

1.14 Heteroepitaxial Growth of MNBA on Lattice-Matched EDT Single Crystals by Organic Molecular Beam Epitaxy

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In an effort to find suitable substrates for heteroepitaxial growth of the highly nonlinear optical organic compound MNBA a systematic scan of the Cambridge Structural Database (CSD) was performed. The organic salt ethylenediammonium terephthalate (EDT) was found to match the structure of MNBA in the (010) plane, with a lattice mismatch of less than 0.3 %.

EDT was synthesized in a solution of dimethylformamide (DMF) from the commercially available starting materials ethylenediamine and terephthalic acid. Single crystals with typical dimensions of several millimeters were grown from aqueous solution by solvent evaporation or temperature lowering techniques. X-ray precession measurements were performed with several samples to identify their (010) faces.

The surfaces of as grown crystals were cleaned in the organic deposition chamber by a thermal desorption procedure at 120 °C for 30 min, prior to the growth experiment. Optimum heteroepitaxial growth conditions were determined experimentally by varying the deposition parameters systematically. Long range ordered island growth was observed on (010) faces of EDT, at a substrate temperature of 80 °C, and a growth rate of 0.1 Å/s. Below 70 °C no epitaxial growth of MNBA on EDT substrates was observed, and above 90 °C the desorption of MNBA dominated the growth process.

Nonlinear optical second harmonic generation (SHG) was demonstrated in heteroepitaxially grown MNBA thin films. EDT crystallizes centrosymmetrically (space group P21/n), so that no SHG contributions arise from the substrates. As MNBA features only one major nonlinear optical coefficient ($d_{111} = (175 \pm 20) \text{ pm/V}$ @ $\lambda = 1064 \text{ nm}$), the intensity of the SHG signal was theoretically predicted to follow a $\cos^4\theta$ dependence on the angle θ of the linear polarization plane of the fundamental laser beam. The calculated intensity dependence was confirmed experimentally, and thus the long-range order in the heteroepitaxial films was evidenced.

MNBA: 4'-nitrobenzylidene-3-acetamino-4-methoxy-aniline

EDT: ethylenediammonium terephthalate