FeS_2 Monolayer: An Unusual High Valence Ising Ferromagnet

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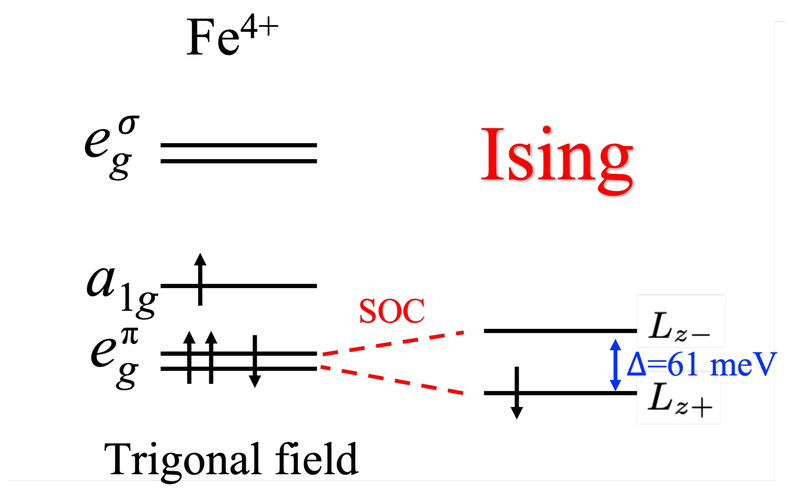

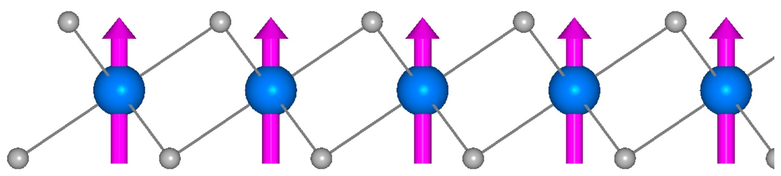

ABSTRACT

Two-dimensional magnetic materials are of great interest for their promising applications in spintronics. Here we propose the unusual high valent FeS_2 hexagonal monolayer as a candidate for a strong Ising ferromagnet. We find the high valent Fe^{4+} ion is in the low-spin state ($t_{2g}^4, S = 1$) rather than the high-spin state ($t_{2g}^3 e_g^1, S = 2$). The low-spin state allows to carry a large perpendicular orbital moment and produces a huge single ion anisotropy of 25 meV/Fe. Moreover, strong Fe 3d-S 3p hybridization and small band gap associated with negative charge transfer effect help to establish a strong ferromagnetic (FM) superexchange. We predict the FM T_C is 261K and could be increased to 409K under 5% compressive strain. Therefore, FeS_2 monolayer could be a promising strong Ising ferromagnet.

RESULTS

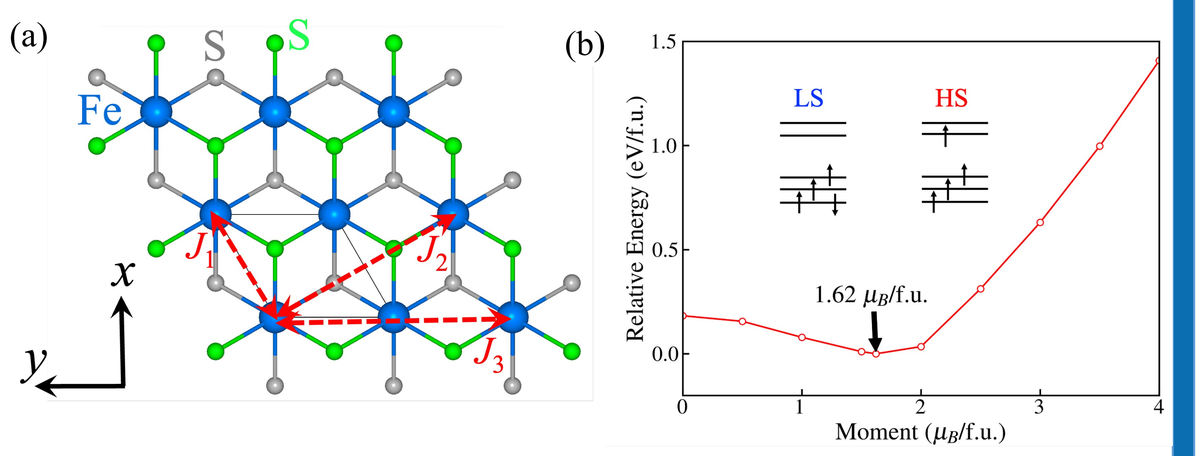

Fig.1. (a) The crystal structure of hexagonal FeS_2 monolayer. (b) Fixed-spin-moment calculations imply the low-spin state with $S = 1$ for Fe^{4+}.

