



<b>Experiment title:</b> Impulsive nanoparticle melting - Surface melting versus ablation	<b>Experiment number:</b> SI1152	
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<b>Shifts:</b> 12	<b>Local contact(s):</b> Dr. Michael Wulff	<i>Received at ESRF:</i>
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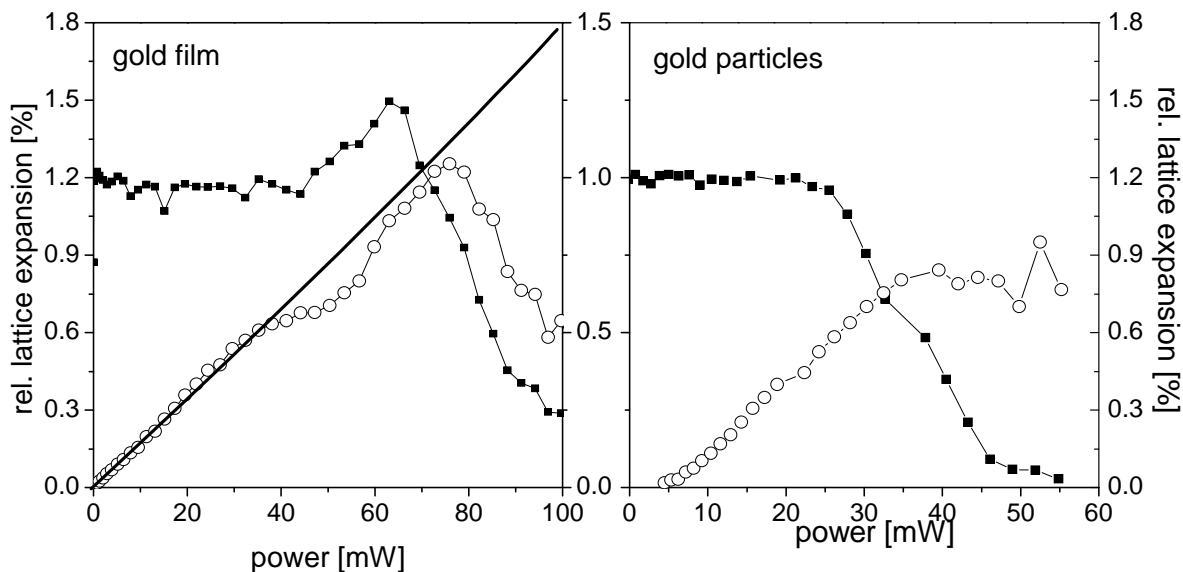
## Report:

The aim of the present experiment was to record lattice changes in nanoparticles, that are exposed to intense femtosecond laser pulses. The questions are driven by the comparison of the thermal dynamics of excited gold particles in a water suspension to quasi free adsorbed particles, that only have contact to a substrate[1-6].

Gold particles in water display lattice expansion immediately after laser excitation (100ps), that can reach the lattice parameter of the bulk melting transition [1]. However structural relaxations of the water phase and the particles are observed before the melting transition. The water can evaporatively boil and form transiently nanobubbles [2,3] and the particles display as well nonreversible transformations. This is manifested by ablation from the particles and size reduction [4]. Free particles on a surface show thermal dynamics, which are distinctly different from the embedded or suspended particles [5]. Firstly, the cooling kinetics are considerably longer due to the reduced contact area to the substrate. Secondly the lattice expansion before particle destruction is considerably lower as in the embedded case. While in past experiments mostly stroboscopic excitation of the same particles was recorded, within this experiment we tried to reveal the threshold for particle destruction both with multiple shot excitation (ME) and single shot excitation (SE).

For the SE a fast translation stage was built to replace the illuminated surface region within two laser shots (1 kHz), which worked successfully. Indeed it was found, that while for ME the threshold of lattice deterioration was at about 0.8 % expansion (equivalent to a lattice temperature rise of 500 K), for the single case an expansion up to 1.6 % (melting transition at 1.8 %) was observed for the present gold particles of 64 nm diameter. The same is true for continuous gold films of 30nm on glass substrate. The film showed expansion up to the melting point value (and slightly above due to relaxation of strain) for the SE case. This shows that the particle destruction process is an additive reaction, whereby a small fraction of the particle mass is ablated for each laser shot. This accumulates to a complete particle destruction in the ME case. While the SE experiment is important to determine the elastic and plastic behaviour of the particles, the ME experiment is very sensitive on the exact threshold of the ablation process. In fig. 1 the data from the ME experiment is compared for the film and the particles. The higher expansion for the film is clearly visible, showing the relative stability against ablation to higher

temperatures. In spite there are modifications to the theoretical expansion visible starting above 40 mW of laser power, which may indicate the start for relaxation mechanisms such as surface melting of grain boundary movement within the film. This is visible by the increase of the (111) integrated peak intensity. For the particles no such effect occurs and the peak intensity disappears at very low excitation density.



**Fig. 1:** Change of the lattice parameter of a gold film of 30nm thickness (left) compared to 64 nm gold particles adsorbed on a glass surface after excitation with 400nm, 100fs multiple laser pulses at a laser x-ray delay of 60ps together with the integrated peak intensity of the (111) reflection of gold. The expansion (open symbols) is almost linear at very low laser power, which is expected for simple heating of the gold lattice by the dissipated energy. Above a threshold in the range of 40 to 60 mW the expansion deviates from the theoretical calculation, which indicates nonreversible processes. At the same time the peak intensity (full symbols) decreases, which shows the disappearance of crystalline lattice ordering. The full line is the expectation, if the laser energy is completely converted to heat and thus thermal expansion.

By analysing the full time dependence of the reaction and the scattering patterns in view of peak profiles and relative orientation to the surface we hope to clarify the irreversible reaction pathway, which should be important for a number of applications in materials processing by laser excitation.

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