Saturation of a Ce:Y₃Al₅O₁₂ scintillator response to ultra-short pulses of extreme ultraviolet soft X-ray and X-ray laser radiation

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Abstract: Investigations of radioluminescence saturation in monocrystalline cerium doped yttrium aluminum garnet (Ce:YAG) exposed to intense extreme ultraviolet (XUV), soft X-ray and X-ray radiation delivered from three free-electron lasers are described in this article. The measurements were performed with wavelengths of 98, 25.6, 13.5 and 0.15 nm. We have found that saturation of the photon yield occurs at an excitation level of 2.0 x 10²⁰ eV/cm³ resulting in an emission of 1.5 x 10¹⁸ visible photons per cubic centimeter. This number is much smaller than the concentration of Ce³⁺ in the scintillator that was equal to 2 x 10²⁰ cm⁻³.

Because the internal radiance efficiency η does not depend strongly on the irradiating photon energy, i.e., η = 0.035 ± 0.015 in the range 10 to 100 000 eV, the results presented here could be used to predict saturation effects in scintillator slabs placed in imaging systems of bright XUV, X-ray and particle beams. The saturation of the Ce³⁺ emission is explained by mutual quenching of excitons created at high densities, preceding the stage of energy transfer to the Ce³⁺ ions.

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1. Introduction

Inorganic scintillators are widely used for diagnostics and visualization of x-ray and charged particle radiation [1]. The recent advances in high-brightness sources of energetic photons, esp. plasma-based and free-electron short-wavelength lasers, set new challenges for diagnostic tools, particularly the saturation effects in phosphor screens. Ce:YAG (cerium doped yttrium aluminum garnet, YAG = Y₃Al₅O₁₂) scintillators have been used and are planned to be used for imaging bright electron, proton and photon beams [2–5].
The concerns regarding the saturation of Ce:YAG applied for diagnostics of various beams of ionizing radiation were expressed in several publications [2–4]. In refs [2,3], the authors reported the saturation of radioluminescence emitted by Ce:YAG crystals exposed to an electron beam. In ref [4], the TESLA Test Facility Free Electron Laser (TTF FEL) photon beam coherence measurements had to be corrected to take into account a nonlinear response of the Ce:YAG crystal. However, in the mentioned papers the saturation of the Ce:YAG scintillator yield was treated only indirectly, without a quantitative description of the phenomenon. In this paper we present systematic measurements of the luminescence of Ce:YAG crystals excited by the ultrashort, intense pulses of short-wavelength laser radiation produced at the TTF FEL [6] and Free Electron LASer in Hamburg (FLASH) [7,8] at DESY (Deutsches Elektronen-Synchrotron) and at the Linac Coherent Light Source (LCLS) at SLAC National Accelerator Laboratory [9,10]. These measurements allow a quantitative description of the dependence of scintillation yield on the absorbed dose of XUV, soft x-ray and x-ray radiation. It is known that the recombination of electrons and holes results in the formation of self-trapped excitons (STEs) in pure YAG crystals [11], which contribute to the transfer of energy to dopant and defect centers present in the host [12]. Rare-earth doped phosphors have been investigated under XUV FEL excitation and both the emission spectra and decay kinetics of various rare earth ions (Ce, Pr, Eu) have been shown to stay unchanged upon excitation density variation. However, all luminescence centers have shown a non-linear response to pulse energy as reported in ref [13].

2. Experimental

The luminescence experiment was carried out at the TTF FEL [6] and FLASH [7,8] facilities (both at DESY, Hamburg, Germany) and the LCLS [9,10] (SLAC National Accelerator Laboratory, CA, USA) tuned to wavelengths of 98 nm, 13.5 nm and 0.15 nm respectively. The FEL devices provided up to $10^{13}$ photons per < 100 fs pulse with a repetition rate of 1 Hz and higher. The single crystal Ce:YAG (0.2 at. % Ce) slab was polished to optical quality and mounted on the sample holder in the vacuum interaction chamber [14,15] at the TTF FEL and FLASH.

Fig. 1. The layout of the experimental setup. At the TTF FEL and FLASH, the excitation fluence was varied by moving the chamber together with the sample in and out of the focus of the incident beam.

