Density functional theory calculations of single-molecule magnets $Co_3(SALPN)_2(O_2CCH_3)_2 \cdot R_2$

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In search of candidate magnetic molecular systems that emulate Majorana zero modes, we perform density functional theory (DFT) calculations for Co trimers $Co_3(SALPN)_2(O_2CCH_3)_2 \cdot R_2$, where R is an OCH_2, $OCHNH_2$, or $OCHN(CH_3)_2$ solvent molecule. The three Co atoms form a one-dimensional chain, and each Co atom exhibits axial spin with S=3/2. We extract the exchange coupling constant and the local axial and rhombic zero-field splitting (ZFS) parameters based on DFT total energies. According to our calculations, the ratio between the exchange coupling constant and the local axial ZFS parameter is 7.3 for the end Co atoms and 5.7 for the center Co atom; the local magnetic easy axis of the center Co atom differs from that of an end Co atom by 42.9 degrees. These results provide a starting point for mapping the spin-3/2 Hamiltonian to an effective spin-1/2 Hamiltonian, which indicates the existence of Majorana zero modes. We will also analyze the d-orbital occupation matrix and discuss the necessity to control it for obtaining reliable results.

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