Supporting Information

Synergistic Electrical Bistability in a Conductive Spin Crossover Heterostructure

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Experimental Details

Synthesis and Fabrication A microcrystalline powder of **Fe-trz** was synthesized by following methods described in the literature[39]: methanolic solutions (5 mL) of $Fe(BF_4)_2 \cdot 6H_2O$ and 1,2,4-triazole were mixed in a stoichiometric ratio (2 mmol : 6 mmol). The obtained suspension was stirred for an additional 3 hours, and a purple powder of **Fe-trz** was collected by centrifugation and dried in N_2 (yield ≃40%). Then, in typical cases, 50 mg of **Fe-trz** was mixed with 50 mg of **PC** and 2 mL $CHCl_3$ under magnetic stirring to yield a viscous suspension. The suspension was gently warmed on a 75°C heating plate and concentrated to ≃0.5 mL to reduce aggregation by the shearing force; then, an additional 0.5 mL of $CHCl_3$ was added to restore the fluidity of the suspension. Finally, the suspension was drop-casted into monoliths on glass slides or into the SCO-active heterostructure on pre-fabricated polyimide/constantan alloy/polyimide plates. Following the deposition process, a micrometer was used to identify a thickness (including 70 μm for the blank plate) of 150 μm for the **Fe-trz**/**PC** heterostructure and 170 μm for the **PC** compensator.

General Characterization The IR spectra were recorded using a Thermo Nicolet AVATAR 330 FT-IR spectrometer (**Figure S5**). PXRD for the **Fe-trz** and VT-PXRD for the **Fe-trz**/**PC** were performed on a Bruker D8 X-Ray diffractometer with Cu Kα radiation (**Figure S6**). The optical photos were captured with an Olympus SZX7 microscope on a Linkam THMS600 heating and freezing stage. The TEM images were obtained using a JOEL JEM-2010HR TEM, where a thin slice of a **Fe-trz**/**PC** monolith (which was taken from the same batch as the sample used for the optical observation) was prepared by ultramicrotomy after embedding the sample in epoxy hardeners.

Magnetic and Electrical Measurements The magnetic susceptibility measurements were performed on a Quantum Design PPMS VSM Option by sweeping at 2 K/min with an applied field of 5 kOe. The electrical resistivity was measured using a PPMS Resistivity Option by sweeping at 1 K/min. The Wheatstone bridge was constructed by trigger-linking a Keithley 2400 source meter and a Keithley 2182A nanovoltmeter operating in DELTA mode to cancel out the thermoelectric voltage. Temperature control was achieved using a Linkam THMS600 heating and freezing stage or an IKA RET heating stage operating in settle mode. An additional heat shield was fabricated from aluminum foil to homogenize the temperature. The corresponding V, I, T data were recorded by a computer with RS-232 interfaces running a customized program (**Figure S7**). For each series of consecutive cycles, the first cycle, which was regarded as the "run-in" process, was omitted.

