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# Optical and dosimetric properties of variously doped $SrF_2$ crystals

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**Abstract**

Optical properties and irradiation effects of $Nd^{3+}$-, $Pr^{3+}$-, $Tb^{3+}$- and $Tm^{3+}$-doped $SrF_2$ crystals and their possible application to solid-state dosimetry were studied and compared to those induced in pure $SrF_2$ crystals. Optical absorption, thermoluminescence (TL), X- and light-induced luminescence (XL and PL) as well as optically stimulated luminescence and phototransferred TL (OSL and PTTL) were measured in the various samples. Special attention was given to effects of monochromatic vacuum ultraviolet (VUV) radiation. TL was excited in the pure and doped samples by X and $\beta$ rays as well as by VUV radiation. TL peaks appeared after VUV irradiation at the same temperatures and with the same thermal activation energies as after X or $\beta$ irradiation, indicating that they are due to the same processes. The VUV excitation spectra showed two maxima at about 145 and 130 nm. Comparison of the TL sensitivities of the various TL materials, showed that the sensitivity of $SrF_2$: $Pr^{3+}$ was by more than an order of magnitude higher than that of the known LiF:Mg,Ti (TLD-100) phosphor. The sensitivity of pure and of the Nd, and Tb-doped $SrF_2$ were by a factor of 2–4 higher than that of the TLD-100 and that of $SrF_2$:Tm was slightly lower. The main emission bands of $SrF_2$: $Pr^{3+}$ are located in a convenient spectral region between 460 and 640 nm, where most of the standard photomultipliers are sensitive. The dose dependence of the 460 K TL peak in $SrF_2$: $Pr^{3+}$ is nearly linear in a wide range up to above 27 000 Gy.


*Keywords:* Luminescence; Radiation effects; $SrF_2$; Dosimetry

## 1. Introduction

Irradiation effects in $SrF_2$ and other alkaline-earth fluorides have been studied for several decades (e.g. Tzalmona and Pershan, 1969; Hayes, 1974; Lindner et al., 2001). Radiation-induced absorption bands in $SrF_2$ crystals near 434 and 325 nm have been attributed to electron and hole centers, respectively; it is also well known that the formation of color centers in these fluorides is enhanced by the presence of trivalent rare-earth ions. Most previous studies dealt with effects of higher energy radiation such as $\gamma, \beta$ or X-rays and only relatively few works reported also effects of VUV radiation (e.g. Kirsh and Kristianpoller, 1976; Radzhabov, 2001). Much interest has recently been given to the search of new materials, which can be used as efficient dosimeters. Variously doped fluorides, for example TLD-100 (LiF:Mg,Ti), are frequently used for radiation detectors and efficient dosimeters (e.g. Prokić and Bøtter-Jensen, 1993). For a few decades, mainly thermoluminescence (TL) methods have been applied for solid-state dosimetry. However, more recently, methods of phototransferred TL (PTTL) (e.g. Alexander and McKeever, 1998) and optically stimulated luminescence (OSL) have been applied to dosimetry. For application of a phosphor to solid-state dosimetry, the luminescence efficiency as well as other spectral and thermal properties of the material have to be considered.

In the present work, the effects of X, $\beta$ and UV irradiations on pure and variously doped $SrF_2$ crystals were studied. XL, PL, TL, PTTL and OSL as well as optical absorption were investigated. The dependencies of the luminescence efficiencies of the various materials were

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compared and their dependence on the radiation dose was investigated.

## 2. Materials and experimental methods

The variously doped $SrF_2$ crystals, used for these investigations were grown by the crystal growing laboratory of the Hebrew University in Jerusalem and the pure single crystals were from Harshaw. The optical absorption was measured with a Cary 17 spectrophotometer in the 200–1000 nm region and at shorter wavelengths with a 1m normal-incident VUV monochromator. The far UV irradiations were also carried out with this VUV monochromator and an $H_2$ arc lamp. For the monochromatic near UV illuminations, a 150 W high-pressure Xe lamp was used. Further experimental details are given elsewhere (Kristianpoller et al., 2002).

