Geometrically frustrated magnetism of spins on icosahedral clusters: The $Gd_3Au_{13}Sn_4$ quasicrystalline approximant

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By investigating the magnetism of spins on a quasiperiodic lattice, we present an experimental study of static and dynamic magnetic properties, specific heat, and magnetic entropy of the $Gd_3Au_{13}Sn_4$ quasicrystalline approximant. The magnetic sublattice of $Gd_3Au_{13}Sn_4$ is a periodic arrangement of nonoverlapping spin clusters of almost perfect icosahedral symmetry, where gadolinium localized *f* magnetic moments are distributed on equilateral triangles. The absence of disorder on the magnetic sublattice and the antiferromagnetic (AFM) interactions between the nearest-neighbor spins distributed on triangles result in geometrical frustration of spin-spin interactions. Thus, the $Gd_3Au_{13}Sn_4$ phase can be viewed as a prototype site-ordered, geometrically frustrated spin system on icosahedral clusters. The zero-field-cooled and field-cooled magnetic susceptibilities, the alternating current susceptibility, the thermoremanent magnetization, the memory effect, the magnetic specific heat, and the magnetic entropy all show that the spin system undergoes at low temperatures a transition to a nonergodic state at the spin freezing temperature $T_f \approx 2.8$ K. Below this, the ergodicity is broken on the experimental timescale, because the thermally activated correlation times for the spin reorientations become macroscopically long. The magnetic state achieved at low temperatures by continuous cooling in low magnetic fields is likely a superposition of metastable states with randomly frozen spins that have no long-range order yet undergo gradual spin-freezing dynamics and an AFM-like magnetically ordered state with critical slowing dynamics. The magnetic properties of the site-ordered, geometrically frustrated $Gd_3Au_{13}Sn_4$ system are discussed in comparison to site-disordered spin glasses that contain both randomness and frustration.