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## New detecting techniques for a future calorimetry

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**Abstract.** In the last forty years, application of crystalline materials in homogeneous Electromagnetic Calorimeters has played a crucial role in the discovery of matter properties and promoted a continuous progress in the detecting technique. The detection systems progressed from small detectors based on NaI(Tl), CsI(Na), BaF<sub>2</sub>, PbF<sub>2</sub>, and Bi<sub>4</sub>Ge<sub>3</sub>O<sub>12</sub> to giant Electromagnetic Calorimeters of CMS, ALICE Collaborations at LHC and PANDA Collaboration at FAIR, where the systems consisted of thousands lead tungstate PbWO<sub>4</sub> scintillation crystals. Lead tungstate (PWO) became the most extensively used scintillation material in high energy physics experiments. PWO possesses a unique combination of scintillation properties including high energy and time resolutions in the detection of high energy particles. Here, we report on the results of the two photon absorption in PWO crystals obtained by pump-probe technique using ultra short laser pulses. The results demonstrate that the relaxation processes in PWO offer capability of this material to be used in detection systems to make a time stamp with precision close to 10<sup>-12</sup> s or even better.

### 1. Introduction

There is an upcoming demand for a new generation of detectors enabling a high time resolution in high energy physics experiments. To develop the new generation of ultrafast detectors, especially in experimental particle physics at high luminosity colliders, the processes permitting the measurement precision better than 10-20 ps for the interaction time between ionizing radiation and detecting medium need to be identified. In order to reduce the interaction region and have a compact experimental setup in high energy physics experiments, detectors with short radiation  $X_0$  and nuclear interaction  $R_H$  lengths and small Moliere radius  $R_M$  are on demand. Among the variety of inorganic scintillation materials meeting these requirements, lead tungstate (PbWO<sub>4</sub>, PWO) self-activated scintillation material possesses a unique combination of scintillation and other physical properties [1].



It ensures high energy and time resolutions, especially in electromagnetic calorimetry [2-4]. However, even the self-activated scintillation materials like PWO with fast scintillation decay (10 ns) and rise (0.1ns) time constants has limited time resolution due to stochastic origin of luminescence processes. The available data indicate that scintillation materials can not provide a better time resolution than 50 ps. The authors of [5] considered another options to improve the time resolution by exploiting of the transient phenomena, which appear at the early stages of the interaction between ionizing radiation and crystalline material. These processes occur when the density of states has peculiarities, such as a gap in the bottom of the conduction band. In such conditions, hot intra-band radiative and absorption transitions occur in the conduction band. These processes take place before free carriers are captured by the luminescent centers and/or shallow electron traps. The time scale of these processes is considerably shorter than the scintillation rise time of a self-activated scintillation crystal and does not exceed 10-30 ps. However, these transient phenomena can be reliably observed at high free carrier density, thus they can be utilized at the conditions of a large and high speed energy deposit in the material. Such conditions are achievable in collider experiments at LHC or ILC [6,7]. Thus, a search for fast processes with a weaker or indirect dependence on the free carrier density is a necessary. Two photon absorption in crystalline media is one of such processes. The effect is proportional to the laser pulse power squared and is relatively easy observed by picosecond laser spectroscopy in many media [8]. It is expected that electromagnetic shower might induce the polarization of the crystalline media and change the conditions for two photon absorption.

In the present work, we studied two photon absorption in PWO single crystals. The band gap  $E_g$  of PWO at room temperature is 4.33 eV, so a defect-free PWO crystal does not absorb in the visible range. Two photon absorption in the range of photon energy between 3 and 4 eV results in excitation to the electronic states in the bottom part of the conduction band. The bottom of the conduction band is formed by d orbitals of tungsten ions. Moreover, the density of electronic states has two peaks near 4 and 6 eV [9]. The first peak corresponds to the states of triplet terms  $^3T_1$ ,  $^3T_2$ , whereas the peak near 6eV is formed with strong admixture of  $^1T_1$ ,  $^1T_2$  states. Thus, effective two photon absorption due to the transitions from the ground state which is the upper  $^1A_1$  state of the valence band to the  $^1T_2$  state of the conduction band is expected in the range of 6eV.

