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GROWTH MECHANISM OF POLAR ORGANIC NONLINEAR OPTICAL CRYSTALS

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Organic crystals with large second-order non-linear susceptibility attract much interest because of their potential applications, such as second harmonic generation, frequency mixing, optical parametric oscillation and spatial soliton generations. However the difficulty in growing such crystals of large size and high perfection has been the major obstacles for practical applications. 3-methyl-4-methoxy-4'-nitrostilbene (MMONS) crystals show one of the largest second-order non-linear susceptibilities, d_{eff}=37pm/V at 1.064 ㎛. MMONS crystals belong to space group Aba2 (point group mm2 Z=8) with lattice parameters a=15.750Å, b=13.470Å, c=13.356Å. The MMONS molecule has a permanent dipole moment along the charge transfer axis. When it is crystallized, the molecular dipole moments are summed up to cancel out each other in the crystallographic a- and b-axes, but the net dipole moment remains along the polar c-axis, which manifests strong polar nature of the crystal. This polar nature not only strongly affects the crystal growth process but also overall linear and nonlinear optical properties.

A number of different solvents were attempted to grow single crystals of MMONS. The morphology and quality of the grown crystals varied significantly depending on the strength of solvent polarity. The overall morphology is represented by the truncated octahedra covered with the well-facetted {111}, {21$\bar{1}$}, {120}, {100} and (001) faces, which is a consequence of the unidirectional growth along a direction of the polar axis. The observed polar morphology and growth behaviour can be explained by the molecular recognition concept and the interplay of the dipole-dipole interactions between MMONS molecules, and between solvent and MMONS molecules. Single crystals of a large size ($40 \times 40 \times 40 mm^3$) and of high perfection were grown by the low temperature solution growth method using methyl ethyl ketone as a solvent.

The perfection of the crystal was characterized by synchrotron X-ray topography using 002 and 004 reflections, which indicates that the crystal is highly perfect. Optical characterization was made by measuring second harmonic conversion efficiency and laser damage threshold.