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Letter to the Editor

Stimulated recovery of the optical transmission of $PbWO_4$ scintillation crystals for electromagnetic calorimeters after radiation damage

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ABSTRACT

In this paper we describe the phenomenon of the stimulated recovery of radiation damage in lead tungstate scintillation crystals achieved via illumination by visible and infrared light. It allows fast and efficient in-situ recovery of the optical transmission either during beam-off periods or on-line during data accumulation. The application can substantially improve or extend the running period of the experiment by keeping the damage at a tolerable level.

1. Introduction

The operation of inorganic crystalline scintillation materials for detector applications in an environment of strong irradiation requires resistivity to damage due to ionizing radiation. This problem becomes crucial for electromagnetic calorimetry in high-luminosity hadron accelerator experiments. However, the perfection of the synthetic crystals has physical, technological and economical limits and therefore relies on the search for methods to counteract the radiation damage during the operation of the calorimeter. Point structure defects as well as traps for electrons and holes create in the crystal a variety of color centers with absorption bands in a wide spectral region. After the irradiation stops a spontaneous relaxation of the color centers takes place via thermo-activation. In fact, this is a thermo-dynamical process in an open system, which can be further accelerated by an injection of energy in an appropriate manner. When most of the carriers, after being released from the color centers, recombine and are not recaptured again, the relaxation process of the color center i can be described by

$$n_i = n_0 \exp\left(-w_T^i - \sum_j b_j I_j\right) t \quad (1)$$

where n_0, and n_i are the initial and current concentrations of the color center of type i, $w_T^i = A_i \exp(-E_{TA}/kT)$ is the spontaneous relaxation probability, E_{TA} the thermo-activation energy of the color center, k the Boltzmann constant, T the temperature, A_i the normalization factor, I_j the specific energy flux, and b_i describes the interaction of the color center with a flux of specific energy. As a direct consequence of the definition the relaxation can be accelerated by heating the crystal exploring the exponential dependence of w_T^i on temperature. However in practice, the heating in between the experimental phases of an entire experimental setup comprising up to thousands of crystals would require an enormous amount of energy, which is excluded in most of the cases for practical reasons. Alternatively, energy can be delivered to a crystal via photons of selected wavelengths but requires high b_j factors.

There exist two processes that are initiated by photons: the ionization of color centers and the transport of the captured electron from ground state of the color center to a radiating excited level. The first process (sometimes called 'optical bleaching') depends on the energy width of the conduction band and the location of the ground state of the color center in the forbidden zone. It may be initiated in a wide spectral range from UV to visible light. However, ionization requires in an inefficient manner that the energy E_f of the applied photons is much higher than E_{TA}. The second possible process—which we call 'stimulated recovery'—is an intra-center resonant transition that can be initiated by photons with an energy even as low as $E_f \sim E_{TA}$, favorably in the infrared region. In this case, stimulation can even be applied simultaneously with ionizing irradiation,

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in particular, if the used photosensor is blind for the chosen wavelength region. As a consequence, the level of the dynamic saturation of the induced optical absorption in the crystal will be reduced. The stimulation of the recovery becomes effective when two conditions are fulfilled: there is at least one energy level in the center located slightly above the radiating state providing fast recombination and re-trapping processes within the crystal should be strongly suppressed. The latter is achieved by minimization of the number of color centers of type j (ideally $j=1$). By varying the intensity of light one can even control the rate of stimulated recovery.

2. Stimulation of the recovery with visible and infrared light in lead tungstate

Lead tungstate ($PbWO_4$, PWO) became the most widely used scintillation material in electromagnetic calorimetry in modern collider experiments [1,2]. It combines in a unique manner physical and scintillation properties [3], which allow the construction of an affordable compact electromagnetic calorimeter. Further perfection of the PWO technology during the last decade resulted in the development of PWO-II [4] with doubled light yield at room temperature. PWO-II has been chosen for the construction of the electromagnetic target calorimeter of PANDA at the future FAIR facility at Darmstadt (Germany). To achieve further increase in light yield, the detector will be operated at a reduced temperature of $T=-25^{\circ}C$ extending electromagnetic calorimetry with sufficient energy resolution even down to 10–20 MeV photon energy. But at the same time, significantly longer time constants for spontaneous relaxation of the color centers have to be taken into account and lead to an accumulation during irradiation. It prevents from reaching a dynamical saturation level of induced absorption in a reasonable time range at a typical dose rate of 0.02–0.05 Gy/h and leads to a nearly continuous degradation of the energy resolution of the detector [5,6]. Therefore, extremely radiation hard crystals have to be selected in order to keep the reduction of the optical transparency at a tolerable level.