The relevant features of the experimental set-up are presented in Fig. 1. The decay curves of the emissions, spectrally selected by suitable filter combinations, were detected for each FEL pulse by a fast XP2020Q photomultiplier (PMT) and stored using a digital oscilloscope. In addition to the inherent fluctuations of the FEL radiation due to the nature of the self-
amplified spontaneous emission from the FEL, the excitation density was varied through the range of 0.01 to 10 GW/cm² by moving the chamber together with the sample in and out of the focus of the incident beam. All presented decay curves result from crystals at a room temperature (300 °K) excited by a single FEL pulse much shorter (< 100 fs) than the instrumental response of the detection system (~1 ns).

The measurements performed at LCLS were done using the Direct Imager (DI) diagnostic tool described in detail elsewhere [16]. The direct imager’s Ce:YAG crystal was exposed to an unfocused LCLS photon beam. The X-ray intensity profile was recorded by a CCD camera. The flux of the 8.3-keV photons in the LCLS beam was controlled by solid Be attenuators and measured by gas ionization detectors [17].

In addition, radioluminescence and emission decay studies were performed under 25.6 nm excitation from FLASH using the setup described in [18]. Emission spectra were recorded using an ARC Spectra 300i spectrometer with a Princeton Instruments CCD detector, and luminescence decay was recorded using an XP2020Q PMT with suitable color filters. In these experiments, the pulse energy was changed using FLASH’s gas attenuator.

3. Results

Typical data acquired at the TTF FEL facility are shown in Fig. 2. It represents five data sets measured at different sample–focus distances: 104 mm, 14 mm, 11 mm, 9 mm, and 5 mm. The saturation of the scintillation emission is clearly seen in this figure. The lines represent the calculated number of photons using a simple linear saturation model for the local radiance:

$$\phi_l = \eta \left\{ \begin{array}{lll} e & \text{if} & e \leq e_{sl} \\ e_{sl} & \text{if} & e > e_{sl} \end{array} \right\}$$

where \(\phi_l\) is the local energy of visible photons radiated per unit volume (local energy density emitted in visible band), \(e\) is the local energy density of absorbed energetic photons, \(\eta\) is the low excitation internal radiance efficiency defined as the ratio \(\phi/e\) and \(e_{sl}\) is the saturation energy density in this linear model. The local radiance formula was integrated over the sample volume with the absorbed energy distribution given by the parameters of the FEL beam and the focusing optics, position of the sample with respect to focusing mirror, and the Ce:YAG optical constants [19]. After approximating the angular distribution of the FEL beam by a Gaussian beam distribution, one derives the following relation describing number of photoelectrons reaching the PMT anode \(N_t\):

$$N_t \sim \left\{ \begin{array}{lll} \hat{E} & \text{if} & \hat{E} \leq e_{sl} \\ e_{sl} \left[ 1 + \ln \left( \frac{\hat{E}}{e_{sl}} \right) + \frac{1}{2} \ln^2 \left( \frac{\hat{E}}{e_{sl}} \right) \right] & \text{if} & \hat{E} > e_{sl} \end{array} \right\}$$

Here \(\hat{E} = ET(\pi r_0^2 \Lambda)^{1/4}\) is the maximum value of absorbed energy density at the spot center where \(E\) is the short-wavelength pulse energy, \(T\) is the transmission coefficient, \(r_0\) is the Gaussian beam radius at the sample position and \(\Lambda\) is the attenuation length. \(T\) and \(\Lambda\) have been obtained by solving the Fresnel equations [20] for p-polarized light at an incident angle of 55° using the YAG complex refraction index of \(n = 1.3 + 0.82i\) measured at a wavelength of 98 nm [19]. Using these parameters, the the experimental data measured at focus-sample distances of 5 mm, 9 mm, 11 mm and 14 mm were fit to the linear model of saturation described by Eq. (2). It was found that single value \(e_{sl}\) of 38 ± 6 J/cm³ makes excellent predictions of the PMT signal as a function of XUV photon beam energy for all focus-sample distances.
A similar procedure was applied to measure the saturation energy density at a wavelength of 13.5 nm (FLASH), except $\lambda$ was calculated using Henke’s Tables \[21\] as appropriate for the soft x-ray (and x-ray) FEL radiation. The experimental data are plotted in Fig. 3. Here the quality of the data is lower in comparison with data plotted in Fig. 2, due to technical differences between the experimental stations used for the measurements at 98 nm and 13.5 nm. At 13.5 nm the interference filter in the decay data acquisition system had a smaller bandwidth resulting in a larger scatter in the data. Also the pulse energy monitor had a considerably larger scatter at 13.5 nm wavelength due to electronic disturbances resulting from long wiring. In addition the range of beam spots sizes used was much smaller at 13.5 nm and the beam profile showed considerable internal structure in the smallest spots. This means that an additional variation of the excitation density within the focal spot and an inherently larger signal scatter occurs as the detector records an average over the whole exposed area. Even so, the fit of the 13.5 nm data to Eq. (2) resulted in a saturation energy density of $e_{sl} = 25 \pm 15 \text{ J/cm}^3$, which is within the error range of the value obtained for 98-nm photons. Of course for higher energy densities the crystal can be damaged. The damage thresholds in Ce:YAG illuminated at the TTF FEL and FLASH facilities have been investigated \[22,23\], and it was found that damage dose is approximately 3 order of magnitude higher that luminescence saturation dose.
LCLS data was collected by a CCD camera having a resolution of 20 microns per pixel. The intensity profile, was a Gaussian of 200 micron rms beam size. The measurements were done at an average x-ray pulse energy of 1.8 mJ. The fluence on the Ce:YAG crystal was controlled by inserting one or more solid Be foils to vary the total attenuating thickness in the beam from 0 to 16 mm. At a given transmission we recorded multiple CCD images and the average intensity in the brightest pixel was analyzed as a function of fluence. The experimental normalized average intensity is plotted as open circles in Fig. 4.

