

**Theory of Chemical Bonds in Metalloenzymes XIII:  
A GSO Study of non-collinear spin structures in iron-sulfur clusters**

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Abstract

We present a first principle study of non-collinear spin structures of iron-sulfur clusters ([3Fe-4S] and [4Fe-4S]) employing the density functional theory with generalized spin orbital approach (GSO-DFT) [1-3]. Calculations of a [3Fe-4S] cluster show that non-collinear spin (NC) state is more stable than either the antiferromagnetic (AF) or the ferromagnetic (FM) states. Fig 1 shows the lowest non-collinear spin state (NC). It is found that the calculated energy at the NC state is slightly lower than an expected value estimated by applying a Heisenberg spin Hamiltonian model:  $H = -\sum_{i,j} 2J_{ij}S_iS_j$ . This

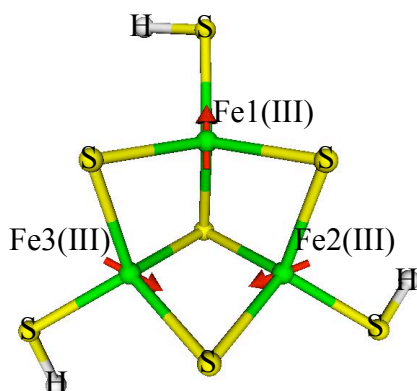


Fig1. Calculated non-collinear spin state of a [3Fe-4S] cluster ([3F(III)-4S](SH)<sub>4</sub><sup>3-</sup>). Arrows indicate atomic spin densities.

indicates that some orbital relaxations are occurred at the NC state. Chemical bond indices and atomic charge changes are discussed in this study.

[1] S. Yamanaka, et al, Int. J. Quantum Chem. 80, 664 (2000).

[2] S. Yamanaka, et al, Int. J. Quantum Chem. 91, 376 (2003).

[3] M. Shoji et al. Polyhedron 26, 2335 (2007).