A femtosecond X-ray / optical cross-correlator: X-ray induced transient optical reflectivity in solids

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For a fundamental understanding of ultra fast dynamics in chemistry, biology and materials science it has been a longstanding dream to record a movie, where both the atomic trajectories and the chemical state of every atom in matter is followed in real time. Free-electron lasers (FEL) provide this perspective as they deliver brilliant femtosecond X-ray pulses in a wide photon energy range, which is necessary to gather element-specific and chemical-state-selective information with femtosecond time resolution. One of the key challenges lies in synchronizing the FEL with separate optical lasers. We have now established X-ray pulse induced transient optical reflectivity changes ($\Delta R/R$) in GaAs as a powerful tool for X-ray/optical cross-correlation [1], using the peak brilliance of the FEL at Hamburg (FLASH) [2]. This constitutes a breakthrough en route to a wave packet movie and equally important opens the novel field of femtosecond X-ray induced dynamics, only accessible with high brilliance X-ray free-electron lasers.
At the PG2 beamline at FLASH [3], extreme ultraviolet radiation pulses of 39.5 eV photon energy in pulses shorter than 50 fs impinged on a clean, undoped GaAs surface, depositing fluences up to 16 mJ/cm^2. The induced change in the optical reflectivity was monitored by a time-delayed optical pulse (800 nm or optionally 400 nm), measured by a fast photodiode. The optical laser (pulse length around 120 fs) was operated at twice the repetition rate of the FEL to compare the pumped and unpumped reflectivity signal. With this setup, we were able to normalize the signal directly and analyze changes in the time structure of a bunchtrain of 30 pulses, separated by 2 μs (Fig. 1 shows a schematic setup).
We observe reflectivity changes $\Delta R/R$ induced by the FEL X-ray pulse, occuring on time scales from femtoseconds to many hundreds of picoseconds. Fig. 2 shows this evolution for different delay ranges. The time scale of the initial drop of the reflectivity change is only limited by the pulse length of the optical laser. Thus the reflectivity change follows directly after the creation of Ga 3d core vacancies by the FEL pulse. Those are converted to valence excitations via Auger transitions on a femtosecond time scale. For optical excitations, often a Drude type Ansatz can adequately describe the changes in the dielectric function. For X-ray excitations, screening and electron many-body effects are important and have to taken into account by a full theoretical description.
Using a tilted beam geometry, the temporal evolution of the reflectivity change can be mapped on a spatial coordinate to cross-correlate the pulses even in a single shot measurement (see contribution by T. Maltezopoulos).
In conclusion, we have established the technique of X-ray induced transient optical reflectivity on a GaAs surface as a powerful tool for cross correlation between femtosecond optical and X-ray pulses, spanning the energy range of present and future X-ray free-electron laser sources. Furthermore, X-ray induced non-equilibrium dynamics opens a new field of time resolved studies of matter which is highly relevant for X-ray induced chemistry in biological systems, solids and interfaces e.g. present in atmospheric and interstellar dust. Thus our findings pave the way towards time resolved structural dynamics in chemistry, biology and materials science.