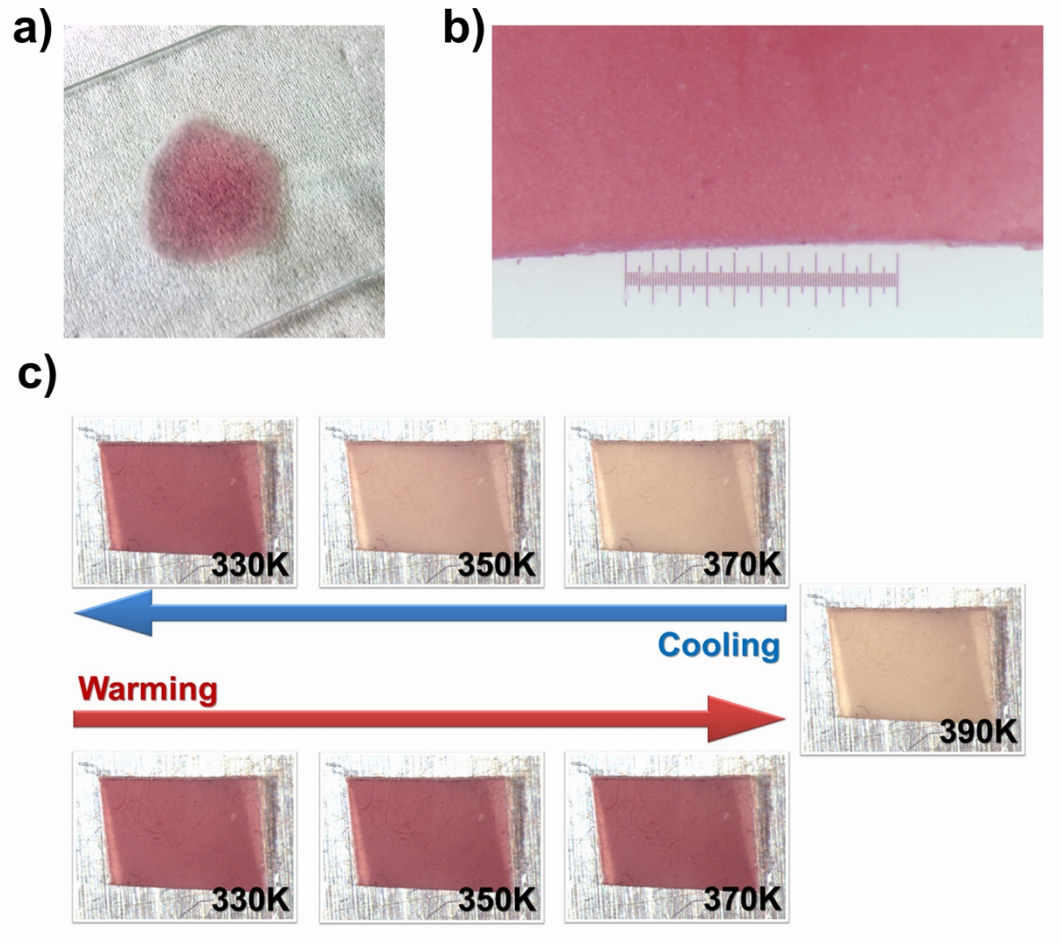

Figure S1 a) An **Fe-trz/PC** monolith on a glass slide at room temperature b) An optical microscopic photo of an **Fe-trz/PC** monolith next to the ruler (l = 1 mm, d = 10 μm. c) Color change and bistability of an **Fe-trz/PC** monolith during SCO observed under an optical microscope.

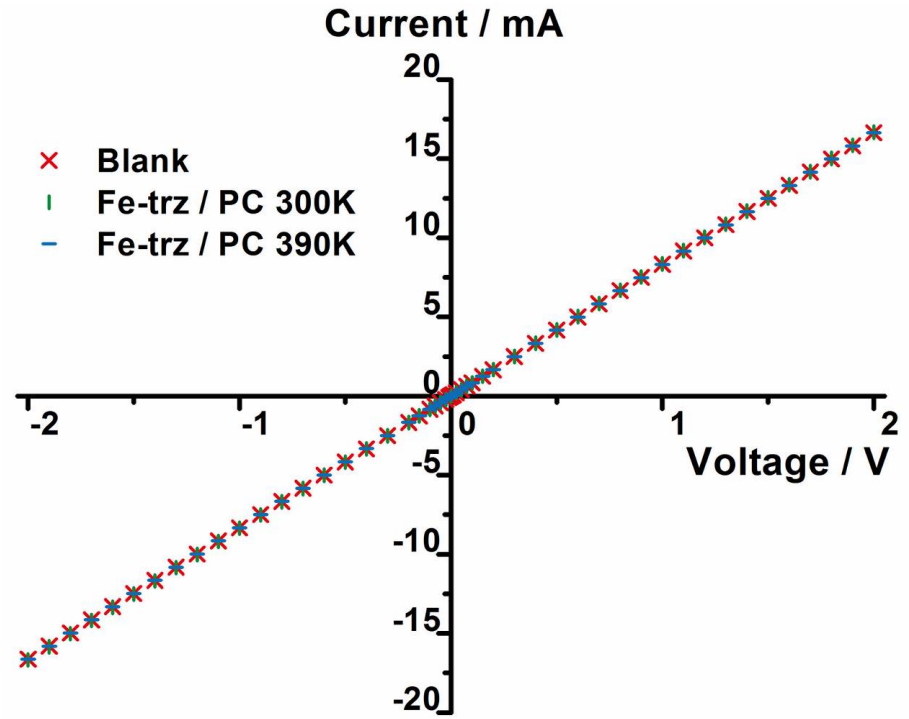

Figure S2 Current (*I*) to voltage (*V*) response of the blank plate and the heterostructure.

