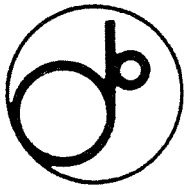

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Radiation Damage of a Cerium-doped Lutetium Oxyorthosilicate Single Crystal

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Abstract

Degradation in optical transmittance due to irradiation with ^{60}Co γ-rays is less than 1% per cm below 10^6 rad, about 2.5% at 10^7 rad, and 7% at 10^8 rad. Spontaneous recovery occurred only partially with a time constant of the order of days to tens of days. Neither recovery nor damage was sizable upon exposure to UV light. Almost full recovery occurred by thermal annealing at 300°C for one day.

1. Introduction

Cerium-doped lutetium oxyorthosilicate ($Lu_2(SiO_4)O$:Ce or "LSO:Ce")has recently been discovered as a promising dense, fast, and efficient inorganic scintillator.[1,2] It has a density of $\rho = 7.4$ g/cm^3 with a unit radiation length of $X_o = 1.14$ cm, a fast decay of approximately 40 ns, and light output as large as 75% of NaI:Tl. These characteristics, together with the absence of hygroscopicity and a peak emission wavelength in the visible range (420 nm), are excellent for the use of LSO:Ce in γ-ray detectors in various fields such as positron emission tomography, nuclear physics, high energy physics, nuclear well logging, etc., in spite of a few drawbacks such as the high cost of raw materials and natural radioactivity coming from lutetium (78 Bq/g).

In some applications, however, high radiation damage hardness is also required. Recently we measured radiation damage of LSO:Ce caused by ^{60}Co γ-rays. In addition, the recovery of the damage was studied as a function of time, exposure to UV light, and thermal annealing. This report presents the results of these investigations.

2. Experimental

An LSO crystal doped with 0.25 mol% cerium was grown by Schlumberger-Doll Research with the Czochralski technique. A 1 x 1 x 2 cm^3 sample was cut from an ingot of 2 cm diameter and 5 cm length and then polished to an optical grade. Optical transmission spectra across a 1 cm thickness were measured with a spectrophotometer (Hitachi 330) before and after γ-ray irradiation.

Irradiation with ^{60}Co γ-rays was carried out at the Japan Atomic Energy Research Institute (JAERI). Six cycles of irradiation followed by transmission measurements were carried out, covering an accumulated dose range of 10^3 to 10^8 rads in factors of ten.[3] Each cycle took about a week including mailing of samples between KEK and JAERI. The irradiation period was one hour for doses of 10^3, 10^4, 10^5, and 10^6 rads; 16 hours for 10^7 rads; and 125 hours for 10^8 rads by mounting the sample at different distances from a calibrated ^{60}Co source. Transmission measurements were carried out within a few days following irradiation, i.e. 37, 61, 35, 38, 39, and 106 hours after irradiation by 10^3, 10^4, 10^5, 10^6 , 10^7, and 10^8 rads, respectively.

The sample was not exposed to strong UV light, including sunlight, at any time during the course of the irradiation-measurement cycles. The sample was wrapped in aluminum foil except during a short period (~ 10 minutes) during each transmission measurement.

3. RESULTS

Figure 1 shows the transmission spectra of LSO:Ce following different accumulated radiation doses. The decrease in transmittance at the emission peak wavelength of 420 nm is <1% for doses $\leq 10^6$ rads. It reaches 2.5% at 10^7 rads and 7% at 10^8 rads.

In order to see slow spontaneous recovery, i.e. without heating or optical bleaching, the transmission spectrum was measured 4.5, 18, 24, 51, and 107 days after the 10^8 rad irradiation (see Fig. 2). Although a small recovery of about 1% was seen between 4.5 and 18 days, further recovery between 18 and 51 days was less than the experimental uncertainty of the measurement (0.5 - 1 %).

The effect of exposure to UV light was examined 51 days after the 10^8 rad irradiation by placing the crystal very close to a UV fluorescent lamp (National GL-10, 10W) for 1.5 hours. Neither bleaching nor darkening was clearly observed although there may have been a slight darkening tendency at a level of 0.5%. The transmittance was again measured 56 days after the UV exposure, i.e. 107 days after irradiation. As seen in Fig. 2, recovery to some extent (~1.5%) occurred between 51 and 107 days after irradiation. It is, however, not clear whether the observed recovery is a pure spontaneous recovery or a recovery triggered by the UV exposure.

