Jahn-Teller solid, liquid and gas in spin-crossover system Mn(taa)

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Jahn Teller (JT) distortions play important roles in many spin-crossovers (SCO) in molecule-based magnets. Long-range ordering of the JT distortions in the crystalline molecular magnet Mn(taa) has been shown to lead to magnetoelectric coupling, which is the interplay between magnetism and electric polarization or dielectric properties. Our theoretical study based on first-principles calculations and Monte Carlo simulations on Mn(taa) reproduces the SCO and magnetoelectric coupling. More importantly, a low-temperature Jahn-Teller ordered (solid) phase, an intermediate-temperature dynamically correlated (liquid) phase, and an uncorrelated (gas) phase are identified in the high-spin region. In particular, the Jahn-Teller liquid phase arises from competition between mixing with low-spin impurities, which drives the disorder, and inter-molecular strain interaction. The latter is a key factor in both the spin-crossover phase transition and the magnetoelectric coupling. Jahn-Teller liquids may exist in other spin-crossover materials and materials that have multiple equivalent Jahn-Teller axes.

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