ESRF	**Experiment title:** Real-time studies of picosecond particle vibrations	**Experiment number**: HS 1729
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Shifts:	**Local contact(s)**: Dr. M. Wulff, Dr. K. Scheidt , Dr. G. Naylor	*Received at ESRF:*

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Report:

The aim of the present experiment was to explore the picosecond lattice and shape changes of metal nanoparticles exposed to femtosecond laser pulses. It is well known that internal energy redistribution from the excited free electron gas in metal particles can lead to coherent lattice excitations. These lead to oscillatory changes in the shape of highly symmetrical nanoparticles, oscillation which were deduced from laser spectroscopic analysis. Up to now there is no direct proof of the lattice vibrations by scattering techniques.
We have performed in the past pump-probe experiments with an ID09 laser pump and the x-ray probe pulses emitted by the synchrotron in a way to follow the lattice kinetics on a 100ps time scale (i.e. the bunch length of the synchrotron).
Hereby the lattice heating and thermal relaxation can be precisely followed, which enables to set up the energy balance and characteristic time scales of phonon-phonon interactions. The coherent lattice motion nevertheless has characteristic periods from 5 to 50 picoseconds, a temporal resolution that the conventionnal ID09 set-up doesn’t allow. Therefore an x-ray streak camera, installed and prepared under the supervision of Keith Scheidt and Graham Naylor, was used to improve the time resolution in the picosecond scale.
This instrument meanwhile has a quite low detection efficiency for high energy x-ray photons. This is in particular important for nanoparticle assemblies with filling factors of 10^{-4} with respect to the surrounding matrix or monolayered particles on surfaces. In addition, the efficient x-ray flux in the pump-probe experiment accounts for only 1/400 of the primary flux in single bunch mode or 1/ 6000 in 16-bunch mode, this low efficiency is due to the low repetition frequency of the experiment.
The only way to get enough scattering signal was to use the scattering from high coverage of gold colloids adsorbed on a silicon wafer. We used the (111) reflection of the powder scattering pattern (it is important to remember that the colloids are disordered on the surface) guided through the streak camera aperture. Despite of these improvements in the signal, the monochromatic x-ray flux was still to weak to be detected by the

improved the detected flux considerably. In contrast this method had the disadvantage of a reduced resolution due to energy spread and of course less sensitivity to lattice changes.

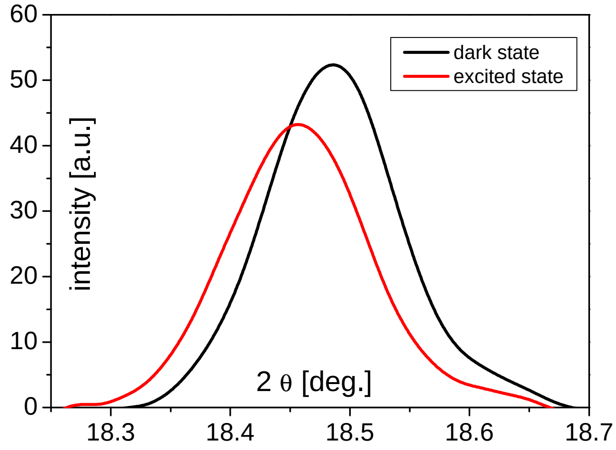

Fig. 1. *Debye Scherrer profiles as derived from conventional pump-probe scattering, where the excited state from 60nm gold particles is characterizd by a lattice expansion of about 0.15 %. In scattering with polychromatic light of 3 keV bandwidth at 15.6 keV peak this profile is smeared out. The working point of the streak camera is at the rising edge of the dark state profile, at about 18.45 degrees for highest sensitivity to lattice changes.*

We were able to detect the trace of the particle scattering in the streak camera. In order to overcome the noise of the setup (image intensifier for photoelectrons and CCD camera at 0°C) we had to accumulate hours for one frame. The resulting time traces of detected x-ray photons are displayed in figure 2.

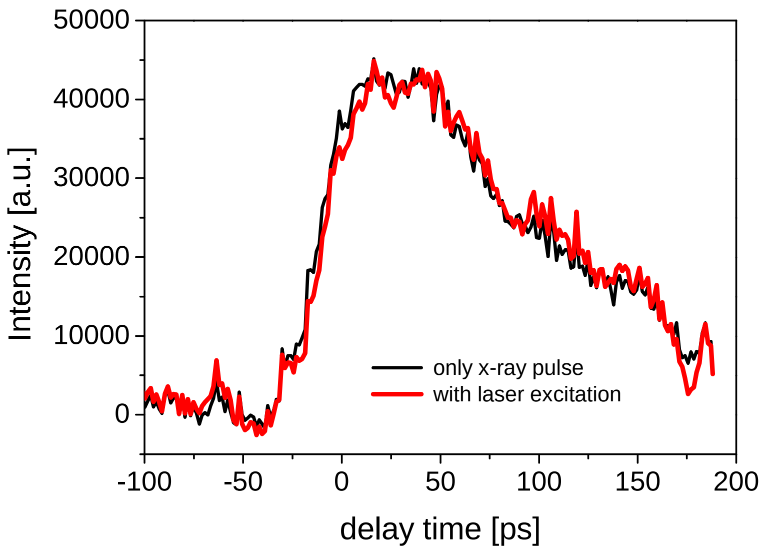

Fig 2: Traces of the scattering from 60nm gold particles supported on a silicon wafer as function of time. There is a slight change in scattering after the laser impact on the particles (delay time 0), which may reflect the lattice disturbance.

Unfortunately no clear indication of coherent motion of the lattice is observed. The measured time resolution of the setup was 3-4 ps, which should be sufficient to follow lattice oscillations with periods of 18 ps. Clearly the signal-to noise ratio limits the resolution, keeping in mind that due to the polychromatic x-ray beam the contrast ratio of excited to dark state is considerably reduced.

The prospect of this studies will critically depend on finding an optimized trade-off in between time-resolution and detection efficiency of the streak camera. It is as well desirable to improve the noise level on the CCD side to allow detection ot thinner details. Samples will have to be optimized for higher scattering cross section as well as favorable scattering geometry.