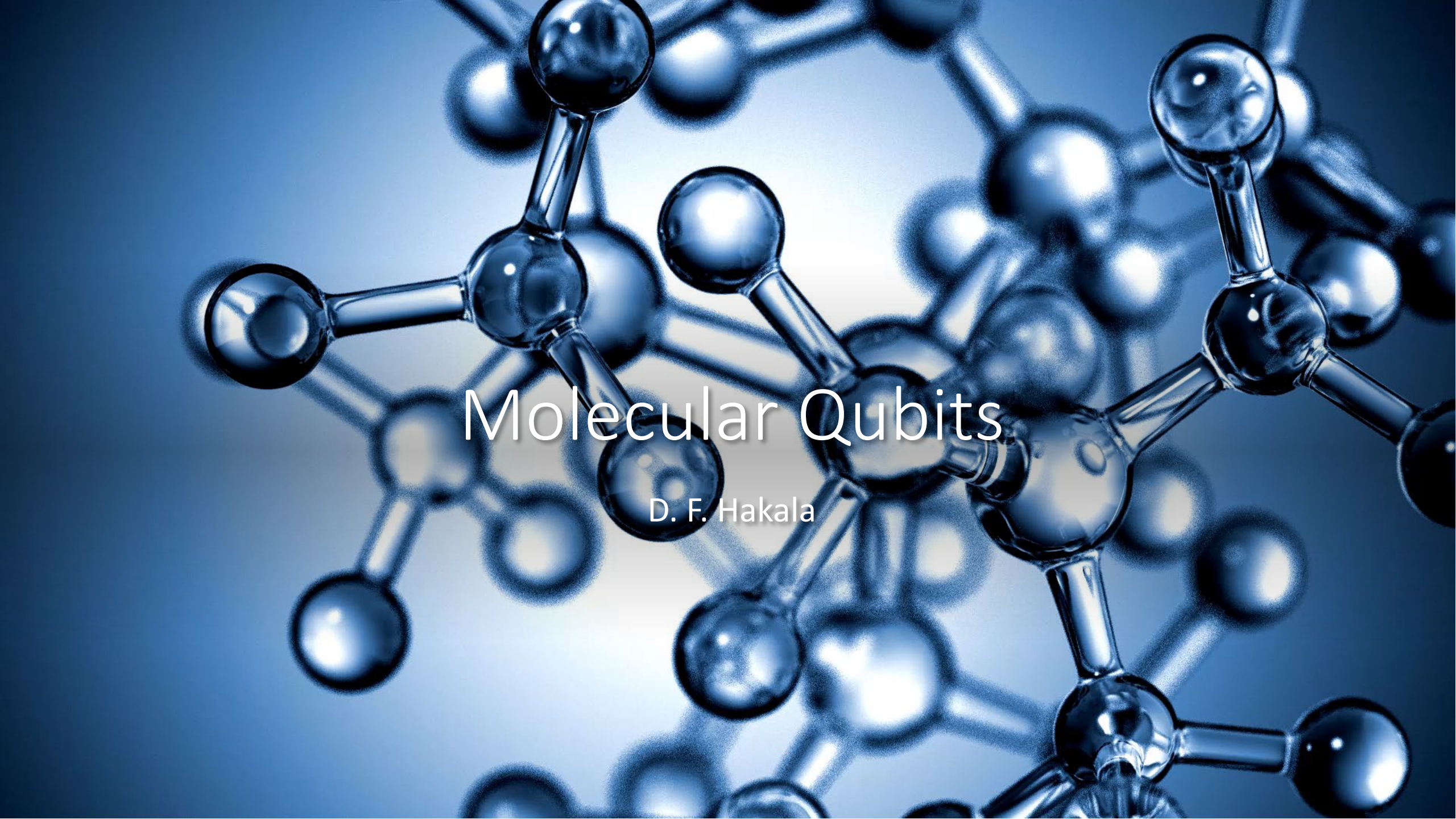
