ESRF	**Experiment title:** Ultrafast melting of metal nanoparticles	**Experiment number**: HS 2192
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Report:

The aim of the present experiment was to induce the melting transition in metal nanoparticles by femtosecond laser excitation and observe the lattice dynamics via time-resolved x-ray scattering. As we have previously observed, single shot laser excitation might damage the particles even at relatively low powers and therefore stroboscopic experiments can not give full account for the time resolved irreversible changes in nanoparticles.
In consequence true single shot experiments have to be performed. The scattering power of these samples is very low so that at least 1000 shots have to be accumulated in order to resolve the powder scattering of the particles. This can be done by scanning a nanoparticle covered surface stepwise (see HS1918) or by exciting nanoparticle sols that are pumped continuously through a capillary. We used a large amount of gold particles in the range of 40 to 120 nm diameter that have been synthesized from a 0.9 mM solution of gold ions by the well known citrate technique. The (111) Debye Scherrer ring has been recorded at different femtosecond laser excitation levels as well as for different delays in between laser and x-ray pulses. This allows to resolve the lattice temperature and the thermodynamic state of the particles.

Fig. 1 displays the radially integrated profiles for the set of parameters. It can be seen clearly that at a relatively low power the peak shifts to lower angles indicating lattice heating, while relaxing towards the cold state within the nanosecond range [1]. At the doubled power the peak vanishes within the time resolution of 100ps giving a clear proof for ultrafast particles melting. At thermal equilibrium 1 µs after excitation the reflection has almost completely recovered. It is interesting to note that the peak reappears only with a low lattice expansion. This can be interpreted as a nucleation limited freezing transition. The crystal lattice does not reconstitute immediately after the particle temperature has crossed the melting point through environmental cooling, but several nanoseconds of incubation time are needed.

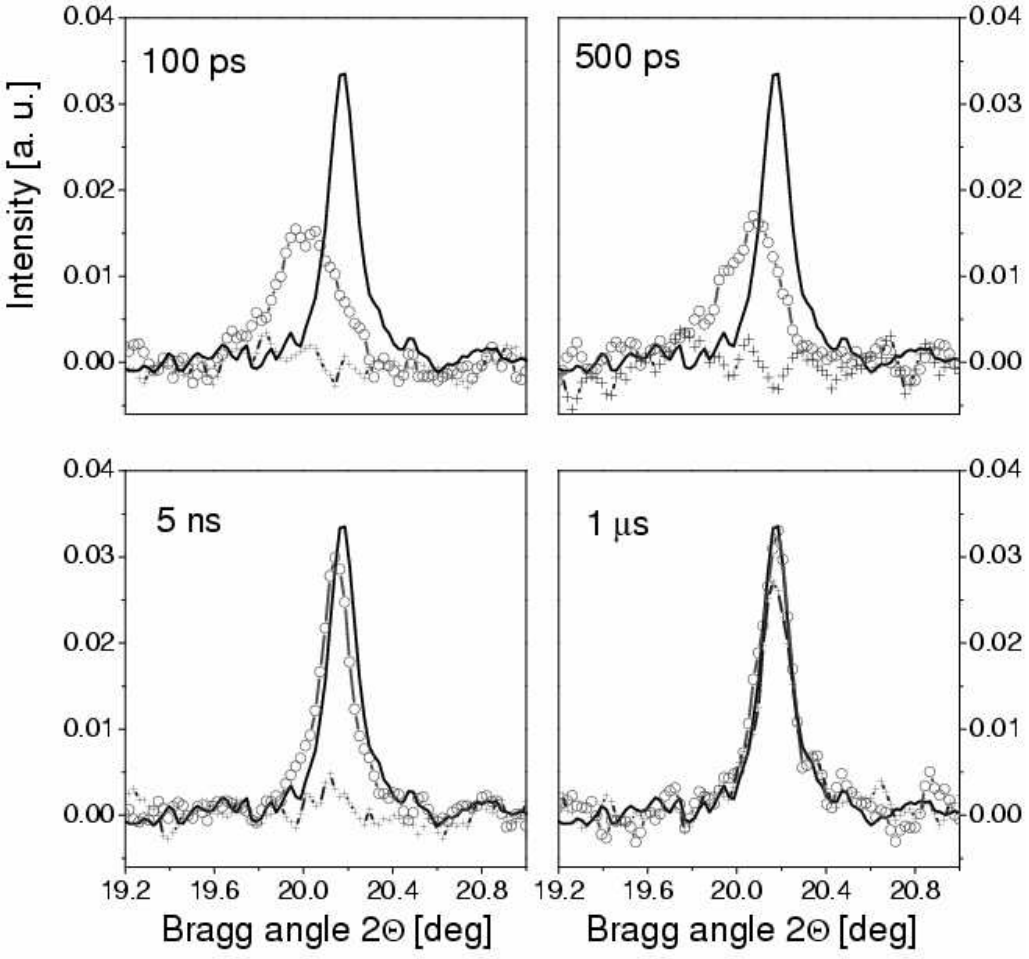

Fig. 1*:Profiles of the (111) Debye Scherrer reflections from crystalline gold nanoparticles of 100 nm diameter at a set of time delays in between laser pulse and x-ray probe pulse. The black lines mark the non-excited state, whereas the red circles are characterized by a laser power corresponding to more than 500 K lattice heating. For the blue crosses the power has been doubled thus crossing the melting transition.*

This experiment prooves the melting transition in gold nanoparticles induced by femtosecond laser excitation in contrary to an earlier optical study where no indications for melting were found at even higher energies [2]. We hope to further investigate the melting dynamics in nanoparticles by this method.

[1] A. Plech, V. Kotaidis, S. Grésillon, C. Dahmen and G. von Plessen, submitted.

[2] Gregory V. Hartland, Min Hu and John E. Sader, J. Phys. Chem. B **107** (2003) 7472.