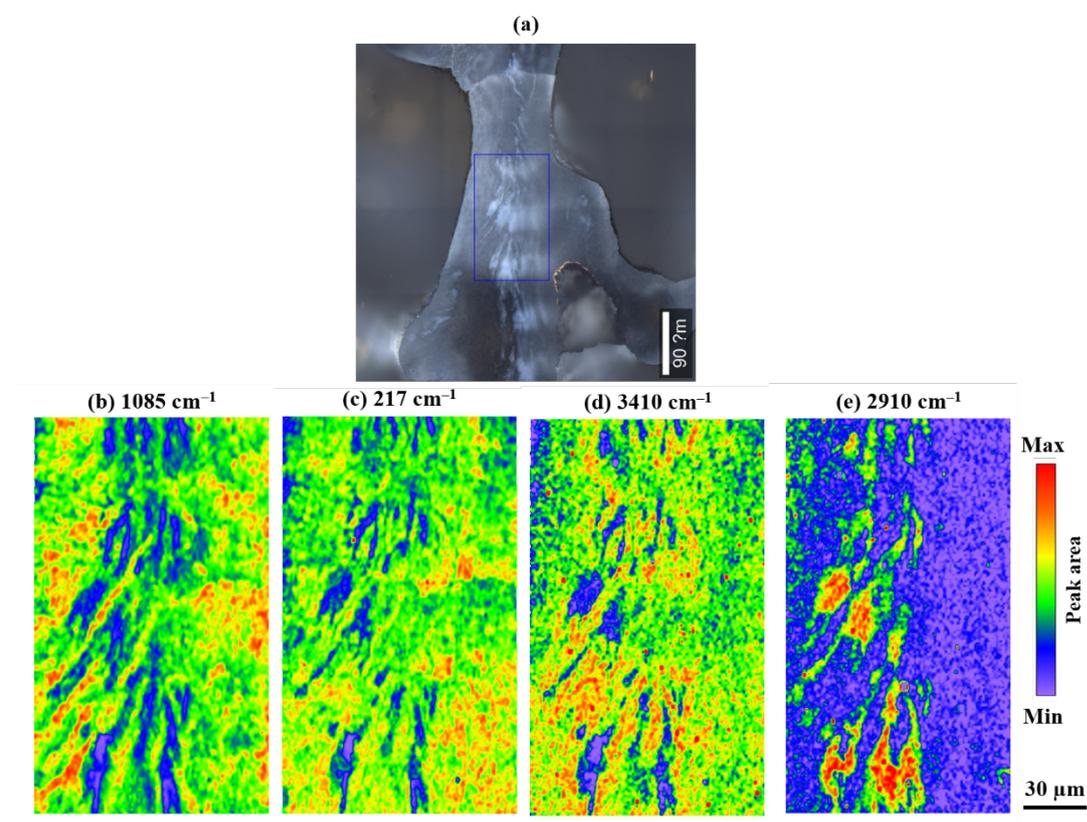


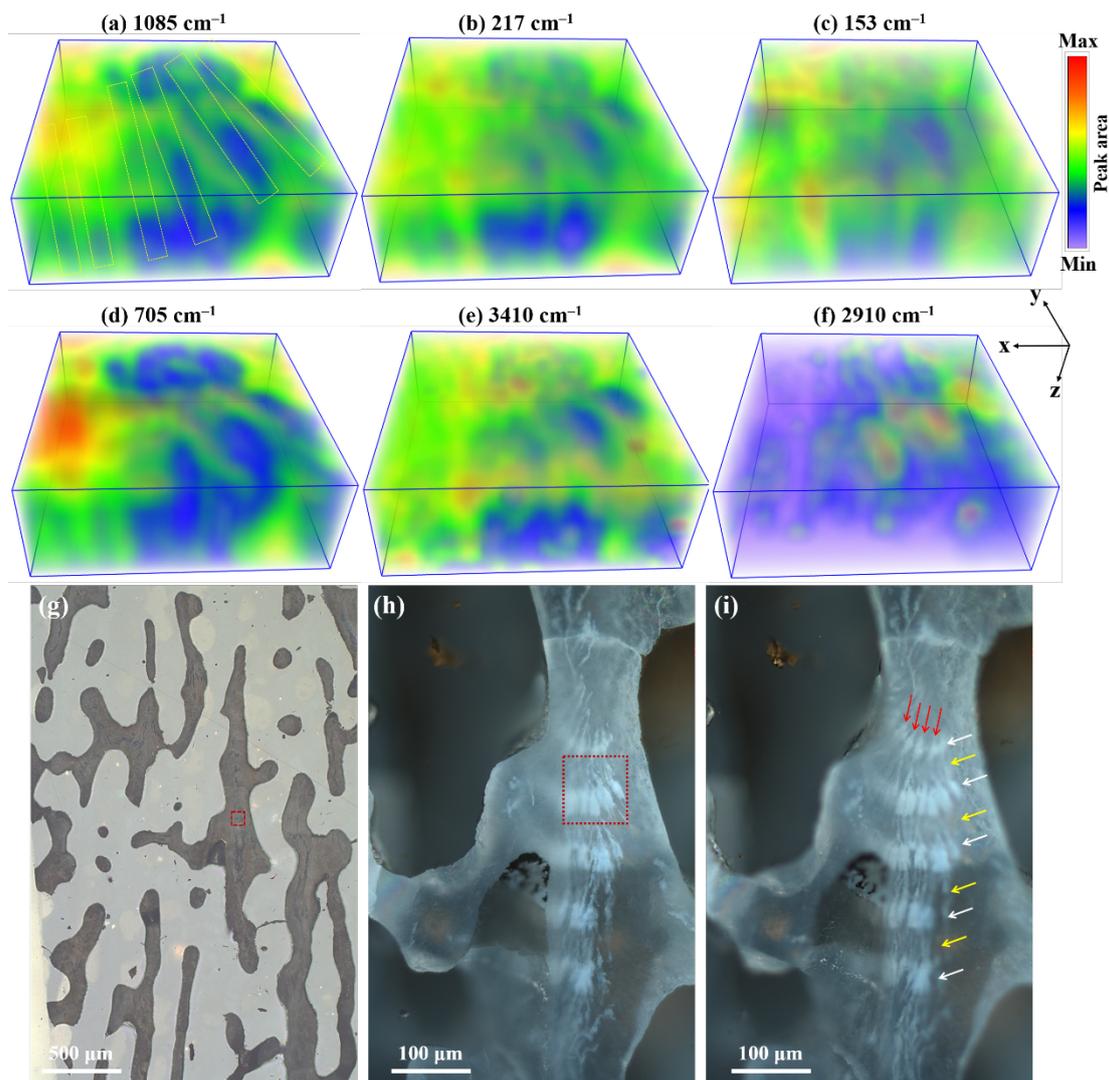
Supplemental material for Biological control of ultra-skeleton mineralization in coral

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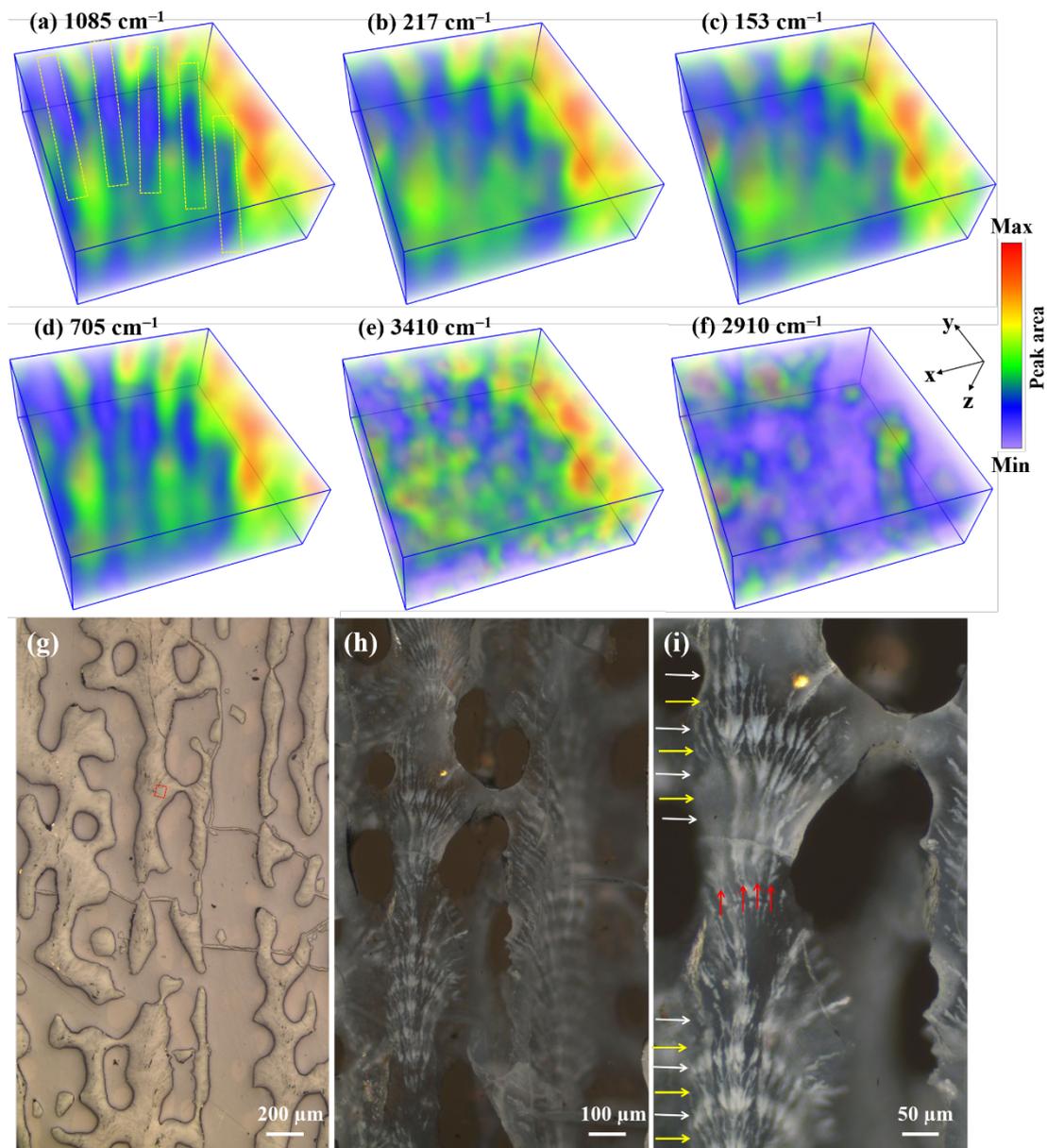
State Key Laboratory of Isotope Geochemistry, CAS Center for Excellence in Deep Earth Science, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China



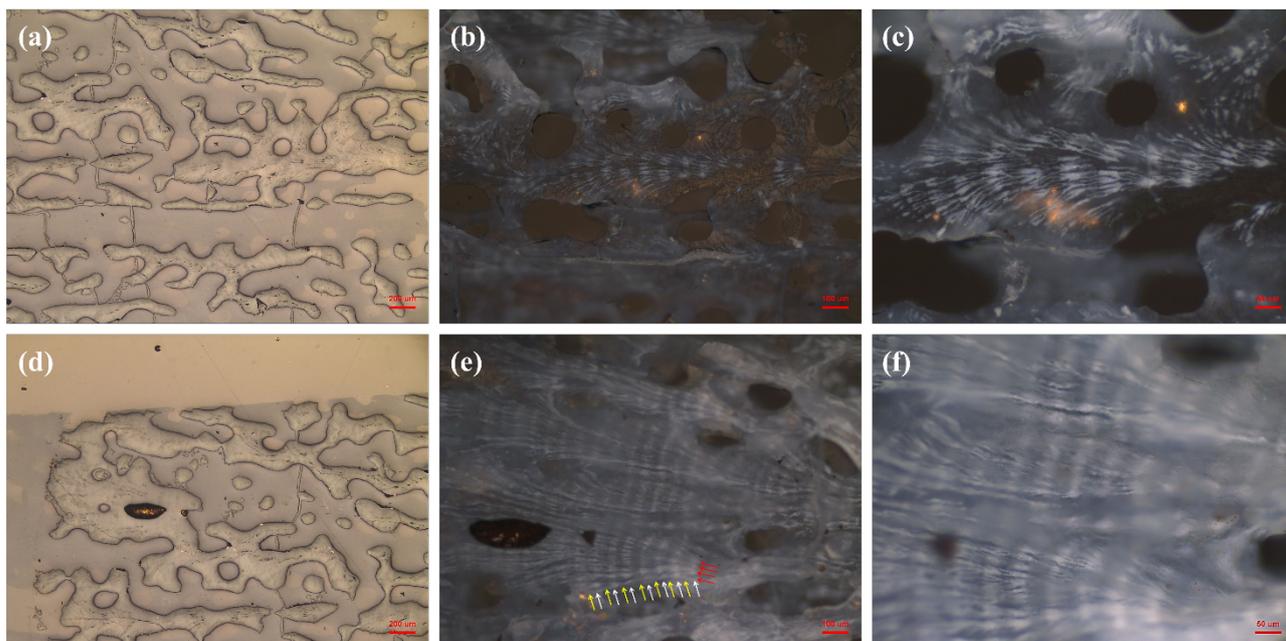
Supplementary Fig. 1 Two-dimensional images of the second coral skeleton over a $106 \times 180 \mu\text{m}$ area. (a) Cross-polarized reflected light (CPRL) microscopy image; and (b–e) images obtained from the 1085, 217, 3410, and 2910 cm^{-1} Raman bands, respectively.



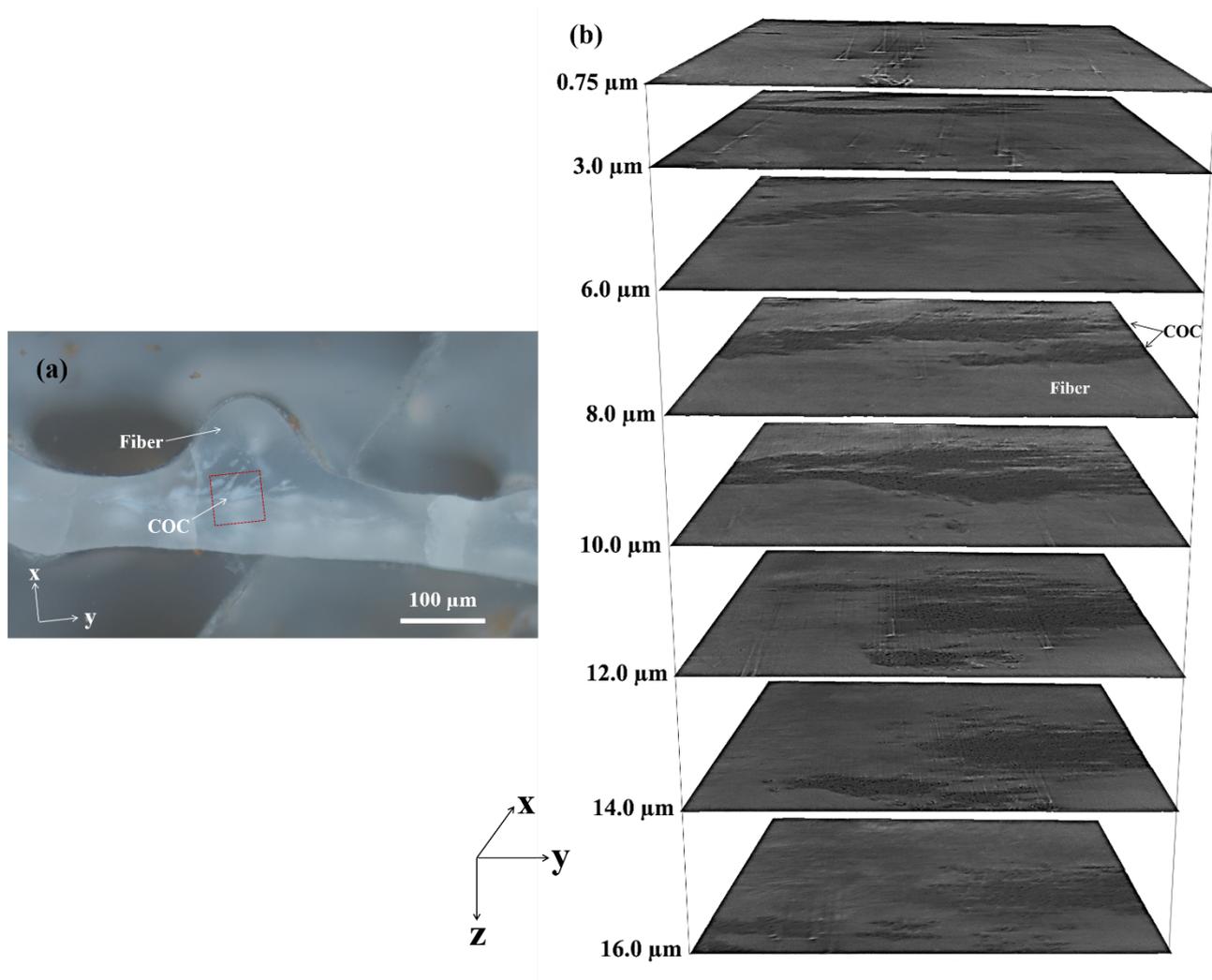
Supplementary Fig. 2 (a–f) Three-dimensional Raman imaging of the Raman bands at 1085, 217, 153, 705, 3410, and 2910 cm⁻¹ in the second coral skeleton. The imaged skeleton of optical (g) and cross-polarized reflected light (CPRL) microscopy (h-i) images with modification of the focus length, and the red dotted rectangle is the imaged area. Blue–purple regions in (a–e) correspond