About two weeks after the last transmission measurement shown in Fig. 2, we began thermal annealing experiments. The crystal was heated in a silica tube of 3 cm diameter in a slow gas flow of nitrogen, first at 300°C for one day, then 360°C for one day, and finally at 460°C for one day. The transmittance was measured after each annealing cycle soon after the sample had cooled to room temperature. As seen in Fig. 3, the radiation damage was almost fully removed by annealing at 300°C for one day. The recovery by thermal annealing suggests that the trapping centers may lie relatively close to the conduction band. In this case, however, one might also expect recovery by UV exposure but this was not observed. A possible explanation for the absence of significant recovery with UV exposure could be that the energy of the UV light was too large. When the energy of the light is greater than the band gap E_g (between valence and conduction bands), both annealing and damaging (darkening) effects may

compete with each other, resulting in the net effect of neither significant damage nor recovery. As seen from the absorbance spectrum of undoped LSO shown in Fig. 4, the band gap of LSO is about 195 nm (6.4 eV). The UV lamp used in our experiment had some output at wavelengths < 195 nm.

A similar effect has been reported for BGO. The radiation damage due to γ-ray irradiation is fully removed by thermal annealing at 400°C for two hours.[4] The trapping centers are expected to be only ~0.4 eV below the conduction band.[5] The success of optical bleaching depends on the wavelength of the illuminating light, i.e. darkening for UV[6] and recovery for 633 and 514 nm.[5] Optical bleaching of LSO:Ce with visible light remains to be studied.

The above results indicate that LSO:Ce is radiation hard at least up to 10^6 rads and shows only a small amount of damage even at 10^8 rads. Since the radiation hardness frequently depends on the crystal quality, we have examined the quality of the present sample by measuring its scintillation performance.

Fig. 5 shows the excitation and emission spectra of LSO:Ce, measured with a spectro-fluro-photometer (Shimadzu RF-510) in a right-angle configuration as sketched in the inset. Figure 6 gives the decay time spectrum of the scintillation emission measured by the conventional single photoelectron method[7] in a separate scintillator scheme.[8] The spectrum can be fit with two exponential decay constants of 11 ns (62% intensity) and 37 ns (38% intensity). Figure 7 shows a pulse height spectrum of a ^{137}Cs γ-ray source in which a 1 x 1 cm^2 face of the crystal was coupled to a Hamamatsu R329 bialkali photomultiplier tube (50 mm dia.). The FWHM of 16% for the 662 keV γ ray is worse than expected on the basis of photon statistics alone and somewhat worse than the FWHM of 10.3 - 12.4 % previously reported for smaller crystals.[1,2] Laue photographs for different planes of the crystal showed a consistent single crystal structure (monoclinic, space group C2/c). Infrared transmission spectra were measured for different positions of the sample. Fig. 8 gives a typical infrared spectrum measured with a FTIR spectrometer (Japan Spectroscopic, WS/IR7300). The cutoff which is observed around 2000 cm^{-1} (5 μm) is similar to both pure and Ce-doped GSO.[9] We conclude that, in general, the excitation and emission spectra, the decay constants, the pulse height spectrum, and the transmission

spectrum indicate the crystal quality is roughly comparable to that of smaller LSO:Ce crystals described previously.[1,2,10]

4. SUMMARY

We note the following observations:

(1) Radiation damage due to low energy γ-ray irradiation is less than 1% below 10^6 rads, as small as 2.5% at 10^7 rads, and 7% at 10^8 rads.

(2) Partial spontaneous recovery of radiation damage seems to occur with a time constant of the order of days to tens of days. Neither recovery nor damage was significant after exposure to UV light. The damage was almost fully removed by thermal annealing at 300°C for one day.

ACKNOWLEDGMENTS

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FIGURE CAPTIONS

Fig. 1 Radiation damage of LSO:Ce due to ^{60}Co γ-ray irradiation. Transmission spectra through a 1 cm sample thickness, measured with a bandwidth of 2 nm, are given for various accumulated doses.

Fig. 2 Recovery of radiation damage with time (at room temperature) and after UV exposure for 1.5 hours.

