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Radiation-induced color centers in La-doped $PbWO_4$ crystals

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Abstract

This report presents the result of a study on radiation-induced color center densities in La-doped lead tungstate ($PbWO_4$) crystals. The creation and annihilation constants of radiation-induced color centers were determined by using transmittance data measured for a $PbWO_4$ sample before and during ^{60}Co γ-ray irradiation at a dose rate of 15 rad/h. Following a model of color center kinetics, these constants were used to calculate color center densities under irradiations at 100 rad/h. The result was found to be in good agreement with experimental data, indicating that the behaviour of $PbWO_4$ crystals under irradiation can be predicted according to this model.

1. Introduction

Because of high density and fast decay time, lead tungstate ($PbWO_4$) crystals were chosen by the CMS experiment to construct a precision electromagnetic calorimeter at LHC [1]. $PbWO_4$ crystals have been extensively studied by several groups [2–21]. Our previous studies on $PbWO_4$ samples concluded that most $PbWO_4$ crystals suffer from non-negligible radiation damage after a dose as low as 10 rad [19–21], but the scintillation mechanism of $PbWO_4$ is not affected by the radiation, i.e. the loss of light output is only due to absorption by radiation-induced color centers [22]. Following a kinetic model [23], we also proposed that the level of the radiation damage in $PbWO_4$ crystals should be dose rate dependent because of the damage recovery observed [24]. Our experimental measurements later confirmed this prediction [25].

In this report we describe an experimental study on the kinetics of radiation-induced color center densities in La-doped $PbWO_4$ samples. The creation and annihilation constants were determined by using transmittance data measured for a $PbWO_4$ sample before and during ^{60}Co γ-ray irradiation at a dose rate of 15 rad/h. Consequently, these constants were used to calculate color center densities under different dose rates, and a good agreement was found with the data. Section 2 gives a brief summary of color center kinetics. The $PbWO_4$ sample and experiment are described in Section 3. Finally, Section 4 gives a brief summary and discussions.

2. Model of color center kinetics

It is well known that color centers are created in $PbWO_4$ crystals under irradiation. It is also known

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that radiation-induced color centers annihilate under room temperature. During irradiation, both annihilation and creation coexist, the color center density will reach an equilibrium at a level depending on the dose rate applied. Assuming that the annihilation speed of color center i is proportional to a constant a_i and its creation speed is proportional to a constant b_i and the dose rate (R), the differential change of color center density when both processes coexist can be written as [23]

$$\mathrm{d}D = \sum_{i=1}^{n} \{-a_i D_i + (D_i^{\mathrm{all}} - D_i) b_i R\}\,\mathrm{d}t \tag{1}$$

where D_i is the density of the color center i in the crystal and the summation goes through all centers. The solution of Eq. (1) is

$$D = \sum_{i=1}^{n} \left\{ \frac{b_i R D_i^{\mathrm{all}}}{a_i + b_i R} [1 - \mathrm{e}^{-(a_i + b_i R)t}] + D_i^0 \mathrm{e}^{-(a_i + b_i R)t} \right\} \tag{2}$$

where D_i^{all} is the total density of the trap related to the center i and D_i^0 is its initial density.

The color center density in equilibrium (D_{eq}) thus depends on the dose rate (R):

$$D_{\mathrm{eq}} = \sum_{i=1}^{n} \frac{b_i R D_i^{\mathrm{all}}}{a_i + b_i R}. \tag{3}$$

This prediction of dose rate dependence has been confirmed in all $PbWO_4$ samples [25].

3. Experiment

Sample L6 was used in our investigation, which is an La-doped sample produced at Shanghai Institute of Ceramics (SIC). It has a rectangular shape with a dimension of $2 \times 5 \times 2$ cm. This particular sample was used because of its good initial transmittance and significant radiation damage caused by irradiations, which reduced the uncertainty in the calculation of radiation-induced color center densities. The entire investigation was carried out at Caltech.

A 50 Ci ^{60}Co γ-ray source was used for irradiation. The entire body of the sample was irradiated with its side face facing the source during the irradiation. The transmittance of this sample was measured using a Hitachi U-3210 UV/Visible double beam, double monochromator spectrophotometer equipped with a large sample compartment, including a custom Halon-coated integrating sphere. During irradiation and measurement the sample was kept in the dark and under the same room temperature.

