POLARIZATION RESOLVED COHERENT RAMAN MICROSCOPY

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Coherent Raman microscopy is a nonlinear optical technique that provides label-free imaging thanks to its resonant nature. The interaction of the incident fields with the sample induces a third-order nonlinear polarization whose properties depend on the structure of the medium, characterized by a fourth-rank susceptibility tensor, $\chi^{(3)}$. This tensor contains rich information on (1) the microscopic-scale vibrational symmetry properties; (2) the macroscopic-scale structural information in ordered media.

We present here an extensive approach based on multiple-fields polarization resolved CARS and SRS, in order to unravel the complexity of vibrational resonances up to the fourth order symmetry, at the microscopic scale in isotropic and non-isotropic media.

The CARS polarized signals measured under a continuous variation of the incident Pump and/or Stokes excitation beams are analyzed using a full tensorial picture both in the non-resonant and resonant regimes. This method evidences the strong influence of vibrational symmetries on polarized CARS, and more specifically the occurrence of Kleinman symmetry deviations at the vicinity of the Raman lines frequencies. The technique is illustrated on a cubic symmetry crystal but is general and can be applied to other medium symmetries.

As an example, we applied the multiple-fields polarization resolved CARS to probe the organization of collagen fibers, showing the potential the technique for conformational investigation in biology.

Figure: A: CARS image of rat tail collagen, B: CARS signal resolved in polarization (red: along x, green:along y) in point 1 of image A when the Stokes beam linear polarization is fixed along the x axis and the pump beam linear polarization is rotated, C: deduced orientational distribution of collagen fibres in point 1 of figure A.

References