## 3. Results and discussion

Pure pristine $SrF_2$ crystals are transparent in a broad spectral region from about 130 to 1100 nm. The doped samples showed an increase of absorption below 220 nm and a series of sharp absorption bands typical of the rare-earth ions. The $SrF_2$ :$Pr^{3+}$ crystals showed an absorption band at about 215 nm and additional bands in the VUV region near 187, 168, 155 and 145 nm, followed by a sharp increase in absorption toward the edge of fundamental absorption below 130 nm. Low-temperature irradiation induced in the various $SrF_2$ samples absorption bands at 434 and 325 nm, which have previously been attributed to F and V-type centers, respectively (Hayes, 1974). The pure as well as all doped samples showed after X- or $\beta$ irradiation and during subsequent heating a strong TL emission. Significant differences were, however, recorded in the TL sensitivity of the various crystals and in the spectral composition of the emission as well as in the dose dependence of their TL intensities. After exposure of the various samples to a constant $\beta$ dose of 1.5 Gy, the ratio of the main TL intensities in the 400–470 K temperature region of: (1) pure, (2) Nd-, (3) Tb-, (4) Tm- and (5) Pr-doped $SrF_2$ crystals was 5: 4: 3.5: 0.2: 5 and after exposure to a 90 Gy dose the ratio was: 8: 5: 8: 0.2: 65. The comparison of the TL response of $SrF_2$:$Pr^{3+}$ with that of TLD-100 showed that at a low dose of 1.5 Gy the sensitivity of $SrF_2$: $Pr^{3+}$ was by an order of magnitude higher than that of TLD-100 and at a dose of 90 Gy by a factor of $\sim$ 30. In Fig. 1, a comparison of the TL sensitivities (excited by 45 Gy beta dose) of pure $SrF_2$, $SrF_2$:$Pr^{3+}$ and TLD-100 is given. The 410 K TL peak is the most sensitive one at low doses but at higher doses, the TL peak near 460 K becomes dominant in $SrF_2$:$Pr^{3+}$. This strong TL peak showed also almost linear dose dependence up to high doses above 27 000 Gy, while the lower temperature peaks reach saturation at much lower doses. The dependence of the 460 K peak on the radiation dose is shown in Fig. 2.

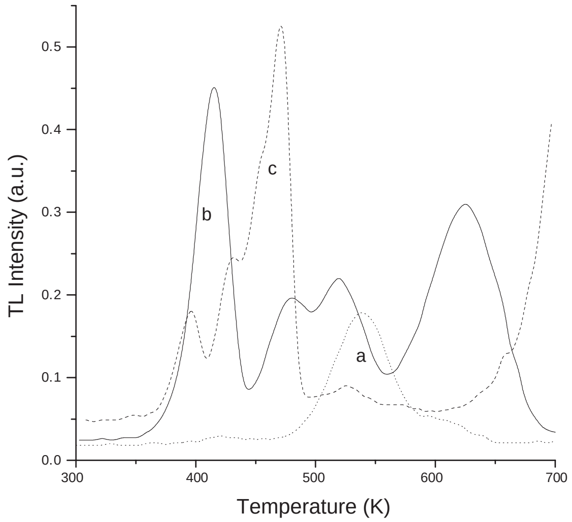


Fig. 1. Thermoluminescence excited at RT by a 45 Gy $\beta$ dose in: (a) pure $SrF_2$, (b) $SrF_2$:$Pr^{3+}$, (c) LiF:Mg,Ti (TLD-100) [enlarged by 10].

