o.m1.p21 What crystallography can bring to the determination of cubic optical nonlinearities in non-centrosymmetric crystals.

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Keywords: ferro-electricity, NLO materials.

We present the measurement of two elements of the third order dielectric susceptibility tensor $\chi^{(3)}$ of $KTiOPO_4$ (KTP). These two coefficients, $\chi^{(3)}_{16}$ and $\chi^{(3)}_{24}$, are the ones involved in phase-matched 4-wave optical frequency conversion interactions. Because of the weak magnitude of the $\chi^{(3)}$ coefficients in the case of non-resonant processes, phase-matching is the only way to achieve efficient 4-wave frequency conversion. From this point of view, KTP is a very interesting crystal because phase-matching conditions exist over a wide spectral range.

In order to achieve accurate measurements, the $\chi^{(3)}$ coefficients are deduced from the optical power generated during a phase-matched interaction because the associated conversion efficiency is maximum. In our experiments, we choose a direct third-harmonic generation (THG) interaction $\omega+\omega+\omega=3\omega$. Because KTP is non-centrosymmetric, the second order dielectric susceptibility tensor $\chi^{(2)}$ is non-zero, so that 3-wave interactions are allowed ; in particular, the cascading of second-harmonic generation $\omega+\omega=2\omega$ and sum-frequency generation $\omega+2\omega=3\omega$ also generates the third harmonic of the fundamental incident laser beam. Even though these quadratic interactions are non-phase-matched, the resulting third harmonic power may be of the same order of magnitude whenever compared to that generated by the cubic THG process because of the relative amplitude between the $\chi^{(2)}$ and $\chi^{(3)}$ elements. Thus, the determination of $\chi^{(3)}$ coefficients requires to precisely quantify the respective contributions of the cubic and cascaded quadratic interactions to the third harmonic power ; in particular, one has to know the relative sign between the quadratic and cubic dielectric susceptibilities.

For that we calculate the $\chi^{(2)}$ and $\chi^{(3)}$ elements using a bond charge model[1] that we generalize for $\chi^{(3)}$. The model, based on a decomposition of the macroscopic dielectric susceptibility into the bond polarizabilities, is suitable for the modelling of the different orders of the dielectric susceptibility of mineral materials. The calculated values of $\chi^{(1)}$ and $\chi^{(2)}$ are in satisfying agreement with the measured coefficients. In the case of $\chi^{(3)}$, the calculated polarizabilities of all the different Ti-O and P-O bonds are very similar, and the deduced coefficients mainly depend on the geometrical factors due to the projection of each bond over the optical frame axes. These geometrical factors calculated from the crystal structure of KTP are all positive. From the previous calculations, all $\chi^{(2)}$ and $\chi^{(3)}$ elements of KTP are positive.

This conclusion is very important since it allows us to determine $\chi^{(3)}_{16}$ and $\chi^{(3)}_{24}$ from our THG experiments[2]. Moreover, the relative value of all the $\chi^{(3)}_{ij}$ elements deduced from the bond charge model is also in very good agreement with our measurements and with the other coefficients measured by z-scan[3,4].

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o.m1.p22 Piezo-electric materials. X-ray diffraction from crystals under electric fields. A first test on alpha quartz.

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The aim of our work is to analyse by diffraction techniques the correlations between structural and physical properties of crystals onto which an electric field is applied.

In the laboratory in Nancy we have, based on the ideas of previous work [1,2], build a device using a field switching technique. It consists of a high voltage supply, the electronics for switching the field, and synchroneous counting on four chains combined with a control for step-scanning the diffraction profiles (details are given the poster by P. Allé *et al*.). By using this 'stroboscopic' technique, it is possible to measure very small changes in the Bragg angles due to the strain resulting from the converse piezoelectric effect, and also to measure minute changes in the Bragg intensities due to polarisations of atomic structure and electron density.

A first test of the instrument was carried out at LURE with the 4-circle diffractometer WDIF-4C. For this test we have chosen alpha quartz, since perfect crystal samples can easily be obtained. Furthermore, quartz is a well-characterised piezo-electric material. Electrodes were vapour deposited onto the (011) extended faces of a crystal plate of dimensions 10 x 5 x 0.5 mm. We have been able to measure changes in the Bragg angles as small as 0.0005°. These changes are, in good agreement with the known piezo-electric tensor coefficients of alpha quartz.

Future work will mainly concentrate on studies of the effect of electric fields on the structure of ferro-electric materials at temperatures close to phase transitions.

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