ESRF	**Experiment title:** Ultrafast excitation of coherent phonons in embedded nanocrystals	**Experiment number**: HS1423
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Report:

Nanoscopic metal particles can be excited selectively via plasmon bands in the optical spectrum. From transient optical spectroscopy it has been shown, that due to electron-phonon interaction tha particles can be haeted up and vibrational modes with frequencies of several tens of picoseconds can be excited.

In a first experiment at ID09 (august 2000) we were able to quantify the expansion of Ag particles embedded in a glass matrix after femtosecond laser excitation (amplitude and relaxation time). It was found that the amplitude of the thermal expansion was less than what has been calculated from optical spectroscopy. In addition elongated particles displayed a characteristic anisotropy, that can be understood in the framework of matrix damping.

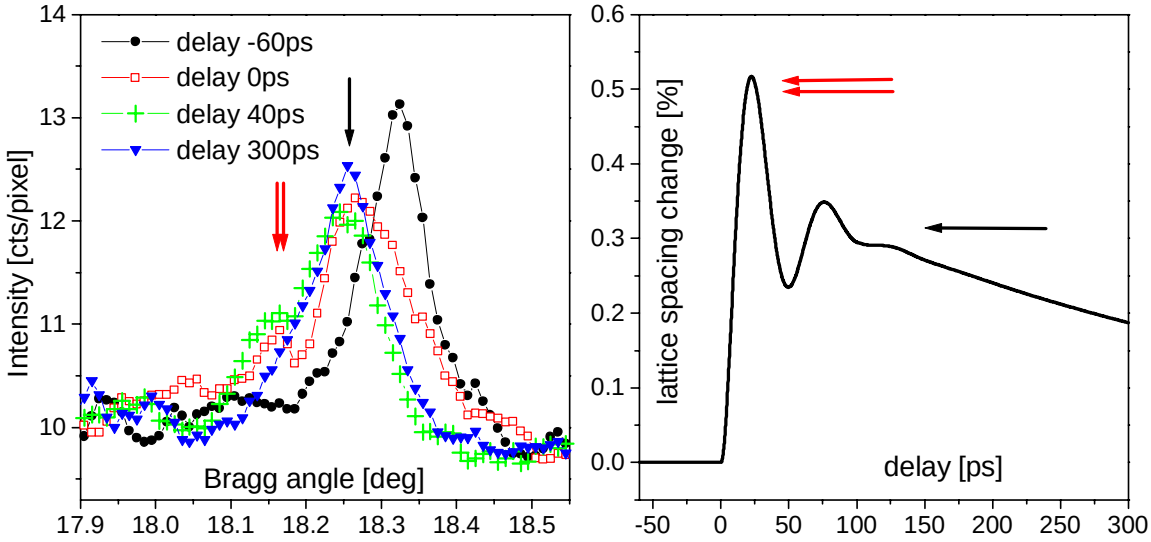
This second experiment was devoted to the determination of the elastic limit of thermal expansion in the view of results from nonreversible plastic deformations of particles at high laser power as well as the relation of nonthermal (vibrational) to thermal motion in the very first steps of structure reaction.

As the oscillation period of the 20 – 100 nm sized particles is smaller than the pulse length of individual x-ray pulses at ESRF in 16-bunch mode (80 – 120ps), we were limited in the tracking of the lattice parameter within the coherent motion of the particle. Therefore we used used a technique called fine slicing to improve the time resolution. The delay in between the laser excitation and x-ray probe can be fine tuned to an accuracy of much better than the x-ray pulse length. If data at delays smaller than the pulse length are taken it is possible to derive transients that are shorter than the pulse length. In addition the position of the Debye-Scherrer ring on the detector is separated for the different states of lattice expansion, so that the line profile can be fitted to models that average over several periods of the oscillation.

Compared to the first experiment in 2000 it was possible to take larger and more reliable datasets of delays in between laser and x-ray beam. This is due to:

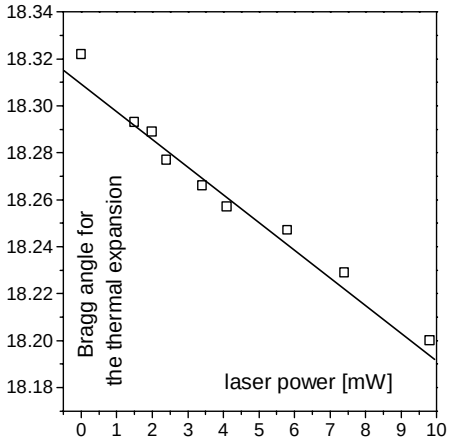
- An improved stability of the optical part of the beamline (monochromator before the toroidal mirror reduced thermal drifts)
- An improved sample quality (better crystallinity, a dispersion of sizes on one wafer allows the study of sizes without realigning samples)
- Several experimental details that allow better control of the excitation (automated laser power adjustment, coaxial microscope for better footprint control)

From analysing the transient profiles of the Debye-Scherrer rings within the first 100ps of excitation we could for the first time prove a nonthermal motion of the lattice. It can be seen from fig. 1, that a second maximum of lattice spacing probability (double arrow) appears at the low angle side of the (already shifted) main peak (single arrow) within the first 40 ps. This can be explained by a relatively large contribution of the average lattice spacing at the turning point of the oscillation (simulated curve from optical data).



By proper fitting we will be able to improve the simulations based on transient optical absorption experiments. This will help to quantify the coupling parameters of the colloid particles to the matrix.

Data showing the elastic response with increasing laser power are still to be further analysed, but already show the large range of linear response of the particles, as indicated in fig. 2. We see that a broadening occurs, that smears out the nonthermal part with the thermal expansion, which complicates the resolution of nonthermal motion. Any further experiment will demand for the use of better time resolution (streak camera).



Additionally the present ultrafast system allows to check the performance of the time resolved mode at ID09.
It was still uncertain for the ultrafast operation to what extent the time resolution is compromised by unevitable jitters of the delay electronics and the uncertainty of the determination of time zero (combination of GaAs photoconductor + 3 GHz oscilloscope, peak width 150ps). From our fine sliced data we could prove, that the timing stability is reliable within at least 30ps, much better than the pulse length, but maybe still critical for fine slicing methods. Time zero (laser incident after 50% of the x-ray intensity) can be defined to better than 50ps (at that level already phase shifts of the x-ray pulses relative to the high frequency clock available at ID09 matter). Nevertheless we found indications of small delay drifts in the order of 100ps/day with continous operation of the phasing electronics.