4$ is characterized by nano-scale magnetic correlations below $\sim 18$ K. Using the muon-spin rotation and relaxation ($\mu$SR) techniques, we have evidenced that the compound displays a phase transition at $T_c = 9.2 (2)$ K below which a coherent oscillation of the muon spins is detected. Remarkably, the magnetic specific heat $C_m$ exhibits a rounded low-temperature maximum at $\sim 13$ K, i.e. at a temperature above $T_c$. This is consistent with the theory of Kawamura and Miyashita which attributes the transition at $T_c$ to the unbinding of $Z_2$ vortices. NiGa$_2$S$_4$ is characterized by a wide spectrum of magnetic fluctuations as inferred, for example, from the difference in the temperature scale deduced from $\mu$SR and neutron scattering.

The investigation of the physical properties of triangular antiferromagnets has a long history; see for example the review of Collins and Petrenko [1]. But only quite recently NiGa$_2$S$_4$ was discovered as the first quasi two-dimensional Heisenberg antiferromagnet with an exact triangular lattice [2]. This compound is a chalcogenide magnetic insulator with the Ni$^{2+}$ magnetic ions in the $S = 1$ state sitting on a regular triangular lattice. The interactions are of the Heisenberg type, referring to the isotropic Curie constant [3] and the identical temperature dependence of the spin-spin and spin-lattice relaxation rates measured by nuclear magnetic resonance (NMR) above 10 K [4]. The magnetic specific heat has a quadratic thermal dependence below $\sim 7$ K [2, 5], suggesting the existence of gapless excitations at low temperature (Goldstone modes). However, despite its large Curie-Weiss temperature, $\Theta_{CW} = -80 (2)$ K, NiGa$_2$S$_4$ does not order magnetically in a long-range manner, at least down to $\sim 0.35$ K. It displays only nano-scale magnetic correlations below $\sim 18$ K according to neutron scattering (see insert of Fig. 4 in Ref. [2]).

We have recently reported muon-spin rotation and relaxation ($\mu$SR) measurements performed at the GPS spectrometer of the S$\mu$S laboratory of the Paul Scherrer Institute (PSI, Villigen, Switzerland) [5]. Here we shall further discuss these results, in reference to published results obtained by other experimental techniques and theoretical predictions.

We first recall the main results derived from the $\mu$SR technique, an experimental method which allows to probe the static and dynamical properties of magnetic materials; see for example published reviews [6, 7]. The measurements were carried out on a powdered sample which was characterized by neutron powder diffraction at 50 K performed with the DMC diffractometer of the SINQ facility at PSI, zero-field specific heat and low-field magnetic
susceptibility measurements [5]. These characterization measurements were found consistent with published data [2]. Our key result is the observation of a well defined damped oscillation, corresponding to a spontaneous magnetic field, below $T_c = 9.2 (2)$ K. Since, apart in pathological cases, the spontaneous field in insulators is proportional to the ordered moment, our result indicates that the ordered moment vanishes at $T_c$. We are therefore entitled to define $T_c$ as the critical temperature of NiGa$_2$S$_4$. Note that this $T_c$ value is at variance with other results in the literature [8, 4]. We shall come back to this point later. The spin-lattice relaxation rate $\lambda_Z$ found in our $\mu$SR measurements is appreciable below $T_c$ pointing to the persistence of spin dynamics down to the lowest temperature at which the experiments have been performed, i.e. 2.3 K. Now, concerning the region above $T_c$ the relaxation is exponential above $T_{\text{cross}} = 12.6$ K: $\lambda_Z$ increases monotonously as the sample is cooled from $\sim 200$ K as it is expected. A fit to a random phase approximation model is consistent with a transition at $T_c$. Below $T_{\text{cross}}$ (but above $T_c$), whereas the muon signal relaxes faster, the lineshape which is no longer found exponential can be fitted to a stretched-exponential function. This behavior is attributed to the unbinding of the $Z_2$ vortices.

Concerning the lack of spontaneous muon spin precession in the previously published $\mu$SR spectra, we note that the time resolution available at the $\mu$SR facility used to record one set of data does not allow to evidence the precession that we have observed [4]. On the other hand, the absence of precession in the first ever published spectra is more surprising [8]. It may reflect either an instrumental limitation of the spectrometer used for these measurements or the presence of an appreciable magnetic disorder in the investigated sample.

In Fig. 1 is plotted the thermal dependence of the maximum of the local field at the muon site and the measured magnetic specific heat $C_m$ at low temperature. The plot shows clearly that $C_m(T)$ displays a maximum at $\sim 13$ K, i.e. around 4 K above $T_c$ at which the local field at the muon site vanishes. Hence the maximum in $C_m(T)$ is not a signature of a phase transition. This is consistent with the important width of the peak in $C_m(T)$. In Fig. 2, we display $C_m(T)$ deduced from Monte Carlo simulations for a triangular Heisenberg antiferromagnetic system. In agreement with these numerical simulations, the measurements locate the phase transition below the temperature at which $C_m(T)$ has a maximum. However, the experimental specific heat is more rounded than expected from the simulations. A possible reason for this difference might be in the incommensurate nature of the short-range correlations which is not accounted for in the available simulations. In addition, one should remember that biquadratic Heisenberg interactions may influence the physics of the system [9].

