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Interdependent scaling of long-range oxygen and magnetic ordering in non-stoichiometric Nd2NiO4.10

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Hole doping in Nd2NiO4.00 can be either achieved by substituting the trivalent Nd atoms by bivalent alkaline earth metals or by oxygen doping, yielding Nd2NiO4+δ. While the alkaline earth metal atoms are statistically distributed on the A-cation sites, the extra oxygen atoms in interstitial lattice remain mobile down to ambient temperature and allow complex ordering scenarios depending on δ and T. Thereby the oxygen ordering, usually setting in far above room temperature, adds an additional degree of freedom on top of charge, spin and orbital ordering, which appear at much lower temperatures. In this study, we investigated the interplay between oxygen and spin ordering for a low oxygen doping concentration i.e. Nd2NiO4.10. The presence of a complex 3D modulated structure related to oxygen ordering already at ambient was evidenced by single crystal neutron diffraction, the modulation vectors being ±2/13a±*3/13b*, ±3/13a±*2/13b*and ±1/5a±*1/2c* with satellites up to fourth order. The coexistence of oxygen and magnetic ordering below TN ≅ 48 K was evidenced, with magnetic satellite reflections adapting the same modulation vectors as found for the oxygen ordering, evidencing a unique coexistence of 3D modulated ordering for spin and oxygen ordering in Nd2NiO4.10. Temperature dependent measurements of magnetic intensities suggest two magnetic phase transitions below 48 K and 20 K, indicating two distinct onsets of magnetic ordering for the Ni and Nd sublattice, respectively.

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