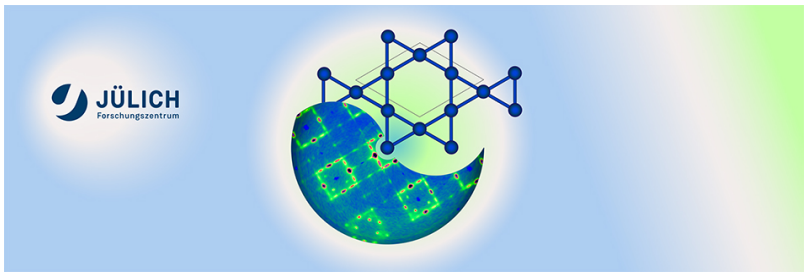

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Transition metal carbodiimides –A new playground for solid-state physics?

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Transition metal oxides, particularly those with partially filled 3d shells, have been the playground for solid-state physics for many decades. These correlated oxides exhibit a competition between coulombic repulsion, which tends to localize electrons, and hybridization, which promotes delocalization, that leads to numerous many-body effects such as metal-insulator transitions, colossal magnetoresistance and superconductivity, to name but a few. Traditionally the modification of such phases, to imbue different physical properties, has relied on the doping of one cation for another to adjust both the electron count and degree of correlation. However, an alternative strategy may also be employed via anion substitution. Enter the divalent carbodiimide anion, [−]N=C=N[−], which is well considered a pseudo-oxide, but with enhanced covalent character [1]. Indeed, numerous quasi-binary transition metal carbodiimides, M_x(NCN)_y, have now been prepared, including CuNCN which exhibits spin-liquid behavior and a resonating-valence bond ground-state [2]. Here, however, we will take a closer look at MnHf(NCN)₃ and FeHf(NCN)₃, which are the first examples of transition metal carbodiimides with a perovskite-like AB(NCN)₃ composition [3]. These quasi-ternary phases adopt a chiral crystal structure, with P6₃22 symmetry, and magnetometry measurements on MnHf(NCN)₃ evidence strong AFM coupling of Mn²⁺ centers, but no evidence for long-range order. This suggests a degree of magnetic frustration in MnHf(NCN)₃ and highlights that the types of quantum behavior observed in correlated oxides may also be accessible to their carbodiimide analogs.

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