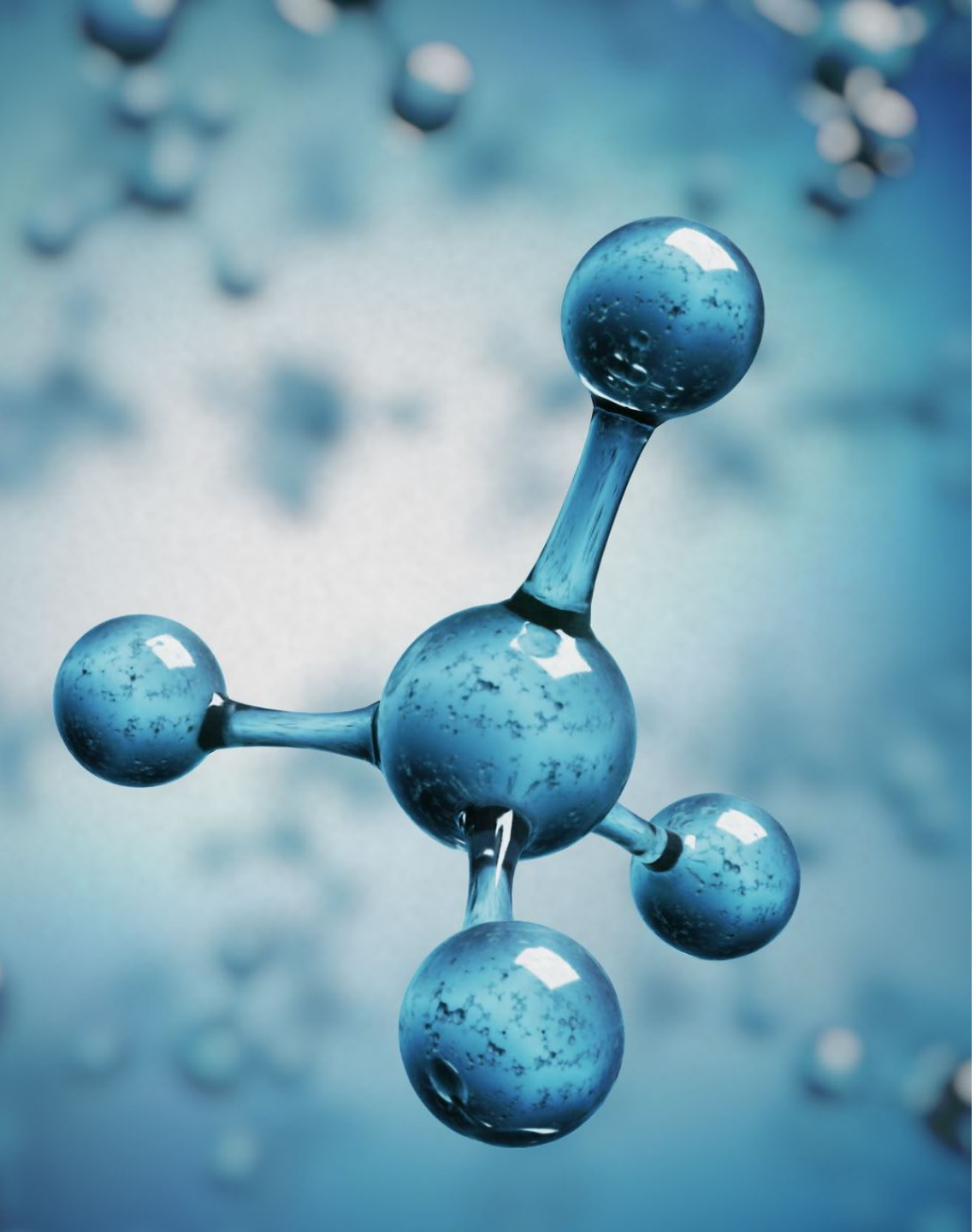


