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Bonding and Magnetic Exchange in Metal-[TCNE] Magnets[1]

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The correlation between indirect magnetic exchange and bonding is an outstanding question in condensed matter physics and is also directly related to understanding the mechanism that governs magnetic ordering temperature, electron delocalization and Coulomb correlations resulting in high carriers' spin-polarization in molecular-based magnetic solids. The relationship between this magnetic exchange pathway and the chemical bonding that holds the solid together is yet unclear. In the first room-temperature molecule-based magnetic semiconductor V[TCNE]$_x$ (TCNE = tetracyanoethylene; $x \sim 2$), ferrimagnetism was attributed to a strong antiferromagnetic exchange between unpaired $V^{II} 3d$ t_{2g} electrons and a delocalized unpaired electron residing on the singly occupied TCNE π^* molecular orbital, while both these states do not participate in chemical bonding, yet they exhibit a strong indirect (e.g., super, double) magnetic exchange. Resolving this problem can provide an indispensible information allowing to achieve a long term goal of direct spin polarization injection into a semiconductor, and presents an exciting opportunity for incorporating organic-based magnets into contemporary data processing systems.

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