	Experiment title: Parametric down-conversion of x-rays into ultraviolet in ferroelectric crystals	**Experiment number**: HC 3473
Beamline: ID20	**Date of experiment**: from: 24.01.18 to: 30.01.18	**Date of report**:
Shifts: 15	**Local contact(s)**: Blanka Detlefs	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Sharon Shwartz[1]*

Or Sefi[1*]

Edward Strizhevsky[1*]

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In this report we describe the main results of the experiment on parametric down-conversion of x-rays into ultraviolet radiation in the ferroelectric crystal $LiNbO_3$ performed at beamline ID-20.

Introduction:

The objective of this proposal was to pursue the development of a new inelastic spectroscopy technique based on the effect of parametric down conversion (PDC) of x-rays into ultraviolet (UV) radiation. In order to achieve that goal, the effect was explored in a ferroelectric lithium niobate (LiNbO3) single crystal. PDC is a second order nonlinear process in which an x-ray pump photon decays into two photons, where one is an x-ray photon and the other is a UV photon. Recently, it has been shown that the effect can be used to study excitations of valence electrons and structures of chemical bonds with atomic scale resolution. Since the generated photons are completely correlated this is done by measuring only the generated x-ray photons. So far, the effect has been measured mostly in semiconductors and the extension to other materials demonstrated the potential for the measurements of properties of valence electrons in complex materials.

Experimental procedure:

The experimental setup is shown in Fig. 1. The nonlinear media was a $LiNbO_3$ single crystal. The pump beam was at 10 keV. We moved the crystal and the two detectors to the phase matching positions. We used a Si (4 4 0) double-bounce analyzer to suppress the Compton background and the tail of the elastic at the x-ray detector. For detection we used a Medipix multipixel detector with a pixel size of $55\times55\mu m^2$. We measured the PDC signal at different reflections and idler

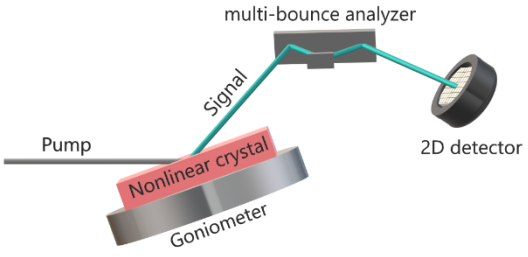

Fig.1: Experimental setup

[1] Physics Department and Institute of Nanotechnology, Bar Ilan University, Ramat Gan, 52900 Israel.

energies by scanning the angle of the crystal and the energy of the analyzer.

Summary of results:

The dependencies of the efficiencies on the idler energies and on the inter – planer spacing is shown in Fig. 2. The efficiencies behave differently than the Bragg efficiencies, which deacrease for smaller inter–planer spacing. The measured efficiencies are extremely high, about three orders of magnitude higher than in diamond crystals that we measured before at ID-20. Additionally, the efficiencies along the polarized direction of the crystal, (0 0 6) and (0 0 12) reflections, are much higher than the efficiencies along the un-polarized directions. In fact, the ratio between the efficiencies in the polarized and un-polarized directions fits the efficiencies ratio for second order nonlinearities in the optical regime. These results suggest a new underlying physical mechanism, which dominates this process.

