Structural modifications in laser-annealed PdSi alloys characterized by total scattering measurements

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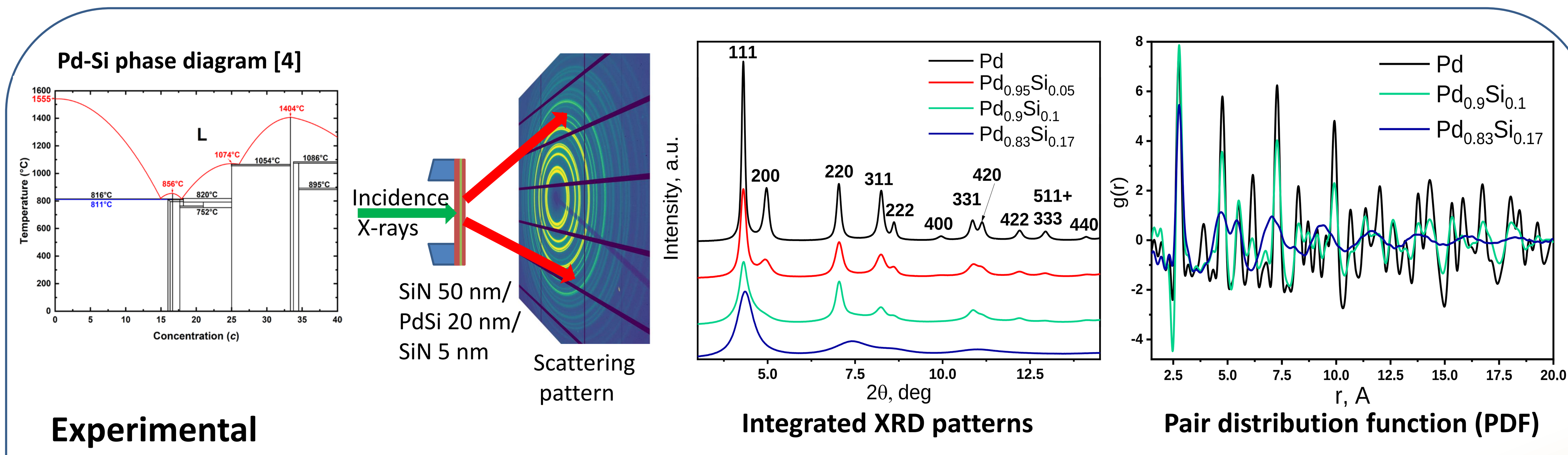
Motivation

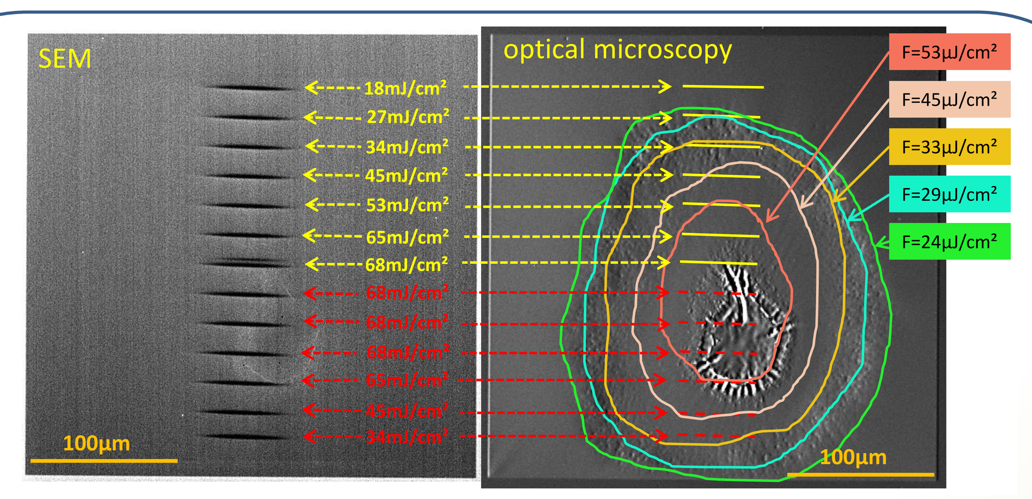
Amorphous metallic glasses are widely investigated nowadays due to their prominent properties: corrosion resistance, high strength, large elastic limit and good damage tolerance[1]. Pure metals were found to amorphize only when quenched at a rate of ~10^{14} K/s, while some metallic alloys with composition located near deep eutectics vitrify on quenching at ~10 K/s. The binary Pd-Si system is one of the most remarkable examples of such a dependency. While pure Pd has been found to vitrify only in computer simulations[2] on quenching at an ultra-high rate, the eutectic $Pd_{81}Si_{19}$ (composition in at. %) alloy can be easily cast into glass, e.g., by air or water cooling (1-10 K/s) [3].