The sample was fully annealed to eliminate all the residual radiation-induced color centers, which set the initial value of color center density (D_i^0 in Eq. (2)) to zero. In this case Eq. (2) can be written as

$$D = \sum_{i=1}^{n} \frac{b_i R D_i^{\mathrm{all}}}{a_i + b_i R} [1 - \mathrm{e}^{-(a_i + b_i R)t}]. \tag{4}$$

The sample was first irradiated under a dose rate of 15 rad/h. Its transmittance was measured when the accumulated dosage was up to 50, 100, 325, 800 and 1600 rad. Its radiation-induced absorption

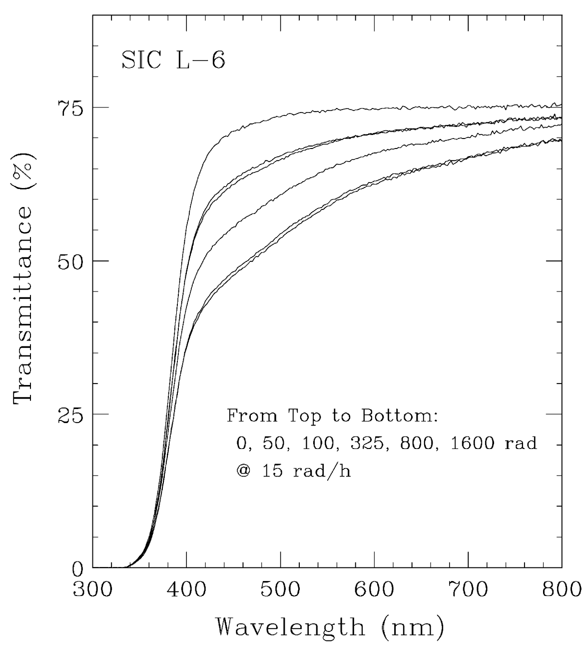

Fig. 1. The transmittance before and after irradiation is shown as a function of the wavelength.

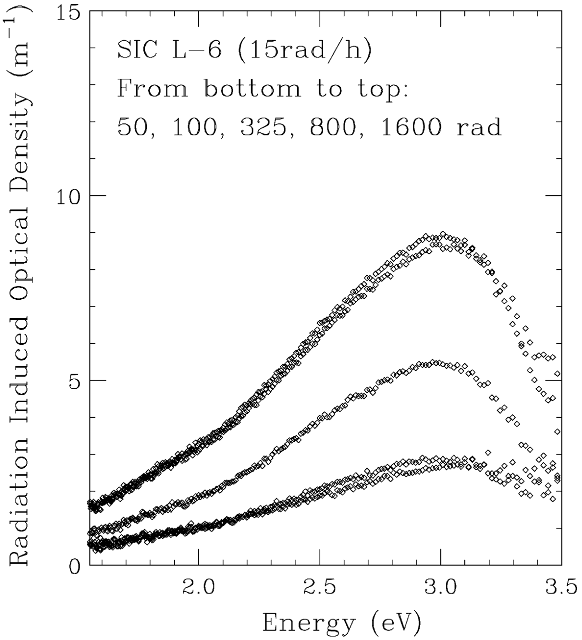

Fig. 2. Radiation-induced absorption is shown as a function of photon energy.

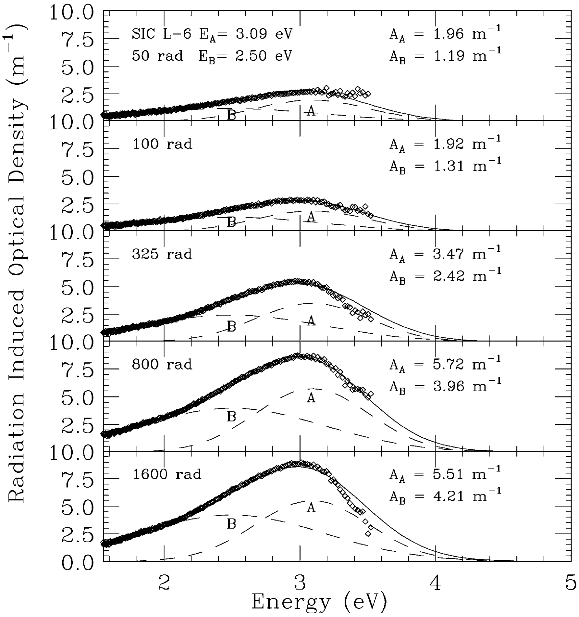

Fig. 3. Radiation-induced absorptions are shown as a function of photon energy for five cumulative dosages.

coefficient (D) is defined as

$$D = 1/\mathrm{LAL} - 1/\mathrm{LAL}_0 \quad (5)$$

where LAL_0 and LAL are the average light attenuation lengths before and during irradiation, which were calculated by using the longitudinal transmittance data according to Eq. (1) of Refs. [19–21]. This calculation takes care of multi bounces of light in the crystal.

