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Computational Electronic Structure of Photoinduced Spin Transitions in Molecular Photoswithches TEEPANIS CHACHIYO, JORGE H. RODRIGUEZ, Department of Physics, Purdue University — Minimum Energy Crossing Points (MECPs) are the energy barriers associated with radiationless spin-crossover. Understanding the mechanisms of such spin-forbidden processes is of great fundamental and practical interest. Methods of locating MECPs have been developed by several authors with, in general, a poor convergence behavior which increases the computational cost. We present a new method based on spin density functional theory (SDFT) that exhibits fast logarithmic convergence and is suitable for large molecular systems. Spin photo-switchable compounds are becoming a growing area of research since, in principle, it is possible to exploit their spin degrees of freedom to store bits of information. For instance, the photoswitch $[Fe(ptz)_6](BF_4)_2$ ($ptz = 1-propyltetrazole$) has a singlet (S=0) ground state and, upon optical excitation, becomes trapped in a quintet state (S=2) as long as T $<$ 50K. We applied our MECP methodology to study its geometrical and spin dynamics during spin crossover. Our results are useful for elucidating the geometric rearrangements and microscopic mechanisms that control the lifetime and the critical temperature of the meta-stable high spin (quintet) state. For example, the MECP-SDFT calculations showed that the transition from singlet ground state to a triplet "intermediate" state is accompanied by an unusually large 0.3Å bond length elongation of the iron axial ligands.

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