Aim of the experiment is exploring of structural changes in Pd-Si alloys (Si concentration 0, 3, 5, 10, 17 at. %) induced by ultrashort pulsed laser annealing



Experimental

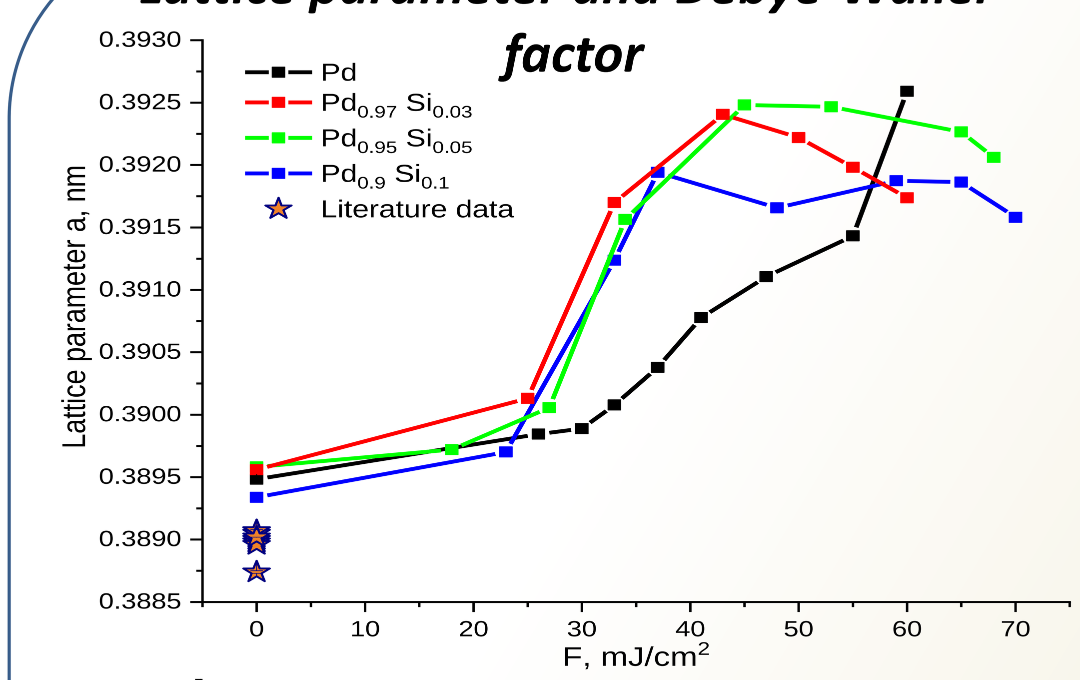
The samples were thin (20 nm) films deposited on 50nm thick amorphous silicon nitride membranes. Upper cup layer was 5-nm thick SiN. Single 0.25-picosecond optical laser pulses (λ=515nm) were used for local heating of the metallic layers. High-energy XRD measurements were performed on PETRA III P07 line (E=73.47keV) in transmission geometry. Rietveld refinement was performed using Profex [5] software

