Time-resolved X-ray diffraction study of ultra-fast structural dynamics in laser-excited solids

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Abstract. Time-resolved X-ray diffraction using sub-300 fs, multi-keV x-ray pulses from a laser-produced plasma has been used to study ultrafast melting of Germanium.

1. Introduction and Experimental Methods

Ultrashort x-ray pulses offer a unique combination of *atomic-scale* spatial and temporal resolution, which permits direct measurements of structural transients on a femtosecond time scale. Using femtosecond, multi-keV x-ray pulses from a laser-produced plasma we have performed time-resolved diffraction experiments during ultrafast phase transitions in optically excited germanium.

Short bursts of Ti-K_α-radiation at 4.51 keV were produced by focusing laser pulses of about 100mJ in energy and a pulse duration of 120 fs onto the surface of a moving titanium wire. Part of the emitted radiation was collected by a toroidally bent Si (311)-crystal to form a monochromatic, 1:1 image of the plasma that was generated on the surface of the Ti-wire. Single-crystalline thin films of germanium, grown by surfactant-mediated heteroepitaxy on crystalline silicon substrates, were placed in the image plane under the appropriate Bragg-angle, and the transient changes of the x-ray diffraction after optical excitation were measured.

2. Non-Thermal Melting of Germanium

In covalent semiconductors there is extensive evidence from time-resolved optical spectroscopy [1-3] that a solid-to-liquid phase transition occurs *non-thermally* in a few hundred femtoseconds, as a result of strong optical excitation. This process represents an ideal case to be studied by time-resolved x-ray diffraction [4-6]. The data displayed in the left viewgraph of Fig. 1, obtained on a 170 nm thick, (111)-oriented Ge-film, show the angular integrated diffraction efficiency as a function of pump-probe time delay for two different excitation fluences.

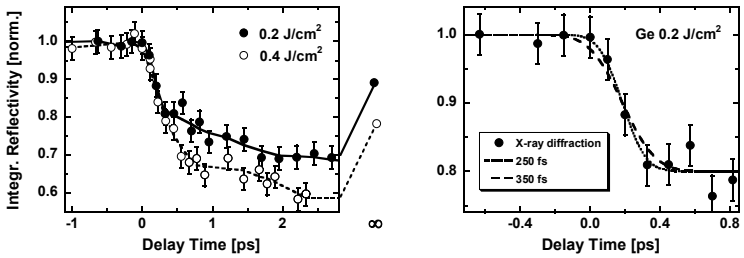

Fig. 1. Left: angular integrated X-ray reflectivity of a 170 nm Ge-film (111-reflection) as a function of time delay for two different pump fluences. Right: fits of the diffraction data (0.2 J/cm^2) assuming a step-like material response and Gaussian-shaped X-ray pulses.

For both fluences the diffraction efficiency exhibits an fast drop within a few hundred femtoseconds, followed by a more gradual decrease on a picosecond time-scale. Analysis of the data shows that a loss of order over a depth of 40 nm has occurred in less than 300 fs, a clear indication of *non-thermal* melting. Explanations of the observed decrease of diffraction as being due to plasma formation and/or ablation can be ruled out, mainly because of the nearly complete recovery of the diffraction efficiency after long times (data points "∞" in Fig. 1).

These results permit also a reliable estimate of the duration of the Ti-K_α-emission. As is demonstrated by the right part of Fig. 1, deconvolution of the data assuming an *instantaneous,* step-like material-response (which *overestimates* the X-ray pulse duration) leads to an x-ray pulse width of about (300 ± 50) fs.

3. Transient Thermo-Acoustic Effects

Additional insight into the physical processes following ultrafast melting can be obtained from an analysis of the angular dependence of diffraction, the so-called *rocking curves*. Examples for different delay times measured at a pump fluence of 0.2 J/cm^2 are displayed in Figure 2. The solid and dashed curves represent the transient rocking curves and the rocking curves of the unexcited material, respectively.

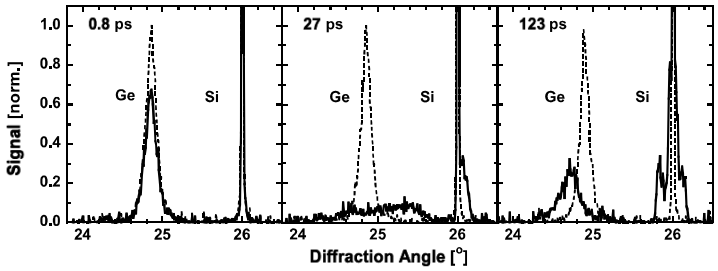

Fig. 2: Rocking curves of the laser-excited Ge/Si-herterostructure for different delay times. Pump fluence: 0.2 J/cm2, solid: transient, dashed: reference of the unpumped sample.

The x-rays, incident from the crystal mirror onto the sample, cover an angular range of about 2.5 degrees. Therefore, we are able to simultaneously measure diffraction from the Ge overlayer as well as of the Si substrate. At early time delays (0.8 ps) only a drop of the diffraction efficiency of the Ge line without any change in shape is observed, because this time-scale is too short for any volume changes to occur.

At later time delays the non-thermally, impulsively generated hot and pressurized liquid layer expands, leading to a compression of the non-molten material (Ge and Si) underneath. Such compressive strain is indicated by the appearance of diffraction contributions at larger angles with respect to the original Bragg-angles (Ge: 24.88^0; Si: 26^0). Measured maximum compressive strains of about 1.5-2 % in Ge and 0.5 % in Si correspond to transient pressures of about 2-3 GPa in Ge and 1 GPa in Si, respectively. Simple acoustic considerations allow then to roughly estimate the maximum pressure in the liquid layer at the surface to about 5-10 GPa.

At even later times (123 ps) the acoustic perturbations of the Ge layer have damped out, leaving behind the relaxed, thermally expanded material, as indicated by a shift of the Ge line to smaller angles. The measured expansive strain of 0.8-1% corresponds to a temperature of approximately 1.200 K, close to the melting temperature of Ge, suggesting that liquid and solid have come to an equilibrium near the melting point.

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