Fig. 3 Recovery of radiation damage after thermal annealing carried out 120 days after irradiation by 10^8 rads of ^{60}Co γ-rays.

Fig. 4 Absorbance of undoped LSO (0.19 mm thick) showing the band gap of 6.4 eV (195 nm).

Fig. 5 Excitation and emission spectra measured with the crystal face polished on the excitation side while frosted on the emission side.

Fig. 6 Decay time spectrum of the scintillation emission. The solid curve gives the fit with two exponential components plus a constant background term.

Fig. 7 Pulse height spectrum for ^{137}Cs after pedestal subtraction. The output of the PMT was analyzed by an ADC (Lecroy QVT) in the charge mode.

Fig. 8 Infrared transmission spectrum of the 1 x 1 x 2 cm^3 LSO:Ce crystal.

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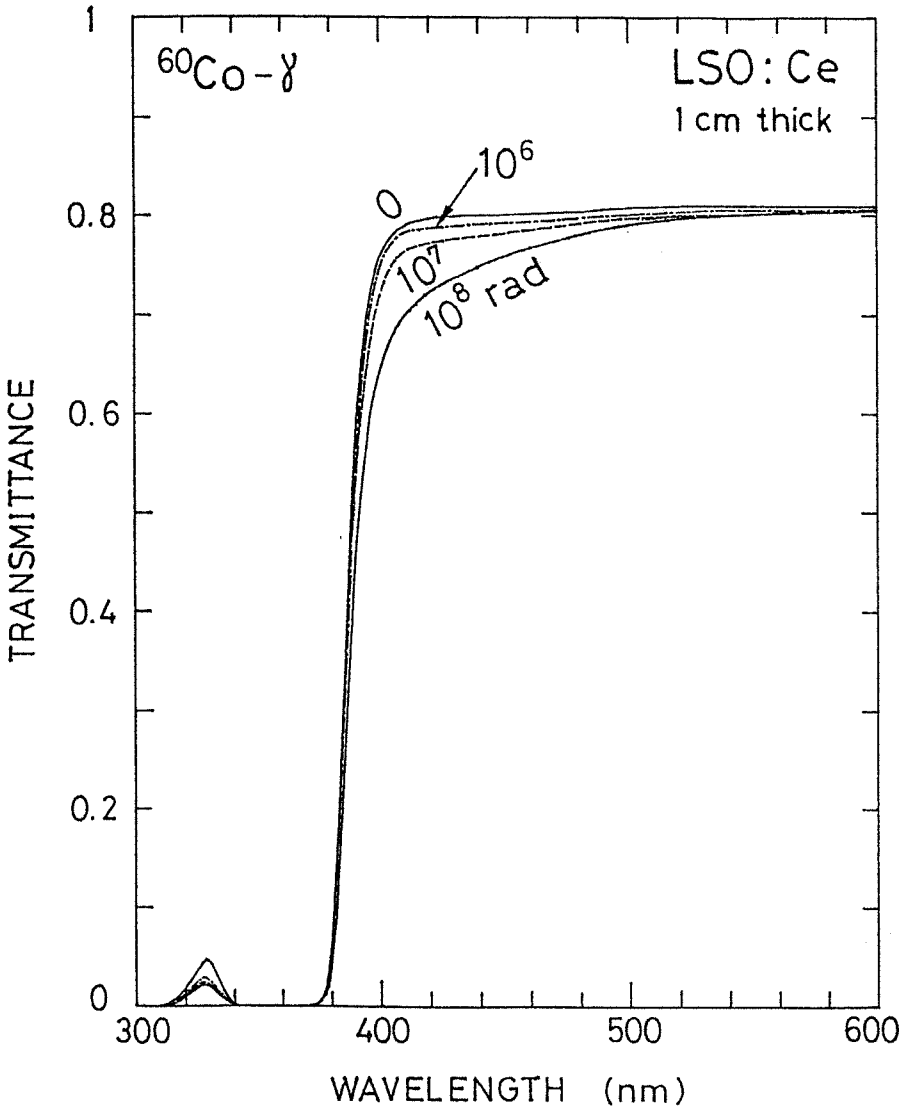
60Co-γ
LSO:Ce
1 cm thick
0
10^6
10^7
10^8 rad
TRANSMITTANCE
WAVELENGTH (nm)
1
0.8
0.6
0.4
0.2
0
300
400
500
600