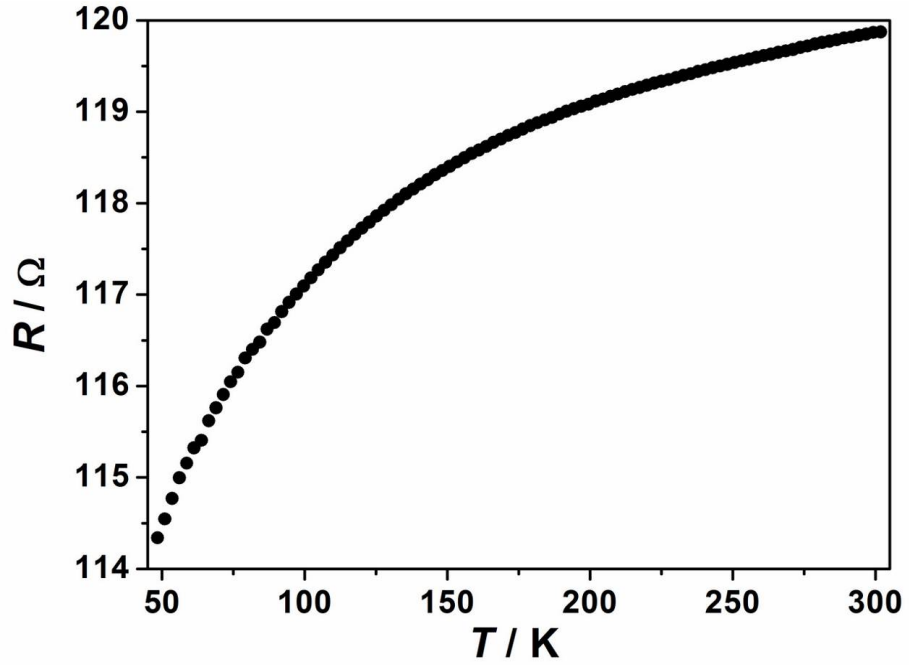

Figure S3 Temperature-dependent conductance for a blank plate measured in warming mode.

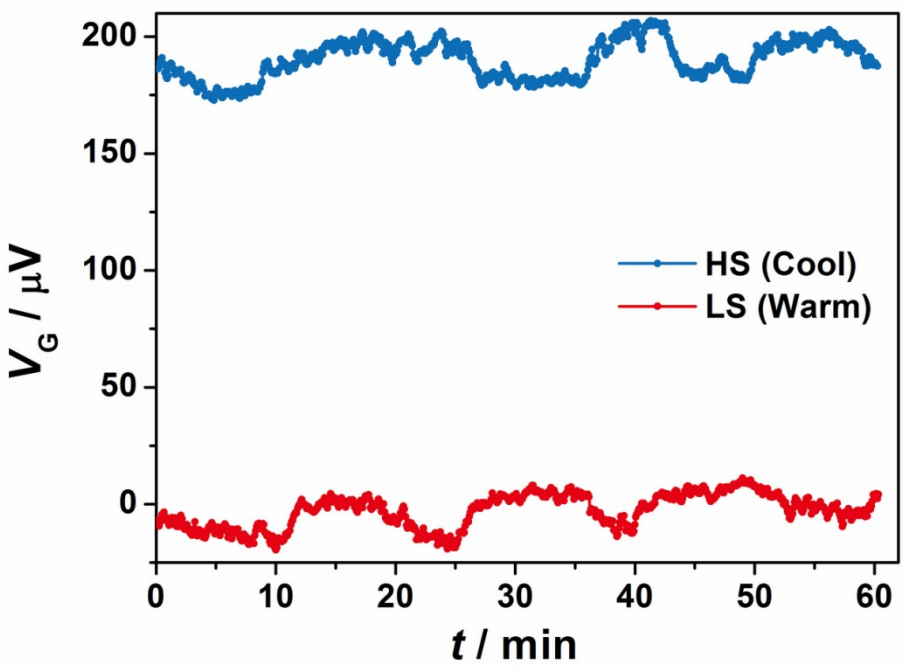

Figure S4 The Wheatstone bridge voltages stabalized at 360 K in LS state (after warming from 320 K) and HS state (after cooling from 380 K) measured over time.

