Invited Lecture

Magneto-structural correlations in Ni^{2+}-halide···halide-Ni^{2+} chains from electron charge density analysis

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Hybrid organic inorganic materials are ideal platforms for the realisation of low-dimensional quantum magnets and the study of model Hamiltonians. In this context, the magnetic properties of the novel S = 1 NiX_2(3,5-lutidine)$_4$ materials with X = Cl, Br, or I have recently been reported by some of us.[1] Interestingly, in this family of materials a through-space two-halide exchange supports significant antiferromagnetic interactions over a large X···X distance for X = Br and I. However, a model based on elongation/compression of the Ni^{2+} octahedra cannot explain the observed single-ion anisotropy. Here, we present results from X-ray charge density determination and density functional theory calculations that describe the nature of chemical bonding and all possible pathways for the magnetic superexchange interactions. We show how chemical bonding descriptor obtained from charge density analysis may be able to capture correlations between structure and magnetic properties beyond the bond distance. These correlations will allow to engineer halide···halide interactions in quantum magnets.

[1] Blackmore, W.J., Curley, S.P., Williams, R.C., Vaidya, S., Singleton, J., Birnbaum, S., Ozarowski, A., Schlueter, J.A., Chen, Y.S., Gillon, B., Goukassov, A., Kibalin, I., Villa, D. Y., Villa, J. A., Manson, J. L. & Goddard, P. A. (2022) *Inorg. Chem.* **61**, 141−153.