Time-resolved x-ray diffraction study of ultrafast acoustic phonon dynamics in Ge/Si-hetero-structures

K. Sokolowski-Tinten [1], A. Cavalleri[2], C.W. Siders[2,6], F.L.H. Brown[2], D.M. Leitner[2,7], C. Toth[3], M. Kammler[8], M. Horn von Hoegen[1], D. von der Linde[1], J.A. Squier[4], C.P.J. Barty[5], and K.R. Wilson[2]

[1] Institute for Laser- and Plasmaphysics, University of Essen, 45117 Essen, Germany
E-mail: kst@ilp.physik.uni-essen.de
[2] Department of Chemistry and Biochemistry, [3] Instiute for Nonlinear Science,
[4] Department of Electrical Engineering, [5] Department of Applied Mechanics and Engineering Sciences, The University of California San Diego, La Jolla, CA-92093-0339, USA
[6] School of Optics/CREOL, University of Central Florida, Orlando, FL-32816
[7] Department of Chemistry, University of Nevada, Reno, NV-89557
[8] Institute for Solid State Physics, University of Hannover, 30167 Hannover, Germany

Abstract. Using time-resolved x-ray diffraction the ultafast strain dynamics in fs-laserexcited Ge/Si-heterostructures has been studied. A fluence dependent, anharmonic damping of the impulsively generated acoustic phonons and vibrational transport across the buried Ge/Si-interface are observed.

Short pulse optical excitation of semiconductors creates highly non-equilibrium states of the irradiated material. To re-establish equilibrium a complex sequence of relaxation processes is necessary, starting with intra-band thermalization of the hot carriers, carrier relaxation via phonon-emission and recombination, vibrational acoustic transport into the bulk, and eventual complete thermalization within the phonon-subsystem. The processes involving the electronic sub-system and Raman-active optical phonons have been extensively characterized with time-resolved optical techniques. On the other hand, access to the ultrafast acoustic phonon dynamics has been indirectly achieved only at surfaces.

By combining the temporal resolution of ultrafast laser spectroscopy with the structural sensitivity of x-ray scattering, direct *quantitative* studies of ultrafast atomic motion deep inside the bulk of matter have recently become possible [1-6]. In this contribution, we wish to report on ultrafast x-ray measurements of strain oscillations in fs-laserexcited Ge/Si-heterostructures. Excitation-dependent damping times and vibrational transport across a buried interface are measured simultaneously, thereby revealing different acoustic phonon decay mechanisms.

Single crystalline Ge thin films with 111-surface orientation, grown on 111-oriented Si substrates by surfactant-mediated heteroepitaxy [7], have been optically excited with 30fs laser pulses at 800nm. Evolution of the impulsively generated transient lattice strain is observed by diffraction of short bursts of Cu K_α−line radiation at 8 keV from a laser-produced microplasma in a symmetric Bragg-configuration. Due to the different lattice constants of the two diamond-like materials the Cu K_α-radiation is diffracted at two distinct Bragg-angles. Therefore, we could separate diffraction from the film and the substrate and measure the strain dynamics in *both* components of the heterostructure .

In contrast to bulk crystalline samples [3], where a splitting of the Bragg-line is observed, the rocking curve of the thin film is shifted as a whole towards smaller diffraction angles, without changes of its shape. This indicates homogenous expansion of the film over its entire thickness. In Fig. 1 the time-dependent shift of the centroid of the rocking curve of the Ge-film (left) and the Si-substrate (right) are plotted for four different excitation fluences (15-40mJ/cm^2).

