

## Supplementary Information

### Evidence for Surface Effects on the Intermolecular Interactions in Fe (II) Spin Crossover Coordination Polymers

Thilini K. Ekanayaka,<sup>1\*</sup> Hannah Kurz,<sup>2</sup> Kayleigh A. McElveen,<sup>3</sup> Guanhua Hao,<sup>1,4</sup> Esha Mishra,<sup>1</sup>  
Alpha T. N'Diaye,<sup>4</sup> Rebecca Y. Lai,<sup>3,5</sup> Birgit Weber,<sup>2</sup> Peter A. Dowben<sup>1</sup>

1. Department of Physics and Astronomy, Jorgensen Hall, University of Nebraska, Lincoln, NE 68588, U.S.A.

2. Inorganic Chemistry IV, University of Bayreuth, Universitätsstrasse 30, NW I, 95447 Bayreuth, Germany.

3. Department of Chemistry, Hamilton Hall, University of Nebraska, Lincoln, NE 68588, U.S.A.

4. Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, U.S.A.

5. Nebraska Center for Materials and Nanoscience, University of Nebraska-Lincoln 68588

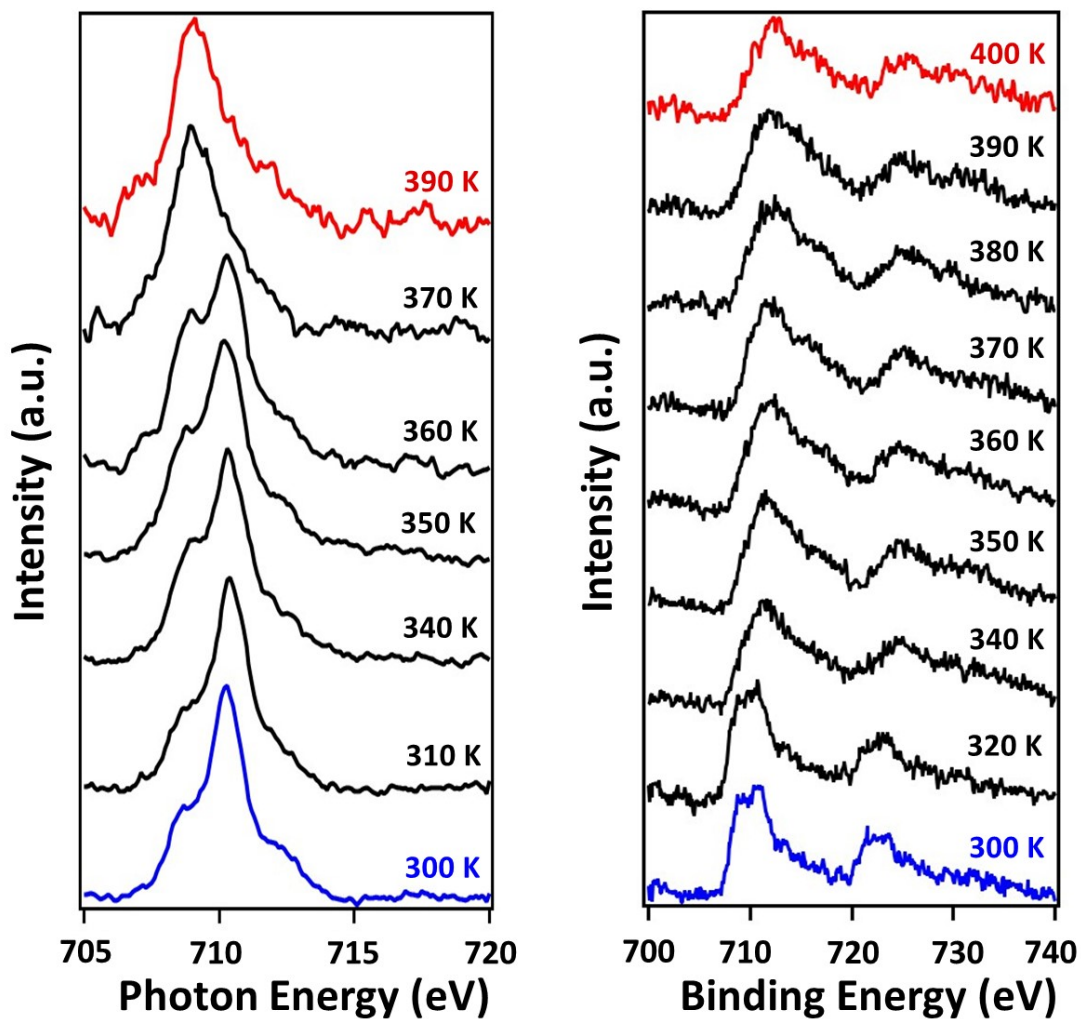


Figure S1: The temperature dependent X-ray absorption spectra and X-ray photo emission spectra of  $[\text{Fe}(\text{L1})(\text{bipy})]_n$  (complex **1**) for temperature increasing, demonstrating reproducibility of these measurements, as these results closely resemble those in the main paper. Blue indicates the Fe  $2p_{3/2}$  - L3 edge spectrum of the low spin (LS) state and red indicates the spectrum of the high spin (HS) state

