

COHERENT RAMAN MICROSCOPY OF CRYSTALS: FROM MOLECULAR SYMMETRIES TO POLYMORPHISMS

Carsten Cleff, Hervé Rigneault, Patrick Ferrand, Virginie Chamard and Julien Duboisset*

Aix Marseille Univ, CNRS, Centrale Marseille, Institut Fresnel, Marseille, France

Obtaining information on the matter organization on the micrometer scale still remains a challenge in physics, chemistry or biology. In this context non-linear vibrational processes such as Coherent Raman Scattering (CRS) are powerful approaches for 3D imaging with chemical specificity without sample preparation. Most often used in the context of biology or soft matter, CRS is more rarely used to study molecular crystals. However, the bonds density and the symmetry properties of a repetitive pattern allow exploiting all CRS specific strengths, in particular regarding spectral resolution or light polarization.

In the first presented study, we have implemented polarization-resolved scheme to retrieve the molecular symmetries and thus target more specifically the vibration to improve the signal contrast. We have used the irreducible tensor formalism to describe both the susceptibility tensor and the polarization of the electromagnetic fields [1]. By choosing a specific set of circular polarizations, the symmetries of the crystals could be highlighted, whatever the crystals orientation in the sample plane was [2-3].

In a second study, we have used the specific spectral signatures of crystalline polymorphs to investigate the biomineralization process of an oyster shell, made of calcium carbonate crystals [4]. We could image the spatial distribution of the free carbonate and the crystalline carbonate at the growing edge of the shell. Those detailed polymorph maps, combined with a proteins image, allow us to propose a biomineralization scenario.

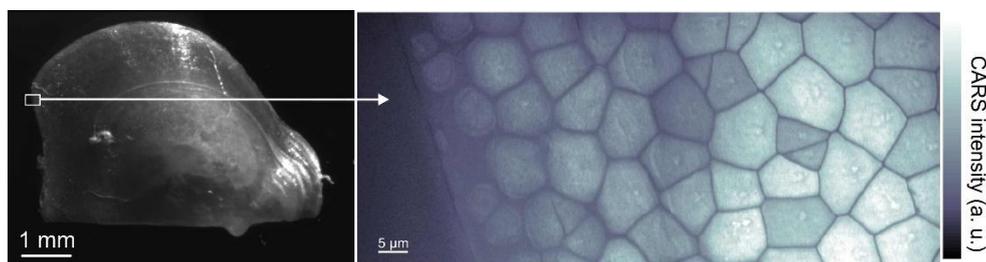


Figure 1: Structure of a calcium carbonate shell imaged with Coherent Raman scattering.

- [1] J. Duboisset, Phys. Rev. A. **90**, 063827 (2014)
- [2] C. Cleff, Nature Commun., **7**, 11562 (2016)
- [3] C. Cleff, Phys Rev A., **96**, 013851 (2017)
- [4] F. Mastropietro, Nat materials, **9** 946 (2017)