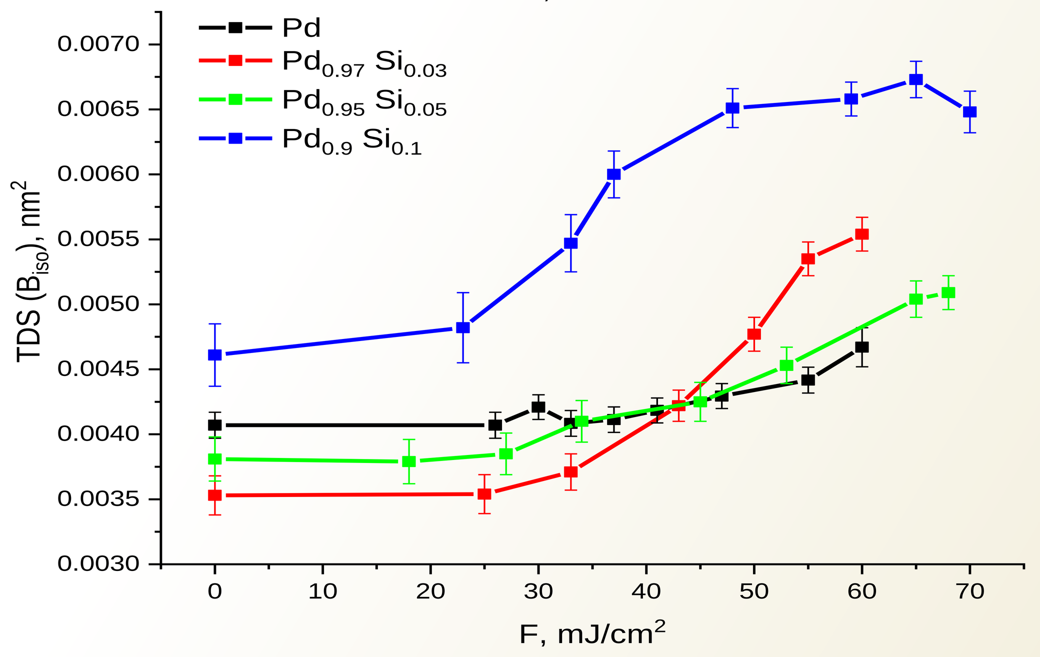


Optical microscopy & SEM

Analysis of the irradiation imprints at various pulse energies allowed for characterization of the spatial intensity/fluence profile for each laser pulse (right). Surface contamination due to X-ray beam enabled identifying the position of the measurements vs. to the center of the laser irradiated spot (left).