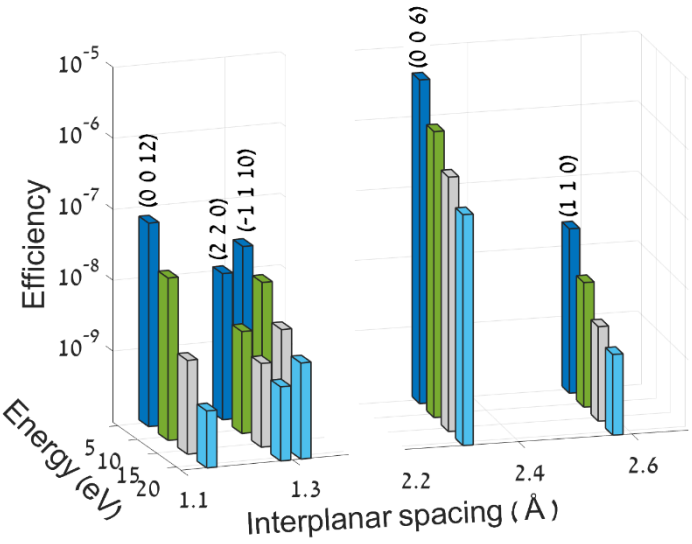

Fig.2: Comparison between measured efficiencies of PDC in $LiNbO_3$ for several atomic planes for signals, which correspond to idler energies at 5 eV, 10 eV, 15 eV, and 20 eV. The horizontal axis represents the interplanar spacing, which corresponds to the Miller indices of the various atomic planes.

In order to improve the understanding of the effect and in order to show the possibility for its application for spectroscopy, we measured the spectral dependence of the effect. The spectra for the (1 1 0), (3 3 0), (-1 1 10) and (0 0 6) are shown in Fig. 3. Each peak in each spectrum can be attributed to an electronic transition. The peak at 5.9 eV, which appears at the (1 1 0) and (3 3 0) atomic planes and the broad structure we observed for the (-1 1 10) atomic planes, which are shown in Fig.5 (a)-(c) respectively, can be attributed either to the direct band transition or to the Li-2s atomic resonance. The peak at 7.5 eV, which appears in Fig. 3 (a)-(b) can be attributed either to the transition between the valence and the second conductance bands or to the Nb-4s atomic resonance. The peaks at 12 eV and 15 eV, which appears for the (-1 1 10) and the (3 3 0) atomic planes relate either to higher band transitions or to a deeper atomic level. Fig. 5 (d) shows the spectrum for the (0 0 6) atomic planes. In these measurements the atomic planes that participate in the effect are directed along the polarized direction on the crystal. This spectrum shows no prominent features. The main implication of the measured spectra presented here is that the effect of PDC of x-ray into longer wavelengths can be used as a powerful tool to investigate phenomena in solid-state physics and in atomic physics.

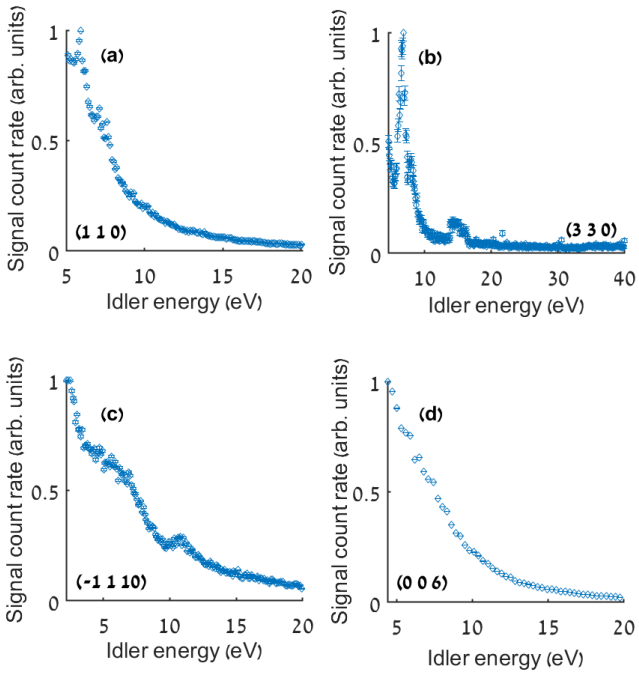

Fig.3: Spectral measurements for PDC signal in $LiNbO_3$ for the (a) (1 1 0) atomic planes, (b) (3 3 0) atomic planes, (c) (-1 1 10) atomic planes and (d) (0 0 6) atomic planes. The features in (a)–(c) can be attributed to either band transitions or atomic resonances. See further detail in the text.

In conclusion, we have measured the x-ray signal of PDC of x-rays into ultraviolet radiation and have shown its possible application for spectroscopy.

Future & perspective:

We will repeat this experiment in more exotic materials and explore the possibility for the development as a spectroscopy tool for the valence charges.