The Spin-dependent Jahn-Teller Effects of Re^{-1}_{Mo} in MoS_2

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Point defects in crystalline materials present unique and spatially isolated electronic states. These localized electronic states allow for breaking energetic degeneracies and pseudo-degeneracies via the Jahn-Teller effect and the pseudo-Jahn-Teller effect, respectively. Spontaneous symmetry breaking in point defects is analogous to symmetry breaking in molecular transition metal complexes. In many transition metal complexes, the electron occupations can change to yield different net spin on the molecule. Changes in electron occupation can also apply to the solid state. Isolated electronic states associated with point defects may change occupation as in molecules. As the electronic ordering in a point defect changes, so can the orbital energies. Therefore, energetic degeneracies do not break identically across different spin states. The Jahn-Teller and pseudo-Jahn-Teller effects will necessarily change with the spin state. Here we present the case of symmetry breaking from Re^{-1}_{Mo} in MoS_2. We observe both a high and low spin state for this point defect and that the high spin state anomalously has a lower total energy. The mode of symmetry breaking is identified as the pseudo-Jahn-Teller effect through electronic structure analysis. The adiabatic potential energy surface differs between the two spin states. Therefore, the vibronic coupling that drives the pseudo-Jahn-Teller effect in both systems must arise from different vibrational modes. We perform a vibrational mode analysis to improve our understanding of the interplay between spin-crossover and the pseudo-Jahn-Teller effect.