

Molecular designing of organic radicals and their quantum magnetic states

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Purely organic magnets with π -electron spins have essentially negligible small spin-orbit couplings and are attractive materials because they are archetypical $S = 1/2$ Heisenberg spin systems in which quantum fluctuations play an important role. The antiferromagnetic frameworks of isotropic $S = 1/2$ spins connected by the small number of the nearest neighbors are the key factors of the novel magnetic states arising from quantum fluctuations. Most extensive study has been done on one-dimensional (1D) Heisenberg antiferromagnets. Our recent study on a quasi-1D Heisenberg antiferromagnet, F5PNN, has established the direct 3D long-range order without the development of 1D short-range order near the saturation magnetic field [1]. Besides the study on 1D Heisenberg antiferromagnets, growing attention is being paid to the effect of quantum fluctuations in 2D or 3D Heisenberg antiferromagnets with the small number of the nearest neighbors (z), but the experimental realization is still rare.

The molecular designing toward high-dimensional magnets is presented. The key is the multidirectional magnetic exchange pathways by the twist of π -conjugated planes and polyradicals. By the usage of twisted monoradicals, 2D and 3D magnetic networks are realized. 3D network of pentagonal lattice and its novel state near the saturation field induced by frustration is described [2]. Planar or non-planar biradicals forming 2D and 3D lattices are also presented. A new organic biradical with twisted π -planes, F4BIPBNN, forms a new 3D honeycomb lattice with $z = 4$ [3]. The magnetic interactions were evaluated in the range of 4.3 to 6.6 K. The three-dimensional long-range order has been observed below $T_N = 2.7$ K. The effect of the quantum fluctuation appears in the concave shaped magnetization curve and the field dependence of T_N . The quantum fluctuation was evaluated as the shrinkage factor of spin to 28 % with respect to its classical value. This is the first observation of the quantum fluctuations among 3D magnetic lattice without spin frustration.