Laser Induced Liquid Phase Instabilities: Transition From Single Particle Coalescence To Multi-Particle Breakup of Nickel Nano-Rivulets and Programming Instabilities

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The synthesis of nanoparticles and nanostructures with unique spatial and morphological characteristics is increasingly important in science and technology, where new fabrication routes are crucial for future advances. Conveniently the liquid phase properties of a metal, such as low viscosity and high surface energy, make it an ideal platform to experimentally study the governing liquid state dynamics, liquid instabilities, and mass transport of self- and directed-assembly metallic nanostructures.

*Nickel Nano-Rivulets***:** It is experimentally observed that liquid rivulets below critical dimensions collapse to form single particles rather than break up into multiple particles, Fig. 1. A competition happens between two fluid dynamic phenomena: (a) capillary-induced edge retraction and collapse/coalescence and (b) break up due to varicose mode instability. We focus on the unique spatial and temporal transition region between the two competing regimes. To investigate this competition we utilize nanofabrication techniques and pulsed-laser induced dewetting (PLiD). With these methods it is experimentally possible to precisely control the initial far-from-equilibrium geometry and study the resultant fluid dynamics and assembly phenomena.

Programming Instabilities: Classic Rayleigh-Plateau (RP) stability theory for a free liquid jet predicts a fluid column to break into droplets (particles) when perturbing surface modes are above a critical wavelength, $2\pi R$, albeit with dispersion. By selectively harnessing specific surface perturbations through

experimentally imposed initial fabrication conditions (programming) it is possible to significantly increase the fidelity of particle-particle spacing and size. We have taken this capability beyond the 1-D case and into 2-D using PLiD.[1] By controlling the various parameters of a square-wave geometry, Fig. 2(a), it is possible to access many different spatial array outcomes. We have also shown it to be possible to access particle spacing below the critical wavelength predicted by RP theory. In addition, Volume-of-Fluid (VoF) simulations support experimental results, and suggest several new and unusual morphologies are possible. Precisely controlling both metallic nanoparticle size and inter-particle spacing in multiple dimensions is an important capability, physically significant for various research areas; for instance, spintronics[2] and plasmonics.[3]

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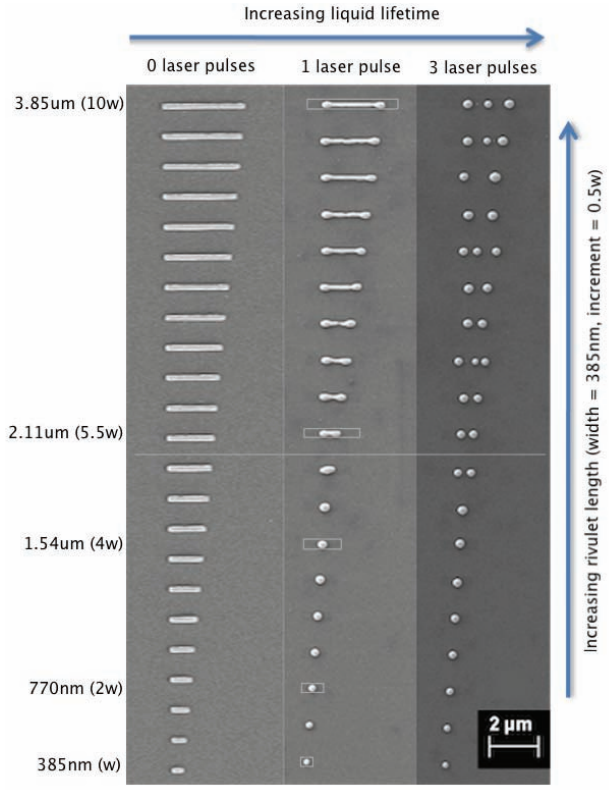

Figure 1. SEM image showing the transitional boundary (horizontal line) between single particle coalescence and multi-particle break up. We focus on the unique spatial and temporal transition region between the two competing regimes for a liquid metal rivulet.

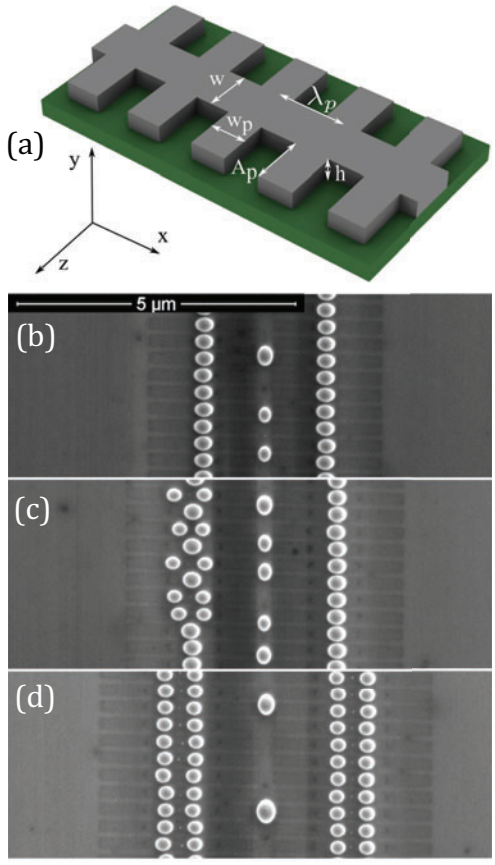

Figure 2. SEM images showing various 2-D array end-states after 3 laser pulses (250mJcm^{-2}), $h = 20nm$ of nickel (a) Square-wave geometry
(b) $\lambda_p = 300nm, w = w_p = 200nm, A_p = 2\mu m$
(c) $\lambda_p = 300nm, w = w_p = 200nm, A_p = 2.4\mu m$
(d) $\lambda_p = 300nm, w = w_p = 200nm, A_p = 2.9\mu m$

[1] N. Roberts et al., Appl. Mater. Interfaces **5**, 4450 (2013)
[2] A. Bernand-Mantel et al., Appl. Phys. Lett. **89**, 062502 (2006)
[3] N. J. Halas et al., Chem. Rev. **111**, 3913 (2011)