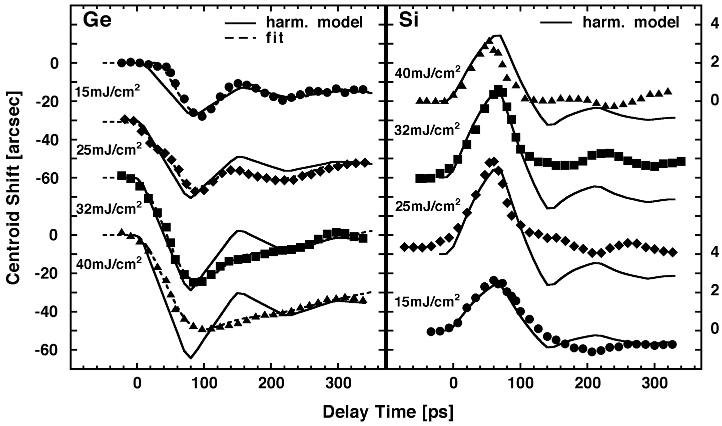

Fig. 1. Time dependent shift of the centroid of the measured rocking curves in the Ge-overlayer (left) and in the Si-substrate (right) for different fluences. Solid lines: harmonic model; dashed lines: anharmonic fit.

From maximum *negative* shifts of the Bragg-angle in the Ge-film between 25-50arcsec, values for the peak strain of 0.05-0.1%, corresponding to an increase in the lattice spacing of about 150-300fm, are obtained. Maximum expansion is reached 80-100ps after excitation, followed by damped oscillations at later times, indicating periodic expansion and compression of the film. Damping of these oscillations is apparently fluence dependent, becoming stronger for higher excitation. In the Si-substrate mainly *positive* line shifts, indicating compression of the material, are observed. The peak of the measured compression corresponds to a 20-40fm change in the average lattice spacing (a few nuclear diameters!).

Interpretation of the experimental data proceeds along the following lines. Irradiation of the sample leads to the generation of a dense e-h-plasma ($N \approx 10^{21}cm^{-3}$) in the Ge-overlayer, but negligible photo-excitation of the Si-substrate. Before complete energy relaxation between the electrons and the lattice can take place (via direct electron-phonon scattering and delayed Auger-heating), fast carrier diffusion [8] distributes the energy over the entire thickness of the Ge-film. X-ray measurements with samples of different film-thickness and on a bulk Ge-crystal further confirm the importance of fast, diffusion-mediated heat transfer in Ge. On the other hand, electronic energy transport into the Si-substrate is inhibited due to the 0.43eV difference of the energy gap in both materials.

Impulsive and homogenous heating produces an initially highly stressed state of the Ge-film. Acoustic relaxation of the stress leads then to vibration of the film with a period of $2d/c_L \approx 140$ps (d=400nm±20nm: film thickness; c_L=5560m/s: sound velocity). In harmonic approximation, damping of the film vibrations and the compression of the Si-substrate can be attributed to acoustic transmission across the Ge/Si-interface. However, in a real crystal other processes, (defect-, surface- and phonon-phonon-scattering), lead to additional contributions to the decay of the oscillations. While damping due to acoustic transmission and defect- and surface-mediated scattering should be independent of the degree of excitation, the anharmonic phonon-phonon interaction scales with the population of the individual modes and thus with temperature. Our data allow us to distinguish between these mechanisms: the observed fluence dependent damping is clear indication of lattice anharmonicity.

For a quantitative estimate we first compare the data to a fully harmonic description. The initial stress distribution was obtained from a numerical solution of the coupled transport equations for the e-h-plasma and the lattice taking into account Auger recombination and carrier diffusion. The one-dimensional elastic equation was then used to calculate the time-dependent strain in the two-layer system. Finally, dynamic diffraction theory yields the transient x-ray diffraction patterns. The solid curves in Fig. 1 represent the results of these calculations. As expected, the model predicts a fluence-independent damping, originating solely from acoustic transmission across the Ge/Si-interface. For the lowest fluence the calculated curves follow the measured data very closely, but with increasing fluences the harmonic calculations progressively deviate from the measurement, as would be expected for an anharmonic, temperature dependent damping process. To describe lattice anharmonicity we fit the measured Ge-data to a phenomenological response function (dashed curves), which includes the fluence-independent damping due to acoustic transmission and an additional fluence-dependent contribution. The obtained anharmonic damping times range from 500ps at low fluences/temperatures down to 50-70ps at high fluences/temperatures and are consistent with estimated four-body, elastic dephasing times (T_2) for 7-GHz LA-phonons in Ge [9]

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