The temperature dependence on crystal structure of chiral perovskite thin film and the estimation of corresponding 2nd order nonlinear susceptibility

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Chirality is a key characteristic in many physical, chemical, and biological systems, profoundly influencing their properties and behaviors in chiral environment. This research specifically focuses on the effects of temperature and crystal structure on chirality in perovskite (PVK) materials, a class of compounds with immense significance in optoelectronics. Besides the crystal phase transfer during film fabrication, the work posits the estimation of second-order nonlinear susceptibility ($\chi^{(2)}$), a critical property in nonlinear optics which encapsulates all pertinent information about the material's chirality for chiral PVK films.

Employing PVK synthesized by chiral methylbenzylamine, circular dichroism (CD) spectra is applied to evaluate the chiral intensity, while X-ray diffraction (XRD) and second harmonic generation CD (SHGCD) are employed to characterize the chirality difference as well as $\chi^{(2)}$ based on structural symmetry of PVK thin films fabricated under different fabrication parameters. We found the chirality of thin films annealed under different temperature exhibit different CD, indicating the chirality's temperature dependence. Based on the results of powder XRD, we have ascertained the crystalline structures of thin films at various temperatures (from 80°C to 120°C), all of them possess the symmetry of C_∞ macroscopically, while the microscopic symmetry changes by temperatures, such as C_2 (phase A) and D_2 (phase B) shown in fig a. Combined with the SHGCD intensity generated by Nd: YAG laser with wavelength of 1064nm, the relative magnitude of $\chi^{(2)}$ for C_∞ symmetry, fabricated under 120°C have been quantitatively computed in fig b as $\chi_{zxx} = 1, \chi_{xyz} = 0.142 - i0.213, \chi_{zzz} = 5.015 + 1.414\chi_{xxz}$ (using χ_{zxx} as reference value), among which we can infer the role of each tensor that affecting the chirality, e.g., imaginary part of χ_{xyz} is particular sensitive to the difference on SHG intensity between L- and R- circular polarization. As all the information regarding symmetry and chirality is embedded in the $\chi^{(2)}$ tensors and combinations, this experiment significantly contributes to the understanding of how the $\chi^{(2)}$ tensors changes during crystal phase transition and further influent the film's chirality. This relationship can provide the potential approaches to choose or design the chiral electronic materials with desired chirality based on their $\chi^{(2)}$.

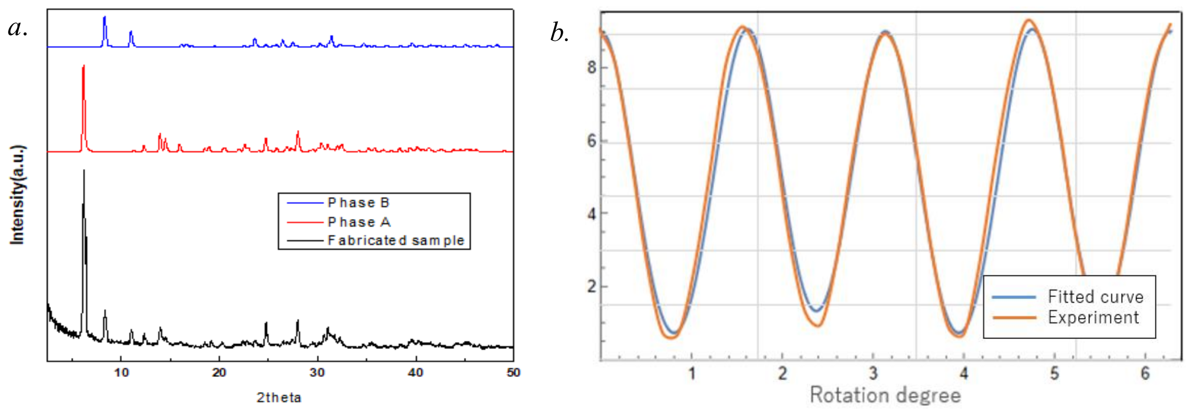

Fig a. XRD fitting for film crystal structure under 120°C, the film contains two structures (phase A and B)

Fig b. The fitted result of SHG curve for the sample with C∞ point group.