Fig. 1

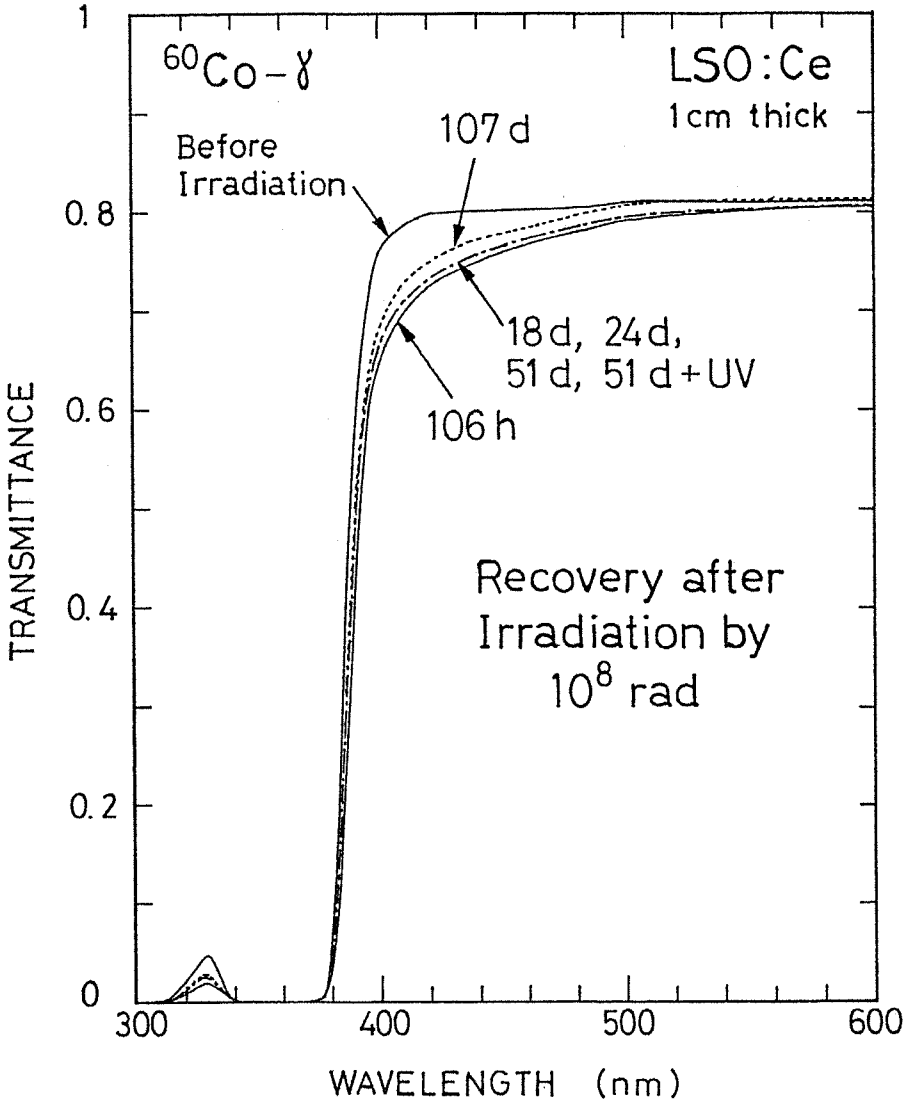
60Co-γ
LSO:Ce
1 cm thick
Before Irradiation
107 d
18 d, 24 d,
51 d, 51 d+UV
106 h
Recovery after
Irradiation by
10^8 rad
TRANSMITTANCE
WAVELENGTH (nm)
1
0.8
0.6
0.4
0.2
0
300
400
500
600