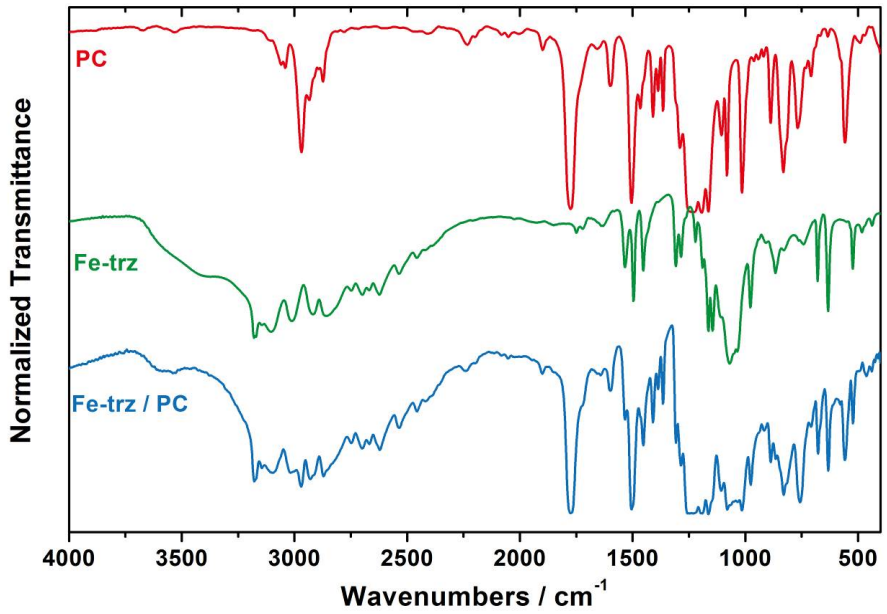

Figure S5 IR spectra of pure **PC**, as-synthesized **Fe-trz** and **Fe-trz**/**PC** composite.

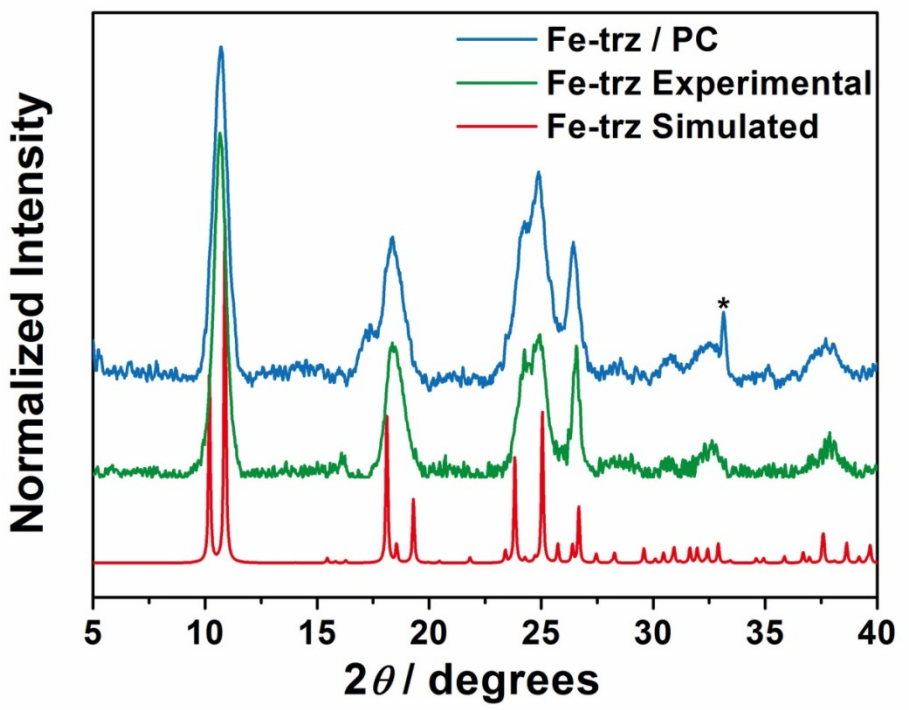

Figure S6 PXRD patterns of as-synthesized **Fe-trz** and **Fe-trz/PC** composite compared with a simulated pattern; **PC** is in amorphous state when deposited from solvent; the peak marked with an * is from the VT-PXRD sample holder.

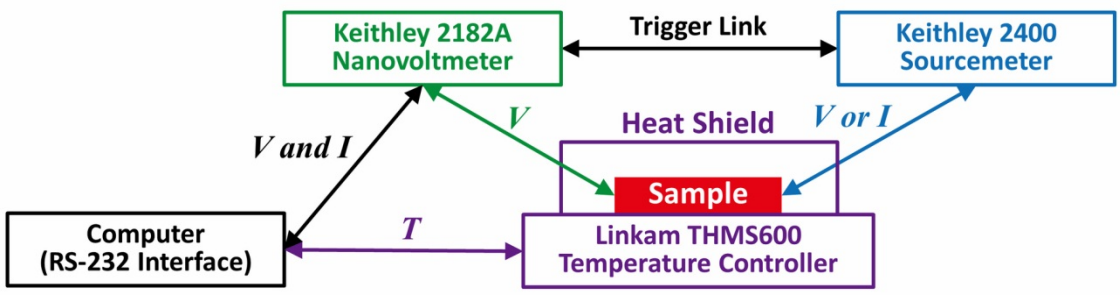

Figure S7 Schematic of the experimental apparatus for variable-temperature electrical measurements and data acquisition.