Molecular Qubits

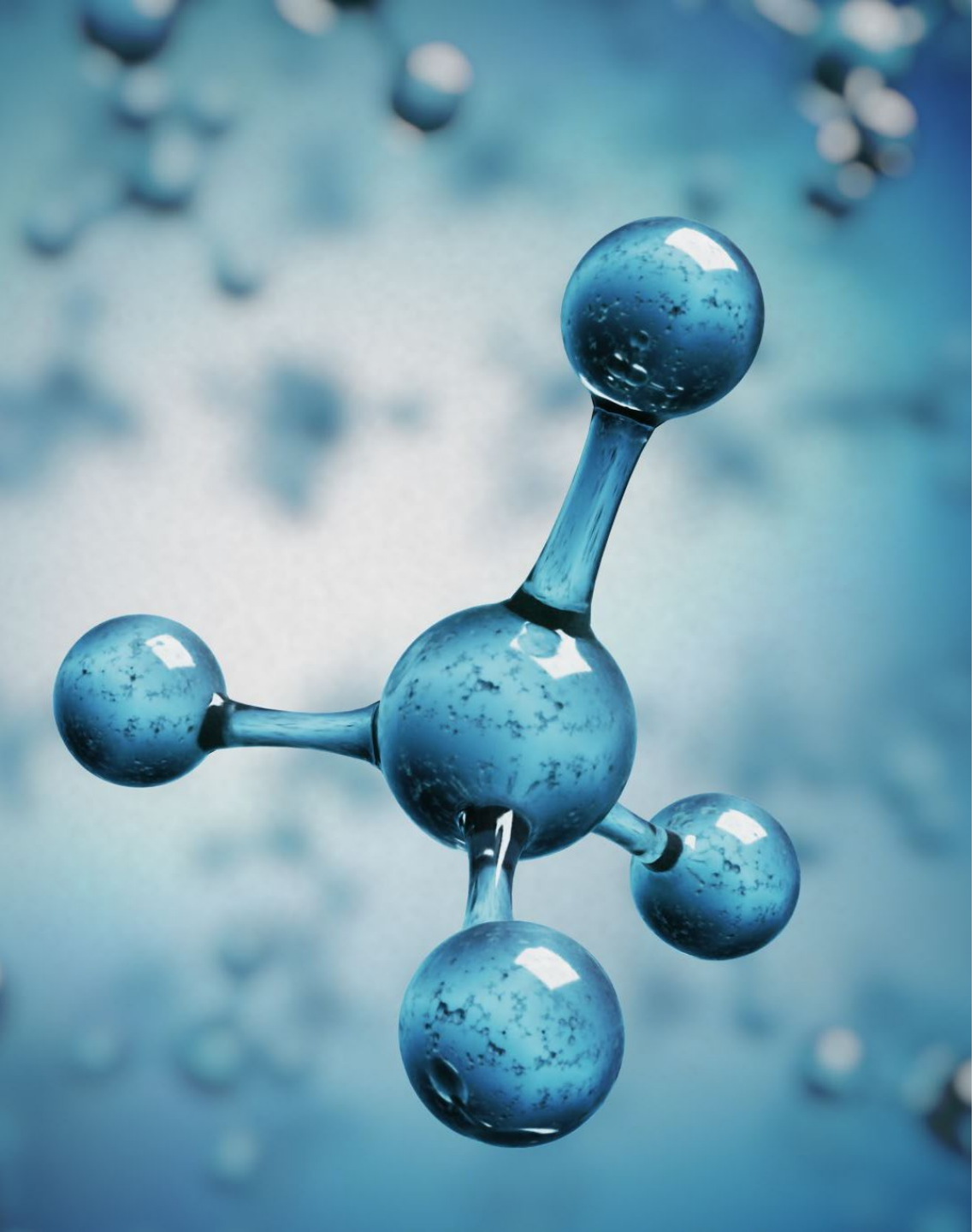
D. F. Hakala



Introduction-Molecular Qubits

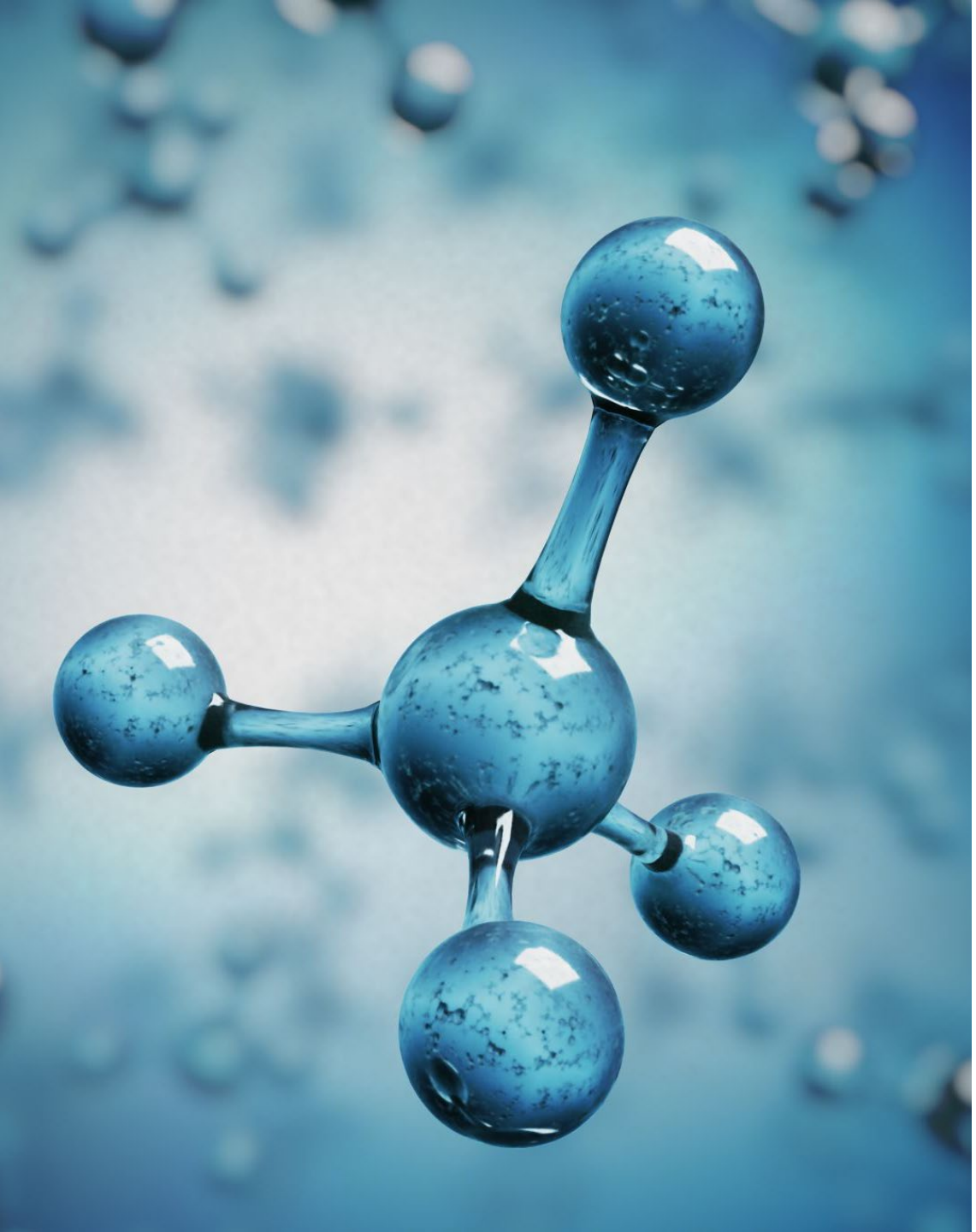
- A good relatively recent review related to molecular qubits is
- **Recent Innovations in Solid-State and Molecular Qubits for Quantum Information Applications**

  J. Phys. Chem. Lett. 2021, 12, 10742-10745
- There are over 40 references in this article, many of them with respect to molecular qubits.
- This is an overview for a Virtual Issue of J. Phys .Chem.Lett. on this subject.



Molecular Qubits

- A qubit is an object that has two states (generally referred to as |0> and |1>) and all their superpositions. There are many possible physical implementations of the qubit. One of these that is attractive is the molecular qubit.
- The attractiveness is for several reasons.
  - They are small which will help in scaling up to higher device densities
  - They can be engineered or designed with respect to a number of different properties.
  - They are capable of being encoded, addressed, and read by means such as optical or microwave radiation.
- A typical molecular qubit is one that is spin based using the spin of unpaired electron(s). This energy of the molecule will both change and split in response to a magnetic field.