Fig. 2

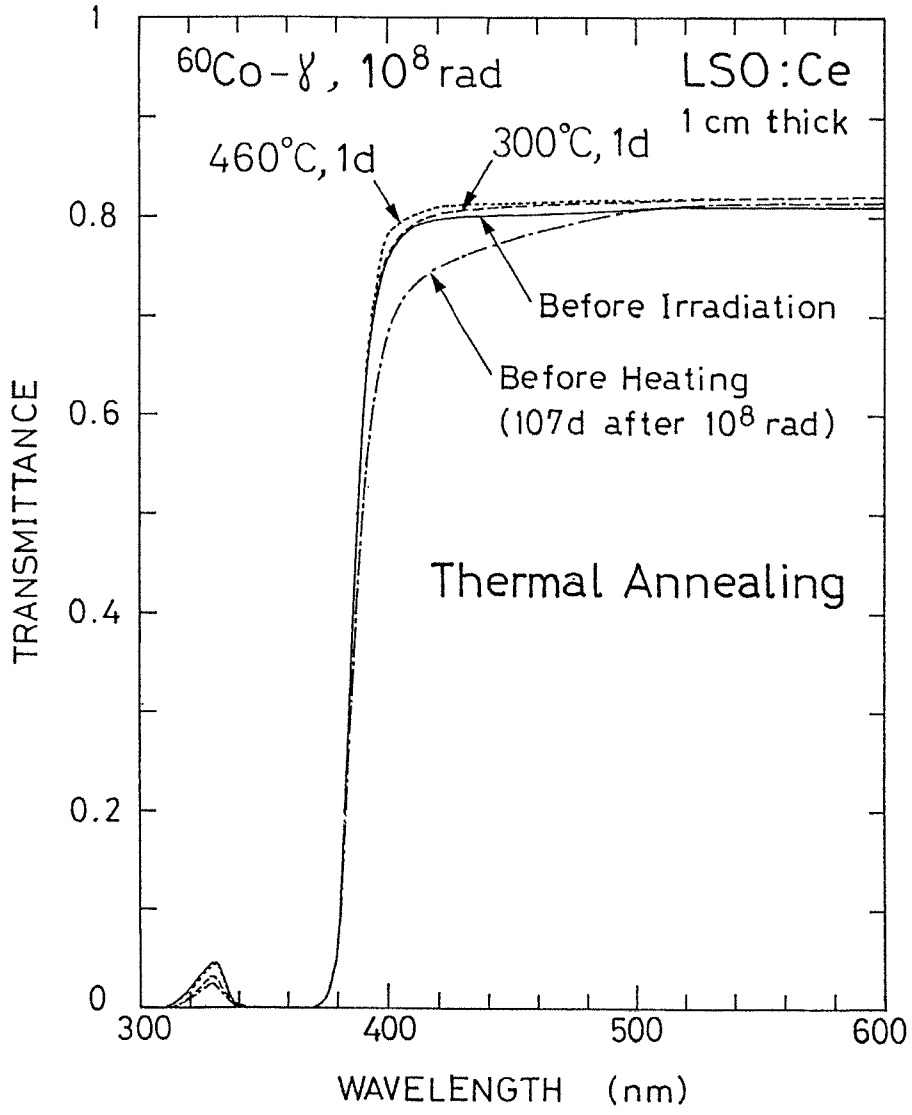
60Co-γ, 10^8 rad
LSO:Ce
1 cm thick
460°C, 1d
300°C, 1d
Before Irradiation
Before Heating
(107d after 10^8 rad)
Thermal Annealing
TRANSMITTANCE
WAVELENGTH (nm)
0
0.2
0.4
0.6
0.8
1
300
400
500
600