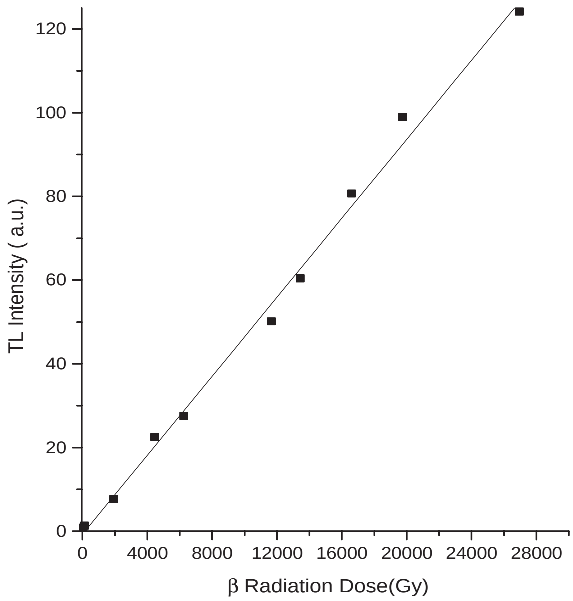


Fig. 2. Dependence of the 460 K TL peak intensity in $SrF_2$:$Pr^{3+}$ on the $\beta$ dose. (The solid line is a best-fit of the experimental results to a linear dependence).

TL could also be induced by monochromatic VUV radiation. The excitation spectrum of the TL showed maxima at about 145 and 130 nm (Fig. 3). The first maximum is located at a weak absorption band at 145 nm and the second on the long wavelength tail of the fundamental absorption;

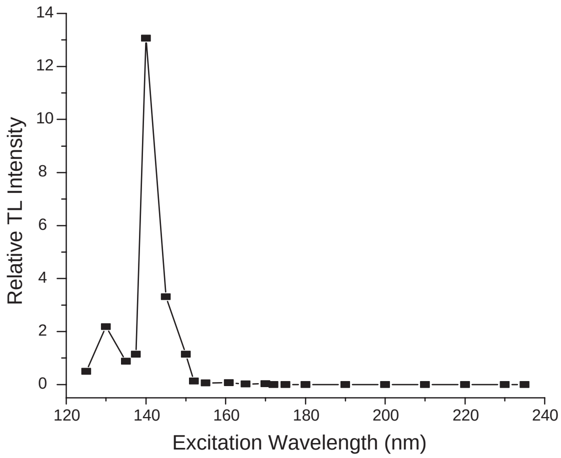


Fig. 3. TL excitation spectrum of $SrF_2:Pr^{3+}$.

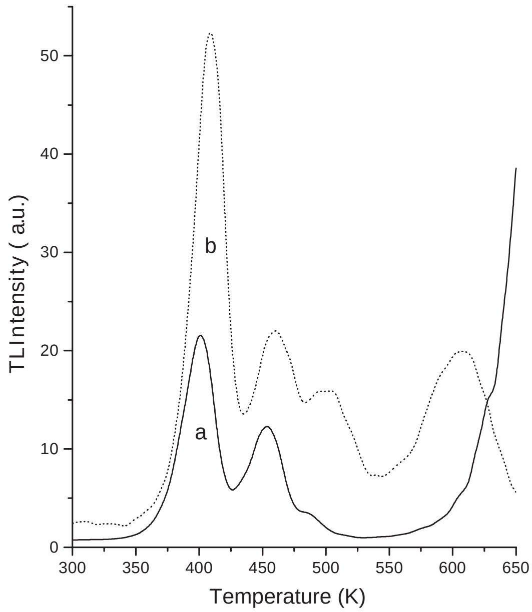


Fig. 4. TL induced in $SrF_2:Pr^{3+}$ at RT by: (a) 145 nm VUV radiation, (b) $\beta$ radiation of 90 Gy.

