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Structure of Kaolinite and Influence of Stacking Faults: Reconciling Theory and Experiment Using Inelastic Neutron Scattering Analysis

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Kaolinite [kéiələnàit]

- 1:1 layered clay mineral
  $Al_2Si_2O_5(OH)_4$ or $Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O$

- Name derived from Chinese village Kao-Ling
  (高岭 / 高嶺 , Gāolǐng)

- Used in ceramics / porcelain, toothpaste, cosmetics, paint, production of paper, etc.

Kaolinite Structure

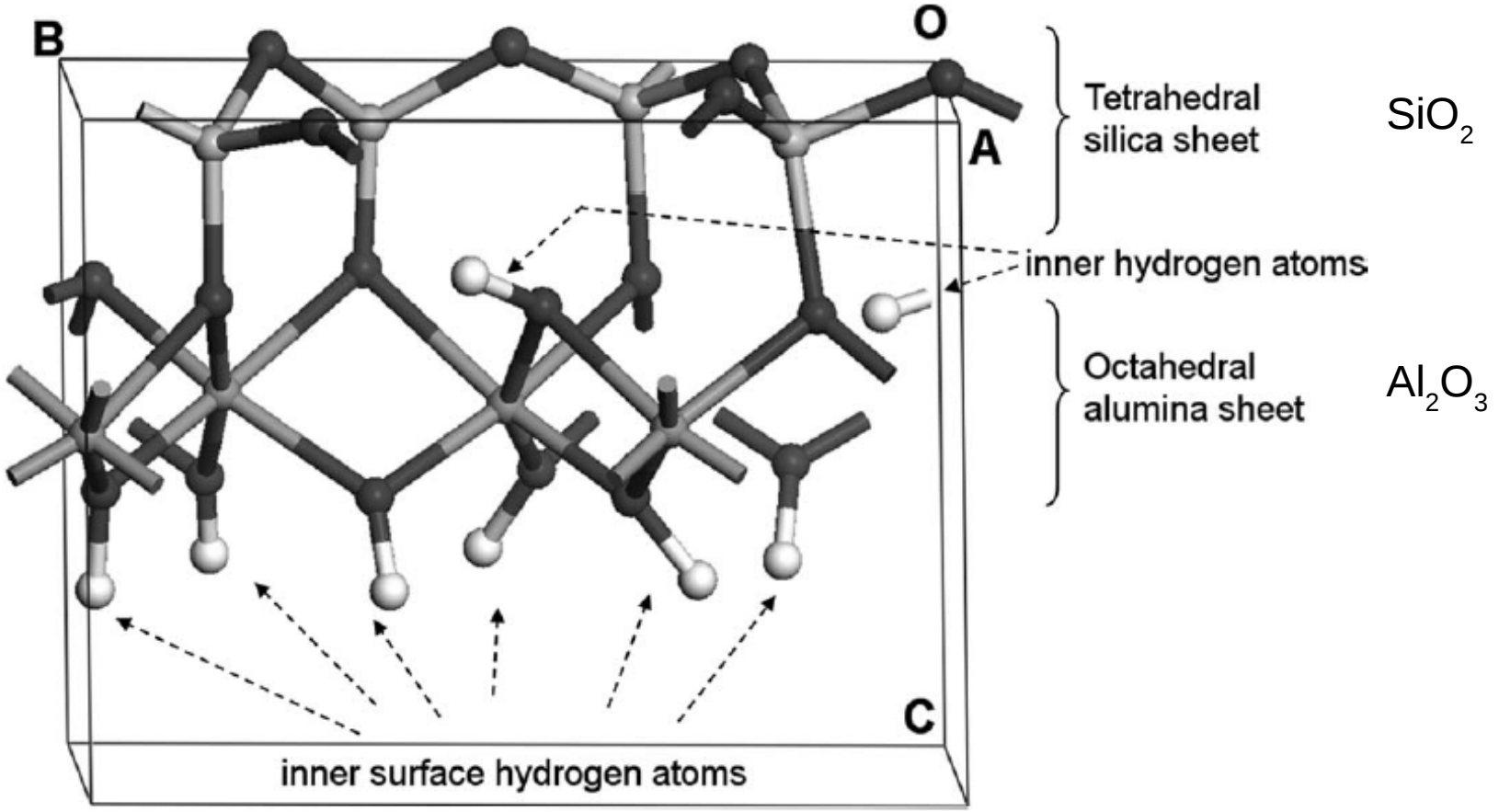


FIG. 1. Schematic representation of kaolinite (depicted as a P1 unit cell). The 1:1 layering of silica and alumina sheets is labeled, as are the two types of H-atoms present (*inner* H-atoms and *inner surface* H-atoms).

