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Orbital ordering and magnetic dimensionalities in the p-orbital spin-1/2 CsO₂ and Cs₄O₆ DENIS ARCON, Jozef Stefan Institute — The materials containing magnetic O₂⁻ anions, i.e., alkali superoxides, AO₂ (A = Na, K, Rb, Cs), and alkali sesquioxides, A₄O₆ (A = Rb, Cs), exhibit two key features that make them appealing for investigation of the coupling between lattice, orbital and spin physics as an alternative to the more established *d*-orbital materials. First, the O₂⁻ dumbbells can easily reorient down to the low temperatures, thereby modulating the overlaps of *p* orbitals. And second, as the *S* = 1/2 spin is localized in a pair of *p*-derived π* orbitals, their original degeneracy can be removed by the cooperative tilting of O₂⁻ dumbbells. Here we report on our studies of CsO₂ and Cs₄O₆ using ¹³³Cs nuclear magnetic resonance and electron paramagnetic resonance techniques. In CsO₂ we find the structural phase transition occurring at 61 K on cooling associated with the freezing out of the O₂⁻ librations. The transition also includes π* orbital ordering that is responsible for the quasi-one-dimensional low-temperature magnetism. Clear signs of the spin Tomonaga-Luttinger liquid state are found from the spin-lattice relaxation and spin susceptibility data. On the other hand, the mixed valence Cs₄O₆ shows much more complex phase diagram with several transitions depending on the exact cooling protocol.

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