Understanding quantum thermalization through entanglement build-up in isolated many-body systems addresses fundamental questions on how unitary dynamics connects to statistical physics. These studies also constitute a novel way to investigate strongly correlated quantum systems. A promising pathway is being opened by ultracold atomic systems featuring internal levels that can be initialized in pure states and coherently evolved with controllable long-range interactions. Experiments have been limited so far to small systems (hundreds or fewer particles), or to dilute disordered molecular ensembles. In contrast, magnetic atoms offer a unique possibility to investigate truly macroscopic and well-ordered arrays of spins [1].

We report experimental study of the dynamics and approach towards thermal equilibrium of a pure macroscopic ensemble of spins initially tilted compared to the magnetic field, under the effect of dipole-dipole interactions [2]. The experiment uses a unit filled array of $\approx 10^4$ chromium atoms in a three dimensional optical lattice, realizing the spin-3 XXZ Heisenberg model. We monitor the population of the seven spin components after a collective rotation of an initially polarized ensemble, as a function of the angle between the initial coherent state with respect to the magnetic field. We find that the approach to thermal equilibrium is increasingly driven by quantum correlations as the angle approaches $\pi/2$. The quantum dynamics is benchmarked by comparison with an improved numerical quantum phase-space method [3] which also enables us to compute the dynamics of the Renyi entanglement entropy. Although the measurements only allow us to reconstruct the entropy generated by the diagonal components of the average local density matrix, the excellent agreement with simulations supports entanglement build-up and the corresponding approach to local thermal equilibrium, and provides a first benchmark of our experiment as a quantum simulator.