Methods

- INS (inelastic neutron scattering) data collected at 30 K at ISIS, Rutherford Appleton Lab, UK

- DFT calculations by VASP code with PBE
  - DFT-NCA (normal coordinate analysis)
  - DFT-MD

- INS spectrum calculated by CLIMAX program from vib. frequencies and atomic displacements

INS Spectra

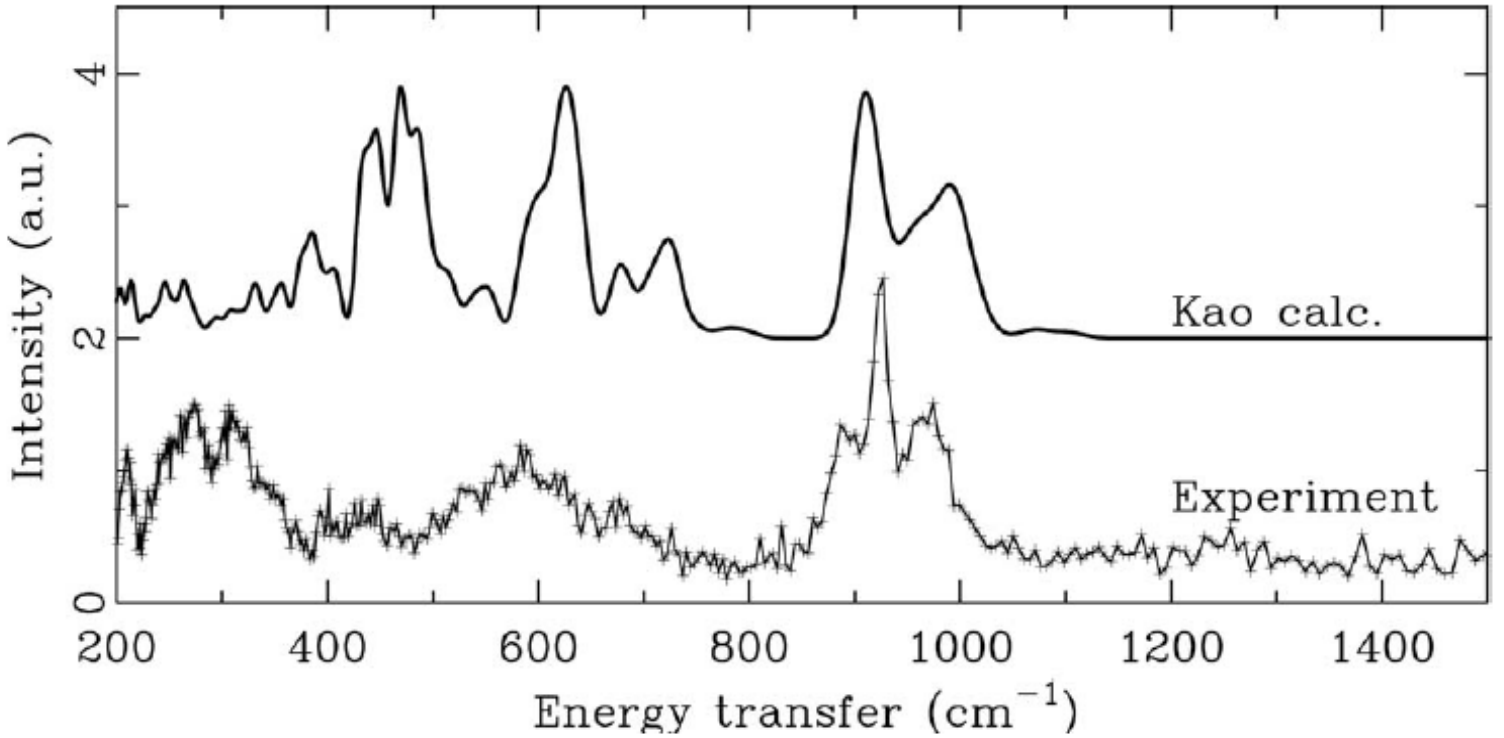
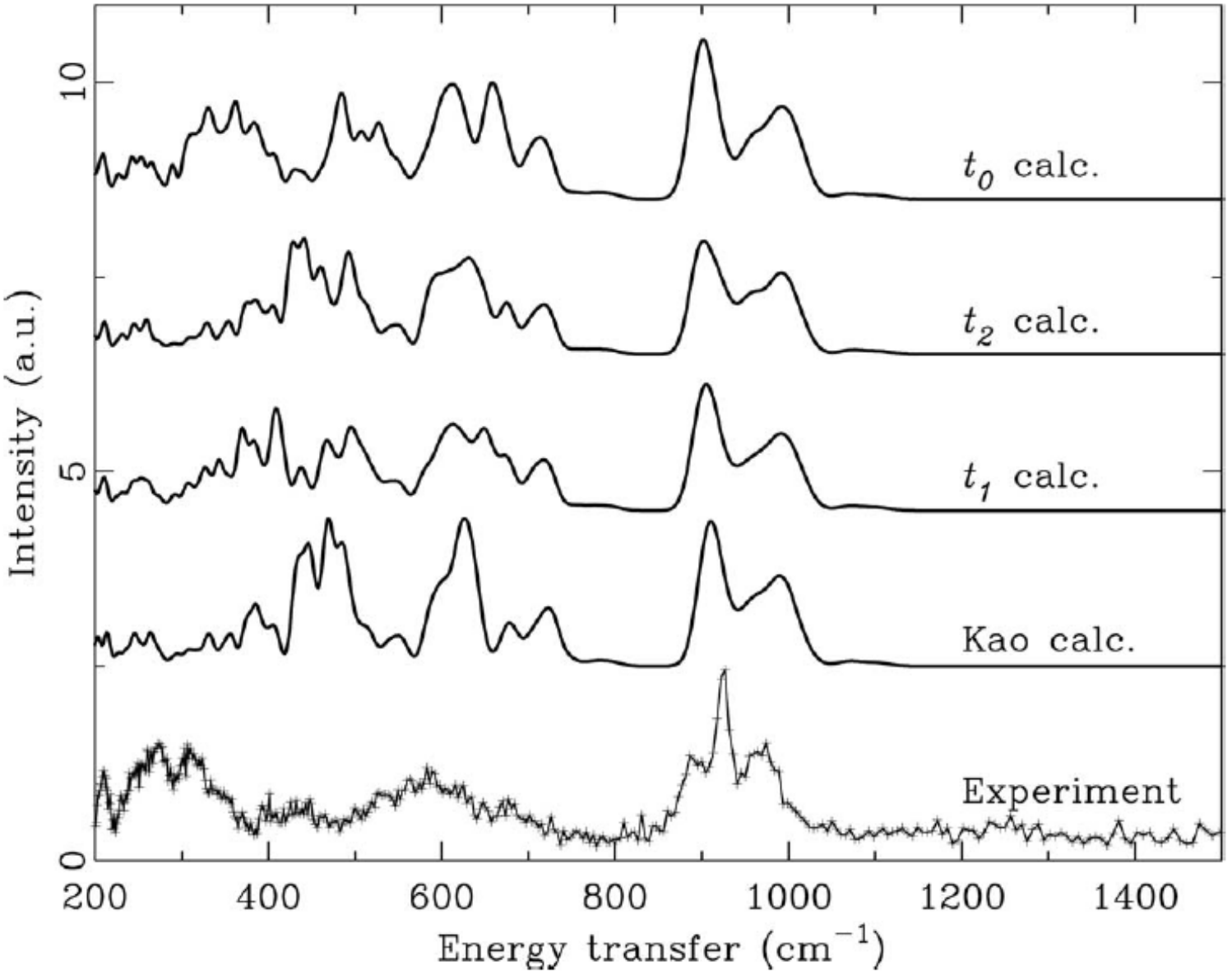


FIG. 2. Experimental INS spectrum of kaolinite (KGa-1b) and the calculated spectrum of the standard kaolinite structure[3] using normal coordinate analysis with force constants calculated by DFT (DFT-NCA).

