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Emergence of cooperativity in the coverage-dependent spin transition of an Fe(II) complex on a graphite surface

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The spin-state manipulation of spin crossover molecules (SCMs) adsorbed on a surface between a paramagnetic high-spin state and a diamagnetic low-spin state has attracted significant interest due to its potential for application in spintronic devices. The absence or presence of cooperativity in the spin transition and their characteristics is one of the most important issues for research, since it is responsible for the bistability in the spin states of SCMs. We present temperature- and coverage-dependent x-ray absorption spectroscopy measurements obtained at the VEKMAG and the high-field diffractometer end stations at BESSY II of the complex $[Fe(H_2B(pz)_2)_2(bipy)]$ deposited on a highly oriented pyrolytic graphite surface, investigating the ultimate scale limit at which cooperative spin switching becomes effective. This system exhibits a complete thermal- and light-induced spin transition. On increasing the coverage, the width of the temperature-induced spin transition curve narrows significantly, evidencing the buildup of cooperative effects. While the molecules at submonolayer coverage exhibit an apparent anticooperative behavior, the multilayers starting from a double-layer exhibit a distinctly cooperative spin switching. These observations will serve as useful yardsticks in designing SCM-based devices.

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