Fig. 1 shows the longitudinal transmittance data measured before and after the irradiations. The corresponding radiation-induced color center density as a function of photon energy is shown in Fig. 2. There are two radiation-induced color centers in sample SIC-L6. A fit with two Gaussians found the energy, width and amplitude of these two color centers. The result is shown in Fig. 3. Five plots from top to bottom show the fit result after five cumulative dosages: 50, 100, 325, 800 and 1600 rad, respectively. Although the amplitude of the radiation-induced absorption is a function of the cumulative dosage, two common radiation-induced absorption peaks were found at 3.09 eV

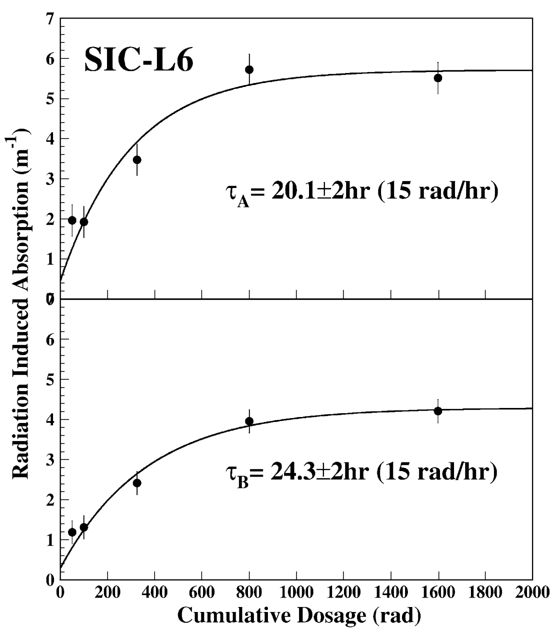

Fig. 4. The amplitude of the radiation-induced absorption centers is shown as a function of the cumulative dosage.

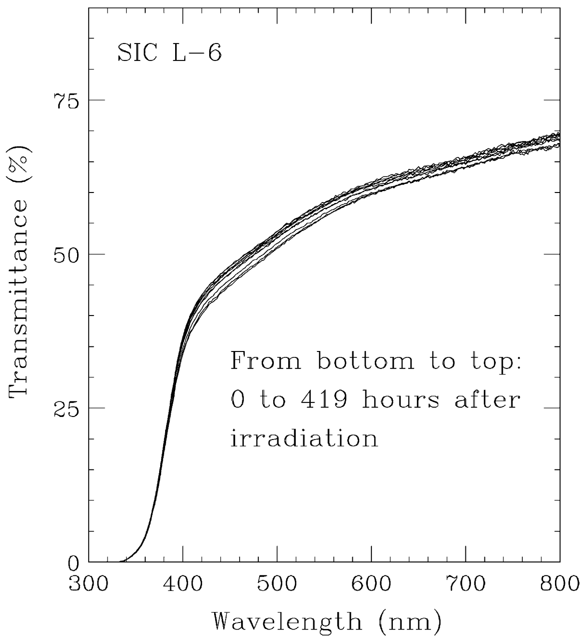

Fig. 5. Transmittance recovery at room temperature.

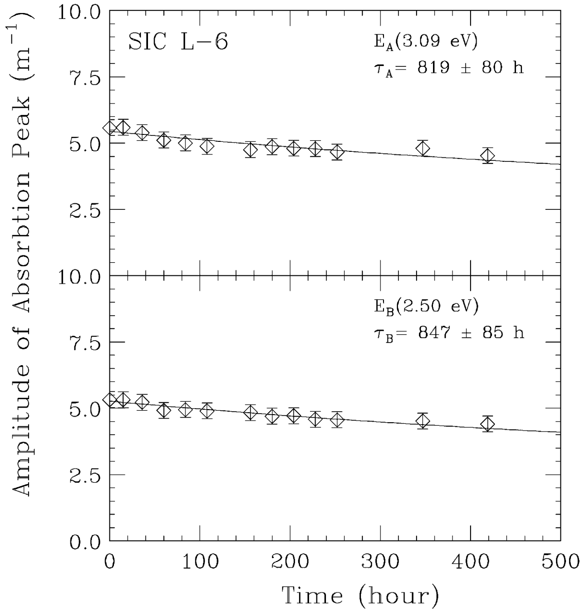

Fig. 6. The amplitudes of the absorption bands is shown as a function of recovery time.

(color center A) and 2.50 eV (color center B). The amplitudes of these two centers (A_A and A_B) for five AC cumulated doses are also listed in these plots.

The right side of Fig. 4 shows the peak amplitudes of color center A (A_A, top plot) and B (A_B, bottom plot) as a function of the cumulative dosage. Eq. (4) can be used to determine the time constant (τ), which is also shown in the figure. This time constant equals $1/(a_i + b_i R)$.