Fig. 3

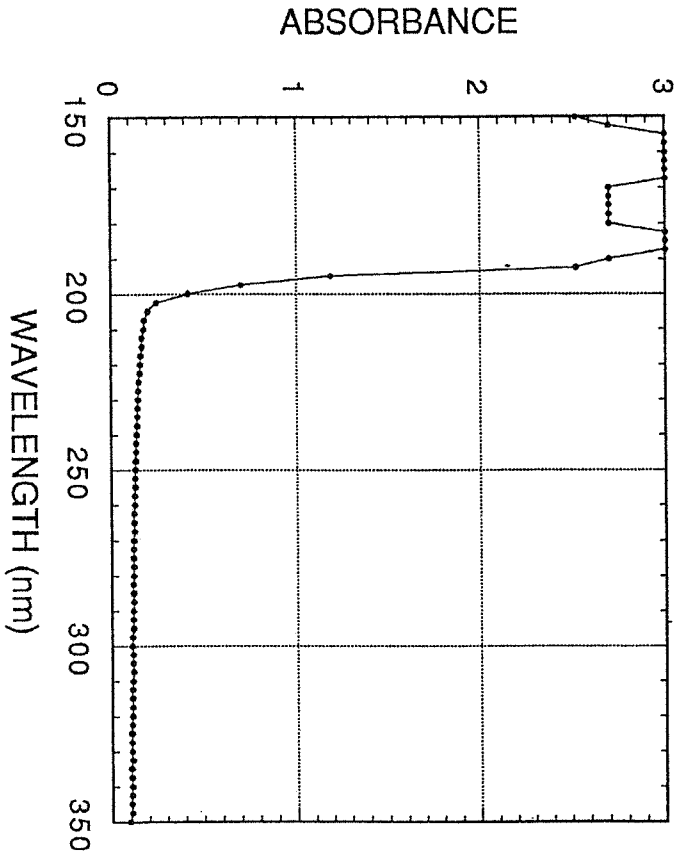
ABSORBANCE
WAVELENGTH (nm)
0
1
2
3
150
200
250
300
350

