Abstract Submitted
for the MAR13 Meeting of
The American Physical Society

First-Principles Modeling of Bonding and Magnetic Exchange in the Metal-TCNE Magnet Family[1] CHRISTOPHER OLSON, The Center for Nanoscale Science and Engineering, North Dakota State University, SHRUBA GANGOPADHYAY, SVETLANA KILINA, The Department of Chemistry and Biochemistry, North Dakota State University, KONSTANTIN POKHODNYA, The Center for Nanoscale Science and Engineering, North Dakota State University — The chemical bond and its role as a mediator of magnetic exchange interaction remains a crucial aspect in the study of molecular magnetism. Within the M-TCNE (M=3d metal; TCNE=tetracyanoethylene) class of organic-based magnets, only V[TCNE]$_x$ (x$\sim$2) orders magnetically above room-temperature (T_c $\sim$400 K), while structural factors underlying this exceptional behavior remain elusive. Conversely, Mn-TCNE complexes of diverse crystal structure, e.g., 1D-chain MnTPP[TCNE] (T_c $\sim$10 K), 2D-layer [Mn(TCNE)(NCMe)$_2$]SbF$_6$ (T_c $\sim$75 K), and 3D-network [Mn(TCNE)$_{1.5}$](I$_3$)$_{0.5}$(T_c $\sim$170 K) have recently become available. Using this structural data, hybrid DFT simulations has been performed and the spin-polarized electronic structures resolved. The nature of bonding and non-bonding orbital interactions crucial for understanding magnetic behavior was revealed. Orbital ordering, hybridization, and trends in spin-density transfer (bonding/backbonding) as well as the formation of exchange/superexchange pathways have been identified and interpreted in terms of the dimensionality of magnetic interaction. The role of these and additional factors in establishing high-T_c magnetism in the broader M-TCNE class will be discussed.

[1]Support from NSF DMR-CMP-1005882 is acknowledged.

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Date submitted: 18 Nov 2012 Electronic form version 1.4