

Pressure-induced reentrance of ferroelectricity in a molecular magnet

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Multiferroics have attracted tremendous research interests with their rich physics and potential in constructing next-generation multifunctional devices. Metal-organic framework molecular magnets have been a recent focus in single phase multiferroics investigations. These materials are usually bonded by intermolecular forces and their magnetic moments interact through H-bond super-exchange couplings. As a result, pressure induced lattice changes would play a significant role in exploring for complex phase transitions in these 'soft' materials. We investigated the crystal structure, magnetism and ferroelectricity changes of a molecular magnet $(\text{NH}_4)_2\text{FeCl}_5 \cdot \text{H}_2\text{O}$ under pressure. While $(\text{NH}_4)_2\text{FeCl}_5 \cdot \text{H}_2\text{O}$ demonstrates spiral magnet phase induced ferroelectricity at ambient pressure, it first reaches a critical paraelectric phase with a collinear magnetic order applying small pressures. Further increasing pressure, it reenters the spiral-magnetic-order driven ferroelectric phase. Combined with theoretical study, we show that pressure induced quantum tunneling of ammonium ions is the underlying mechanism for generating the critical paraelectric region sandwiched by two magnetic driven ferroelectric phases in this molecular magnet. Such quantum tunneling of ammonium has never been reported in magnetoelectric materials. Our discovery of the unique role of quantum tunneling in controlling magnetism in this molecular material under pressure brings new insights into the study of ferroelectrics, magnetism and molecular magnets in similar systems and contributes to finding new territory of functionable multiferroics.