ESRF	**Experiment title:** Ultrafast melting in metal nanostructures	**Experiment number**: HS 1918
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Report:

The aim of the present experiment was to record lattice changes in nanoparticles that are exposed to intense femtosecond laser pulses. Metal nanoparticles strongly absorb energy via plasmon resonance and interband transitions. It is known that the lattice temperature of particles in the size range of few nanometers to hundreds of nanometers can increase drastically within several picoseconds after irradiation [1]. This energy increase can be sufficient to drive the particle above the melting transition and produce permanent or transient structure changes on short time scales. The transformation of the lattice can in principle be investigated by ultrafast laser and x-ray pump probe scattering.
The critical problem with reversible excitations lies in the fact that nanoparticle melting leads to irreversible structure changes, so that a true single shot technique is required. This implies that each particle can be subjected only to one single laser pulse of 200 fs length and would be probed by one x-ray pulse at a fixed time delay. Given that the study of lattice relaxation times in dilute silver and gold particle assemblies required up to 100 000 shots for one single time delay [2], a new procedure had to be implemented.

The self-assembly of charged gold nanoparticles from solution is known to result in well defined monolayer coverage on eg. silicon substrates [3]. The favorable scattering condition in reflection geometry, together with the availability of large covered surface areas allow to acquire enough shots in order to obtain Debye Scherrer rings.
We performed experiments on gold particles between 100nm and 150nm in size, excited with λ = 400nm wavelength pulses at P = 250mW maximum power. From the elastic lattice expansion we estimate that this energy is in the present geometry just sufficient to achieve temperature rises up to the melting point of bulk gold. Thus the melting is supposed to occur via phonon thermalization, which is determined by the phonon-phonon scattering time of a few tens of picoseconds.

In Fig. 1. typical Debye Scherrer ring profiles are shown in the case where the laser arrives at the sample after the x-ray probe pulse (at $\Delta\tau$ = -1ns, profiles also called ‘dark states’) in comparison with a situation where the laser arrives at $\Delta\tau$ = 100ps before the x-ray pulse (‘excited states’). The excited state (circles) is always shown in comparison with the dark state (crosses) for a better visibility of the irradiation effects.

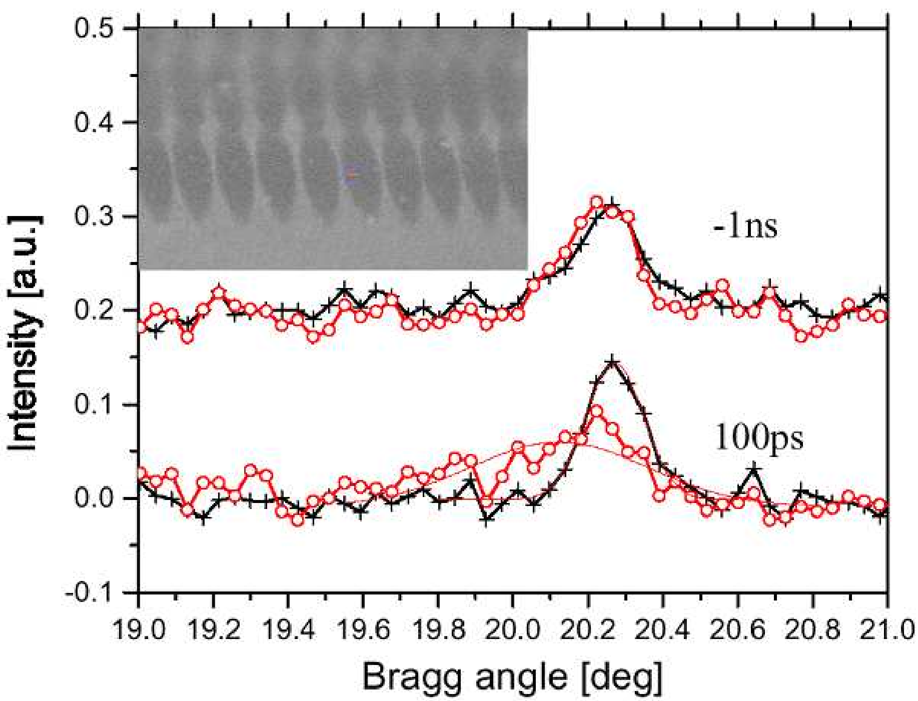

Fig. 1*: Profiles of the Debye Scherrer rings from gold nanoparticles exposed to intense femtosecond laser pulses. The excited state for a 100ps positive time delay between laser pump and x-ray probe is compared to the dark where the laser pump pulse arrives after the x-ray probe pulse. The inset shows typical irradiated areas which forms elliptically formed shapes due to the grazing incidence geometry.*

After the 100 ps delay there is still a signal from crystalline order but with a strongly expanded lattice parameter. We then observed that the Debye-Scherrer ring can completely disappear, which indicates an evident loss of crystalline order. Analysis in progress of the final structure and the lattice temperature will reveal the nature of the disordering process.

[1] M. Kaempfe, T. Rainer, K.-J. Berg, G. Seifert, H. Graener: Ultrashort laser pulse induced deformation of silver nanoparticles in glass; *Appl. Phys. Lett.* **74**, 1200 (1999)

[2] A. Plech, M. Wulff, S. Kuerbitz, K.-J. Berg, G. Berg, H. Graener, S. Grésillon, M. Kaempfe, J. Feldmann and G. von Plessen: Time-resolved X-ray diffraction on laser excited metal nanoparticles, *Europhys. Lett.* **61(6)** xx (2003).

[3] A. Plech and T. Salditt: Structure of self-assembled thin films and polyelectrolyte composites studied by surface sensitive scattering techniques, in: *Handbook of Polyelectrolytes and their Applications*: ed. H. S. Nalwa, S. Tripathy, J. Kumar, American Scientific Publishers (2002); Vol 1: Polyelectrolyte-Based Multilayers, Self-Assemblies and Nanostructures.