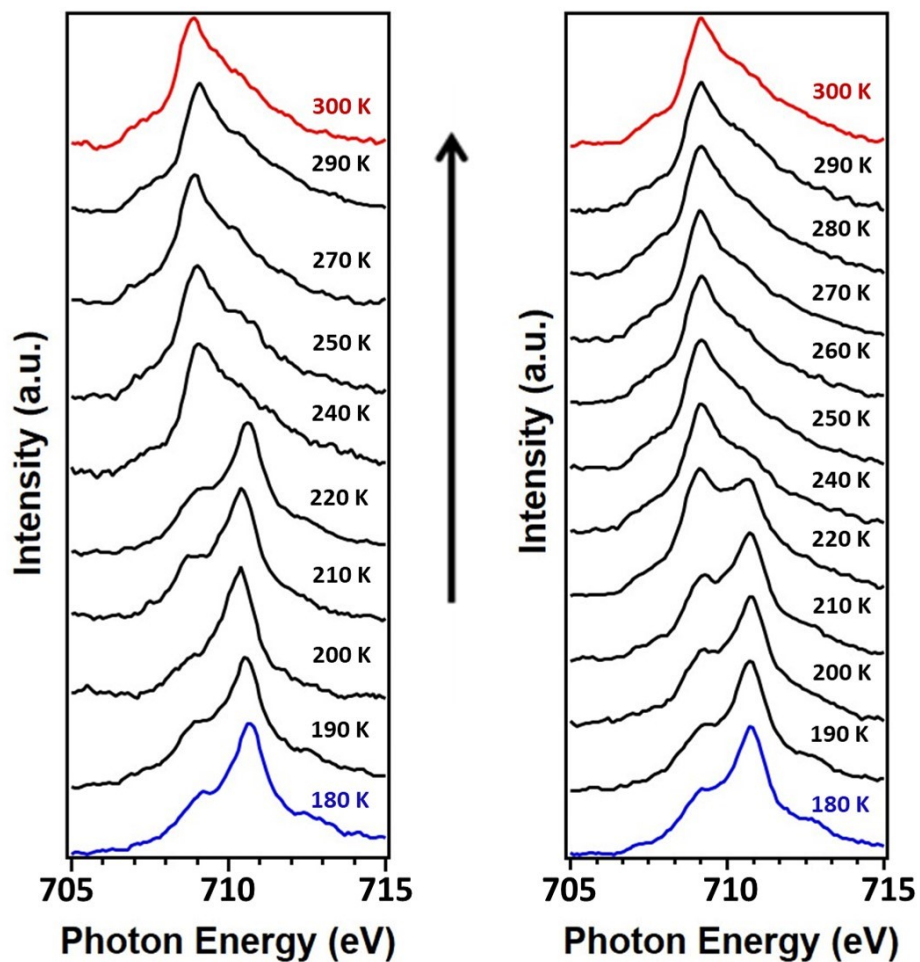


Figure S2: The temperature dependent X-ray absorption spectra of  $[\text{Fe}(\text{L}2)(\text{bipy})]_n$  (complex **2**) for both heating and cooling. Blue indicates the Fe  $2p_{3/2} - L_3$  edge spectrum of the low spin (LS) state and red indicates the spectrum of the high spin (HS) state.

