Structural and surface morphology modifications of $Pd_{100-x}Si_x$ thin film induced by ultrashort pulsed laser annealing

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Over the past years, metallic glasses have been widely investigated due to their outstanding mechanical and magnetic properties[1]. Some pure metals have been found to amorphize only when quenched at a rate of $\sim 10^{14}$ K/s [2], whereas certain metallic alloys with compositions near deep eutectics (so called bulk metallic glasses) vitrify upon quenching at a fraction of a Kelvin per second. The binary Pd-Si system is one of the most notable examples of strong composition dependence of the glass-forming ability. While pure Pd is essentially a non-glass-former and has only been observed to vitrify in computer simulations[3] under ultra-high quenching rates, the eutectic $Pd_{81}Si_{19}$ alloy (composition in at.%) can be easily cast into a glass, e.g., by air or water cooling at rates of 1–10 K/s [4].

In this work, we studied 20 nm thick $Pd_{100-x}Si_x$ thin-film metallic glasses (x = 0, 3, 5, 10), which were deposited on 50 nm thick SiN membranes and covered with a 5 nm thick SiN capping layer. The as-deposited films were either completely crystalline (x=0) or partially crystalline (with a growing amorphous volume fraction as the Si content – x - increased). Structural and surface morphology modifications of the films were induced using ultrashort sub-ps laser pulses of various fluences at a 515 nm wavelength. Local structural modifications were characterized by means of X-ray diffraction in transmission with a micro-focused X-ray beam at the P07 beamline of the PETRA III synchrotron. Analysis of the irradiated spots through optical and electron microscopy enabled to correlate observed structural modifications with the locally deposited energy density from the annealing laser pulses. We observe that with increasing deposited energy density, the average crystal grain size of the layer increased, microstrain reduced, and the volume fraction of the amorphous phase decreased. This indicates that in layers initially in a metastable state, laser heating increased the mobility of atoms, allowing the system to relax towards a lower energy state (i.e. growth of crystalline grains and reduction of lattice strain). At the same time, the final sample state exhibited larger lattice parameters and higher Debye-Waller factor, indicating increased structural disorder. We attribute this to the rapid lattice freezing due to the quick heat diffusion to the substrate. Our analysis provides new insights into the composition-dependent crystallization behavior of Pd-Si thin-films.

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