2.1. Radiation hardness of PWO-II crystals

PWO-II crystals can reach high radiation hardness at room temperature due to the significant improvement and optimization of the production technology. The crystals are grown from melt with a precise tuning of the stoichiometry and co-doping with Y and La with a total concentration up to 40 ppm. As a consequence, the concentration of shallow electron traps is significantly reduced. Moreover, Frenkel type defects (FTD), which cause deep electron centers and appropriate color centers in the crystal ($E_{TA}=0.7$ eV) with an absorption band peaked near the wavelength of 410 nm, are reduced by a factor of two. Therefore, the radiation induced absorption coefficient measured in the scintillation region ($\sim$420 nm) at room temperature can remain as low as $dk=0.2$–$0.6\ m^{-1}$. Nevertheless, this center dominates the induced absorption spectra of PWO-II, fulfilling the second condition (j to be close to 1) mentioned above. Moreover, intra-center transitions in FTD meet the first requirement. The absorption between the lowest stark component of the split $^3T_1$ level, which is a ground state of the electron trap to $^1T_1$, can be used to lift the electron in a recombining state and leads to the possibility to stimulate effectively the relaxation. Only a minimal photon energy of 0.7 eV is needed, which is considerably lower than the energy for FTD ionization between 3.3 and 1.8 eV. These energies correspond to the wavelengths of 380 and 700 nm, respectively. These values are calculated on the basis of spectroscopic data on PWO [7]. The resonant region of the intra-center transition is quite wide, but the upper energy limit for stimulating photons should be the ionization threshold of FTD.

Previous experiments on optical bleaching of PWO had shown two opposite processes: reduction of the radiation induced optical absorption due to illumination by light in the range of 600–700 nm and an additional increase of the optical absorption in the visible range with light of 500 nm [8]. These effects were observed in un-doped lead tungstate crystals, which have in general a variety of defects and did not lead to a practically acceptable procedure for fast crystal relaxation.

2.2. Measurement of the stimulated recovery of PWO-II crystals at different temperatures

Ten 200 mm long PWO-II crystals of PANDA-Endcap geometry [4] obtained from the mass production performed at Bogoroditsk Technical Chemical Plant (Bogoroditsk, Russia) were chosen to study the relaxation of radiation induced absorption via illumination by visible and infrared (IR) light. The radiation hardness of the crystals is characterized by the radiation induced absorption coefficient dk at the wavelength of 420 nm due to an integral dose of 30 Gy imposed by a set of ^{60}Co sources at the radiation facility at the Justus Liebig University at Giessen (applied dose rate of 10 Gy/h). The chosen value corresponds to the expected dose to which the crystals of the forward endcap will typically accumulate during an experimental period of 6 months operation. The coefficient is expressed by $dk=\ln(T_b/T_a)/d$ with T_b and T_a describing the optical transmission measured before and after irradiation, respectively. The parameter $d=0.2$ m is the overall length of the crystal. The measurements are performed at room temperature. The crystals have identical shapes with an induced absorption coefficient in the range of $dk=0.4$–$1.0\ m^{-1}$ at 420 nm wavelength.

The first part of investigations was performed at room temperature. Fig. 1 shows as an example the change over the entire investigated spectral range between 300 and 900 nm, respectively. Obviously only slow thermo-activated recovery sets in immediately after irradiation as long as the crystal is kept in the dark. With the delay of 30 min after irradiation the crystals are illuminated by several external light sources, in case of Fig. 1 with an LED at a peaking wavelength at 940 nm. The significant improvement in the optical transmission documents a very efficient stimulated relaxation over the entire range of wavelength and leads to a simultaneous reduction of the induced absorption in a wide spectral region. Fig. 2 illustrates the typical relaxation of the radiation induced absorption at the selected wavelengths of 360, 420 and 620 nm, which are critical with