## 2. Experimental technique and results

A 10 mm thick sample of  $PbWO_4$  crystal with perfect optical quality and oriented along the crystallographic axis  $a$  was selected for our measurements. The experiment was carried out using a pump-probe spectrometer based on a custom-made original femtosecond Ti:Al<sub>2</sub>O<sub>3</sub> pulsed oscillator and a regenerative amplifier, both operating at 10 Hz repetition rate [10]. The laser pulse duration and energy after amplification were 140 fs and up to 0.5 mJ, respectively, while the wavelength was tunable over the spectral range from 770 to 820 nm. The pulses of the fundamental frequency ( $\omega$ ) at the output of the amplifier (790 nm wavelength was set for the present study) were divided into two parts in a 1:4 ratio (Figure 1). The beam of higher intensity was converted to the third harmonic ( $\lambda \approx 263$  nm,  $E$  up to 12  $\mu$ J) and used as the pump pulse. The pulse energy was chosen to prevent the sample surface from optical damage by the laser pulse. The second beam of smaller intensity was used as a probe. After passing the delay line, it was converted to a white supercontinuum generated by focusing the beam into a 1cm long water-containing cell. By using a semi-transparent mirror, the supercontinuum radiation (360-1500 nm) was subdivided into two pulses (reference and signal) of similar intensity. Both pulses were focused on the sample by mirror optics. The reference pulse is required to eliminate the impact of shot-to-shot instability of supercontinuum. It passes the sample always before the pump pulse. The pump-induced change of the optical density was calculated as:

$$\Delta D(\lambda, \Delta t) = \lg \left[ \frac{E_{sg}(\lambda) / E_{ref}(\lambda)}{E_{sg}^*(\lambda, \Delta t) / E_{ref}(\lambda)} \right],$$

where  $E_{sg}$ ,  $E_{sg}^*$ , and  $E_{ref}$  are the energies of signal pulses passed the sample before and after the pump pulse, and the reference pulse energy, respectively.

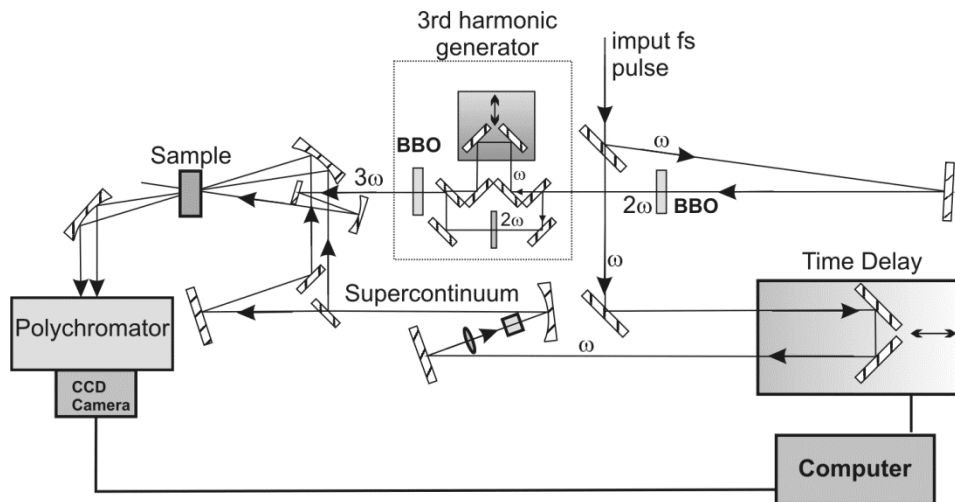


Figure 1: Set up of the femtosecond spectrometer used in this work.

The sample surfaces were carefully polished to prevent the surface damage during the illumination with femtosecond pulses. The beam spot diameter was 2 mm. The spectra of both pulses were recorded for each laser shot by a system including a polychromator equipped with a CCD camera and digitally processed. The absorption spectra of the white supercontinuum were measured in the spectral range from 400 to 730nm.