Molecular Qubits

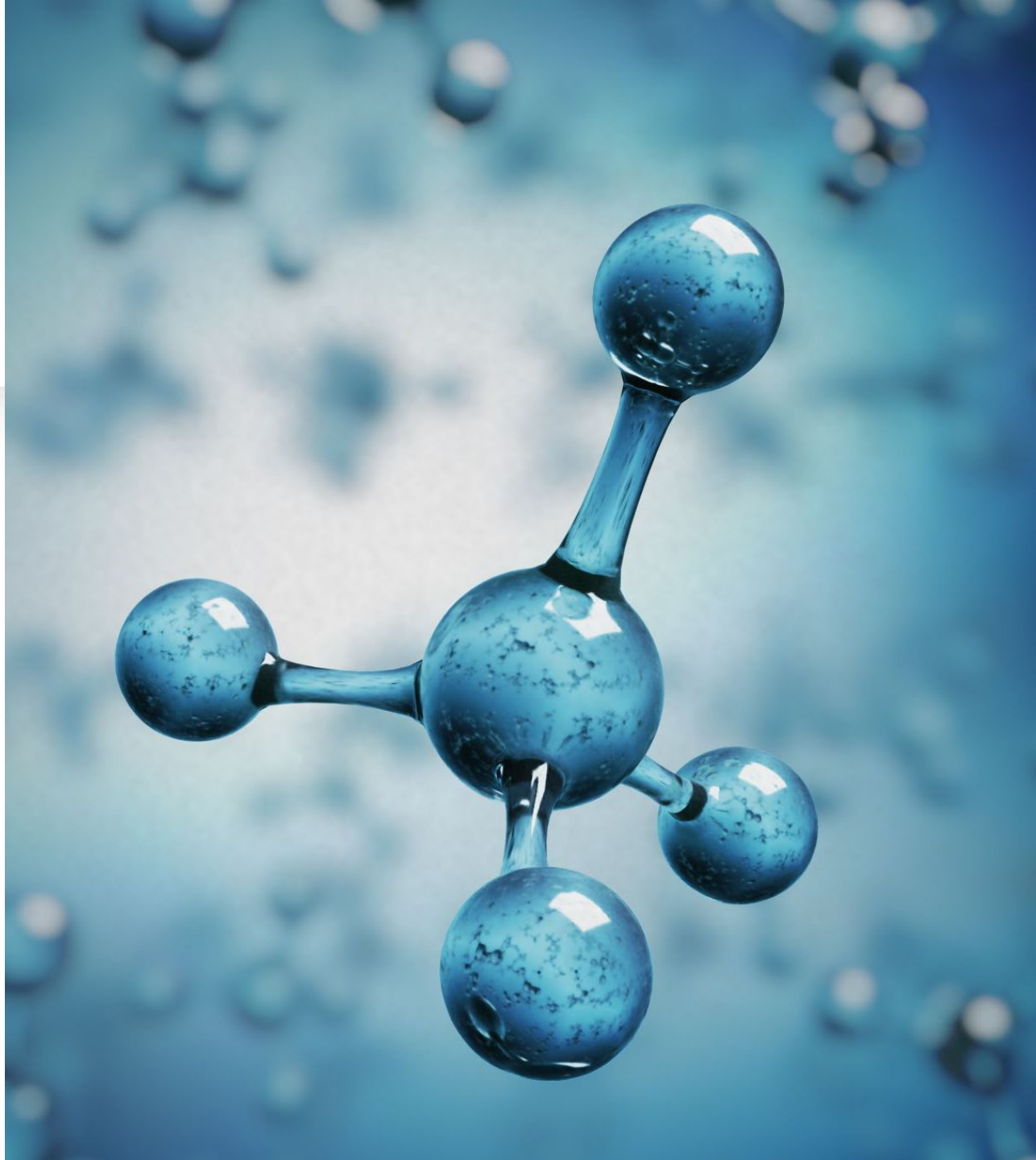
- So, one of the characteristics we want in a molecular spin qubit is that it is easy to achieve a spin state. Coordination compounds of transition metals such as Cr, Cu, Ni, V etc. can be selected to have unpaired spins.
- Another characteristic is that we want to maximize the time it can spend in the targeted coherent superpositions of qubits. Since there can be an I* S interaction between nuclear spins (I) and the electron spin (S) of the transition metal , this interaction can result in a decoherence of the qubit state. By designing molecular qubits such that atoms with net nuclear spins are further from the transition metal atom we should improve the coherence time of the qubit.
- The stiffness of the lattice can also be a factor. A stiffer lattice generally means higher phonon energies. The higher energies are less likely to interact and cause decoherence of the qubit.
- Qubits that have better coherence times at high temperatures are of interest because this could result in a less costly quantum computer. So good room temperature coherence is a desired objective.