Fig. 4

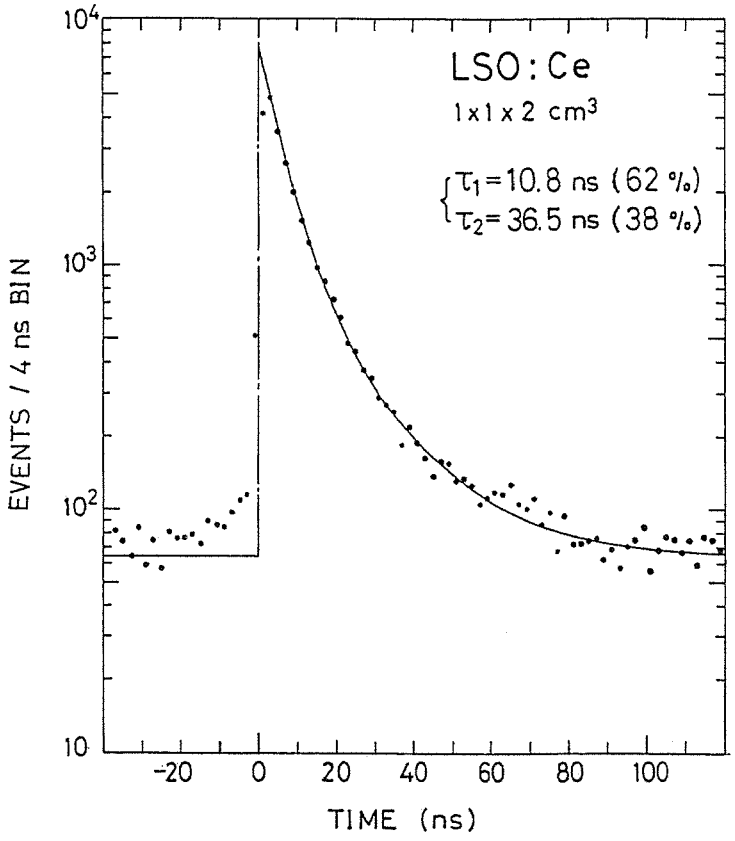

Fig. 6

12

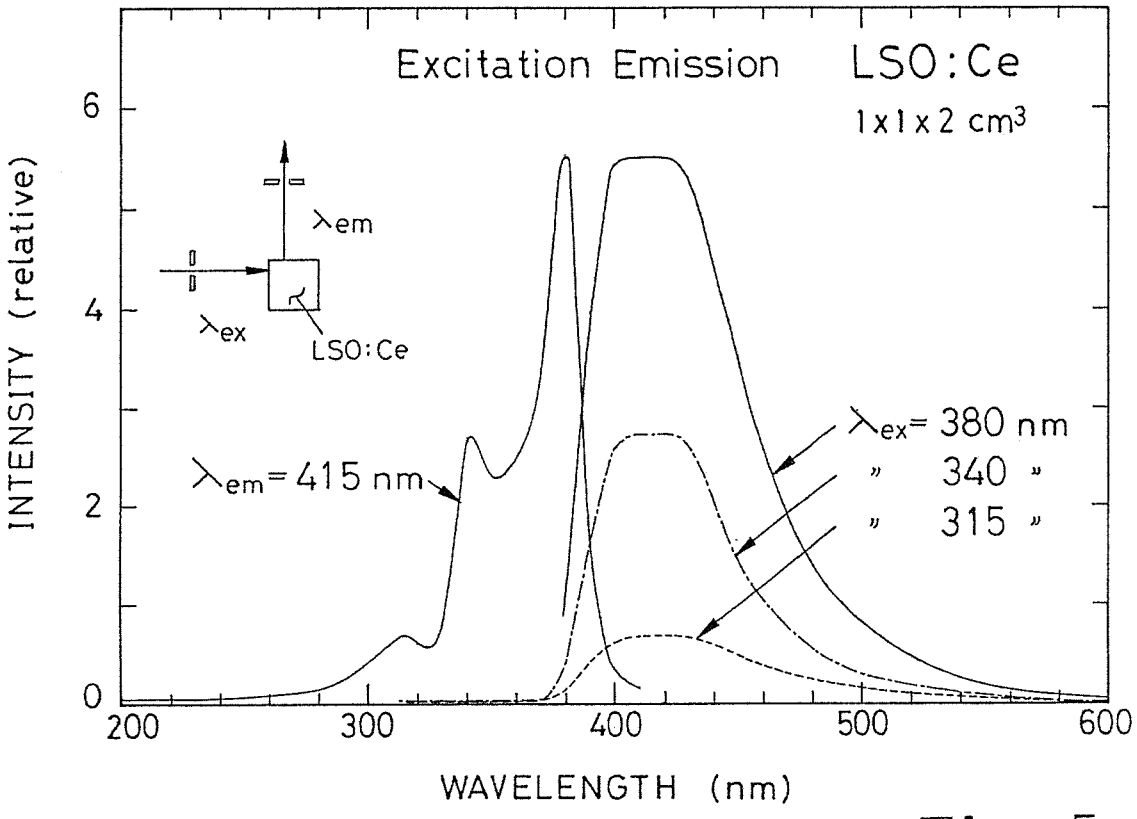

Fig. 5

11

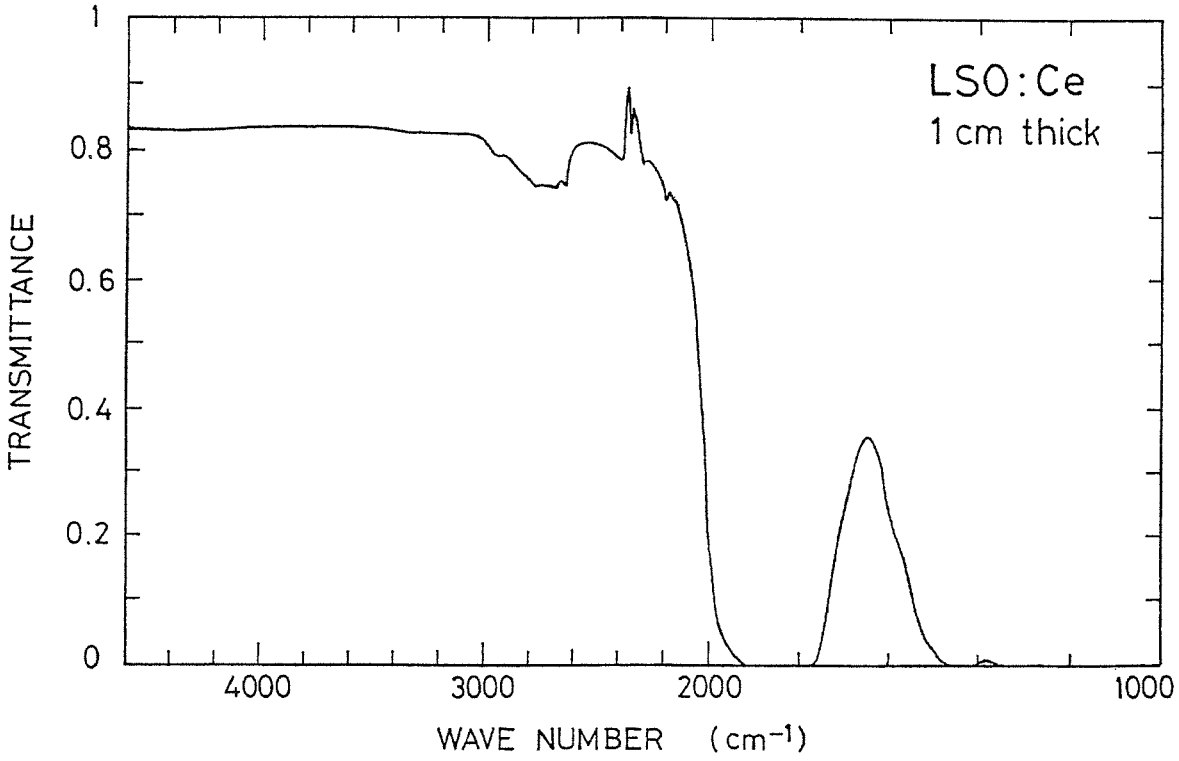
LSO:Ce
1 cm thick
TRANSMITTANCE
1
0.8
0.6
0.4
0.2
0
4000
3000
2000
1000
WAVE NUMBER (cm⁻¹)

