Supplementary Information

for

Unpaired spin populations and spin-pairing tendencies of the nonequivalent vanadium sites of the magnetic metal $\text{NaV}_6\text{O}_{11}$ investigated by electronic band structure calculations and spin dimer analysis

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Figure S1. Energies of the 200 K structure of $\text{NaV}_6\text{O}_{11}$ calculated as a function of the number of unpaired spins per formula unit, N , (with the pseudo-potential method of the VASP code), where the ground state energy of the RT crystal structure was used as the energy reference. The dashed lines refer to results of calculations carried out without restrictions on the wave functions, and the solid lines to those in which the wave function of one N -point calculation were used as the initial guess for the wave function of its neighboring N -point calculation.

Figure S2. Partial DOS calculated for the d-bands of V(2) and V(3) with the pseudo-potential method of the VASP code.

Top: The RT crystal structure $N = 8.25$.

Middle: The RT crystal structure with $N = 5.25$.

Bottom: The 200 K crystal structure with $N = 4$.