Self-quenching effects of excitons in $CaWO_4$ under high density XUV FEL excitation

S. Vielhauer, V. Babin, M. De Grazia[1], E. Feldbach, M. Kirm, V. Nagirnyi, and A. Vasil'ev[2]

Institute of Physics, University of Tartu, Riia 142, 51014 Tartu, Estonia

[1] CEA/DSM/DRECAM/Service des Photons, Atomes et Moleculés, CEA-SACLAY, 91 191 Gif-sur-Yvette, France

[2] Faculty of Physics, Moscow State University, Moscow, 119992, Russia

With the development of powerful FEL sources for the XUV and X-Ray region, new challenges appear also in the field of material science. The photon densities of a XUV FEL are close to the expected damage thresholds of the materials used as optical elements for the beamlines, and extensive research was directed in the last years to investigate their durability under these conditions [1, 2]. Aside from the problem of radiation-induced damage of the optical components, the performance and stability of materials used for the detection of radiation needs to be investigated.

Scintillators are already now widely used to monitor and tune the performance of the FEL at FLASH [3], and future experiments at the XFEL may rely strongly on the performance of scintillators and visualization screens under the intense X-ray excitation. In our experiments, we focused on luminescence from $CaWO_4$ crystals under FEL excitation at beamline BL1 of FLASH. Renewed interest to tungstate crystals as scintillator materials has appeared in the last years due to their radiation hardness and the occurrence of fast intrinsic emission bands. The intrinsic nature of this luminescence, which originates from self-trapped excitons (STE) located on WO_4^{2-} sites, allows the application of a model for self-quenching due to exciton-exciton-interaction [4].

For the experiments, a single crystal $CaWO_4$ sample was mounted on a liquid He cryostat inside a UHV chamber and excited with FEL pulses with a length of ~25 fs and 89.8 eV photon energy. Using the N_2 gas attenuator of the FEL and the intrinsic fluctuations of the pulse energy due to the SASE process, the pulse energies could be varied over a wide range. The maximum pulse energies were 25 μJ, which corresponds to $1.7 \cdot 10^{12}$ photons per pulse at the given photon energy. The spot size of the sample was estimated to be ~ 150 x 350 μm. Luminescence decay spectra were detected using a XP2020Q photomultiplier in combination with suitable band-pass filters to select the desired emission range. Decay curves were recorded as single shot measurements for each FEL pulse using a fast digital oscilloscope. To reduce the noise level of the recorded single-shot decay curves, decay curves were sorted according to the measured pulse intensities and averaged over 10 % intervals of the full pulse energy range.

Decay spectra of $CaWO_4$ for different averaged FEL pulse energies are shown in Fig. 1 for sample temperatures of 8 K and 300 K. In the initial part of the decay spectra, a strong non-exponential behavior is observed, whereas for decay times larger than ~ 5 ns, the decay can be approximated by a single-exponential curve with a radiative life-time τ_r comparable to results obtained with low-density excitation. The initial non-exponential decay becomes more pronounced with increasing pulse energies, indicating a direct influence of the excitation density on the radiative life-time of the STE states in the crystal.

To understand this effect in more detail, we analyzed the data using the model of bimolecular self-quenching of STE under non-uniform excitation densities, which considers energy transfer between two STEs situated at small distance due to dipole-dipole interaction [4]. As a result, one exciton recombines, and its energy is transferred to the second STE. This process can be expressed in form of a rate equation that considers also the radiative decay of the STE. Assuming a non-uniform excitation density due to absorption in the crystal and the spot size of the FEL beam, the decay characteristics of the STE luminescence under these high excitation densities can be calculated.

A comparison between fitting results using this model and the experimental decay curves is given in Fig.1. To achieve these fitting results, only three parameters were adjusted, two of them (the initial number of excitations I_0 and the radiative life-time τ_r of the STE) can be checked against experimental values. While τ_r agrees well with the experiment, the fit values for I_0 are systematically smaller by about one order of magnitude. This discrepancy may be related with additional quenching on a very fast time scale due to triple correlation of excitons. The third parameter, R_{d-d}, which describes the dipole-dipole interaction radius for the energy transfer between excitons, was estimated as 3 – 4 nm. This value corresponds well to the results obtained for $CdWO_4$ under high-order harmonic generation (HHG) laser excitation as reported in [5].

In conclusion, our experiments have shown that the photon densities achieved in the experiment lie above the threshold where excitations can be considered interaction free, and thus non-linear effects come into play when luminescence is considered. Hopefully, measurements with a more collimated beam with a reduced spot size are possible in future to test our model under more extreme conditions.

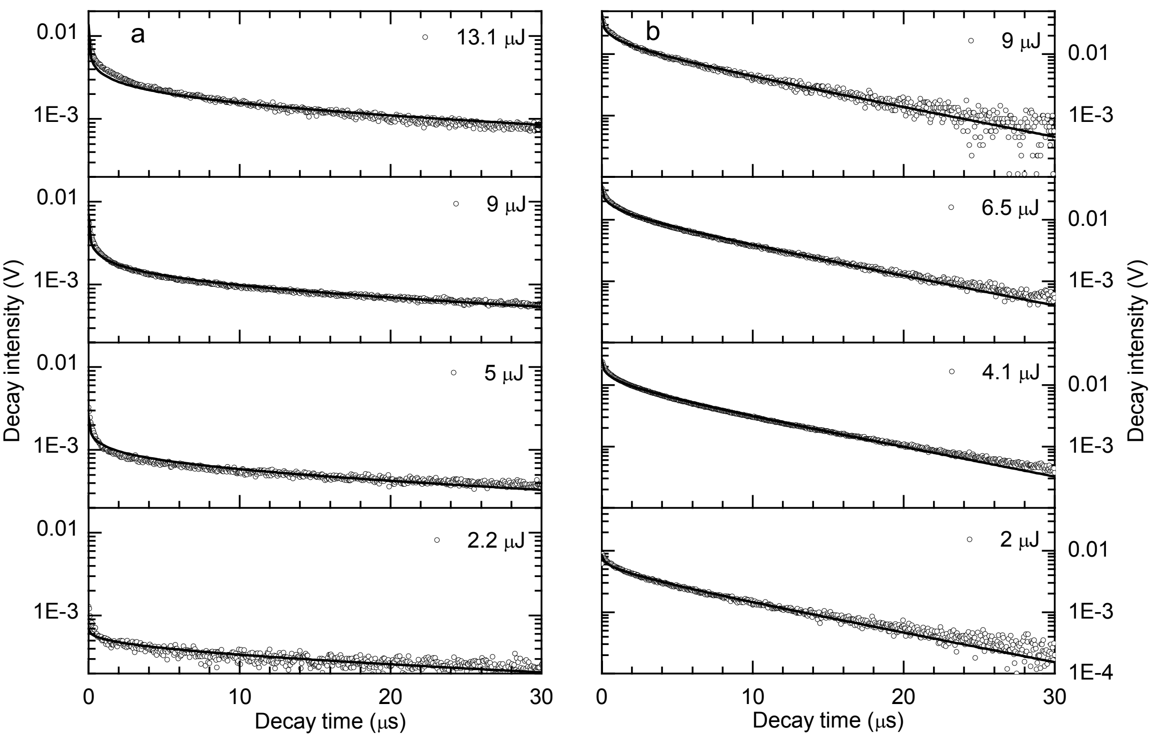

Figure 1: Measured (circles) and modeled (lines) decay curves for $CaWO_4$ at 8 K (a) and 300 K (b) under excitation with 86.8 eV FEL pulses of different pulse energies.

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