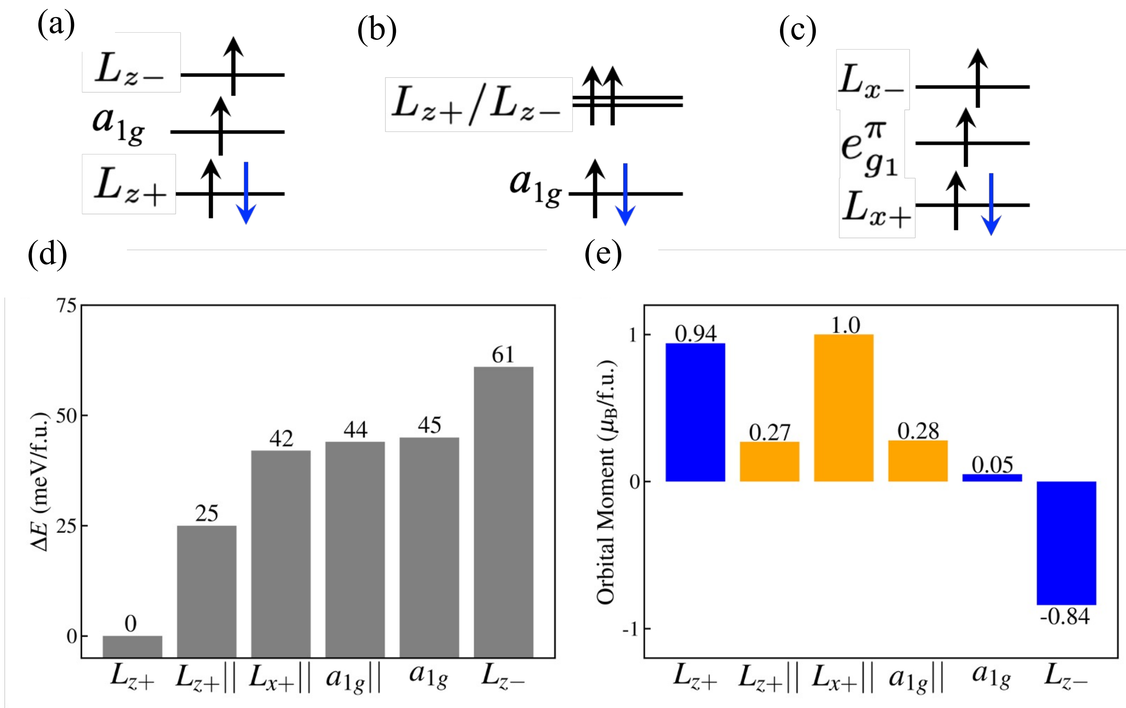

Fig.2. Crystal field level diagrams for the low-spin Fe^{4+} S=1 in different configuration states: the spin-down electron occupies (a) L_{z+}, (b) a_{1g} and (c) L_{x+} states. (d) the relative total energies of each states, which indicate the ground state is L_{z+} state and magnetic anisotropy is 25 meV. (e) orbital moments of each states (blue color for out-of-plane and yellow color for in-plane).

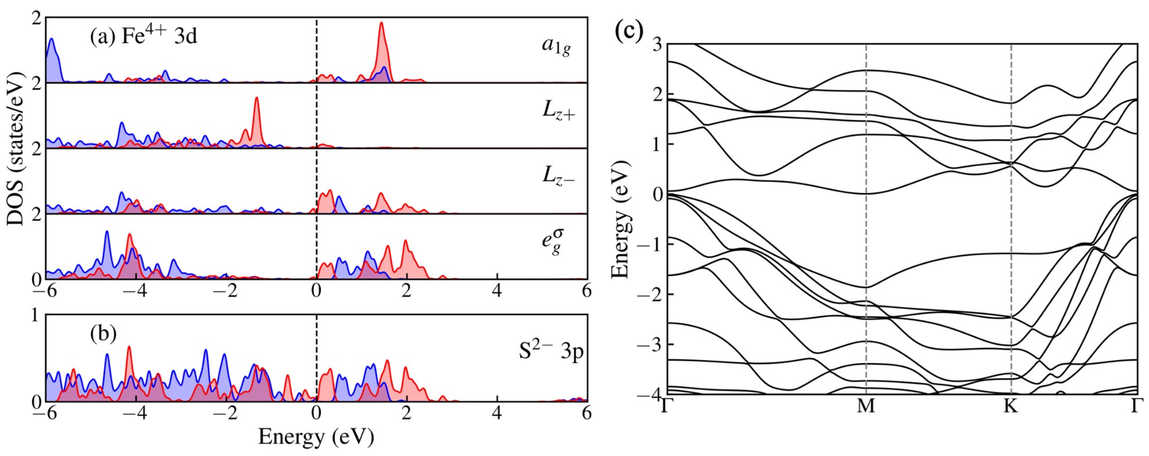

Fig.3. (a) Fe 3d and (b) S 3p DOS of FeS_2 monolayer in the L_{z+} ground state by LSDA+U+SOC. (c) The corresponding band structure, where the band gap is 40 meV.