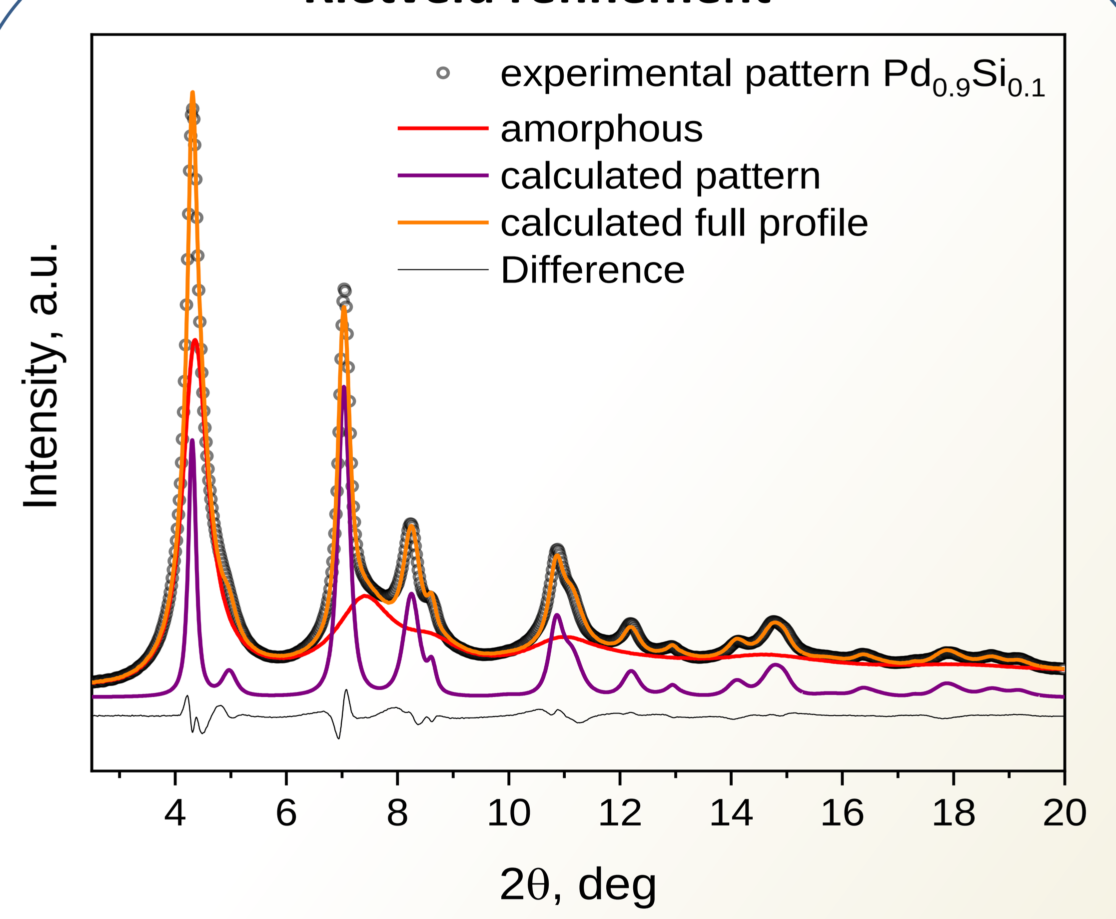
Lattice parameter and Debye-Waller factor





With increasing of annealing temperate (laser intensity) increasing of Debye-Waller factor is observed. This evidences the increase of the structural disorder due to irradiations. Simultaneously the lattice parameter increases, which might be explained by a rapid cooling process – "freezing" of expanded lattice.

