ESRF	**Experiment title:** Time resolved surface plasmon and phonon interactions in nanostructured crystals	**Experiment number**: HS1257
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Report:

The aim of the present experiment was to explore the dynamic behavior of silver nanoparticles in a glass matrix. Optical pump and probe experiments show specific transient transmission change as a function of time. Several steps appear, if the laser pump wavelength is tuned to the surface plasmon band of the colloids.
Fast processes like plasmon damping and electron-electron interactions lead to absorption changes with decay times of several picoseconds. The following relaxation is governed by electron phonon scattering and can display a prominent oscillation within tens of picoseconds followed by a decay up to nanoseconds. This is attributed to structural change of the lattice, namely coherent lattice vibrations and relaxation of the heated lattice to its nominal value due to energy dissipation into the matrix [1].
The instrument for time resolved studies at beamline ID09 has the unique opportunities to study the structural change on a time scale of tens of picoseconds, which is ideally suited for the present studies. We employed a stroboscopic optical pump and x-ray probe experiment to follow the lattice scattering on this time scale.
As detector the MarCCD was used to collect the complete Debye-Scherrer ring from the nanocrystals and providing an excellent resolution. As samples we used spherical and ellipsoidal silver nanoparticles, that can be produced in a very defined and monodispersive way by ion exchange and annealing procedures.
We found that the resolution of the set-up was sufficient to resolve the broadening of the (111) Bragg reflection due to the finite particle size (see fig. 1). The most prominent effect is to be seen with particles of 24 nm diameter with respect to particles of 100 nm size.
The laser energy was set to 400nm slightly off the plasmon resonance. By changing the laser flux, we could determine the elastic limit for the nanoparticle vibration and resolve the transient structure change of the lattice down to a time scale of 50 ps.

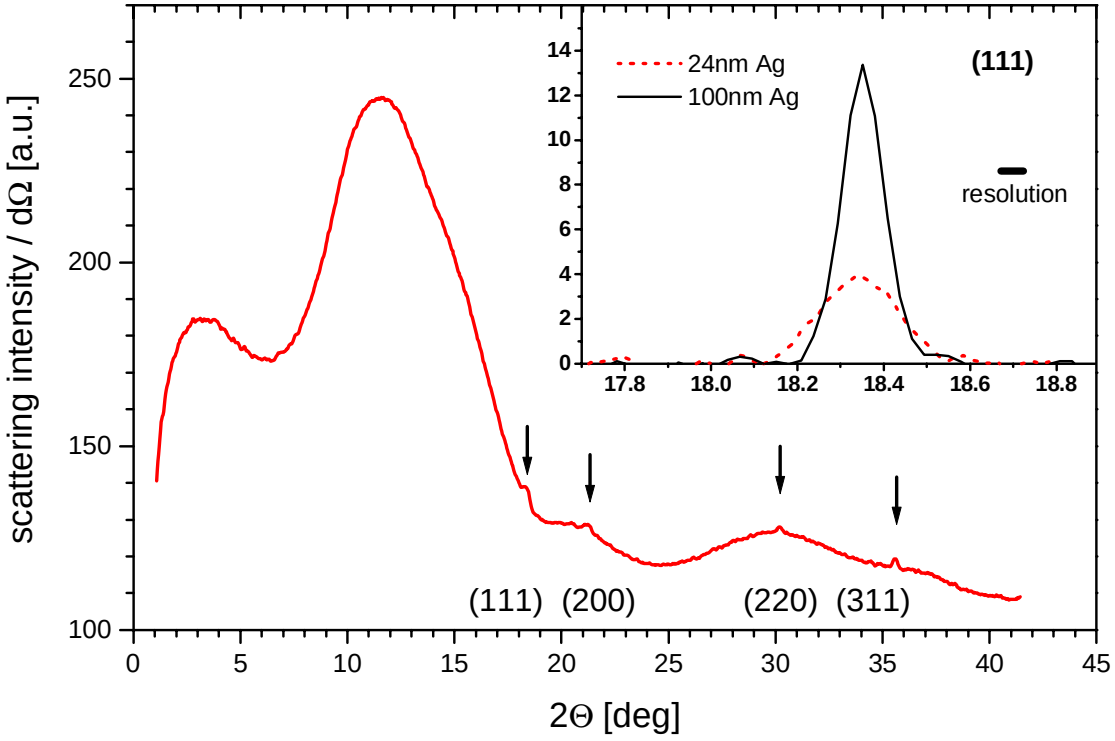
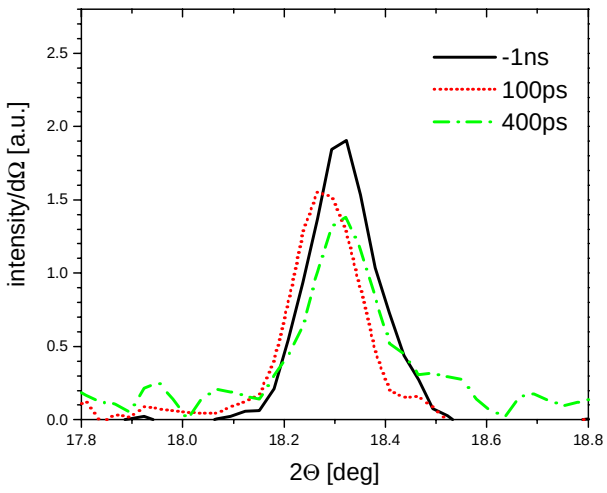


Fig1: Radial scattering distribution on the CCD camera after angular integration, space angle correction and polarization correction. The small peaks are induced by the scattering of the silver particles. The inset shows the reflection profile after substraction of the glass scattering. The width corresponds to crystalline sizes of 21 nm, resp. 49 nm for the nanocrystals.

The Debye-Scherrer rings were collected with single x-ray pulse selection with a defined time after the laser impact on the sample. There is a clear change of the reflection from the nanocrystals with this delay. Changes in position and profile are visible.



In fig. 2 one can see that change for samples with 60nm diameter as a function of time. For t=-1ns the laser is incident after the x-ray pulse, so that this scattering corresponds to the static structure without laser excitation. At t=100ps the x-ray scattering probes the structure after the laser pulse and a clear shift of the peak is visible indicating an expanded lattice. After 400ps this shift has almost disappeared indicating a fast relaxation time. But still the intensity and the profile have not recovered. The relaxation could be followed for a whole time series from 0 to 4ns.

This first analysis of the data already shows, that coherent vibrations of nanocrystals excited by a femtosecond laser pulse can be proved by ultrafast time resolved x-ray scattering techniques. The is still in progress and detailed structural parameters in time and space will have an high impact on the simulation on the optical and structural properties of these new nonlinear materials. Also a deeper insight in relaxation processes and nonelastic behavior is achievable. Further experiments are planned in close cooperation between the different aspects as sample preparation, optical response and structural characterization.

[1] M. Perner, S. Gresillon, J. Maerz, G. von Plessen and J. Feldmann, J. Porstendorfer, K.-J. Berg, and G. Berg; Observation of hot-electron pressure in the vibration dynamics of metal nanoparticles; Phys. Rev. Lett. **85** 792 (2000).