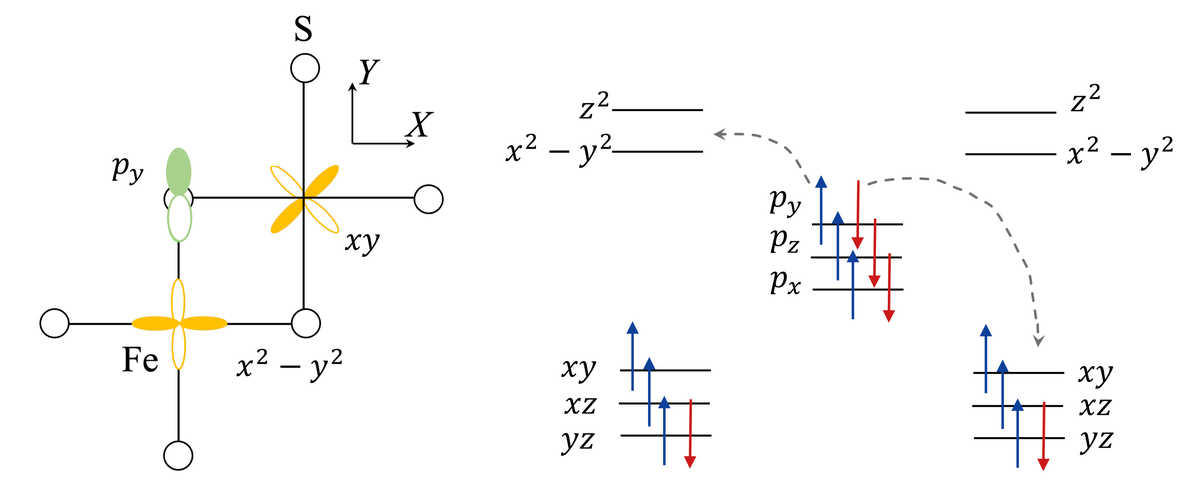

Fig.4. Schematic diagram of FM superexchange channel: $d_{xy} - p_y - d_{x^2-y^2}$

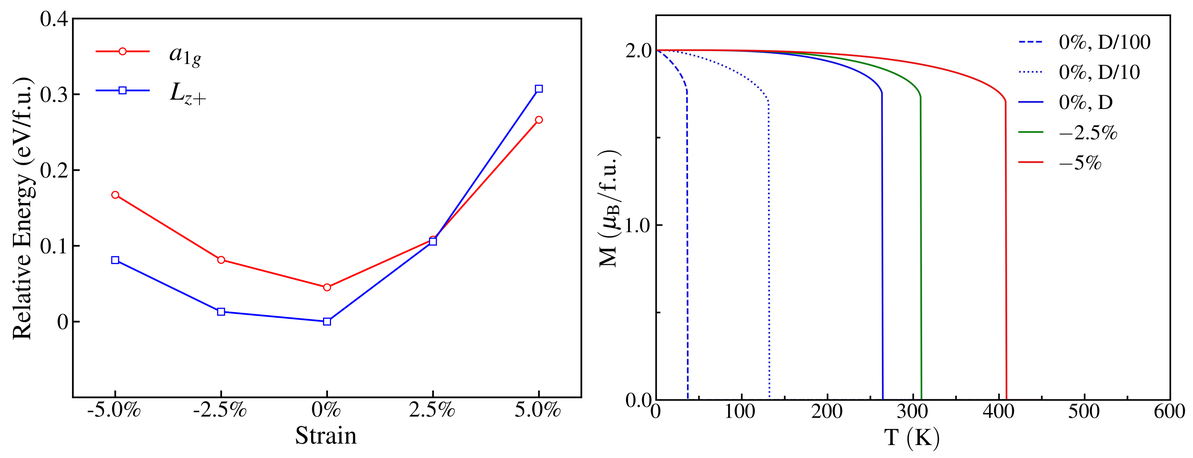

Fig.5. (a) The relative energies of the L_{z+} ground state and the a_{1g} state under different strains. (b) The magnetization as a function of temperature under different compressive strain. The Curie temperature T_C is 261 K, 310 K and 409 K respectively.

CONCLUSIONS

1. FeS_2 monolayer is in L_{z+} ground state with giant out-of-plane magnetic anisotropy.
2. FeS_2 monolayer is a FM insulator with a small band gap.
3. Curie temperature T_C of FeS_2 monolayer increases from 260 K to 409 K under 5% compressive strain.

REFERENCES

1. Jiadong Zhou, *et al.*, Nat. Mater. (2022). doi: 10.1038/s41563-022-01291-5
2. Ke Yang, Yaozhenghang Ma, *et al.*, to be submitted.