

Expanding Possibilities of π electrons in Molecular Conductors

Reizo KATO (RIKEN)

Molecular p-systems: Clean, Soft, Clear

- Small in Quantity and Size, Fragile
- High Quality Measurements: Sensitivity, Accuracy, Resolution
- Multiple Extreme Conditions:

Strong Magnetic Field, High Pressure, Low Temperature



p-frustration system (EtMe₃P[Pd(dmit)₂]₂)



 $p-F \in I$ (k-(ET)₂Cu[N(CN)₂]Br / SiO₂)



Exotic Properties of p Electrons Based on Various Degrees of Freedom in a Mott system with a quasi-triangular lattice



	Cation		Layer 1 a	Layer 2	
Me₄N Me₄P Me₄As	EtMe ₃ N EtMe ₃ P EtMe ₃ As	Et ₂ Me ₂ N Et ₂ Me ₂ P Et ₂ Me ₂ As	for a for the formation of the formation		
Me ₄ Sb	EtMe ₃ Sb	Et ₂ Me ₂ Sb	O J	<u> </u>	

• Spin

Charge

Dimer

Cation

Cation

Orbital (HOMO-LUMO interplay)

Lattice

HOMO-LUMO Level Crossing in the [Pd(dmit)₂]₂ Dimer

Strong dimerization: [Pd(dmit)₂]₂⁻



E. Canadell, et al., Solid State Commun. 75 (1990) 633

Electronic Structure of (Cation)[Pd(dmit)₂]₂

At ambient pressure, most of Pd(dmit)₂ salts behave as Mott insulators where one spin is localized on each dimer. Electron Magnetic frustration $[Pd(dmit)_2]_2$ $\boldsymbol{E}(\boldsymbol{k}) = 2.0 \left[\boldsymbol{t'} \cos \boldsymbol{ka} \right]$ $+t\cos k(a-b)$ prevents long-range $+t\cos kb$] antiferromagnetic order **Transfer integral** Effective on-site Coulomb Deviation from the regular →A≫*t*≥ *t*'→ energy on the dimer triangular lattice (t'/t)M. Tamura and R. Kato, JPSJ, 73, 3108 (2004) Band width *W* **Correlation parameter Frustration parameter**

Mott Insulator with a Quasi-Triangular Lattice

b'-(Cation)[Pd(dmit)₂]₂

Magnetic susceptibility



Valence bond (VB) formation -Transitions towards a singlet state-



Pressure-induced metallic and superconducting states



Pump-probe time resolved spectroscopy for $Et_2Me_2Sb[Pd(dmit)_2]_2$ (T_{CO} = 70 K)



EtMe₃P[Pd(dmit)₂]₂ (Monoclinic): Superconductivity and Valence Bond Solid





The valence bond solid phase neighbors the superconductor and metal phases

Y. Shimizu et al, Phys. Rev. Lett., 99 (2007) 256403



Field effect transistor (FET) behavior of π electrons in an organic Mott insulator

Organic Mott FET





Expanding Possibilities of IMSS for Molecular Conductors

Reizo KATO (RIKEN)

"Advanced Tools" inspire "Advanced Sciences".



p-frustration system Bulk crystal



In collaboration with

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