Upon 1600 rad irradiation, the radiation-induced color center densities reached an equilibrium. Sample SIC-L6 was then kept in the dark and its transmittance during recovery was measured under room temperature. Because of no color center creation, Eq. (1) can be written as

$$dD = \sum_{i=1}^{n} \{ - a_i D_i \} \, dt. \tag{6}$$

The solution of Eq. (6) is

$$D = \sum_{i=1}^{n} D_{eq} e^{-a_i t}. \tag{7}$$

Fig. 5 shows the transmittance data during recovery for sample SIC-L6. The experiment lasted for 420 h. Fig. 6 shows the amplitudes of two absorption peaks, or color centers, (top and bottom) as a function of the recovery time. Eq. (7) can be used to determine the recovery time constant (τ), which is also shown in the figure. This time constant equals $1/a_i$.

By use of the fit result for both damage and recovery processes, the creation and annihilation constants of these two color centers were determined. With these constants, color centers densities under different dose rates as a function of the irradiation time can be predicted by using Eq. (4).

Finally, we annealed SIC-L6 back to its initial condition, and irradiated it under a dose rate of 100 rad/h and measured its color center density during irradiation. The dots with error bars in Fig. 7 show the measured amplitudes of color center density for color centers A and B for sample SIC-L6, and the solid curves are the results calculated by using the constants deduced from data of 15 rad/h. As seen from the figure, the data agree with the prediction rather well. Fig. 8 shows the overall color center densities in equilibrium under 100 rad/h. The experimental data (open circle with error bars) also agree with the calculated prediction (solid curve).

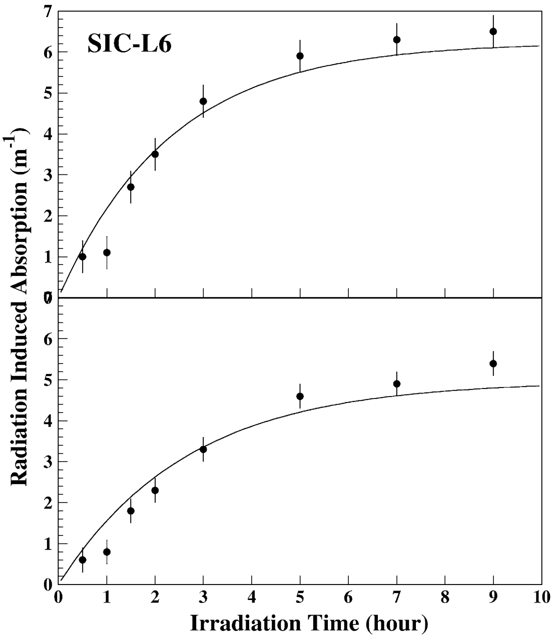

Fig. 7. The experimental data of the amplitudes of color centers A and B as a function of time under irradiation of 100 rad/h is compared to calculation by using kinetic model for SIC-L6.

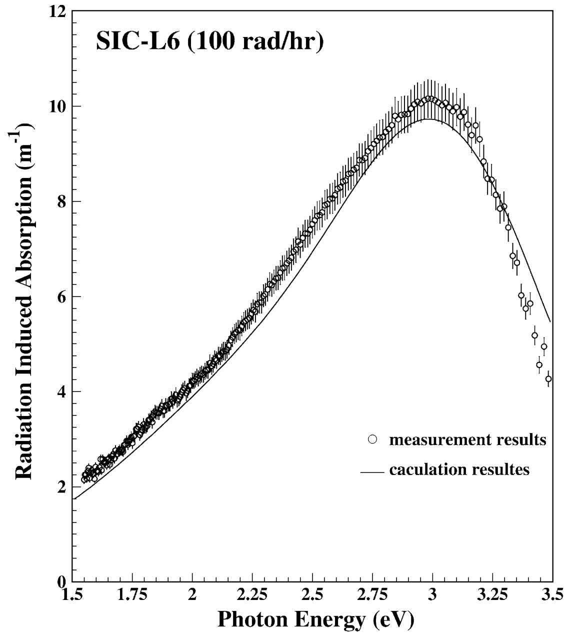

Fig. 8. The measured overall color center density as a function of energy in equilibrium under 100 rad/h is compared to model calculations for sample SIC-L6.

4. Summary

The behaviour of radiation-induced color centers follows a model of color center kinetics, as described in Section 2. This explains the nature of dose rate dependence of radiation damage in $PbWO_4$ crystals [25]. The creation and annihilation constants can be determined by using experimental data obtained under one particular dose rate, and can then be used to predict the behaviour of the same sample under different dose rates.

Following this model of color center kinetics, efforts in developing rad hard $PbWO_4$ crystals may follow two approaches. One is to reduce the total density of traps (D_i^{all}) which is the origin of radiation-induced color centers. This can be achieved by optimisation of crystal growth conditions and purification of raw materials. This approach, however, is usually cost limited. An alternative approach is to transfer shallow traps to deep ones, so that the annihilation constant (a_i) is reduced. This will reduce dose rate dependence, or produce crystals more stable in situ at LHC.

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