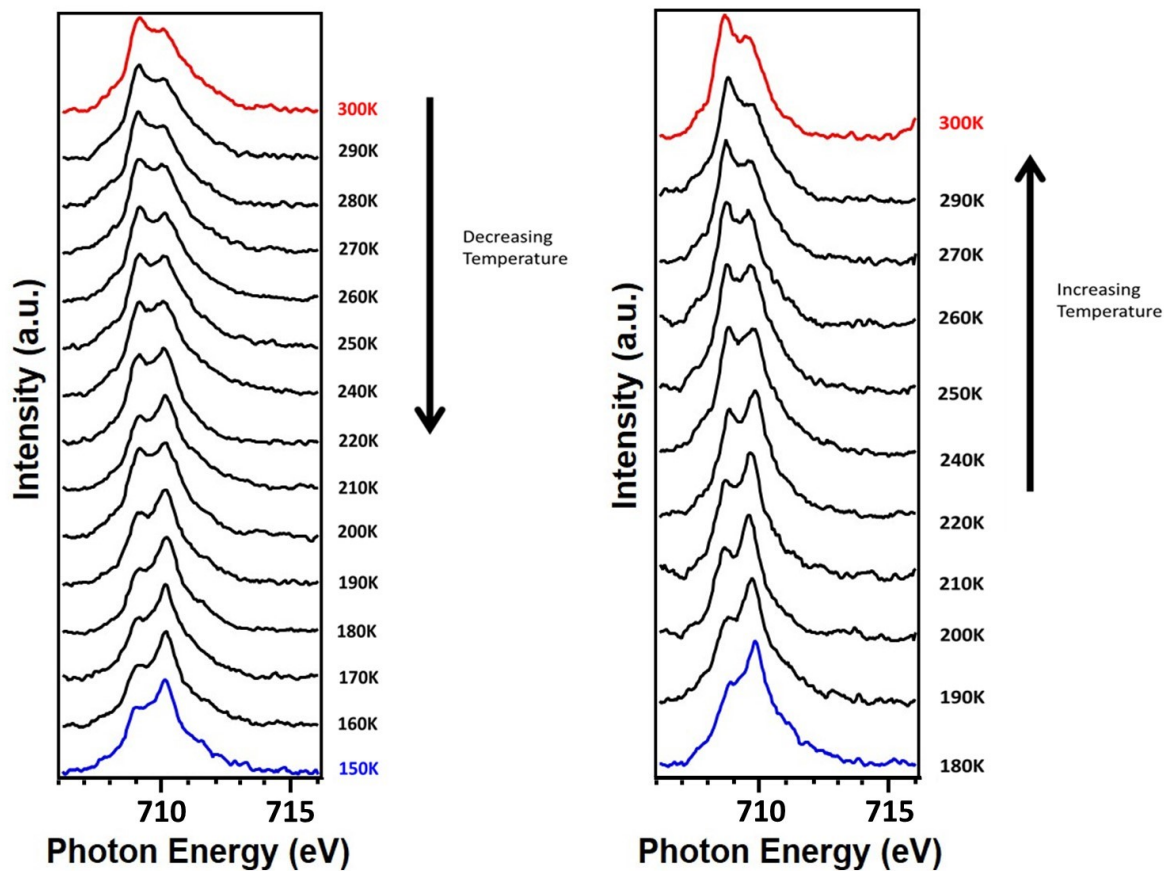


Figure S3: The temperature dependent X-ray absorption spectra of  $[\text{Fe}(\text{L}3)(\text{bpee})]_n$  (complex **3**) for both heating and cooling. Blue indicates the Fe  $2p_{3/2}$  - L3 edge spectrum of the low spin (LS) state and red indicates the spectrum of the high spin (HS) state

