

Supplemental Information:

Modification of Molecular Spin Crossover in Ultra-Thin Films

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This document provides clarifying STM images and conductance maps to support the main manuscript. Figure S1 reproduces the inset of Figure 1 with embellishments that emphasize key points supporting the assignment of these features as high spin molecule pairs. Figure S1a illustrates the fact that the depressions are not aligned with possible crystallographic locations of vacancies. Figure S1b points out the “extra” protrusions around apparent depressions that cannot be associated with any of the low-spin 3-lobed features, but could be consistent with high spin imaging as shown in the calculated STM images in Figure 2 of the main text.

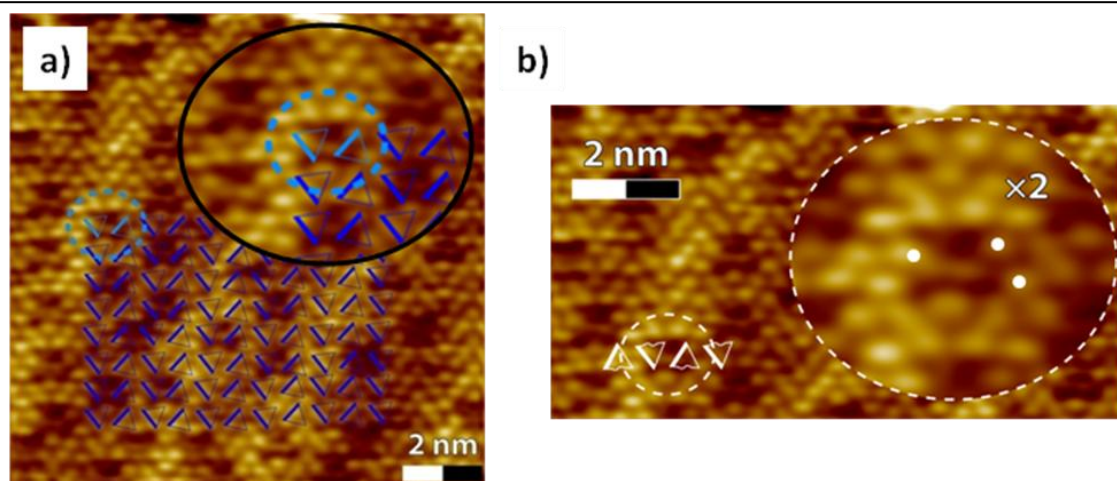


Figure S1. a) Illustration of a tiling of the bilayer surface that shows the lack of coincidence between low spin lattice positions and apparent depressions. This is the first indication that they cannot simply be vacancies; b) Reproduction of inset of Figure 1 that shows the "extra" protrusions indicated with white dots near the depressions that cannot be associated with any low spin molecule.

Figure S2 shows a sequence of bias-dependent STM images and conductance maps measured at 154 K that also supports our assignment of the apparent depressions as high spin molecules. Note that at +0.75 V, there is an abrupt change in imaging condition in the upper third of the frame that is illustrative of the typical situation for unstable imaging in the unoccupied DOS in our experiments.