INS Spectra with Stacking Faults

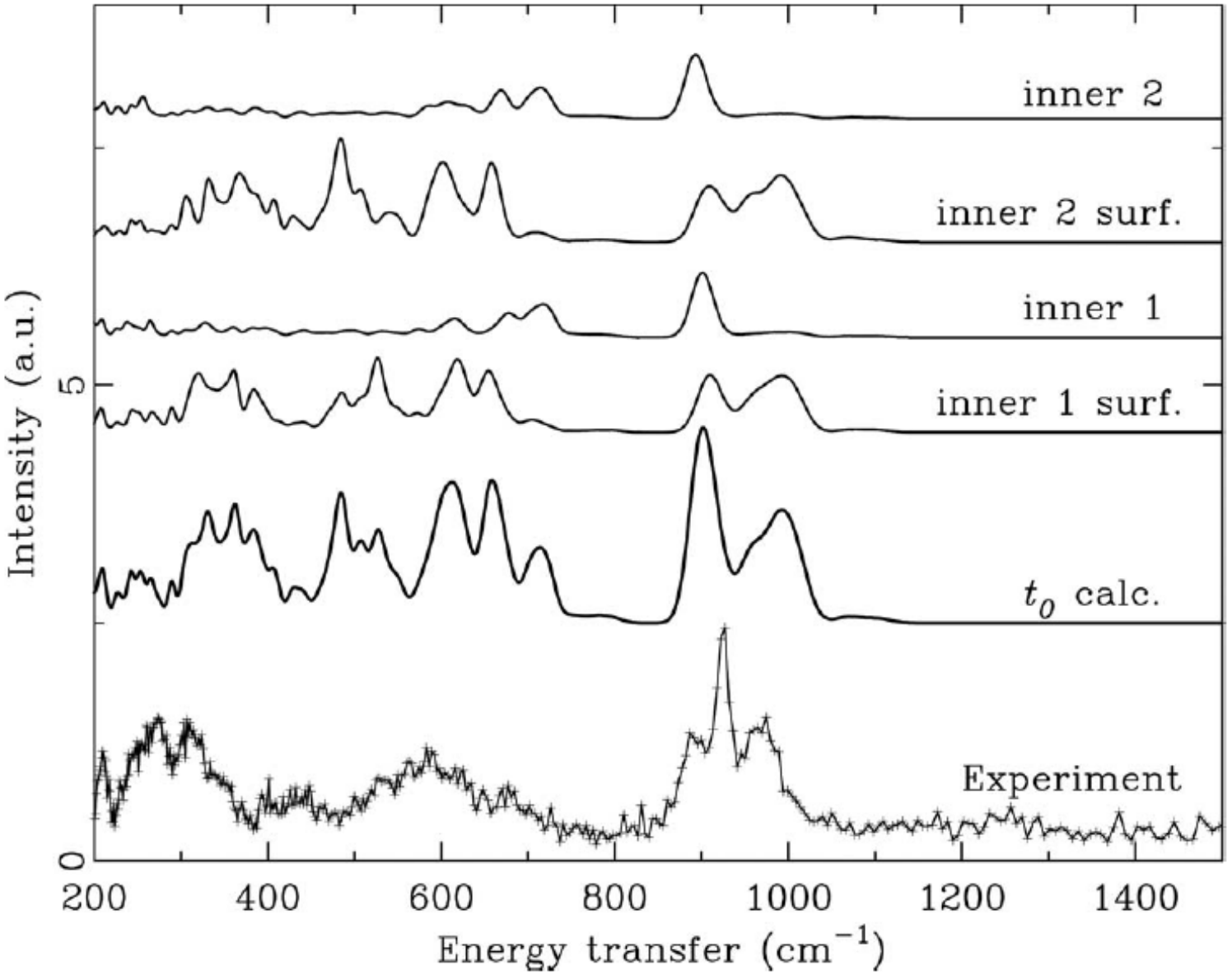


$t_0 = -0.3151a - 0.3151b$

$t_2 = -0.3488a + 0.3051b$

$t_1 = -0.3689a - 0.0223b$

Contributions from Different H-Atoms



Conclusion

- Computed INS spectrum with standard unit cell structure of kaolinite agrees well with experiment between 500 – 1200 cm^{-1}, but shows discrepancies at 200 – 400 cm^{-1}

- Incorporation of stacking faults gives better agreement with experiment
  – 0.3151***a*** – 0.3151***b***