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Optically stimulated luminescence in synthetic quartz

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Abstract

Optically stimulated luminescence (OSL), has been studied in synthetic quartz crystals, which had previously been exposed to β- or X-irradiations. The main emission bands appeared at $\sim$380 and $\sim$440 nm. The excitation spectra of these emission bands were now measured and showed both a maximum at 330 nm. This maximum coincides with an excitation maximum of the phototransferred thermoluminescence (PTTL), indicating that the OSL and PTTL are due to the same traps. The emission intensities were found to depend linearly on the intensity of the stimulating 330 nm light, as well as on the dose of the β- or X-irradiation over a wide range of doses. The results support the possibility of applying OSL in quartz for dosimetry and dating.

1. Introduction

Optically stimulated luminescence (OSL) and its application to dating of geological and archaeological samples has first been reported in 1985 by Huntley et al. [1], and has recently been studied in natural quartz and in various other minerals [e.g. 2,3]. In this method the samples are exposed to ionizing radiation and subsequently illuminated by light of wavelengths, which cannot directly excite luminescence in an unirradiated crystal. This light can however stimulate trapped carriers which may recombine with carriers of opposite sign, yielding the OSL emission. To the best of our knowledge no systematic measurements of OSL as a function of stimulating wavelength, by using a continuous spectrum and monochromator, have so far been published for quartz. Instead most laboratories adopted the use of a single wavelength from an argon ion laser (5.14.5 nm), which was first used by Huntley et al. [1] for excitation of the OSL in natural quartz.

For the present investigations, highly pure synthetic quartz samples were exposed to β- or X-irradiation at room temperature (RT), then illuminated by monochromatic light in a broad spectral range from about 270 to 550 nm and excitation as well as emission spectra of OSL were measured for the first time in these crystals. The OSL is also compared to results recently obtained in the phototransferred thermoluminescence (PTTL) of the same crystals [4]. The PTTL, as the OSL, can be stimulated only in samples which have previously been exposed to ionizing radiation. This radiation causes the trapping of carriers at deep traps; during illumination at lower temperatures part of these trapped carriers may then be transferred to shallower traps. PTTL is observed when during subsequent heating the carriers are thermally released from the shallow traps and recombine radiatively at luminescence centers.

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Effects of thermal pre-treatment on the OSL emission, as well as thermal stability of the radiation induced defects were now also investigated. The dependence of the OSL intensity on the dose of the stimulating UV light were here measured for the first time in pre-irradiated synthetic quartz crystals and applicability to dosimetry is discussed.

2. Experimental procedure

For our measurements single synthetic quartz crystals (Z-growth Premium grade from Sawyer Research Products) as well as polycrystalline powders from the same material were used. The impurity content in these high purity crystals is known to be of the order of 1 ppm Al and a few ppm Ge. Other impurities are at a much lower level. The X-irradiation were performed at RT with a W tube (40 kv, 15 mA) and the β-irradiation with a ^{90}Sr source. For some of the illuminations the 514 nm line of a 2 W argon ion laser was applied. For most of the monochromatic illuminations a continuous light source (halogen or 150 W high pressure Xe-lamp) and a Jarrell Ash 0.25 m double monochromator were used. The OSL was recorded during illumination with monochromatic light at various temperatures, mostly at RT or LNT. Emission spectra were measured with a 0.5 mm Bausch and Lomb grating monochromator, equipped with a fast scanning stepping motor and controlled by an electronic device. The spectra were recorded with a thermoelectrically cooled EMI-9558-QB photomultiplier (S 20: response). Appropriate optical filters were applied in order to exclude straylight and second-order effects. For measurements of excitation spectra of the OSL emission bands, the incident photon flux was monitored by a pyroelectric radiometer. The dependence of the OSL intensities on the dose of the β- and X-irradiations was measured for constant radiation intensities by varying the irradiation time. The dependence of the OSL intensity on the dose of the stimulating light was measured for constant incident photon flux by varying the time of irradiation, as well as for constant time of irradiation and varying photon flux. Absorption measurements were performed with a Cary 17 spectrophotometer.

3. Results and discussion

A notable luminescence emission was observed during near UV illumination of samples, which have previously been irradiated by β- or X-rays. No such luminescence could be detected in samples which had not previously been exposed to ionizing radiation. This fits results of a previous study on high purity synthetic quartz which have shown that luminescence as well as thermoluminescence (TL) can be directly excited in these pristine crystals only by vacuum UV radiation of wavelengths shorter than $\sim$170 nm [5]. Our present results show that the intensity of the OSL emission depends on various factors such as: wavelength and dose of the illuminating light, the dose of the previously absorbed ionizing radiation, and thermal pre-treatment of the samples. Heating the sample to $\sim$400 °C before exposure to β- or X-irradiation caused a notable increase in the emission intensities. The emission intensities of powder samples were relatively higher than those of single crystals at equal conditions. It is assumed that during β- or X-irradiation at RT, carriers were trapped at deep traps, and remain at these traps for long periods. Part of these carriers are then stimulated by light of appropriate wavelength and OSL is emitted as a result of their radiative recombination at luminescence centers. The emission spectra of the OSL were measured and results are shown in Fig. 1. Main emission bands appeared at $\sim$380 and $\sim$440 nm. It can be seen that the intensity of the OSL band is markedly higher by illumination at LNT. Emission bands at the same wavelengths have previously been observed in the TL, PTTL and in the x-induced luminescence (XL) of quartz. The 440 nm XL-emission intensity showed also a similar increase during cooling from RT to LNT. The 380 nm XL emission band has previously been ascribed to the recombination of electrons with holes trapped at Al^{+3}-M^{+} centers and the broad band near 450 nm has been attributed to an intrinsic STE emission [5,6]. The finding that the same bands appeared also in the OSL, indicates that the same luminescence centers are responsible for these emissions in the OSL, XL, TL and PTTL.