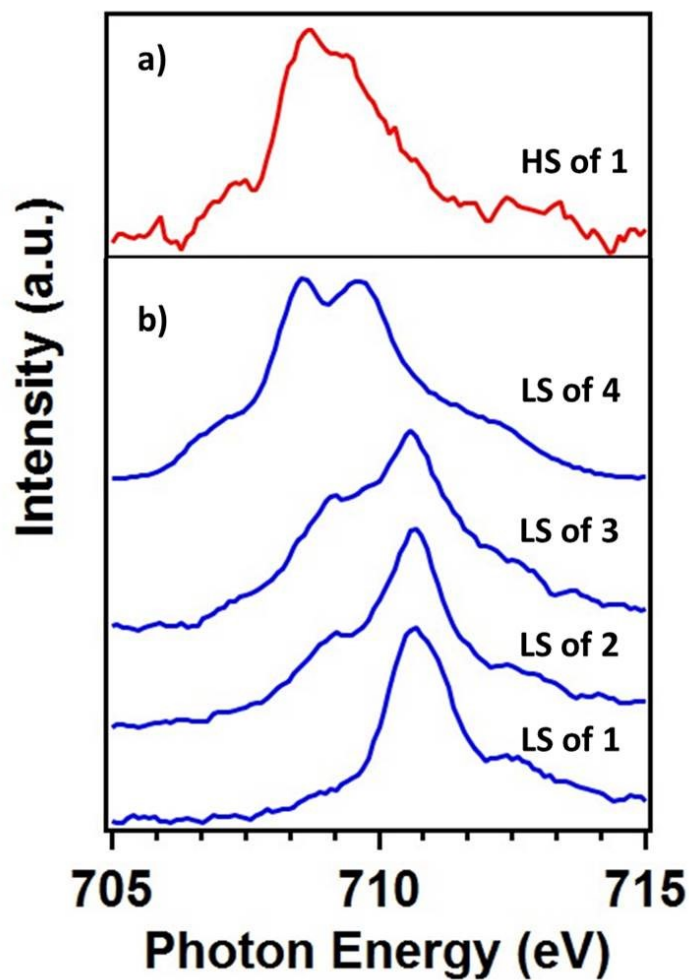


Figure S4: Representative X-ray absorption spectra of the high spin state (a) and the low spin state (b) of molecular complex **1** ( $[\text{Fe}(\text{L1})(\text{bipy})]_n$ ). The nominally low spin state of molecular complexes **2**, **3** and **4** ( $[\text{Fe}(\text{L2})(\text{bipy})]_n$ ,  $[\text{Fe}(\text{L3})(\text{bpee})]_n$ , and  $[\text{Fe}(\text{Htrz})_2(\text{trz})](\text{BF}_4)$ ) are in the mix spin state as is evident in this comparison.

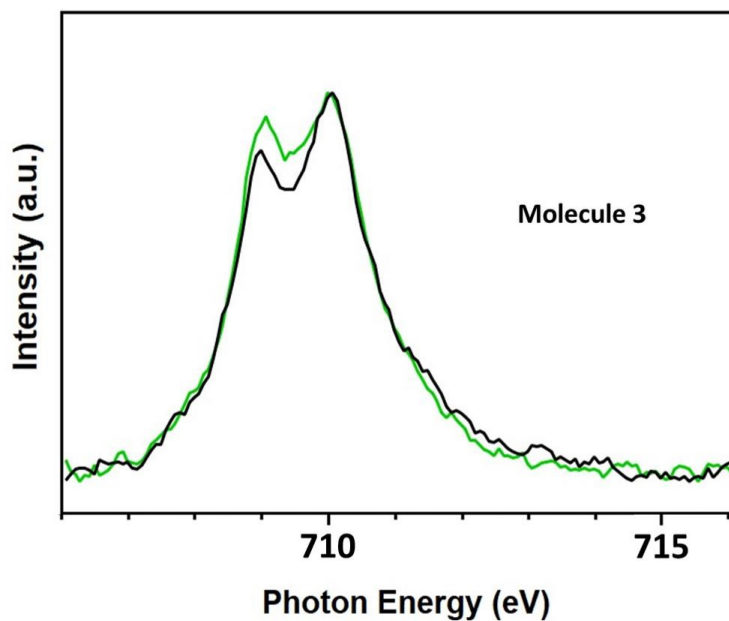
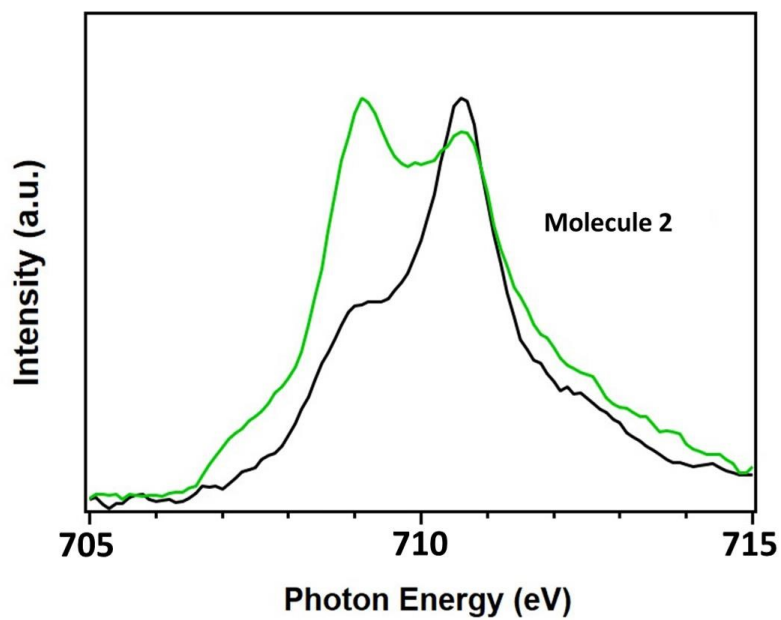


Figure S5: A comparison of X-ray absorption spectra of molecule **2** ( $[\text{Fe}(\text{L}2)(\text{bipy})]_n$ ) and molecule **3** ( $[\text{Fe}(\text{L}3)(\text{bpee})]_n$ ) at 220 K obtained from a sequence of spectra taken from an increasing temperature cycle (black curve) and a decreasing temperature cycle (green curve).