In order to apply the linear model of saturation described by Eq. (1) to the data shown in the Fig. 4 one needs to integrate Eq. (1) along the depth of the crystal. This gives us a relation for number of photons $N_l$: 
where $\tilde{F}_i = \tilde{F} \cdot (e_{\text{sat}} \Lambda)^{-1}$ and $\tilde{F}$ is the fluence in the spot center. The fit obtained with the linear model is drawn as dot-dashed line in Fig. 4. While the linear model describes the experimental data quite well, a more realistic exponential type of saturation has been proposed. In this model the local energy of visible photons $\varphi_\text{v}$ is given by this equation:

$$\varphi_\text{v} = \eta \cdot e_{\text{sat}} \left( 1 - \exp \left( -e_{\text{sat}} / e_{\text{sat}} \right) \right)$$

where similarly to previous model the $e_{\text{sat}}$ is the saturation energy density and $\eta$ is the low excitation internal radiance efficiency. The number of photons $N_i$ in the central pixels is derived by integrating (4) along the Ce:YAG crystal thickness which yields:

$$N_i \sim \left[ \tilde{F}_i \cdot \left( 1 - \exp \left( -L / \Lambda \right) \right) \right] \text{ if } \tilde{F}_i < 1$$

$$\left[ 1 - \tilde{F}_i \cdot \exp \left( -L / \Lambda \right) + \ln(\tilde{F}_i) \right] \text{ otherwise}$$

(3)

where $\tilde{F}_i = \tilde{F} \cdot (e_{\text{sat}} \Lambda)^{-1}$ and $\tilde{F}$ is the fluence in the spot center. The fit obtained with the linear model is drawn as dot-dashed line in Fig. 4. While the linear model describes the experimental data quite well, a more realistic exponential type of saturation has been proposed. In this model the local energy of visible photons $\varphi_\text{v}$ is given by this equation:

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(3)

where $\tilde{F}_i = \tilde{F} \cdot (e_{\text{sat}} \Lambda)^{-1}$ and $E_i$ is the exponential integral.

Both models describe the X-ray experimental data well while the exponential one provides an almost perfect prediction. The best fit to the exponential and linear models are obtained for $e_{\text{sat}} = 25 \pm 5 \text{ J/cm}^3$ and $e_{\text{sat}} = 33 \pm 6 \text{ J/cm}^3$, respectively. To compare the saturation doses measured at different photon energies we use the linear model. It gives closed mathematical formulas for the total number of photons emitted under the influence of incident Gaussian beams. Fitting the linear model to the data results in saturation doses of $e_{\text{sat}} = 38 \pm 6 \text{ J/cm}^3$, $25 \pm 5 \text{ J/cm}^3$ and $33 \pm 6 \text{ J/cm}^3$ at 98, 13.5 and 0.15 nm respectively, which are the same to within measurement errors, suggesting that this constant is independent of photon energy.

Additional experimental data on the saturation of Ce:YAG crystals were obtained under excitation of 25.6 nm where the Ce$^{3+}$ emission spectra at various excitation pulse energies (see Fig. 5 in ref [13].) were measured. The dependence of the total Ce$^{3+}$ emission integrated over the entire band in the range of 450 to 650 nm on pulse energy is shown in Fig. 5. It clearly demonstrates saturation effects analogous to those shown in Fig. 2, 3 and 4.

