The main problem when synthesizing molecular ferromagnetic materials lies in the fact that magnetism is a solid-state property instead of being a molecular property. The magnetic ordering results from the magnetic coupling among intermolecular electronic interactions, which are due to the molecular packing. In turn, such packing is consequence of the molecular structure and electron distribution of the spin-containing radicals. The systematic variation of such radical units is often the only tool that an experimentalist might have in order to synthesize a ferromagnet. Thus, the designing of molecular magnets requires a proper knowledge of the microscopic magnetic interactions present between radical units and a suitable procedure for bringing together such microscopic interactions and the macroscopic properties of interest (e.g. magnetic susceptibility $\chi(T)$, heat capacity $C_p(T)$, ...).

Recently, we have applied a first-principles bottom-up theoretical procedure to the study of the magnetism in molecular crystals [1]. Data from electronic structure computations between pairs of radicals ($J_{AB}$) is used to define the magnetic topology of the crystal and parameterize an algebraic Heisenberg Hamiltonian. The magnetic susceptibility as a function of temperature $\chi(T)$ is, in turn, obtained directly from the computed energy levels of the algebraic Heisenberg Hamiltonian using standard statistical mechanics. Applications to both purely organic and metal-based molecular crystals have proved this first-principles bottom-up approach to be valid to picture the topology of the microscopic magnetic paths. Calculation of the experimental magnetic susceptibility and heat capacity data has been successfully carried out. Here, we present a discussion on the ferromagnetism of p-(methylthio)phenyl nitronyl nitroxide [2], weak ferromagnetism of the $\beta$-phase of the dithiadiazolyl radical p-NCC$_6$F$_4$CNSSN$^-$ [3], and the bistability of the monoclinic and triclinic 1,3,5-trithia-2,4,6-triazapentalenyl polymorphs [4].