Minicolloque n° 26 □ **Oral** ☑ **Poster**

Experimental and theoretical analysis of vibrational modes in $Fe(phen)_2(NCS)_2$ single crystal

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The spin-crossover crystal complex $Fe(phen)_2(NCS)_2$ undergoes a Low spin (LS) to High spin (HS) phase transition driven by temperature, pressure and light. In this compound the relation between phonons modes and photoinduced spin state switching is highlighted by femtosecond experiments [1]. The vibrational entropy difference between the HS and LS states also plays an important role for thermal equilibrium phase transition, as highlighted by Raman and Infrared measurements on $Fe(phen)_2(NCS)_2$ powder, interpreted with Density Functional Theory (DFT) calculations of molecular vibrational modes [2]. However, the unit cell of this crystal contains 4 molecules and the combination of their in-phase or out-of-phase vibrations give rise to 609 optical lattice phonon modes with different symmetry, frequency and polarisation dependency [3].

In this work, we study the lattice phonon modes of $Fe(phen)_2(NCS)_2$ crystal by combining polarization-resolved Raman and IR measurements, allowing selective probing of lattice modes of different symmetry. We compare the measurements with DFT calculations of the lattice phonon modes. For example, we find 8 N↔CS stretching modes (Fig. 1) for the different symmetries and frequencies of the mmm point group or 4 breathing modes. Modes with similar molecular vibration nature and of different symmetries do not exhibit the same frequency change during the HS-LS conversion, which adds a higher contribution to the vibrational entropy.

[1] M.Cammarata et al, Phys. Rev. Lett. 113, 227402 (2014)
[2] K.L. Ronayne et al, Phys. Chemistry Chemical Physics, 10.1039 (2006)
[3,] E.Collet et al, Eur. Phys. J. B, 1140 (2019)

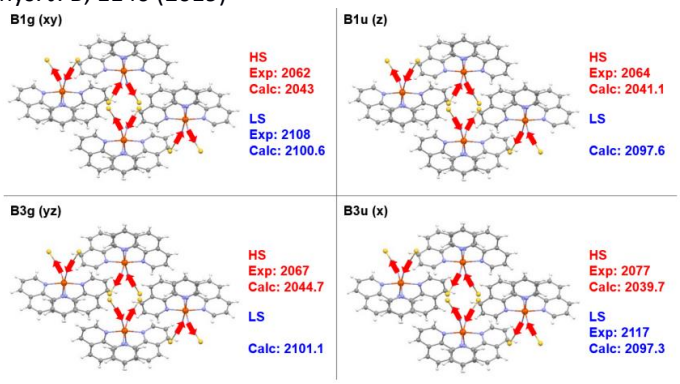

Figure 1 : Representation of 4 N↔CS vibrational modes in the $Fe(phen)_2(NCS)_2$ with their symmetry and frequencies (calculations and measurements in both spin states).