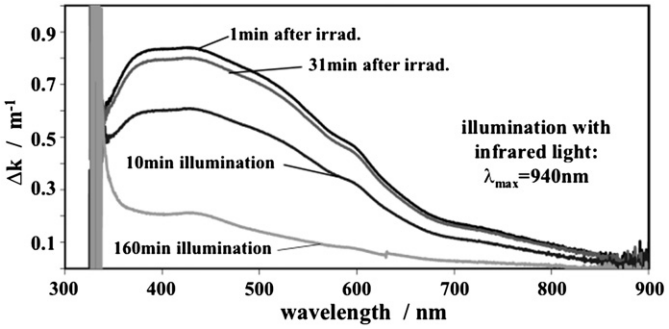

Fig. 1. Change of the entire spectral distribution of the induced absorption coefficient of a PWO-II crystal after irradiation with an integral dose of 30 Gy measured at room temperature. The spectra are measured at 1 and 31 min after irradiation as well as after additional illumination for 10 and 160 min, respectively, with infrared light at 940 nm wavelength.

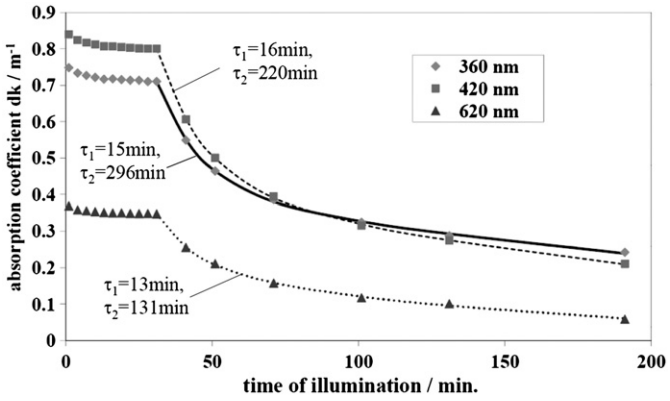

Fig. 2. Stimulated recovery of the radiation induced absorption coefficient dk of a PWO-II crystal measured at the wavelengths of 360, 420 and 620 nm, respectively. The illumination with a LED light source (λmax=940 nm) started 30 min after the end of irradiation with an integral dose of 30 Gy. The measurement was performed at room temperature.

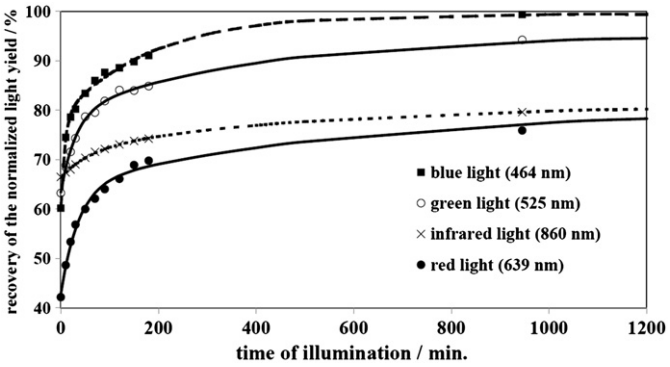

Fig. 3. Stimulated recovery of PWO-II scintillation crystals using LEDs with different wavelengths at an operation temperature of $T=-25^{\circ}$C. The experimental data show the recovery of the signal amplitude of individual detectors normalized to the value before irradiation with an integral dose of 30 Gy. Light sources of four different colors were used. The curves are plotted to guide the eye.

respect to the luminescence spectrum of PWO. At 420 nm the recovery develops finally with a time constant of $\sim$300 min after the slow spontaneous relaxation within the first 30 min without external light. The integral flux of the light source consisting of four LEDs, directly coupled to the end face of the crystal, amounts to 2.2×10^{17} photons per second.

More relevant for the PANDA–EMC application is the behavior at low temperature, which imposes a strong impact on all recovery mechanisms. Therefore, we have integrated into each of the detector modules, consisting of the PANDA-Endcap crystal wrapped in 8 layers of teflon foil as reflector and a photomultiplier tube (Philips XP 1911) as sensor, an additional set of four LEDs as light sources. In total 10 identical detectors were prepared with crystals of different radiation hardness. For the study of the wavelength dependence of recovery LEDs of different colors were used. After cooling down the modules to $T=-25^{\circ}$C the response to low energy ^{60}Co γ-source was measured before and after irradiation with 30 Gy to determine the integral loss of light output. Afterwards, illuminating with light of different wavelengths the stimulated recovery of the light loss was studied as a function of the integral illumination time up to two days in total. The photon flux at the different wavelengths was nearly the same and varied in the range of $(2–6)\times10^{16}$ photons per second.