4. Discussion

The FEL pulse excites electrons either from the valence band or core levels into the conduction band (CB), which is followed by relaxation of the electrons (and holes) through various processes. Absorption of 13.5 nm photons (91.8 eV) creates core holes in the Al 2p levels ($\geq$ 75 eV) [24,25], but is insufficient to cause transitions starting from the Ce$^{3+}$ 4d absorption edge ($\geq$ 115 eV) [26,27]. For harder x-ray photons, however, excitation of electrons from much deeper core levels to the CB takes place. The final energy transfer to the Ce$^{3+}$ ions takes place independently of the exciting photon energy and the relaxation processes involved. As a result, the well-known 5d-4f emission of Ce$^{3+}$ in the range of 500-600 nm is detected in Ce:YAG crystals (see also [19,28]). The decay kinetics of the Ce$^{3+}$ emission hardly depends on exciting photon energy. A single exponential fit of the decay curves under excitation by 98-nm FEL radiation resulted in a life-time value of $\tau \sim 89$ ns [29], which agrees well with $\tau \sim 93$ ns obtained in [12] using excitation by synchrotron radiation.
That the decay time does not depend on the type of excitation suggests that the fluorescence process of the scintillation centers is not significantly influenced by the powerful photon beam despite the observed saturation. This behavior is in contrast to the results obtained for CaWO₄ and CdWO₄ under comparable conditions at FLASH [18] and in experiments with high-order harmonics [30]. For CaWO₄ and CdWO₄, increasing self-quenching of the excitonic emission and changing of its decay curve from exponential to non-exponential is observed at high pulse energies. A model of exciton-exciton interaction due to Förster energy transfer was developed to describe the observed changes in decay profiles. Further studies of this phenomenon under direct creation of excitons using ultra-short pulses of UV laser radiation, where the experimental conditions are better controllable than under FEL excitation, confirmed the validity of the model of excitonic quenching for CaWO₄ and CdWO₄ [18,30–32]. However, for other types of materials such as halides, quenching effects due to the interaction of excited charge carriers has to be considered as well [33].

The internal radiant efficiency $\eta$ for the Ce:YAG scintillator at 98 nm, reported in [34] equals about 0.018. Using this value and the saturation energy density of $e_s \approx 2 \times 10^{29}$ eV/cm$^3$ (32 J/cm$^3$) in Eq. (1) gives a radiance of $1.5 \times 10^{18}$ photons per cubic centimeter, two orders of magnitude lower than the concentration of Ce$^{3+}$ ions: $n_{\text{Ce}} \approx 2 \times 10^{20}$ cm$^{-3}$ (0.2% at., [29]). Thus saturation occurs well before all Ce$^{3+}$ ions are excited. This implies that the saturation observed for ultrashort pulses is not caused by a lack of available scintillation centers, but instead by a reduction in the process of energy transfer to the Ce$^{3+}$ ions.

The mechanism of saturation can be understood from the following considerations. According to [35,36], the relaxation of free carriers in complex oxides is very fast due to the interaction of the free carriers with optical phonons having a large energy. As a result, the free path length of conduction band electrons is small, and electrons and holes quickly form excitons which then decay radiatively or non-radiatively. For that reason, energy losses for charge carrier trapping at impurities and defects are small and self-trapped excitons play a crucial role in the energy transfer from the matrix to the Ce$^{3+}$ ions in Ce:YAG under short-wavelength excitation. Presumably this is due to the Förster transfer at low temperatures or due to exciton hopping diffusion at room temperatures when the excitonic emission is quenched. An efficient excitation of the Ce$^{3+}$ emission from direct exciton creation with 6.7 eV photons has been demonstrated in [12]. The excitonic emission is strongly suppressed, mainly the Ce$^{3+}$ emission and to some extent defect emissions are observed at room temperature. In a simplified form, the probability of excitonic decay may be presented as a
sum of the probabilities of three processes: \( \gamma = \gamma_1 + \gamma_2 + \gamma_3 \), where \( \gamma_1 \) is the probability of radiative decay, \( \gamma_2 \) – the probability of energy transfer to a Ce\(^{3+} \) ion, \( \gamma_3 \) – the probability of energy transfer to other defects. As no excitonic emission is observed at room temperature, \( \gamma_1 << \gamma_2, \gamma_3 \). The ratio between \( \gamma_2 \) and \( \gamma_3 \) defines roughly the portion of STEs transferring energy to Ce\(^{3+} \) ions and thus \( \gamma_2 \) serves as an intensity measure of the observed Ce\(^{3+} \) emission. In the FEL experiments, excitons are created at high densities and the expression for the STE decay probability requires an additional member \( \gamma_4 \) corresponding to the probability of non-linear quenching of excitons due to the Förster dipole-dipole interaction [18,30–32]. The latter process is competitive with the process of energy transfer to Ce\(^{3+} \) centers, thus reducing its efficiency at elevated excitation densities. This allows one to use an approach developed for excitonic emission under high-density excitation (see Eq. (5) in [30]) to obtain a rough estimate of the time dependence of the number of excitons transferring energy to Ce\(^{3+} \) centers at different excitation densities. The main difference would be that the effective time of the exciton radiative transition is replaced in the latter case by the effective time of the energy transfer from an exciton to the Ce\(^{3+} \) ion \( \tau_{\text{tr}} = 1/\gamma_2 \). Integrated over time this equation gives the total intensity of the Ce\(^{3+} \) emission at a given pulse energy:

