Using circularly polarized CARS microscopy to reveal sample symmetries

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The microscopic orientation of a sample within a macroscopic ensemble has a strong influence on the properties of a sample, e.g., in crystals or for lipids forming the membrane of a cell. For coherent anti-Stokes Raman Scattering (CARS) microscopy specific schemes have been developed to gain information about a sample’s symmetry by imaging it at different rotational angles of the linear polarization state of the excitation beams and subsequent analysis of the response of the sample\cite{1}. Here, we present an approach based on circularly polarized excitation and circular polarization-sensitive detection, allowing direct imaging of specific orders of symmetry of a sample which contain rich information on molecular structural behaviors. In contrast to previous works based on circular excitation\cite{2}, the circular polarization-sensitive detection allows to differentiated between individual orders of symmetry. We experimentally demonstrate the symmetry resolving properties of our approach using samples with a well-known symmetry such as HT8 zeolite crystals [four-folded symmetry, fig 1 (a)] and multilamellar lipid vesicles (MLV) [two-folded symmetry, fig 1 (b)]. Both samples are surrounded by water, creating a strong non-resonant image background. Consequently, regular (linearly polarized) CARS microscopy produces low contrast images. On the other hand, symmetry-resolved CARS imaging allows to separate the contribution of an isotropic background and anisotropic signals of a sample such that in case of HT8 zeolite (MLV) a contrast enhancement by a factor of 78 (80) is achieved when a four-folded (two-folded) symmetry is targeted. The achieved contrast enhancement is limited by detector noise and can be much higher. As no post-processing of the images is required to retrieve the symmetry information, this approach is well suited for real time imaging and can be combined with most existing CARS techniques. Moreover, different vibrational modes (bending, stretching, symmetric, asymmetric, ...) show different symmetries such that symmetry-resolved CARS can be used to distinguish them. Consequently, in addition to a strong contrast enhancement a better chemical selectivity can be achieved, as Raman states can be distinguished by their energy as well as by their symmetry.

Fig. 1. Regular and symmetry-resolved CARS images of (a) HT8 zeolite crystals (at 932 cm\(^{-1}\)) and (b) MLV (at 927 cm\(^{-1}\)).