Fig. 3 shows the impressive results for the recovery of the relative light loss achieved with visible light sources centered at emission wavelengths of 464, 525, 639 and 860 nm. In total three crystals were illuminated with the same wavelengths of 464, 525 and 639 nm. Only one detector was exposed to the wavelength of 860 nm. For blue light, nearly 90% of the original signal amplitude was restored within the first 100 min already, which is a shortening of the recovery by two orders of magnitudes with respect to spontaneous processes. One can state that the parameters of the recovery curves practically do not depend on the initial value of the induced absorption coefficient. The different starting points reflect the radiation hardness of the individual crystal sample. At least two components of the recovery—fast and slow—have been observed. Taking into account the difference in light intensity the impact of photons at 464 and 525 nm is within the experimental uncertainties practically the same. However, the deduced time constants of the stimulated recovery (see Table 1) progressively increase towards the IR range, indicating a decrease of the constants b_j. It is in agreement with the reduction of the optical transition oscillator strength of intra-center transitions towards ionization. The presence of at least two components indicates that besides FTD at least one additional center contributes to a b_j constant but smaller compared to the one corresponding to FTD.

Table 1
Time constants of the stimulated recovery processes of PWO-II crystals measured at four different wavelengths at a temperature of $T=-25^{\circ}$C. In case of wavelengths of 464, 525 and 639 nm the constants are average values of three crystals.

LED wavelength (nm)	464	525	639	860
intensity (photons per second)	5×10^{16}	2.4×10^{16}	3.2×10^{16}	7.5×10^{16}
τ_{fast} (min)	10[a]	20[a]	30[a]	60
τ_{slow} (min)	180[a]	350[a]	550[a]	500

[a] Averaged for 3 crystals.

In an additional test we repeated the γ-irradiation of some of the detector modules and illuminated the crystals during irradiation with the above set of LEDs of different wavelengths. There was no visible change in the response function of the detectors.

One can state that stimulation recovery can be applied to PWO crystals operating in a wide range of temperatures.

3. Conclusions

The observation of very efficiently stimulated recovery processes even for detector modules operated at the low temperature of $T=-25^{\circ}$C can have a strong impact on future concepts, how to maintain the performance of electromagnetic calorimeters at accelerator experiments based on lead tungstate. One should only consider that the recovery time increases with the wavelength of the applied light source. This behavior is even more pronounced in case of low temperatures. In-situ recuperation of the optical transmission of the scintillator elements by stimulated recovery during the breaks between or in parallel with data acquisition can substantially improve the detector performance and prolong its lifetime under tolerable conditions. Since commonly used photosensors are blind in the IR region, stimulated recovery could be applied even during full detector operation. The concept of on- and off-line recovery of PWO crystals in a radiation environment and the basic layout are part of a patent in Germany (TM 382_DE).

Presently, detailed studies and correlation with the crystal quality, the most efficient region of wavelengths, the required photon flux for on-line recovery and the characterization of the recovered color centers via EPR measurements are under investigation.

References

[1] F. Pauss (Ed.), The Electromagnetic Calorimeter, Technical design report, CERN, CMS, 1997.
[2] ALICE, Technical proposal for a large ion collider experiment at CERN LHC, CERN, 1995.
[3] A. Annenkov, M. Korzhik, P. Lecoq, Nucl. Instr. and Meth. Phys. Res. A 490 (2002) 30.
[4] Technical design report for: PANDA electromagnetic calorimeter (EMC), ⟨arXiv:0810.1216v1⟩, 2008.
[5] P. Semenov, et al., Nucl. Instr. and Meth. Phys. Res. A 582 (2007) 575.
[6] R.W. Novotny, et al., IEEE Trans. Nucl. Sci. NS-55 (3) (2008) 1283.
[7] M. Korzhik, Physics of scintillation in oxide crystals, Minsk, 2003 (in Russian).
[8] R.Y. Zhu, et al., Nucl. Instr. and Meth. Phys. Res. A 376 (1996) 319.