

Structural study on charge ordered state of DODHT salts

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We have been investigating molecular conductors based on a reduced π donor, DODHT [(1,4-dioxane-2,3-diylidithio)dihydrotetrathiafulvalene], and found pressure-induced superconductors, β -(DODHT)₂X (X = PF₆ and AsF₆). Both the salts showed superconductivity under pressure, but the physical properties such as transport and magnetic properties were apparently different; a clear insulator transition was observed for the PF₆ salt but that of the AsF₆ salt was not clear, and the magnetic susceptibility of the PF₆ salt followed a 1-D alternating Heisenberg chain model but that of the AsF₆ salt did not and decreased abruptly to a non-magnetic state at 90 K. The insulating phase of the PF₆ salt at ambient pressure has already been revealed to be a charge ordered (CO) state with stripe-type CO pattern. In order to clarify the insulating state of the AsF₆ salt including the different physical behaviors, we carried out X-ray structural experiments using synchrotron radiation. From the oscillation photographs of the AsF₆ salt taken below 90 K and the precise structure analyses at 150 K and 250 K, which are just below and above the temperature where the b-axis was significantly increased, we conclude the insulating phase of the AsF₆ salt was also CO but the CO pattern was different from the PF₆ salt. Furthermore, the electronic band structure changed from quasi 2-D to 1-D below 250 K. Although both the salts showed superconductivity under pressure, their insulating states at ambient pressure were completely different; in the AsF₆ salt, CO and spin Peierls transition occurred in stages at 235 K and 90 K, which is in contrast to the case of the PF₆ salt exhibiting only the CO insulator transition at 255 K.