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Exotic magnetism in the alkali sesquioxides Rb_4O_6 and Cs_4O_6

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Among the various alkali oxides the sesquioxides Rb_4O_6 and Cs_4O_6 are of special interest. They comprise two different types of dioxygen anions, the hyperoxide and the peroxide anions. The nonmagnetic peroxide anions do not contain unpaired electrons while the hyperoxide anions contain unpaired electrons in antibonding π^*-orbitals. Electronic structure calculations using the local spin-density approximation reveal strong electron correlations and charge ordering that is due to the mixed valency. The experimental investigation of the temperature dependent magnetization reveals a low-temperature magnetic transition. The mixed valency of both compounds is confirmed using Raman spectroscopy. The time- and temperature-dependent magnetization experiments indicate that both compounds follow a behavior being known from spin glasses and frustrated systems The frustration is explained by first principle calculations that incorporate the correlation between the oxygen $2p$-electrons and deal with the mixed-valent oxygen. This leads to a physical picture where the symmetry is reduced because one third of the oxygen anions in the oxides is nonmagnetic while the remaining anions are antiferromagnetically arranged. A degenerate, insulating ground state with a large number of frustrated non-collinear magnetic configurations is confidently deduced from the theoretical point of view. Further it is shown that the compounds exhibit a variety of interesting physical phenomena under high pressure. Around 75 GPa a transition from the insulating antiferromagnetic frustrated phase to a half-metallic ferromagnetic state takes place. At a pressure of 75 GPa all anionic oxygen molecules (peroxide and hyperoxide) carry magnetic moments. Finally, above 160 GPa a metallic phase appears, where all oxygen molecules become equivalent. It is demonstrated that the bond length differences of O_2^{2-} and O_2^- have a vital effect on magnetism and conductivity of the sesquioxides.