Smart molecular/MoS_2 Heterostructures to Thermally-Induced Strain Driven by Spin Switching

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Layered two-dimensional transition-metal dichalcogenides (TMDCs) have garnered intense attention due to the different properties achieved by modifying their composition and number of layers. One of the most studied members of this family is the MoS_2. This compound is an indirect bandgap semiconductor that turns to direct-gap when is exfoliated from bulk down to a monolayer, which makes it very promising for its integration in electronic, optoelectronic, and photovoltaic devices.[1] Recently, many studies have focused on altering its electrical and optical response via strain engineering.[2]

In this work, we propose an original approach to induce a reversible strain on MoS_2 ultrathin layers. It consists of the 2D surface functionalization with switchable molecular systems that undergo a reversible volume change upon applying an external stimulus. In this context, we propose using spin-crossover (SCO) nanoparticles as mechanical actuators. These SCO materials can change their ground spin state between low spin (LS) and high spin (HS) upon the exertion of various external physical or chemical stimuli, resulting in a variation in the nanoparticles volume depending on the spin state.
In this context, we have achieved the decoration of MoS_2 flakes with SCO nanoparticles (Figure 1a) of the well-known SCO compound $[Fe(Htrz)_2(trz)](BF_4)@SiO_2$.[3] This material presents a transition above room temperature with a broad hysteresis, 40 K, and a change in volume of ca. 10 %. The hybrid composite exhibits an abrupt change in conductivity near the NPs transition temperature (Figure 1b), which can be associated with a decrease in the bandgap. Even more, the narrowing of the bandgap, caused by the spin transition, is monitored by following the composite photoluminescence in both spin states. Corroborating the conductivity results, the expected energy shift of the MoS_2 bandgap is detected after the spin transition (Figure 1c).

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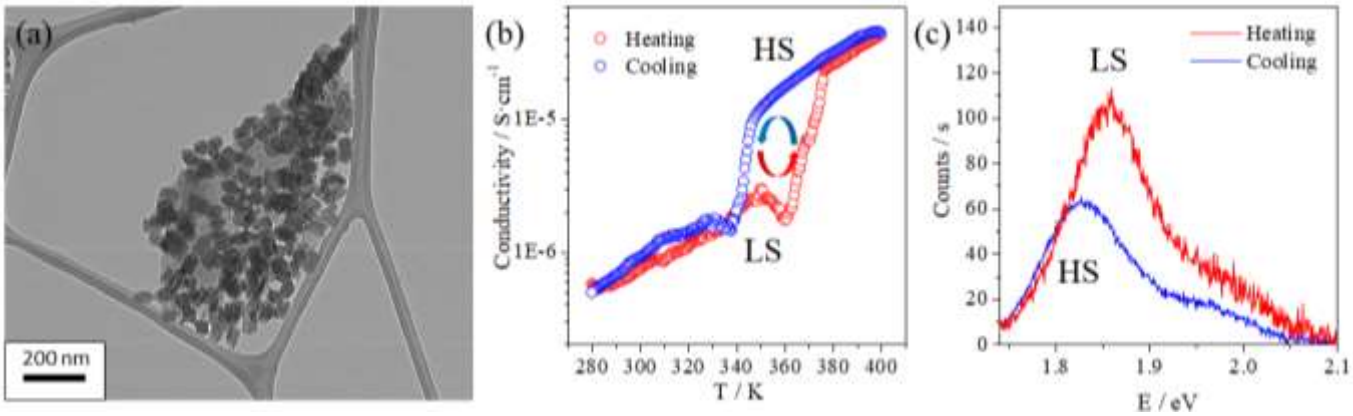

Figure 1: (a) HR-TEM images of the decorated MoS_2. (b) thermal variation of the conductivity in the heating and cooling modes of the heterostructure. (c) PL spectra of the same composite in LS and HS state.