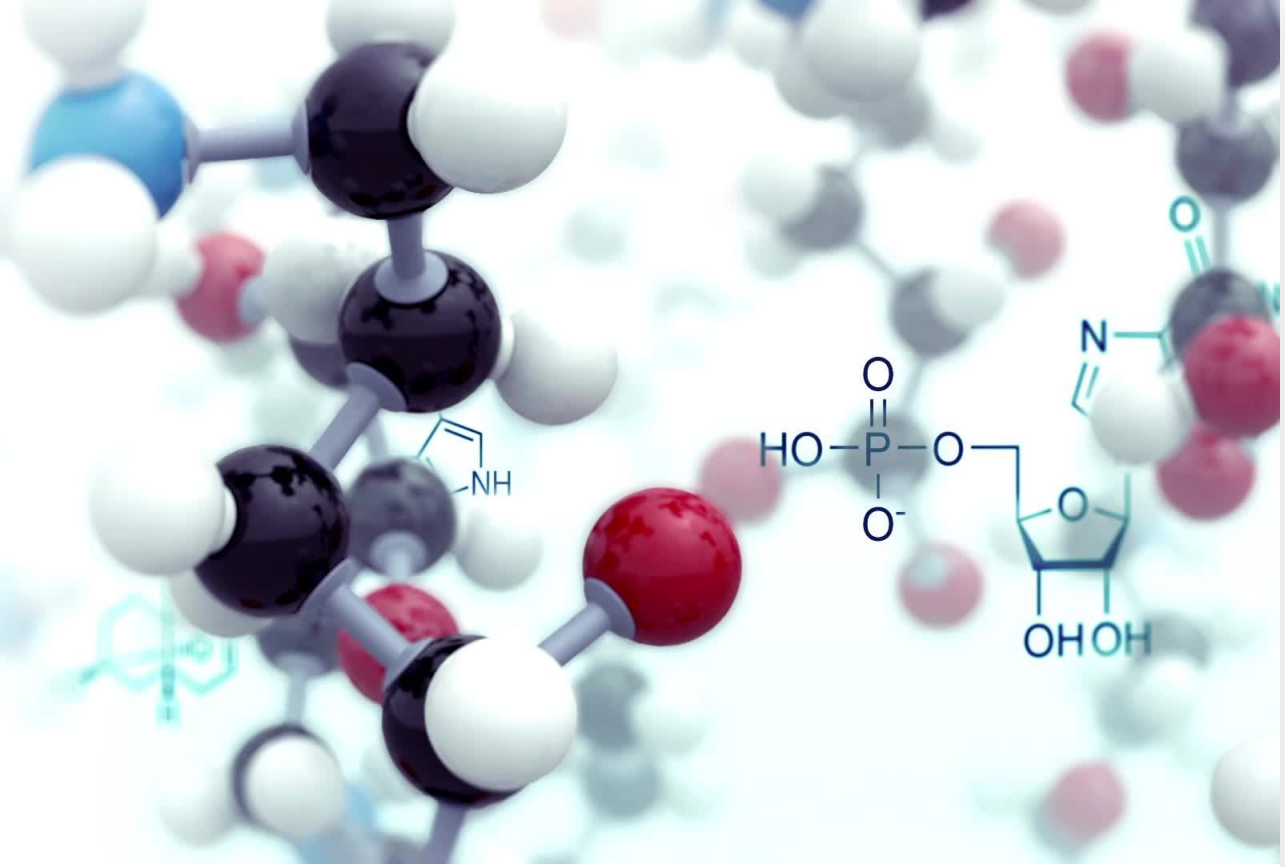
Molecular Qubits

- One type of molecule proposed as a qubit with better room temperature coherence are copper(II) complexes.
    - Room Temperature Quantum Coherence in a Potential Molecular Qubit.

      NATURE COMMUNICATIONS , 5:5304 , DOI: 10.1038/ncomms6304 , www.nature.com/naturecommunications
- An analog for one of these molecules has been studied to better understand the electronic states of the molecule, focusing on the lower lying states which are of most interest in the dynamics of the qubit behavior .
    - Characterizing Excited States of a Copper Based Molecular Qubit Candidate with Correlated Electronic Structure Methods.

      J. Phys. Chem. A 2023, 127, 6764-6770
- The following does initial calculations on the copper qubit analog studied in the last paper.

