Excitation density effects on tungstate scintillator crystals under free electron laser FLASH radiation

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Free electron lasers (FEL) are new generation accelerator based short wavelength light sources, the properties of which (high pulse intensity, femtosecond duration, coherence, *etc.*) are superior to preceding ones enabling new kind of investigations in different scientific fields [1]. Such FEL radiation at 13.9 eV has been applied for a short study excitation density effects of a few scintillator crystals at 300K (BaF_2, $Y_3Al_5O_{12}$:Ce) using the TTF-1 facility. A remarkable shortening of the decay time and quantum yield of luminescence was observed [2]. However, technical and severe time restrictions left many of questions risen, as e.g. nature of emission centres, unanswered. Understanding the mechanisms governing the behaviour of luminescent materials (light yield and saturation effects, radiation damage, *etc.*) under high density excitation is of primary importance for screens used in visualization and diagnostics of focused XUV radiation generated by the FLASH facility and at XFEL laboratories in future. FEL is among a few sources well suited for studies of radiation matter interactions in extreme conditions.

The luminescence experiments were carried out at the FLASH facility of HASYLAB at DESY during 5 experimental shifts in May 2006 [2]. Here we report on our recent results on Ca-, Cd-, Pb-tungstate crystals (taking $CaWO_4$ as example) carried out at BL1, but other kind of insulators (pure and doped oxides, fluorides) were studied as well. The FLASH operated at 25.6 and 13.8 nm in a single bunch mode providing 25 fs pulses at 5 Hz repetition rate. The pulse energies were as high as 30 μJ (~10^{12} photons at 25.6 nm) recorded by a gas monitor detector. In addition to the inherent SASE fluctuation of FEL radiation, a N_2 filled gas absorber was used to attenuate the incident beam by up to two orders of magnitude from the maximum estimated value of ~10^{12} W/cm^2 (the size of a focal spot ~200 μm was estimated from the size of the smallest damage area observed on radiation hard oxides). Luminescence, typically averaged over 2000 FEL pulses, collected through a fiber were analysed by a SpectraPro308i spectrograph equipped with a CCD detector. Single shot decay curves were recorded through band-pass filters using a XP2020Q PMT and stored by a 6 GHz digital oscilloscope. Spectra were not corrected to the sensitivity of CCD and monochromator.

Fig. 1a shows emission spectra, typical for self-trapped molecular excitons (STE) of Frenkel type, of $CaWO_4$ at 300 K excited by fs FLASH radiation being in good agreement with earlier studies. Hence, up to the ~10^{12} W/cm^2 the nature of luminescence centre is similar to that observed under low excitation densities, even though more complex relaxation processes of electronic excitations at

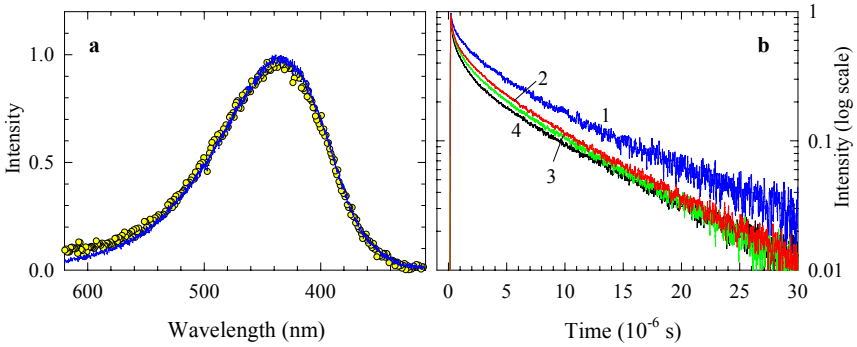
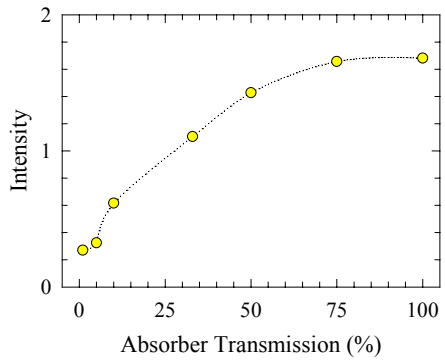


Figure 1: (a) Time integrated emission spectra of a $CaWO_4$ crystal at 300 K excited by 89.8 eV (13.8 nm) photons. Blue line represents the spectrum under maximum fluence (~8 μJ) and symbols 26 % of that. (b) Decay curves of the main emission of the same crystal at 300 K excited by 89.8 eV photons at different fluencies: 1- 16 % (blue), 2 – 26 % (red), 3- 57 % (green), 4 – 100% (black).



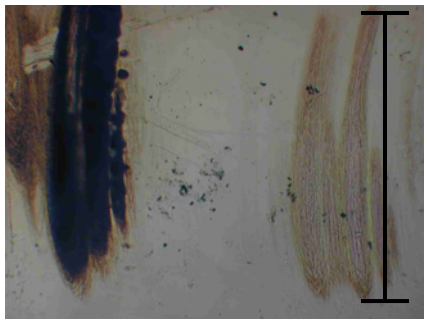


Figure 2: (left) Luminescence intensity of STE emission of $CaWO_4$ single crystal at 300 K as a function of the absorber transmission. The FEL operated at photon energy of 48.4 eV (25.6 nm) with a maximum fluence of ~30 μJ. A microphoto (right) of surface damage of a $CaWO_4$ crystal resulting from the exposure to the FEL radiation. The deeper craters are on the left and the initial erosion on right. The scale shows the length of vertical structures (~ 0.7 mm) due to the sample movement induced by thermal expansion of the cryostat in the temperature range of T=6-300 K.

high densities take place. The decay curves exhibit a strong non-exponential behaviour (see Fig. 1b) within the first ten microseconds because of mutual interaction of elementary intrinsic excitations. A dipole-dipole interaction of closely spaced self-trapped excitons is the main cause for the non-radiative decay channel observed in the initial phase. Thereafter, the decay is described by a single exponential with a characteristic lifetime of ~8.6 μs, typical for $CaWO_4$ under conventional excitation. A detailed quantitative analysis of the decays in different tungstate scintillators is a subject of forthcoming publication based on kinetic equations [3] taking into account non-uniformity of excitating radiation.

Another important issue is the saturation effects in light emission leading to the non-proportional scintillator response and possible distortion of images of fluorescent screens used in visualization of intense FEL beams. Fig. 2 demonstrates a sublinear response of a $CaWO_4$ crystal, behaviour of which was also typical for other studied samples. The 48 eV photons penetrate into the crystal deep enough diminishing the role of surface losses, which contribute significantly to the non-radiative decay of electronic excitations at lower energies. Therefore, such behaviour is resulting from the excitation density effects as discussed above The powerful FEL radiation allows the direct study of such effects in solids using photon in –photon out spectroscopy as luminescence technique is.

The post-experiment photo (Fig. 2, right) shows the irreversible surface damage of $CaWO_4$ crystal induced by the FEL beam. Presently, we cannot give precise answer how it influences scintillator performance and remains open for further studies. However, the luminescence intensity was also monitored as a function of total FEL radiation dose and also by varying incident intensities using the gas absorber. These results indicate that there is some damage depending on the radiation resistance of the material, but in the latter experiment the recovery of luminescence intensities occurred to the level below, but still comparable with the initial yield of the non-irradiated crystal. Obviously, XUV photons are absorbed deep enough in the crystal avoiding influence of surface damage and the power density of ~10^{12} W/cm^2 is still tolerable for tungstates as well as for other crystals. The first results together with short analysis will be published soon [4].

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