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Nuclear Instruments and Methods in Physics Research A 537 (2005) 177–181

NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH Section A

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Light yield improvement in lead tungstate

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Available online 25 August 2004

Abstract

A new approach to the possibilities of increase in the light yield (LY) of scintillations in lead tungstate (PWO) is discussed. The abrupt LY decrease above 200 K is connected with the start of rotations of WO_4 tetrahedra which disturbs electrostatic separation between non-equilibrium electrons and holes, thus accelerating their non-radiative recombination. The logarithmic decrease of LY with elongation of the crystals is attributed to the capture of the emitted light by a new type of the surface light waves, created during the internal total reflection at the surfaces. Two ways of PWO LY increase are proposed: to retard WO_4 tetrahedra rotations and to eliminate the creation of the surface light waves. Two experimental results are attracted to demonstrate the validity of these ways: the relative increase of the light emission at PWO surfaces and LY increase after deposition of transparent quartz coating onto PWO surface, which reduces the internal total reflection.

PACS: 64.70.Kb; 78.20.Fm; 78.60.+t; 81.40.tv

Keywords: Scintallation detectors; Photoluminescence; Polishing; Lead tungstate; Recombination; Surface waves

1. Introduction

Lead tungstate ($PbWO_4$–PWO) is one of the most popular scintillators due to its application for calorimetric detectors at Large Hadron Collider (CERN, Geneva). It is characterized by high density, fast decay time, good radiation hardness and reasonable production cost [1]. But applications of this crystal in the quickly growing market of X-Ray transmission devices for medical diagnostics and various kinds of inspections are problematic due to low light yield (LY) of scintillations, which is more than two orders of magnitude less than LY of NaI (Tl). The increase of LY of PWO by several times would open up a wide field of practical applications for this material. Up to now the search for the possibilities of the LY increase of PWO was concentrated in the traditional ways: to improve the chemical purity of the material and to find a proper

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doi:10.1016/j.nima.2004.08.002

combination of dopants which would decrease the non-radiative recombination of the electron excitations as well as the absorption of the emitted light [2,3]. Studies of the experimental relations between the structural behavior and optical and scintillation characteristics of PWO [4,5] gave us an opportunity to propose a new approach to understanding of the micromechanisms of the abrupt enhancement of the non-radiative recombination in PWO above 200 K and of the anomalous LY decrease with the crystal elongation. This approach leads to practical ways of the increase of PWO LY which are described below.

2. Experimental correlations between scintillation and structural characteristics of PWO

The tetragonal phase of PWO (sheelite) at the temperature range below 200 K demonstrates acceptable value of LY, but the decay time of scintillations is too long. The increase of the temperature above 200 K results in fast decrease of the light yield and simultaneous shortening of the decay time (Fig. 1a) [4].

It is worth noting that PWO can exist in a monoclinic structure and our studies of monoclinic PWO samples showed that the sharp decrease of their scintillation LY and decay time start at lower temperature (150 K) (Fig. 1b) [6]

On the other hand, in the same temperature region between 200 and 250 K several phenomena are observed which reveal certain structural transformations in the tetragonal atomic lattice of PWO: the anomalies in the temperature dependence of the lattice parameter (Fig. 2a) [4],

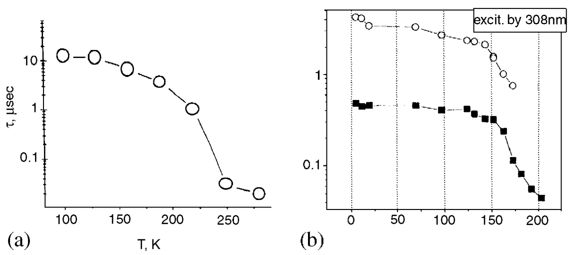

Fig. 1. Temperature dependencies of decay times of scintillations in PWO–sheelite (a) and PWO–raspite (b: two–exponential approximation gives two separate decay times).

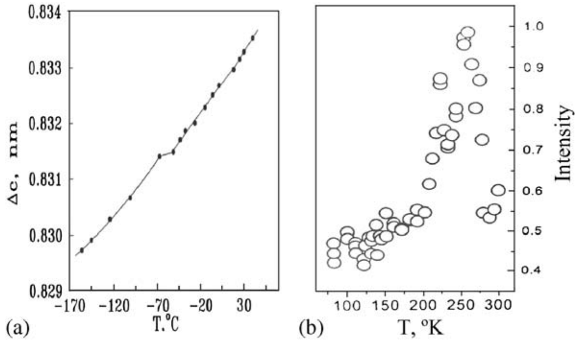

Fig. 2. Temperature dependences of lattice parameter C (along fourth order axis) of PWO (a) and of Raman scattering amplitude (b).

amplitudes of Raman scattering induced by low-frequency phonons (Fig. 2b) [6] and the specific heat [4]. It is important to emphasize that the symmetry of the lattice at 200 K is not changed and remains tetragonal [4]. One more important feature of the temperature behavior of the light emission characteristics of PWO is connected with the change of the direction of the temperature shift of the maximum of the luminescence excitation spectra which takes place at 200 K [4]. When PWO sample is heated from lower temperatures this maximum shifts normally (to the low-energy side, corresponding to the decrease of the band gap), but above 200 K the direction of the shift changes its sign and becomes anomalous: the band gap decreases with the heating but the maximum is shifted to higher energies.

It is well known that lead tungstate crystals having high transparency in the spectral region of scintillations (i.e. not having any traces of the volume light absorption) demonstrate anomalous decrease of the scintillation LY as a function of the crystal length: the LY decrease is proportional approximately to the logarithm of the length [5]. It turns out that LY of a crystal 15 cm in length can be smaller than LY of 1 cm piece cut from the same crystal by 4–5 times. This means that from 70% to 80% of the emitted light in long PWO crystals are lost anywhere. Experimental studies made of the light propagation in PWO crystals showed that these losses are induced by especial surface light waves which are excited when the angle between the lateral surface of the crystal and