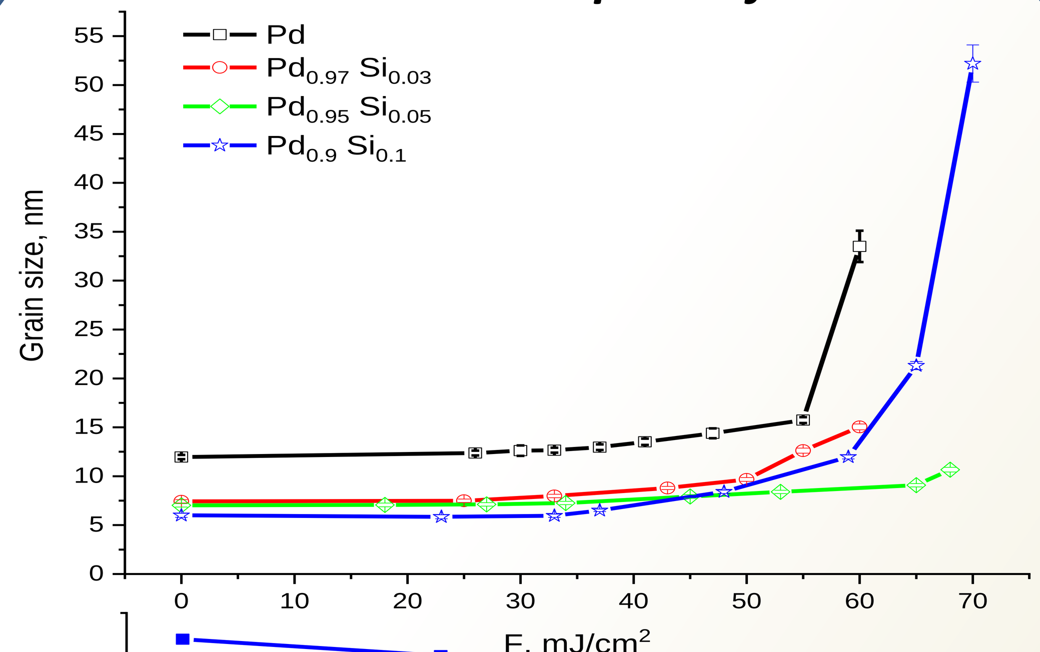
Rietveld refinement

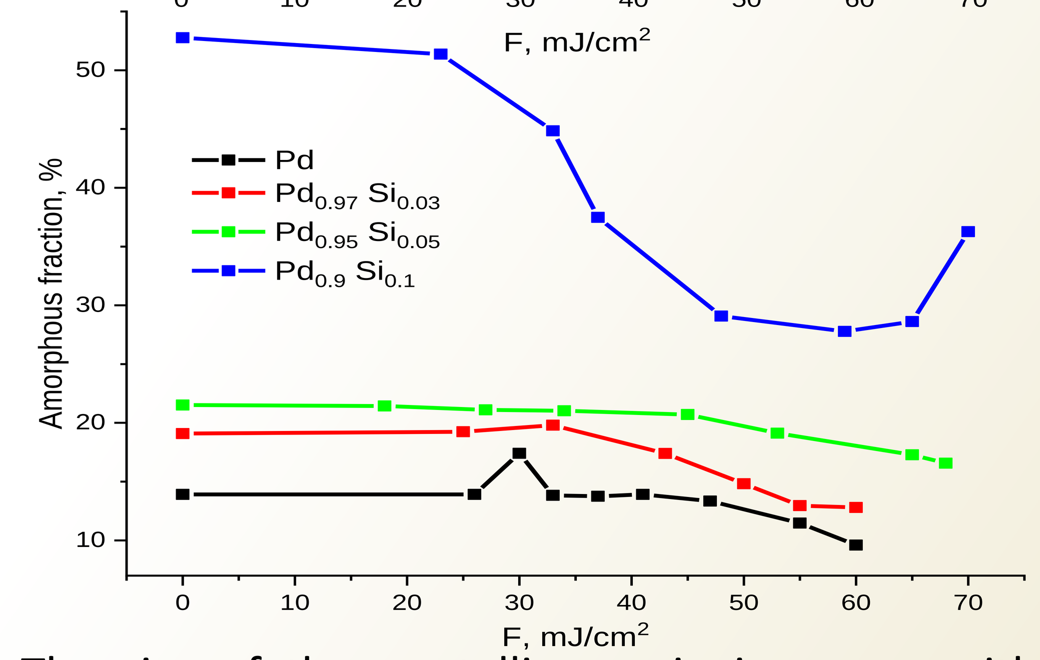


Example of XRD pattern fitting using Rietveld refinement. Before Rietveld analysis, scattering signal from SiN membrane layers was removed from all data. Further, instrumental function was calibrated using CeO_2 pattern.

Pattern from amorphous sample $Pd_{0.83}Si_{0.17}$ was used as marker of the amorphous phase which allow additionally to quantify crystalline and amorphous phase fractions.

