Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting Information for

Spin Crossover Composite Materials for Electrothermomechanical Actuators

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1. *Influence of the cantilever size on the actuating properties*

A second bilayer cantilever with the same composition, but smaller size (when compared to the one described in the manuscript) was fabricated and investigated. The geometry of this cantilever is characterized by its length L = 2300 µm, the thickness of the active layer $a_1 = 60$ µm and the thickness of the conductive layer $a_2 = 30$ µm. The aim of the size reduction was to achieve a faster and more efficient heat transfer to the cantilever. The actuating behaviour of this device was observed when changing the magnitude of the applied ac current, its frequency or the thermostat temperature (Fig. S1 - S3) and it was found very similar to the device described in the main text (Fig. 5 - 7). However, since the geometry and the thus the heat transfer properties are different for the two actuators, the cut-off values of I_{max}, f and T were found to be different for two devices of different geometry.

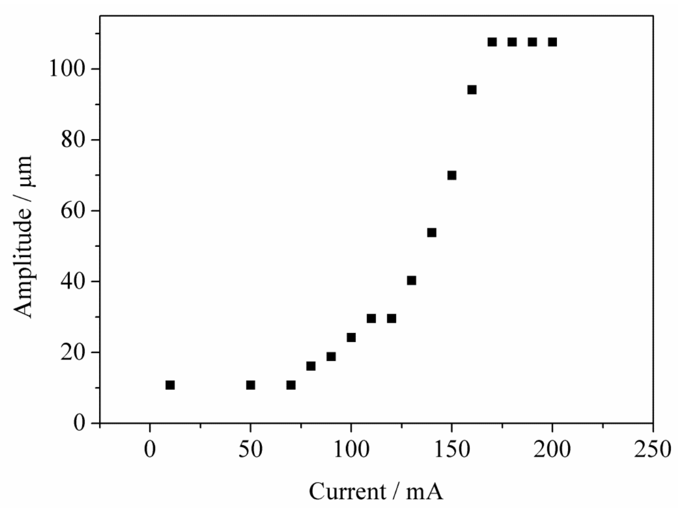

Fig. S1. Amplitude of oscillation of the second bilayer cantilever as a function of the applied ac current amplitude at constant temperature and frequency (T = 20 °C, f = 0.5 Hz).

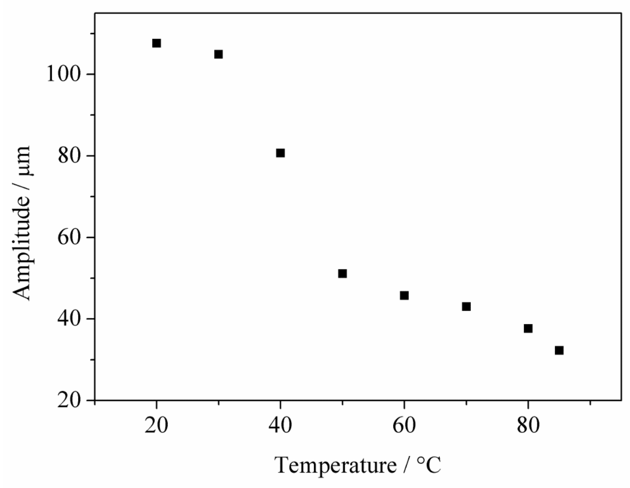

Fig. S2. Amplitude of oscillation of the second bilayer cantilever as a function of temperature. The applied ac current and the frequency were fixed (I_{max} = 180 mA, f = 0.5 Hz).

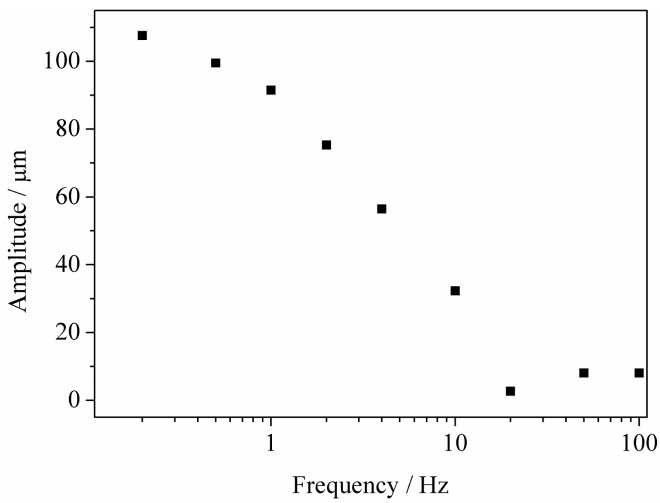

Fig. S3. Amplitude of oscillation of the second bilayer cantilever as a function of frequency at constant temperature and applied ac current (T = 20 °C, I_{max} = 180 mA).

