Spin-liquids are currently attracting a considerable interest because of the numerous novel phenomena related to quantum phase transitions [1]. The compound TlCuCl₃ is a nearly optimal model system showing field-, pressure- and doping-induced transitions to antiferromagnetic (AF) phases, which can be investigated by several experimental techniques and reveal unconventional ground states characterized by condensates of magnetic quasi-particles [2,3].

The two S=1/2 magnetic moments in a pair of Cu²⁺ ions are coupled in TlCuCl₃ by a dominant AF exchange interaction J_intra leading to an effective singlet ground state. The elementary excitations are then triplet states, which can hop to neighbouring dimer sites by residual interdimer interactions J_inter. These interdimer interactions are weak in the related compound KCuCl₃ but strong in TlCuCl₃, which therefore has a singlet-triplet spin energy gap \( \Delta \) of modest 0.7 meV or 8.1 K, see Fig 1. The softening of this gap in an external magnetic field or by pressure application is the basic mechanism causing the quantum phase transitions mentioned above.

However, even the zero-field and zero-pressure spin-liquid state provides an interesting testing ground for many-body quantum theories and needs to be characterized for future studies of the finite temperature properties in the different AF phases. The temperature dependence of the spin energy gap \( \Delta \) is hereby of particular interest, see Fig. 1. Inelastic neutron scattering (INS) experiments have been performed on the thermal triple-axis spectrometer 2T (LLB, Saclay) to investigate the spin correlations in TlCuCl₃ up to \( T=J_{\text{intra}} \). The results can now be compared with the related compound KCuCl₃ [4] and extent previous high-resolution but low-temperature studies carried out on the cold triple-axis spectrometers TASP (SINQ, Villigen) and IN14 (ILL, Grenoble).

Contour plots resulting from 14 constant Q-scans along the \( Q=(qh 0 0) \) [r.l.u.] reciprocal direction are presented in Figure 2 for three representative temperatures. A reduction of the triplet bandwidth is observed, which is even more pronounced along directions including the minimal spin energy gap \( \Delta \). We further report a considerable damping of the excitations, caused by a finite lifetime of the quasi-particles, and a reduction of the inelastic intensity by thermal depopulation of the singlet ground state. The renormalization of the excitation energies is in agreement with predictions from both Troyer-Tsunetsugu-Würtz (TTW-MF) [5] and Bose (Bose-MF) mean field theory at moderate temperatures. At \( T=J_{\text{intra}} \) however, a better description of the new INS data results from TTW-MF, see Fig. 2, which is discussed below.
especially at $T>\Delta$. However, it is a non-trivial problem to describe the resulting interaction among these quasi-particles correctly. Of particular interest is further the implementation of the hard-core constraint, which means that only one of the four possible states, the singlet or one of the three triplet states, can reside on each dimer site. In the dilute limit up to $T=\Delta$, where the triplet quasiparticle density is low, Bose-MF theory [2], which corresponds to TTW-MF in this regime, correctly reproduces the observed renormalization of the excitation energies. TTW-MF theory has been proposed to interpolate between the known low- and high-temperature limits. We report almost perfect agreement with the corresponding theoretical predictions. The complete analysis will be presented elsewhere [6].

To conclude, we have investigated the renormalization of the spin correlations in a quantum spin-liquid up to temperatures close to the dominant energy scale, the dimer exchange interaction, of the magnetic system. The results most probably answer the fundamental question about the correct statistical description of a quantum many-body system, where the particles are on the one hand fermions, the $S=1/2$ moments of each Cu$^{2+}$ ion, but on the other hand hard-core bosons, as a consequence of the dominant correlation within the dimer. The present study at finite temperatures is therefore of general interest for the description of related systems with a spin energy gap as well as for the interpretation of their macroscopic bulk properties.

References