In most previous works the OSL was stimulated in natural quartz crystals by the powerful radiation

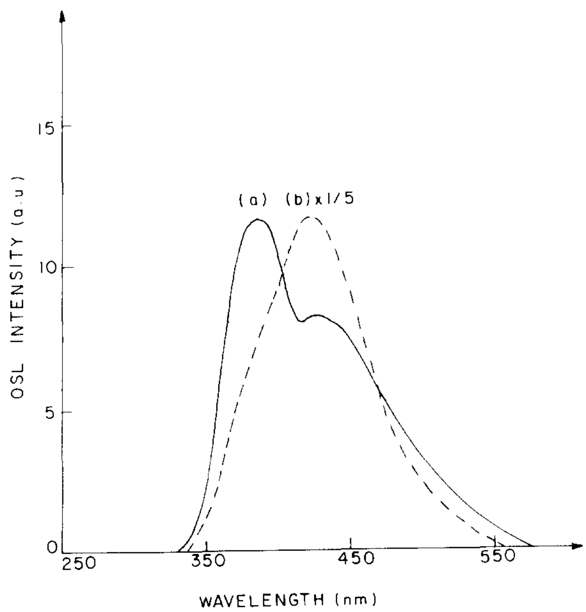

Fig. 1. Emission spectra of the OSL. The samples were exposed to a β dose of ~ 10 Gy and subsequently illuminated with 330 nm light at (a) RT (b) LNT.

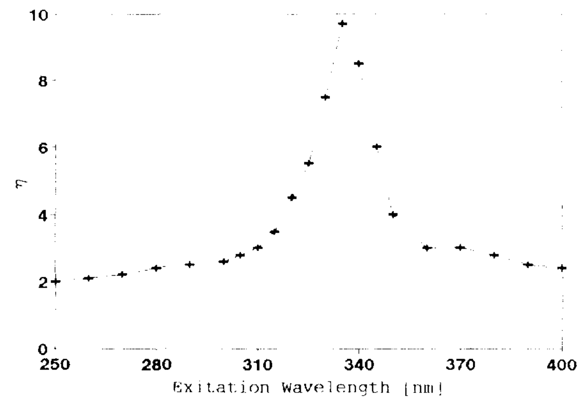

Fig. 2. Excitation spectrum of the ~ 440 nm OSL emission band (data were taken at RT).

of an argon ion laser at 514.5 nm. For some of our preliminary work the same wavelength has been applied for stimulation of the OSL in high purity quartz. Our investigations were then extended to the stimulation with monochromatic light in the 270–550 nm region and excitation spectra of the main OSL emission bands were measured at RT, using continuous halogen or xenon light sources. Results for the 440 nm band are given in Fig. 2. A strong stimulation maximum appeared at 330 nm. The excitation spectrum of the 380 nm emission showed the same stimulation maximum. Except for differences in relative intensities the same stimulation maximum also appeared for UV illuminations at LNT. No absorption band could be detected at 330 nm, and none of the main impurities in these crystals are known to have an absorption band near 330 nm. A comparison with excitation spectra, recently measured in the PTTL of the same samples showed that the PTTL has a stimulation maximum at the same wavelength, indicating that both effects are due to the same traps. This is also supported by recent investigation of the optical bleaching of TL. These measurements have shown that TL effects, which were induced in natural quartz by ionizing radiation, can most efficiently be bleached by sunlight of 330 nm [7]

It should be noted that a weak "B" absorption band at ~ 3.9 eV and half-width of 1.2 eV has previously been reported in fused silica and in heavily γ-irradiated smoky-quartz crystals, and has been attributed to a trapped electron center [8,9]. The observed OSL and PTTL excitation peaks at about 330 nm may possibly be due to this center, although no absorption band was detected at this wavelength region in our highly pure synthetic crystals. Luminescence methods are known to be much more sensitive for detection of impurities and defects than measurements of optical absorption. It has previously been shown that defect concentrations of about 10^{10}–10^{11} cm^{-3} can be detected by luminescence methods, while the lower limit detectable by measurements of optical density was 10^{15}–10^{16} cm^{-3} [10].

Our investigations have shown that heating to above ~ 600 °C between the exposure to ionizing radiation and the UV illumination, caused a decrease in the emission intensities and that no OSL could be excited in samples which were heated to above 800 °C, indicating that the deep traps responsible for the OSL emission are thermally stable up to ~ 600 °C.

The dose dependence of the OSL was now also investigated and the OSL intensities were found to depend linearly on the intensity of the stimulating light, as well as on the dose of the previously absorbed β- or X-irradiation over a wide range of doses. Repeated monochromatic UV illuminations of the same sample caused normally only a slight decrease in the OSL intensities; samples can therefore be reused without a need for additional exposure to ionizing radiation. These results support the possibility of utilizing OSL in quartz for dosimetry and dating.

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