Linking electronic transport through a spin crossover thin film to the molecular spin state using X-ray absorption spectroscopy operando techniques

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One promising route toward encoding information is to utilize the two stable electronic states of a spin crossover molecule. While this property is clearly manifested in transport across single molecule junctions, evidence linking charge transport across a solid-state device to the molecular film's spin state has thus far remained indirect. To establish this link, we deploy materials-centric and device-centric operando experiments involving X-ray absorption spectroscopy. We find a correlation between the temperature dependencies of the junction resistance and the Fe spin state within the device's [Fe(H2B(pz)2)2(NH2-phen)] molecular film. We also factually observe that the Fe molecular site mediates charge transport. Our dual operando [1] studies reveal that transport involves a subset of molecules within an electronically heterogeneous spin crossover film. Our work confers an insight that substantially improves the state-of-the-art regarding spin crossover-based devices, thanks to a methodology that can benefit device studies of other next-generation molecular compounds.

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[1] Studniarek, M.; Halisdemir, U.; Schleicher, F.; Taudul, B.; Urbain, E.; Boukari, S.; Hervé, M.; Lambert, C.-H.; Hamadeh, A.; Petit-Watelot, S.; et al. Probing a Device's Active Atoms. Adv. Mater. 2017, 29 (19), 1606578.