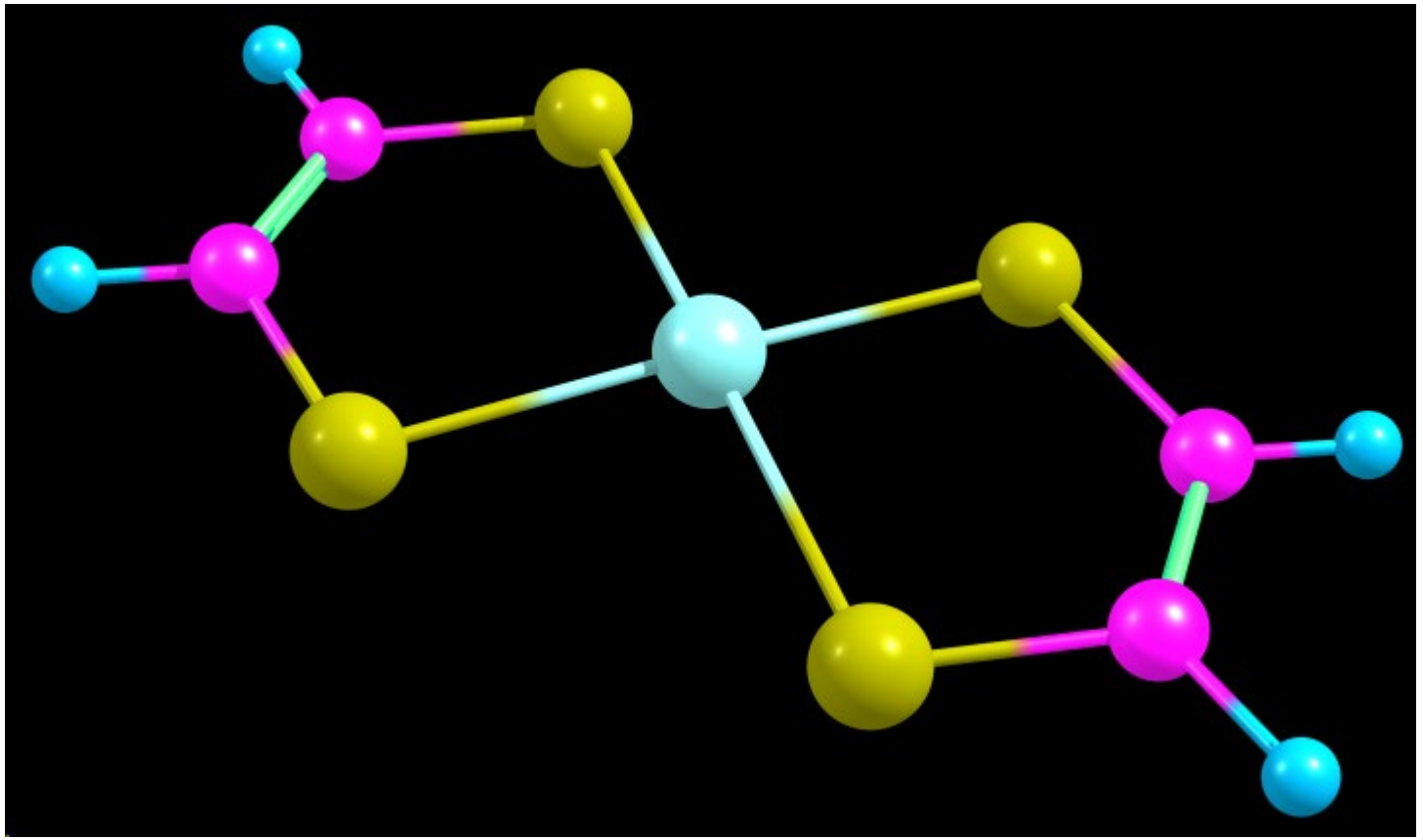




Calculations

- This molecule is an analog for a proposed room temperature molecular qubit. $[Cu(II)S_4C_4H_4]^{-2}$, or $[Cu(II)(edt)_2]^{-2}$ where edt is short for ethylene dithiolate a doubly negatively charged ligand.
- The Cu qubit analog molecule was geometry optimized using a DFT method with the PBEO functional, a basis set of def2/TZVPPD and the auxillary basis set def2/J.
- The molecule was drawn and initially optimized using Spartan 20.
- The resulting data was used to generate an XYZ file that was edited to be suitable for ORCA.
- The DFT calculation was done using ORCA 5.04.

Cu Qubit Analog



This molecule is an analog for a proposed room temperature molecular qubit. $[Cu(II)S_4C_4H_4]^{-2}$

The proposed molecule to which this is an analog has four cyano or N3 groups instead of the four hydrogens. The carbon atoms have no net nuclear spin (as opposed to the hydrogens). The nitrogen has a very weal spin intereaction. They therefore will have less interaction with the Cu(II) spin, reducing environmental interaction of the qubit.

However, the addition of additional electrons from the four cyano or N3 groups significantly adds computational complexity to quantum calculations, So this molecule is easier to study.

ORCA Input File

- ! PBE0 def2-TZVPPD def2/J OPT
- !LargePrint NormalSCF PAL8 Printbasis PrintMOs

- * xyz -2 2
- Cu 0.79423 0.71870 0.00000
- S -1.02180 2.16851 -0.00000
- S 2.14846 2.60707 -0.00000
- S 2.61024 -0.73113 -0.00000
- S -0.55997 -1.16969 -0.00000
- C -2.35657 0.98041 -0.00000
- C -2.16717 -0.38861 -0.00000
- C 3.75565 1.82598 -0.00000
- C 3.94504 0.45693 -0.00000
- H 4.95690 0.07356 0.00000
- H 4.62545 2.46965 0.00000
- H -3.36845 1.36378 0.00000
- H -3.03696 -1.03230 0.00000
- *

