Magnetic monitoring electrochemical properties in strongly correlated Cu-BHT

Yigang Jin, Qi Chen, Kunio Awaga

Graduate School of Science, Nagoya University, Nagoya 464-8602, Japan

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Two-dimensional (2D) π−d conjugated coordination polymers (CPs) have found widespread application as active materials in secondary batteries[1]. Despite their success, the underlying reason for their high charge storage capacity remains unclear. Some studies have suggested a mechanism involving the formation of multiple radicals on a single organic ligand. However, to date, no conclusive evidence supports this mechanism, and it stands in contrast to the established resonance theory[2]. In this study, we employed various magnetometric techniques to track the formation and concentration of paramagnetic species during the electrochemical process of 2D strongly correlated Cu-BHT (BHT = benzenehexathiolate)[3]. Surprisingly, the spin concentration of the fully reduced (discharged at 1.5 V) electrode was found to be very minimal, significantly lower than that expected for a "diradical" form. More intriguingly, we simultaneously observed a significant temperature-independent paramagnetic term. This observation suggests an enhancement of delocalized π electrons in the discharged state, pointing towards a mechanism where π-electron bands accept/donate electrons during the charge/discharge process in this strongly correlated system.

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