to the COC, and the white, yellow and red arrows indicate the nodules, filaments, and COC columns, respectively.



Supplementary Fig. 3 (a–f) Three-dimensional Raman imaging of the Raman bands at 1085, 217, 153, 705, 3410, and 2910 cm⁻¹ in the third coral skeleton. The imaged skeleton of optical (g) and cross-polarized reflected light (CPRL) microscopy (h–i) images, and the red dotted rectangle is the imaged area. Blue–purple regions in (a–e) correspond to the COC, and the white, yellow and red arrows indicate the nodules, filaments, and COC columns, respectively.



Supplementary Fig. 4 Optical microscopy images of the section of the third *Porites* coral skeleton. (a–c) Optical and CPRL microscopy images of two different sections of the surfaced skeleton. (d–f) Optical and CPRL microscopy of the sub-surfaced skeleton. (a) and (d) are optical microscopy images of the coral skeleton. (b–c) and (e–f) are CPRL microscopy images, with (c) and (f) being higher-magnification views of (b) and (e). The white, yellow and red arrows indicate the nodules, filaments, and COC columns, respectively.



Supplementary Fig. 5 Scanning Electron Microscope (SEM) analyses the Focused-Ion-Beam (FIB) polished coral skeleton. (a) the analyzed skeleton under CPRL, where the red rectangle is the FIB-SEM imaged area with size of $60\ \mu\text{m}\times 60\ \mu\text{m}$, slightly smaller than that of Raman imaging (Fig. 4). (b) The quasi three-dimensional reconstruction of skeleton using FIB-SEM. The FIB-SEM three-dimensional reconstruction could offer 3D rendition of the COCs vs. fibers, in which the former appears darker than the latter due to the porosity of morphology of COC crystals, which is basically consistent with 3D Raman images.