\[
I_{\text{tot}} = \int_0^\infty \frac{\sigma I_0}{\tau_\sigma} e^{-t/\tau_\sigma} \frac{-\text{Li}_1(-\xi)}{\xi} \, dt,
\]

where \( \xi = 2\pi^2 N_{\text{max}}^0 R_{\text{int}} \text{erf} \left( \sqrt{t/\tau_\sigma} \right) / 3 \) (6)

In this equation \( I_0 \) is the number of photons in a pulse, \( \sigma \) stands for the number of excitons created by one excitation photon, corrected for non-radiative losses, \( \text{Li}_1(\xi) \equiv \sum_{n=1}^\infty \frac{\xi^n}{k^2} \) is the dilogarythmic function, \( N_{\text{max}}^0 = I_0 \sigma (\pi a^2 \Lambda)^{-1} \) is the maximum of self trapped exciton density where \( a \) is a Gaussian beam radius and \( R_{\text{int}} \) is the characteristic radius of Förster interaction between excitons. This somewhat complicated equation gives a good fit to the dependence of total Ce\(^{3+} \) emission intensity on pulse energy (Fig. 5) providing support for the energy transfer model proposed. The characteristic parameter defining the distance of dipole-dipole interactions between excitons was estimated as \( R_{\text{int}} = 3 \) nm from this fit. Thus, a surprising fact that the saturation of the Ce\(^{3+} \) emission takes place at a radiance value, which is two orders of magnitude lower than the Ce\(^{3+} \) concentration, finds a plausible explanation through mutual quenching of excitons created at high densities, preceding the energy transfer to the Ce\(^{3+} \) ions.

The internal radiance coefficient \( \eta \) reported in the literature, is roughly constant over the range \( 10^7 \) to \( 10^9 \) eV and equals: \( \eta \approx 0.035 \pm 0.015 \) or \( 12 \pm 4 \) visible photons/keV, for a Ce concentration of 0.1 mol % [37]. In other words, the internal radiance depends mainly on the deposited energy dose through electron-hole pair production. The type of ionizing radiation does not play a key role here. Therefore the saturation energy density of \( c_i \approx 2 \times 10^{20} \) eV/cm\(^3\) determined in this work can be used to predict the saturation levels of Ce:YAG scintillators used for imaging of other bright beams of energetic photons and charged particles.

5. Conclusions

Monocrystalline Ce:YAG was irradiated by short-wavelength free-electron laser radiation delivered by XUV (TTF FEL), soft x-ray (FLASH) and x-ray (LCLS) sources to study the dependence of the FEL-induced luminescence signal on energy fluence. At all wavelengths, 98 nm, 25.6 nm, 13.5 nm and 0.15 nm engaged in this study, saturation of the photon yield occurs at the excitation level of \( \sim 2.0 \times 10^{20} \) eV/cm\(^3\). Such excitation leads to emission of \( 1.5 \times 10^{18} \) visible photons per cubic centimeter. The saturation energy density determined in this work can be used for predicting the saturation behavior of Ce:YAG scintillators exposed to high fluxes of energetic photons and charged particles. Saturation of the Ce\(^{3+} \) emission is explained by mutual quenching of excitons created at high densities, preceding the stage of energy transfer to the Ce\(^{3+} \) ions.
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