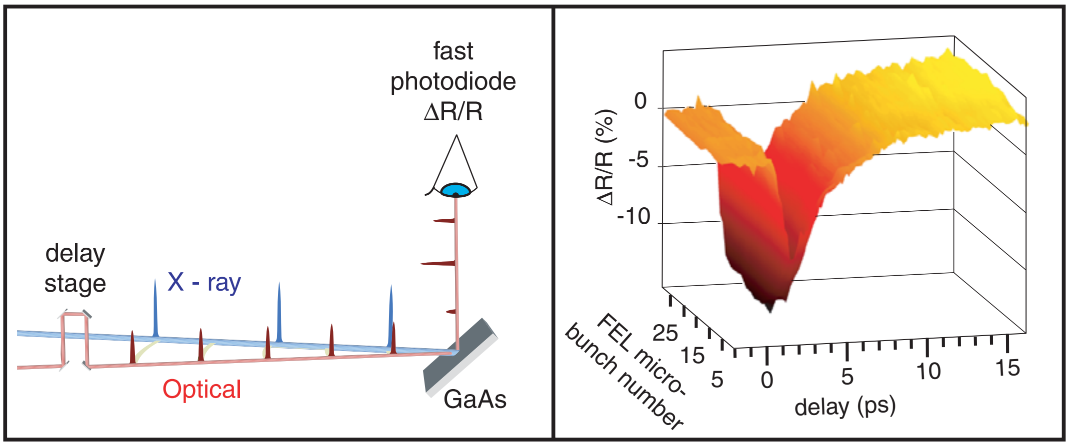

Figure 1: Transient X-ray induced optical reflectivity change ($\Delta R/R$) measurement: schematic overview and experimental data for a pulsetrain of 30 FEL radiation pulses (0.5 MHz repetition rate)

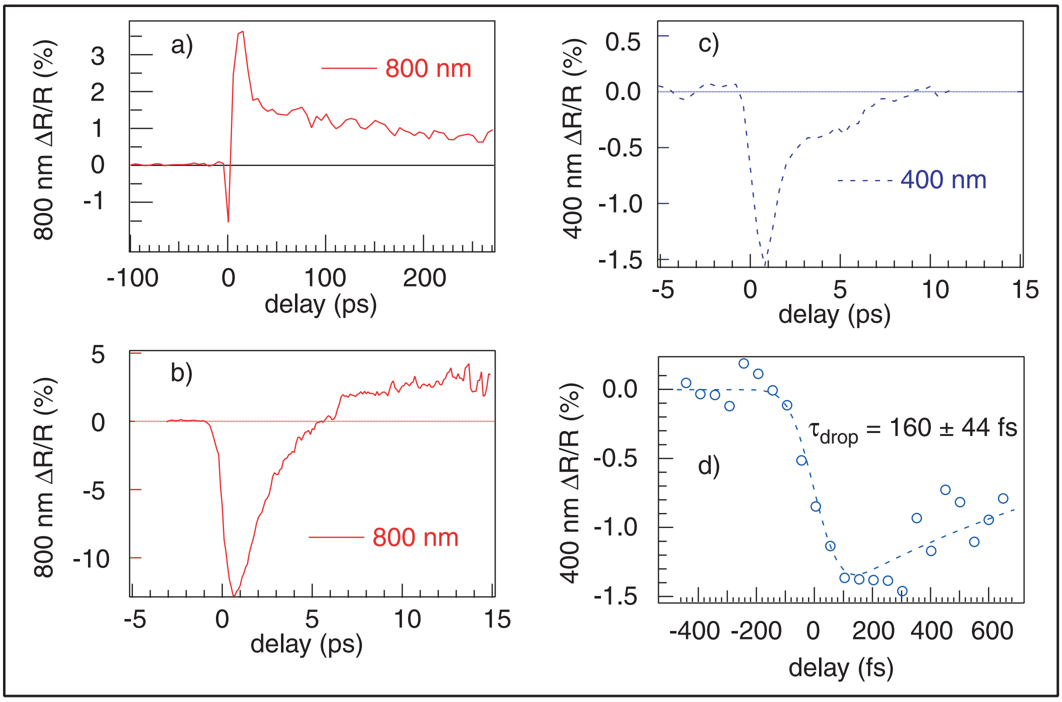

Figure 2: Temporal characteristics of the transient X-ray induced optical reflectivity change ($\Delta R/R$) on different delay range scales

This work we dedicate to our suddenly deceased co-worker Kai Starke. We thank the technical and scientific staff of Hasylab. Financial support by BMBF 05 KS4GU1/8 and 05 KS4GU1/9 and the Helmholtz Joint Research Centre ”Physics with coherent radiation sources” is gratefully acknowledged.

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