Exotic Metals from Geometrical Frustration

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Transition metal compounds with a geometrically frustrated lattice, such as a triangular and a pyrochlore lattice, exhibit a strong tendency toward valence-bond solid (VBS) states. In particular, when the t_{2g} orbitals are partially occupied, the constituent transition metal ions form complex "molecular" clusters in the VBS states: trimer in LiVO₂, heptermer in AlV₂O₄, helical dimer in MgTi₂O₄, and octermer in CuIr₂S₄. The orbital degree of freedom plays an important role of the formation of "molecular" clusters. Thus it is tempting to melt the VBS to see if novel electronic phases appear and how the electronic phase evolves when the orbital degree of freedom is incorporated in the geometrically frustrated systems.

In order to tackle this issue, we focus our attention on a series of compounds, $LiVO_2$, $LiVS_2$, and $LiVSe_2$. By replacing O to S and Se, one can apply "negative" pressure, which destabilize the VBS state of $LiVO_2$. In addition, one can increase the overlap between V 3d and p-orbital (O 2p, S 3p, and Se 4p), thus, decrease the on-site Coulomb repulsion U and increase the band width, which drive the system into metallic side.

We found that the solid solution $\text{LiVS}_{2-x}\text{Se}_x$ with x < 0.3 undergoes a metal-to-insulator (MI) transition at the same time of the VBS formation at T_c . The VBS phase was suppressed at $x_c \simeq 0.3$ and metallic phase was realized down to low temperatures for x > 0.3. In this metallic phase, the Pauli susceptibility showed significant reduction by approaching the MI phase boundary. This behavior contrasts markedly with that of the canonical Mott systems, in which the susceptibility is enhanced near the MI phase boundary. We suggest that the "spin pseudogap metal" is realized in this metallic phase, which might be characteristic of geometrically frustrated systems when the orbital degree of freedom is incorporated.