Grain size and amorphous fraction

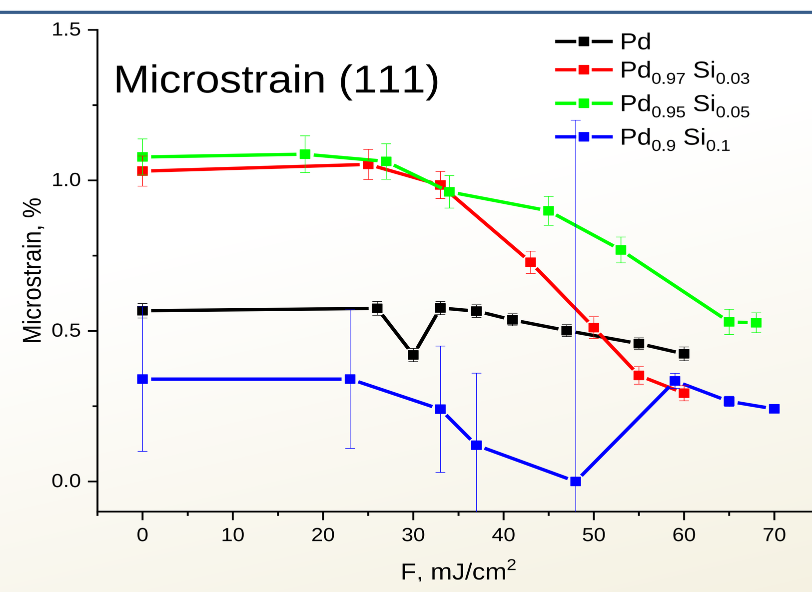


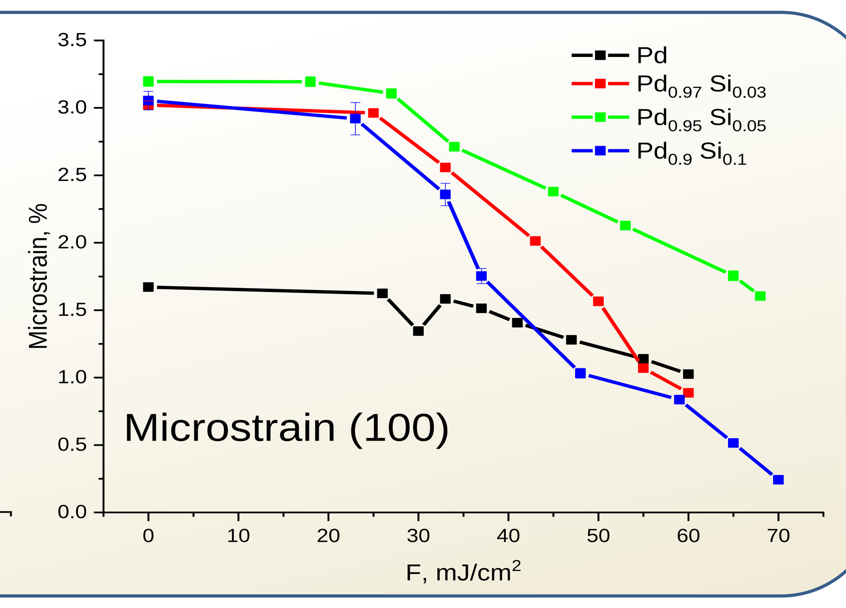


The size of the crystalline grain increases with increasing of fluence. Simultaneously the fraction of the amorphous phase decreases. Both observations indicate on recrystallization process.

Anisotropic microstrain

Presence of anisotropic microstrain is a typical feature of nanocrystalline Pd layers [6]. The strain is bigger in PdSi layers comparing with pure Pd ones. Microstrain decreases with increasing of fluence.





References

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Acknowledgments

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Conclusions

The results for the crystalline films indicate that with an increasing energy density, the mean average crystalline grain size of the film increases, the microstrain is reduced, and the content amount of the amorphous phase (presumably present at the grain interfaces) decreases. Those results are consistent with our predictions regarding the local structural variation of the laser-irradiated films. In those samples that are trapped in a metastable state during film deposition, the laser heating contributes to the transient enhancement of the atomic mobility, which allows relaxation of the system towards the lower energy state (growth of crystalline grains and reduction of the lattice strain).

Additionally, obtained results will be used for reference the results of our MD simulations, we aim to perform total scattering measurements of metallic thin film samples relevant for our previous XFEL experiments.