2. *Calculation of the main actuating properties*

As we have described previously,[11] the curvature (k) of a bilayer strip that is purely related to the spin transition can be described *via* Timoshenko's theory:

$$k = \frac{6 \cdot \frac{\Delta L}{L} \cdot \left(1 + m^2\right)}{h\left[3\left(1 + m^2\right) + \left(1 + mn\right)\left(m^2 + \frac{1}{mn}\right)\right]} \tag{1}$$

where m is a ratio of layer thicknesses expressed as

$$m = \frac{a_1}{a_2} \tag{2}$$

n is a ratio of the Young's moduli (E) for the two materials

$$n = \frac{E_1}{E_2} \tag{3}$$

h is the thickness of the cantilever

$$h = a_1 + a_2 \tag{4}$$

and $\Delta L/L$ is the relative change of the composite linear size related to the spin transition.

In case of a small tip deflection (δ) the corresponding curvature (k) can be obtained as

$$k = \frac{2\delta}{L^2} \tag{5}$$

By considering a tip displacement of 1600 μm the curvature is $7.2 \cdot 10^{-5}$ μm^{-1}.

In case of large displacements that are observed in our experiments the curvature must be calculated more precisely from figure S4.

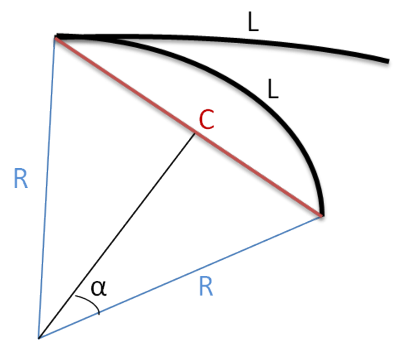

Fig. S4. Geometry of a bent strip with a large curvature.

From this figure the length of the chord C is

$$C = 2R\sin(\alpha) \tag{6}$$

where $R = 1/k$.

The corresponding arc L is

$$L = 2R\alpha \tag{7}$$

Thus

$$\operatorname{sinc}(\alpha) = \frac{\sin\alpha}{\alpha} = \frac{C}{L} \tag{8}$$

From the images of the cantilever in the LS and HS states we determined C_{LS} = 5450 μm and C_{HS} = 5980 μm. This gives α_{LS} = 1.08 rad and α_{HS} = 0.8 rad, hence $k_{LS} = 3.24 \cdot 10^{-4}$ μm^{-1} and

k_{HS} = 2.40·10^{-4} µm^{-1}. Thus the curvature related to the spin crossover $k = k_{LS} - k_{HS}$ = 8.4·10^{-5} µm^{-1}, in reasonably good agreement with the curvature found from the tip displacement (eq. 5).

Even though Timoshenko's theory does not allow the exact determination of the principal mechanical properties of the composites, it still permits the estimation of some of them. Considering the curvature k = 8.4·10^{-5} µm^{-1} and the Young's modulus of the composite (~ 3 GPa), the linear strain effect of the SCO could be estimated as ~1 % (Fig. S5).

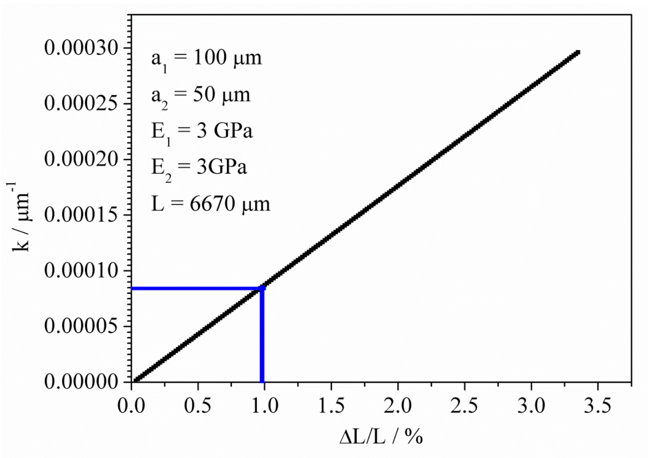

Fig. S5. Theoretical prediction of a cantilever tip displacement as a function of the linear strain induced by the spin transition. The blue line corresponds to the experimentally obtained maximum curvature.

The reactive force at the end of a cantilever (F) can be calculated from

$$F = \frac{3EI\delta}{L^3} \tag{9}$$

where EI is the flexural rigidity

$$EI = \frac{(a_1^2 E_1)^2 + (a_2^2 E_2)^2 + 2a_1 a_2 E_1 E_2 (2a_1^2 + 3a_1 a_2 + 2a_2^2)}{12(a_1 E_1 + a_2 E_2)} \times b \tag{10}$$

and b is a width of the cantilever. These calculations yield a value of $F \approx 20$ mN.

The energetic efficiency of an actuating material can be described *via* the volumetric work density

$$\frac{W}{V} = \frac{E\varepsilon^2}{2} \tag{11}$$

where ε is the linear strain generated in the composite when the SCO takes place. This gives a W/V value of ca. 150 mJ cm^{-3}.

Supplementary Movie SM1 (separate file): Oscillation of the bilayer cantilever (I_{max} = 300 mA, f = 0.1 Hz, T = 20 °C).