This file shows that the ORCA computational chemistry calculation was done using
- A Density Functional Theory (DFT) functional of PBE0
- A basis set of def2-TZVPPD (triple zeta level)
- An auxillary basis set of def2/J

The calculation itself was a geometry optimization (keyword = OPT)

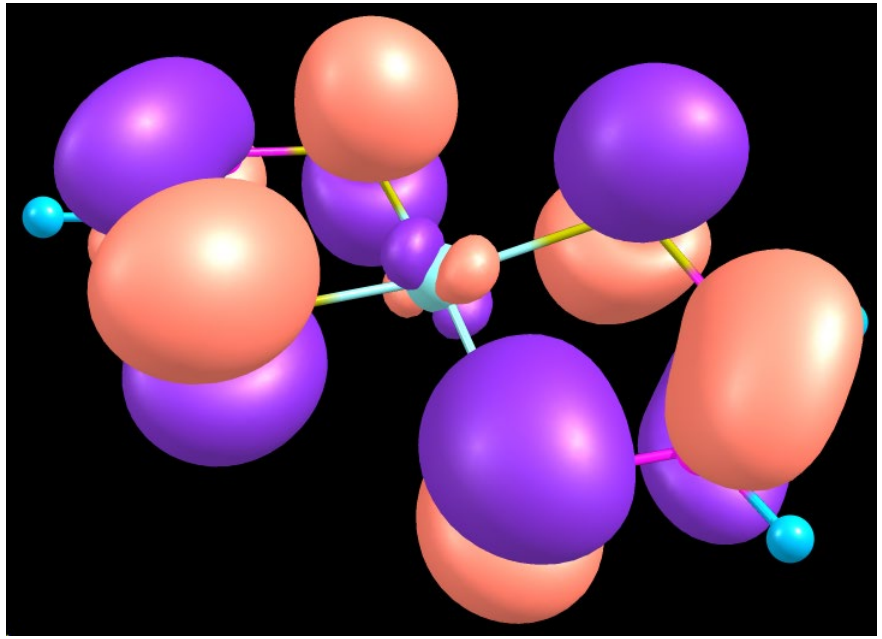
The calculation was done using 8 threads of parallel processing (keyword = PAL8)

The other comments refer to the data printed in the output file to be consistent with what ChemCraft software requires for input to that program for visualization of the Molecular Orbitals.

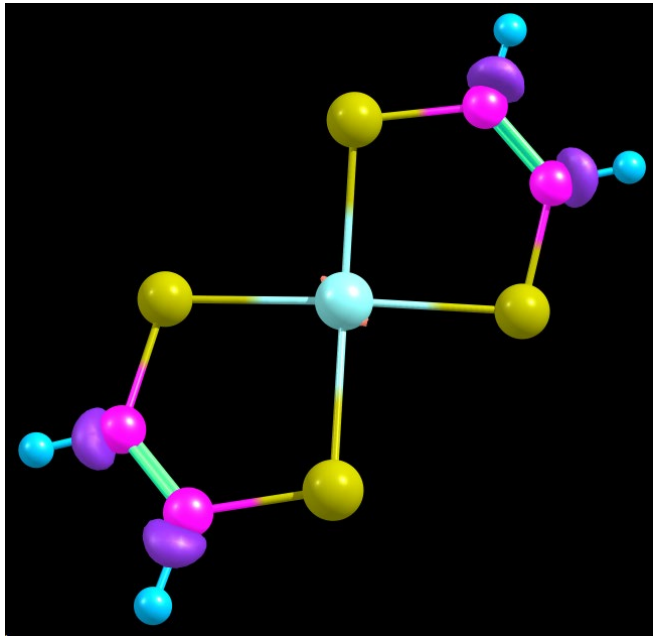
The atomic position are given by the XYZ file in angstroms

The net charge on the molecule is -2
The multiplicity of 2S+1 is 2 since the spin S is ½.

Examples of Molecular Orbitals



This is the Highest Occupied Molecular Orbital (HOMO) for the molecule as calculated.
Energy=0.0941 a.u. (Hartrees)
or 2.5606 eV



This is the Lowest Unoccupied Molecular Orbital (LUMO) for the molecule as calculated.
Energy=0.1991 a.u. (Hartrees)
Or 5.4178 eV

A HOMO -> LUMO transition would be 2.8572 eV , or 23,044 cm-1 , or 434 nm