Fig. 8

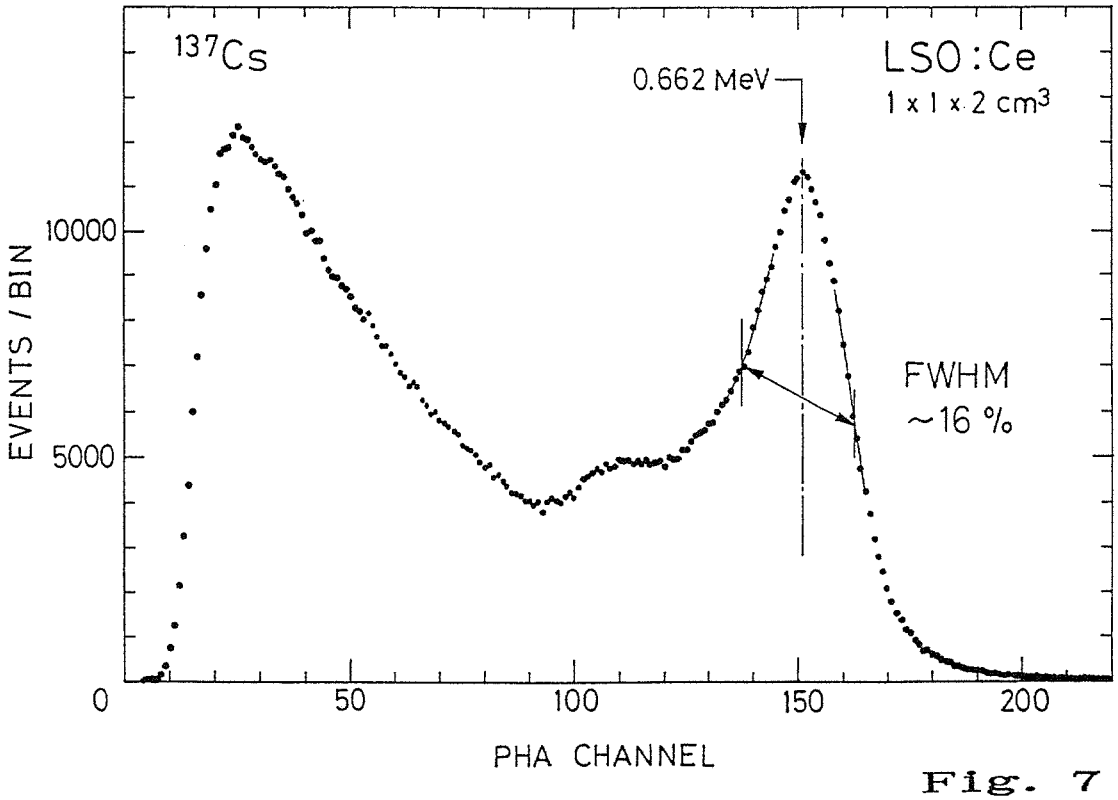
137Cs
0.662 MeV
LSO:Ce
1 x 1 x 2 cm3
FWHM
~16 %
EVENTS / BIN
10000
5000
0
50
100
150
200
PHA CHANNEL

Fig. 7