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Magnetic Long-Range Order and Trimer Formation in the Purely Organic Multiferroic Magnet TNN · CH3CN

Equilateral spin triangles are pivotal in quantum magnetism to explore frustration, spin–electric coupling, and multiferroic ordering. Achieving ideal triangular spin networks is difficult due to structural distortions, but the organic compound TNN·CH3CN overcomes these limitations with isotropic spins free from Jahn–Teller effects, enabling uniform exchange interactions and making it an excellent platform for novel quantum phases. Its magnetic phase diagram exhibits multiferroic behavior—the first example in equilateral spin triangles. However, macroscopic characterization cannot reveal the atomic-scale magnetic structure. Here, we combine experimental and computational methods to fully investigate TNN·CH3CN's magnetic structure. At zero field, single-crystal neutron diffraction detects no direct magnetic signal, but muon spin relaxation (μSR) and density functional theory show that the absence of oscillations in μSR spectra corresponds to a high-symmetry magnetic structure, confirming its multiferroic character. In the 1/3 magnetization plateau (1.25 < B < 8.49 T), where TNN·CH3CN has a twofold-degenerate S=1/2, Sz=1/2 ground state, polarized neutron diffraction and μSR uncover collective spin behavior, where independent paramagnetic spins couple into spin trimers below 5.5 K. In this phase, ferroelectric order emerges below 0.35 K without conventional spin ordering, providing insight into spin–electric coupling in triangular networks.

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