

# Nonlinear Optical Imaging of In-Plane Anisotropy in Two-Dimensional SnS

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Two-dimensional (2D) tin(II) sulfide (SnS) crystals are orthorhombic, semiconducting group IV monochalcogenides, which are characterized by remarkable properties, such as in-plane anisotropic optical and electronic response [1]. This anisotropic response is exhibited along the in-plane armchair (AC) and zigzag (ZZ) crystallographic directions, offering an additional degree of freedom in manipulating their behavior [1]. Here, we perform polarization-resolved second harmonic generation (P-SHG) nonlinear imaging on liquid phase exfoliated SnS containing monolayer and bilayer crystals [2, 3], which lack inversion symmetry and produce SHG [4, 5]. We fit pixel-by-pixel the P-SHG experimental data with a nonlinear optics model, that allows us to calculate and map with high-resolution the AC/ZZ direction of several 2D SnS flakes belonging in the same field of view [2]. It is found that the P-SHG intensity polar patterns are associated with the crystallographic axes of the flakes and with the relative strength of the second order nonlinear susceptibility tensor in different directions. Therefore, our method provides quantitative information of the optical in-plane anisotropy of orthorhombic 2D crystals [2], offering great promise for performance characterization during device operation in the emerging optoelectronic applications of such crystals.

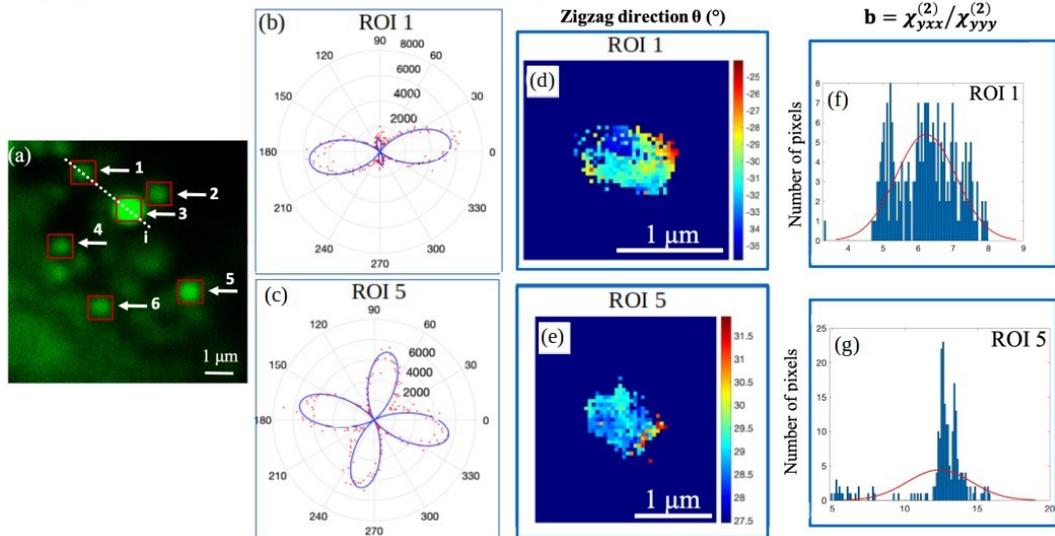


Figure 1: (a) SHG image of 2D SnS crystals. (b, c) Experimental data (in red dots) and fitting (blue line) of the P-SHG intensity, for the ultrathin SnS crystals 1, 5 depicted in (a). The P-SHG intensity is presented in polar plots as function of the linearly polarized excitation angle. (d, e) Pixel-by-pixel spatially resolved mapping of the ZZ crystallographic direction, and (f, g) histograms of the fitted  $\chi^{(2)}$  parameter  $b$ , for the ultrathin SnS crystals 1, 5 depicted in (a).

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