The Directed Assembly of Metallic Nanoparticle Chains by Pulsed Laser Induced Dewetting and Nanolithography

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Directly–assembly via nanolithography and pulsed laser heating was used to assemble *nanoparticle* chains with precise size and spacing. Specifically, liquid–phase, pulsed laser induced dewetting (PLiD) was used to convert metallic thin film strips into nanoparticle chains exhibiting an *unnaturally* low deviation in particle size and spacing.

Two-dimensional thin film strips retracted into three-dimensional fluid rivulets during the initial stages of dewetting. Next, rivulet break-up ensued forming nanoparticle chains according to a process resembling Plateau-Rayleigh fluid jet destabilization. The resulting mean nanoparticle size and pitch *were controlled*, *yet disperse*. However, by imposing a synthetic sinusoidal perturbation onto the edges of thin film strips, the dispersion in the final nanoparticle chain was drastically reduced. Sinusoidal perturbations patterned along the edges of the thin film strips translated into varicose oscillations on the rivulet surface – this following a liquid phase retraction step governed by Stokes fluid flow.

The significant findings reported here are; (1) In the case of un-patterned thin film strips, the break-up of the rivulet into droplets can be understood based on a modified Plateau-Rayleigh instability mechanism - a low fidelity nanoparticle chain result (*figure 1a-b*) (2) Synthetic nanoscale varicose perturbations, characterized by a length-scale larger than a critical wavelength, led to a drastic improvement in nanoparticle chain fidelity (*figure 1c-d*) (3) Linear stability analysis (LSA) predicted both the rivulet instability development and the critical wavelength required for synthetic perturbations to form ordered chains (4) Non-linear hydrodynamic simulations reproduced nanoscale features observed during both liquid phase retraction and nanoparticle formation (figure 1e).



g, nanoparticle formation

Figure 1 (a) An SEM image of a nickel thin film strip, supported on a silicon wafer substrate, following electron beam lithography and metallization. (b) Self-assembly by pulsed laser induced dewetting (PLiD). The same thin film strip, shown in (a), following five KrF (248nm) laser pulses of 25ns duration (melt time per pulse of ~10ns). The thin film strip liquefies and dewets during irradiation flowing into the form of a linear chain of nickel nanoparticles. The energy density of each laser pulse was 420 mJ/cm². The nickel thin film was 23±1 nm thick. (c) A sinusoidal, synthetic perturbation was imposed on a similar thin film strip (same height and width as in (a)). The wavelength of the synthetic perturbation was 1.35 µm. (d) Directed–assembly by nanolithography and *PLiD.* The fidelity in nanoparticle size and pitch is markedly improved by the presence of the initial synthetic perturbation in the thin film strip (same energy density and number of pulses as in (b)). (e) Non–linear hydrodynamic simulations of dewetting for the thin film strip shown in figure (c). Simulations reveal the liquid retraction phase, from thin film strip (e) to the rivulet morphology (f), to the final nanoparticle chain morphology (g).