this fits previous results and conclusions in various alkali halide crystals. where UV excitation of TL was most efficient at the long wavelength tail of the exciton bands and was attributed to an excitonic process (Kristianpoller and Israeli, 1970). In Fig. 4 the TL induced at RT in a $SrF_2:Pr^{3+}$ crystal by 145 nm VUV radiation is compared to that induced in the same sample by a $\beta$ radiation of 90 Gy. Results show that at least some of the TL peaks appear after VUV excitation at the same temperatures and with the same thermal activation energies as after X or $\beta$ irradiation (90 Gy), indicating that they are due to the same processes. In doped as well as in pure samples, which had previously been exposed to X, $\beta$ or VUV radiation at RT and subsequently illuminated at LNT with near UV and even visible light, a luminescence emission was observed during heating from LNT to RT. These wavelengths could not excite any TL in pristine samples and the emission is therefore ascribed to a process of optical stimulation and phototransfer (PTTL). In pure and doped pre-irradiated samples, luminescence could be stimulated with wavelengths that could not excite any PL in pristine samples; this emission is ascribed to a process of optical stimulation (OSL). The emission spectra of the main TL peaks in the pure $SrF_2$ crystals showed a broad composed band between 290 and 330 nm with a shoulder at 382 nm and weaker bands at 485, 520 and 615–640 nm. The emission of the doped crystals was dominated by series of narrow emission bands, characteristic of the different rare-earth ions. In Fig. 5 the emission spectra of TL and XL of pure and doped crystals are shown.

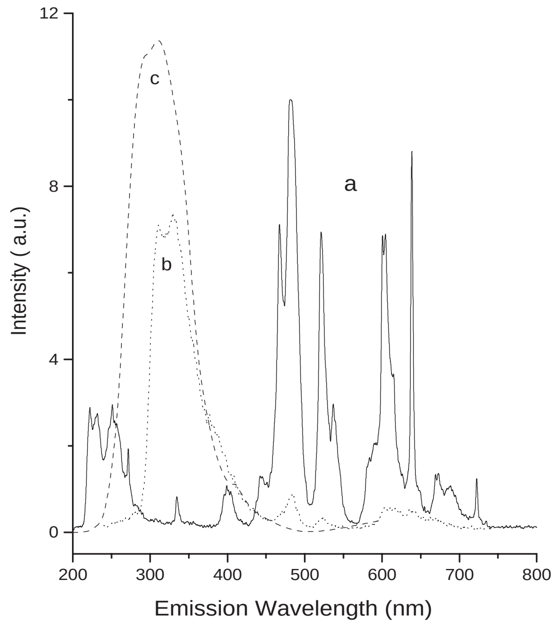


Fig. 5. Emission spectra recorded at 460 K TL peak of: (a) $SrF_2:Pr^{3+}$, (b) pure $SrF_2$ and (c) XL at RT of pure $SrF_2$ crystal.

## 4. Summary of dosimetric properties

For application of a phosphor to dosimetry, the luminescence efficiency as well as other properties of the material such as the temperature of the main TL peaks, the wavelength of the emission, the thermal stability and temperature of thermal annealing, the reproducibility of results and the possibility of re-use of the same sample have to be considered. The results have shown that the TL sensitivity of the $Pr^{3+}$ doped $SrF_2$ crystal was the highest among the various tested $SrF_2$ samples and in particular at the higher temperature peaks. The sensitivity of $SrF_2$:$Pr^{3+}$ was by an order of magnitude higher than that of TLD-100 and than that of the recently examined $CsGd_2F_7$: $Pr^{3+}$ crystal (Kristianpoller et al., 2002). The main emission bands of $SrF_2$: $Pr^{3+}$ are located in a convenient spectral region between 460 and 640 nm, where most of the standard photomultipliers are sensitive. The $SrF_2$ crystals are also stable up to relatively high temperatures and can be re-used for repeated TL measurements. The strong 460 K peak of $SrF_2$: $Pr^{3+}$ is well above RT and normal thermal fading during storage at RT is therefore not expected to occur. The dose dependence of this 460 K peak is nearly linear up to high doses above 27 000 Gy.

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