Charge Separation in Et₂Me₂Sb[Pd(dmit)₂]₂ system under Hydrostatic Pressure

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Et₂Me₂Sb[Pd(dmit)₂]₂ undergoes a novel Mott insulator-charge separation (MI-CS) transition at 70K[1], stabilized by the LUMO-HOMO interplay in [Pd(dmit)₂]₂[2]. This salt exhibits the MI-CS transition in lower pressure region (P<5kbar), and the metal-charge separation (M-CS) transition in higher pressure region (P>5kbar). We have developed the specific heat measurements system under hydrostatic pressure using the differential thermal analysis (DTA) method [3] to quantitatively discuss a mechanism of the MI-CS and the M-CS phase transitions. We observed the large entropy difference ΔS=8 J/mol·K by spin and lattice degrees of freedoms at the MI-CS transition and small one ΔS=5 J/mol·K by the free electron and lattice ones at the M-CS transition, respectively. In the pressure region where the three-phase competes, ΔS rapidly decreases and shows a critical behavior.