We note that the Berezinskii-Kosterlitz-Thouless (BKT) transition which occurs in XY system is also characterized by a rounded peak in $C_m(T)$ located above the temperature of the phase transition; see for example the book of Chaikin and Lubensky [11]. Hence, for both the Heisenberg and XY symmetry, the unbinding of the vortices start at a temperature smaller than the temperature at which $C_m(T)$ exhibits a maximum. A meaningful discussion of the position of the peak of $C_m(T)$ relative to $T_c$ requires first to understand the origin of its large width.

We estimate now the magnitude of the exchange interaction from our data. According to Kamamura and Miyashita’s model for the triangular Heisenberg antiferromagnet with only nearest neighbor interaction taken into account, the thermodynamic phase transition occurs at $k_B T_c = a S(S + 1) J$ with $a = 0.31 (1)$ [10]. We have substituted $S^2$ by $S(S + 1)$ in the original formula recognizing the quantum nature of the spins. The value of $a$ can also be obtained from Fig. 2. The Hamiltonian is written as $\mathcal{H} = J \sum_{\langle ij \rangle} S_i \cdot S_j$ where $J > 0$ is the antiferromagnetic bilinear exchange between two ions and the sum is restricted to the nearest neighbors. In fact, it has been determined that the third-neighbor interactions $J_3$ are antiferromagnetic and dominate since $J_1 / J_3 = -0.2 (1)$ and $J_2 \simeq 0$ [2]. Therefore we need to substitute $J$ by $J_3$ in the formula for $T_c$. Identifying the critical temperature of the thermodynamic phase transition with the
temperature at which the field at the muon site vanishes ($T_c = 9.2(2)$ K), we extract $J_3/k_B = 14.8(8)$ K. This parameter can be also estimated from standard mean-field theory. From $\sum_\alpha z_\alpha J_\alpha = 3k_B|\Theta_{CW}|/[S(S+1)]$, where $z_\alpha$ is the number of $\alpha$-th neighbor magnetic ions to a magnetic ion, we estimate $J_3/k_B = 25(3)$ K. These two estimates of $J_3/k_B$ are not far from each other. This gives weight to our interpretation of the temperature at which the $\mu$SR wiggles vanish, that is $T_c$, as the temperature at which the unbinding of the magnetic vortices start. This is the key result derived from our $\mu$SR study.

The comparison of the results obtained by $\mu$SR and neutron scattering is worthwhile. Whereas the critical temperature is established to be $T_c = 9.2(2)$ K by $\mu$SR, neutron scattering detects nano-scale correlations starting at $\sim 18$ K. Interestingly, this latter temperature value is higher than the temperature at which $C_m(T)$ displays its maximum, and therefore seems to contradict the topological phase transition advocated in this paper. However, the temperature scale as determined by $\mu$SR is nearer to the thermodynamical scale that the temperature scale deduced from the neutron diffraction. This is understandable because the time scale of neutron diffraction is in the range of the picosecond, whereas the $\mu$SR scale in the microsecond range. As already explained elsewhere [5], the two experimental techniques probe different fluctuation scales. We thus deduce that the spectral weight function of NiGa$_2$S$_4$ is quite wide. The fact that the magnetic moment deduced from neutron scattering is only $\sim 75\%$ of the expected Ni$^{2+}$ magnetic moment is an other proof of the wide range of the dynamics characterizing NiGa$_2$S$_4$.

In conclusion, we believe that the continuous transition that we have observed at $T_c = 9.2$ K is the topological phase transition which is predicted to occur in 2D triangular antiferromagnets. We note that it has recently been argued that NMR data recorded on NiGa$_2$S$_4$ uncover a BKT transition [4, 12].

Figure 1. Thermal dependence of the maximum of the spontaneous local field at the muon site, $B_{\text{max}}$, and of the magnetic specific heat, $C_m$, at low temperature. We refer to $B_{\text{max}}$ because the oscillation of the $\mu$SR spectra is Bessel-like, in accord with the incommensurate nature of the nano-scale correlations, and is therefore characterized by this parameter [6].

Figure 2. The magnetic specific heat, $C_m$, versus the normalized temperature as computed for a triangular Heisenberg antiferromagnet by the Monte Carlo technique reproduced from Ref. [10]. The temperature at which the phase transition occurs is also indicated. The different symbols correspond to the different number of cells considered in the simulations. Note the following similarity with Fig. 1: the maximum of the specific heat is located at a temperature above $T_c$. The comparison of the results obtained by $\mu$SR and neutron scattering is worthwhile. Whereas the critical temperature is established to be $T_c = 9.2(2)$ K by $\mu$SR, neutron scattering detects nano-scale correlations starting at $\sim 18$ K. Interestingly, this latter temperature value is higher than the temperature at which $C_m(T)$ displays its maximum, and therefore seems to contradict the topological phase transition advocated in this paper. However, the temperature scale as determined by $\mu$SR is nearer to the thermodynamical scale that the temperature scale deduced from the neutron diffraction. This is understandable because the time scale of neutron diffraction is in the range of the picosecond, whereas the $\mu$SR scale in the microsecond range. As already explained elsewhere [5], the two experimental techniques probe different fluctuation scales. We thus deduce that the spectral weight function of NiGa$_2$S$_4$ is quite wide. The fact that the magnetic moment deduced from neutron scattering is only $\sim 75\%$ of the expected Ni$^{2+}$ magnetic moment is an other proof of the wide range of the dynamics characterizing NiGa$_2$S$_4$.

In conclusion, we believe that the continuous transition that we have observed at $T_c = 9.2$ K is the topological phase transition which is predicted to occur in 2D triangular antiferromagnets. We note that it has recently been argued that NMR data recorded on NiGa$_2$S$_4$ uncover a BKT transition [4, 12].
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