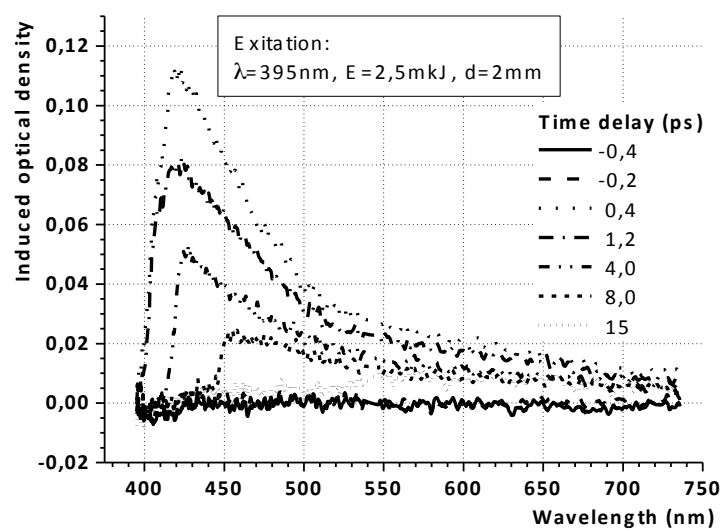


Figure 2: Transient induced absorption ( $\Delta D$ ) spectra of a PWO single crystal at different time delays between the pump and probe pulses. The pump pulse energy is 5  $\mu$ J.

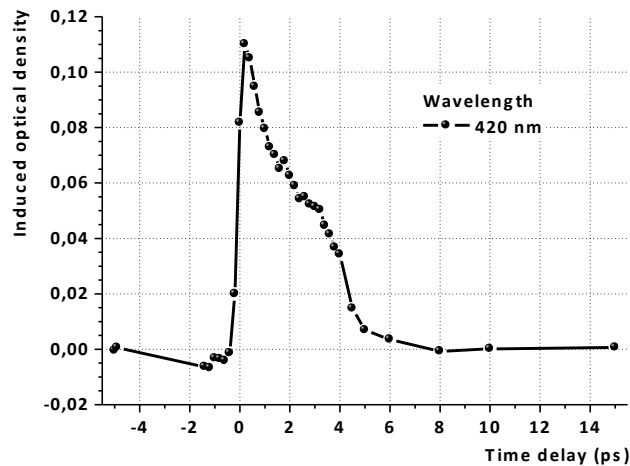


Figure 3: The two photon absorption kinetics of a PWO single crystal at probe wavelength of 420nm.

The simultaneous exposition of the sample to the pump pulse ( the second harmonic at 395 nm, 3.13 eV) and the probe pulse with a supercontinuum spectrum results in strong two-photon absorption.. The effect is practically instantaneous. Thus, the time profile of the induced absorption determined in our experiments by the apparatus function of the detection system. Figure 2 shows the spectra of the induced absorption at different time delays. The time evolution of the probe at 420 nm is presented in Figure 3. This time evolution is connected with the dispersion of the group velocity of the probe pulse in the sample under study.

The data obtained show that the maximal efficiency of the two photon absorption in our experiments corresponds to the combination of the 395 nm ( 3.15 eV) pump pulse and the pulse probe with the closest to the pump pulse wavelength of 400 nm (3.13eV). This perfectly coincides with the second maximum in the spectrum of the density of electronic states described above. As the probe pulse wavelength is increased, the efficiency of the two photon absorption decreases and approaches zero at 500 nm. The energy corresponding to two photon absorption of photons with energies of 3.15 eV (395nm) and 2.5 eV (500 nm) corresponds to the minimum in the spectrum of the density of electronic states in the conduction band of PWO.

To exploit the observed ultrafast two photon absorption in development of precise time stamps related to the interaction of ionizing radiation with the PWO crystal for fast radiation detectors, further study of the fast response of two-photon absorption, especially under ionizing radiation, is required. Combination of the pump and probe experiments with the impact of picosecond X-ray tube emission or electron beam would be highly informative. The PWO scintillation could probably also be used as the probe. In this case, only the pump laser pulses would be required to ensure the time stamp. Moreover, the simultaneous absorption of two photons of the same energy optimally selected in the vicinity of 3 eV would strongly simplify the future application of this technique in the detector systems. In real applications, a set of several hundreds of short laser pulses, 1-5 ps in duration, within the total duration of 500-1000 ps is required, and has to be synchronized with the particle collisions to scale the time intervals between the probe pulses at the level of several picoseconds. The frequency of the laser pulse sets should be similar to the frequency of the particle collisions, i.e., approximately 40 MHz. Such detection systems can be assembled using commercially available lasers and optical components.

### 3. Conclusions

We observed two photon absorption of picosecond laser pulses in the extensively used PWO scintillation crystal. These processes are considered to be sensitive to polarization of the PWO single crystal under ionising radiation. An ultrafast change in the two photon absorption conditions due to the crystal polarisation are prospective to be used for development of precise time stamps of the interaction of ionizing particles with the crystal in high energy physics experiments at a picosecond time scale.

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