

Abstract

Within the scope of this thesis a new strategy was developed to determine the second-order nonlinear optical tensors [d_{ijk}^{SHG}] using the Maker-fringe measurement method for crystals of any symmetry. The theoretical way was realised in a program. For the first time it was possible to determine all components [d_{ijk}^{SHG}] of a triclinic crystal, the sarcosinium tartrate, $C_3H_8NO_2^+C_4H_5O_6$. The largest d_{ijk}^{SHG} value amounts 1.4(4) pm/V. Sarcosinium tartrate is a promising candidate for technical applications: A second harmonic generation in phase-matching directions is possible. In addition, crystals of optical quality are available via crystal growth.

Furthermore, the complete second-order nonlinear optical tensors were determined for the first time of lithium sulfate monohydrate ($Li_2SO_4 \cdot H_2O$) (PG:2), cesium lithium molybdate ($CsLiMoO_4 \cdot 1/3H_2O$) (PG: $\bar{4}3m$) and strontium tartrate antimonate ($Sr[Sb_2\{(+)C_4H_2O_6\}_2] \cdot 2H_2O$) (PG:6). The tensors [d_{ijk}^{SHG}] of the lead tetraborate (PbB_4O_7) (PG:mm2) and yttrium formate dihydrate ($Y(HCOO)_3 \cdot 2H_2O$) (PG:222) were redetermined and are in agreement with literature. The largest SHG coefficient within the scope of this work was measured in PbB_4O_7 with an amount of 4.0 (4) pm/V. The crystals of PbB_4O_7 and of $CsLiMoO_4 \cdot 1/3H_2O$ are not phase-matchable, whereas crystals of $Li_2SO_4 \cdot H_2O$, $Sr[Sb_2\{(+)C_4H_2O_6\}_2] \cdot 2H_2O$ and $Y(HCOO)_3 \cdot 2H_2O$ are phase-matchable. However, the effective values in phase-matching directions are too small to compete with established crystals for applications as frequency converters.

Precision measurements of refractive indices and their dispersion are fundamental for all non-linear optical investigations. These linear optical data are required for both the analysis of the Maker fringes and the calculation of the phase-matching directions.

The theoretical calculation of Bechthold [76Be] forms the basis of the analysis of the non-linear interaction of light within a crystal in form of a plane parallel slab. In contrast to the work of Bechthold numerical methods were used in this work. Based on the theoretical interpretation of the Maker-interference method several programs were developed allowing the calculation of Maker interference curves for all point groups in any orientation.