Fingering instabilities in dewetting nanofluids: Auxiliary Material

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Real-time imaging of nanofluid dewetting

Video 1 shows the formation and evolution of fingering instabilities in a colloidal solution of dodecanethiol-passivated Au nanoparticles (mean diameter $\sim$ 2nm) in dichloromethane. The field of view is 160 μm x 200 μm. Imaging was carried out using the contrast-enhanced Sarfus technique commercially available from Nanolane (www.nanolane.com). This technique involves the use of multilayer substrates which have been engineered to ensure that they do not affect the polarisation state of reflected light. Adsorption of a thin film (such as the Au nanoparticles of interest here), however, strongly affects the polarisation state and thus yields a high-contrast image.

Throughout the video the macroscopic dewetting front is retracting towards the top right hand corner of the frame. In the early part of the video (00:00 - 00:07), fingering patterns *at* the macroscopic dewetting front are observed to form. From 00:19 to 00:28, the macroscopic contact line no longer exhibits transverse instabilities but isotropic fingering patterns nucleate and grow in a thin residual solvent film *behind* the macroscopic dewetting front. Note that at 00:07 the contact line moves out of the field of view. Between 00:08 and 00:13, the field of view is changed so as to relocate the dewetting front.

Calculation of fractal dimension using density correlation function

The fractal dimension of the AFM and simulated images was evaluated via the density-density correlation function, defined as $C(r) =< \rho(r')\rho(r'+r) >$. At intermediate length scales the density-density correlation function behaves as $C(r) \sim r^{-\delta}$ where the decay exponent δ is related to the fractal dimension as follows: $D_0 = d_e - \delta$ (d_e is the Euclidean dimension (i.e. 2 in this case)).

Images were converted into a density profile $\rho(x, y)$ where $\rho = 1$ in the absence of nanoparticles and $\rho = 0$ in the nanoparticle phase. $\rho(x, y)$ was then two-dimensionally Fourier transformed using an FFT algorithm to give $\tilde{\rho}(k_x, k_y)$ and then the spectral density $\sqrt{(}2\pi)|\tilde{\rho}(k_x, k_y)|^2$ was obtained. The spectral density and density-density correlation function form a Fourier pair such that the former can be two-dimensionally inverse transformed to give $C(x, y)$, which is then averaged over all directions to give $C(r)$.

Dependence of fingering on nanoparticle mobility

Fig.1 below shows the dependence of mean finger number on the mobility ratio, i.e. the ratio of the time scales of solvent evaporation and nanoparticle diffusion, in Monte Carlo simulations of dewetting nanoparticle solutions. Each data point represents the spatial mean value (along the direction of front motion) averaged over seven different kinetic Monte Carlo runs of the finger count orthogonal to the direction of front motion. The error bars represent the standard variation.

Video of Monte Carlo simulation

Video 2 is a sequence of frames from a Monte Carlo simulation of finger formation in a dewetting nanofluid. The parameters used in the simulation were: TO BE ADDED. The video comprises 25 frames per sec. A frame was captured every 40 Monte Carlo Steps (MCS) such that the movie rate is 1000 MCS/s. The movie runs for a total of 16,000 MCS (16 seconds). The simulation was carried out on a 1024x1024 grid which was resized to 512x512 for the movie frames.

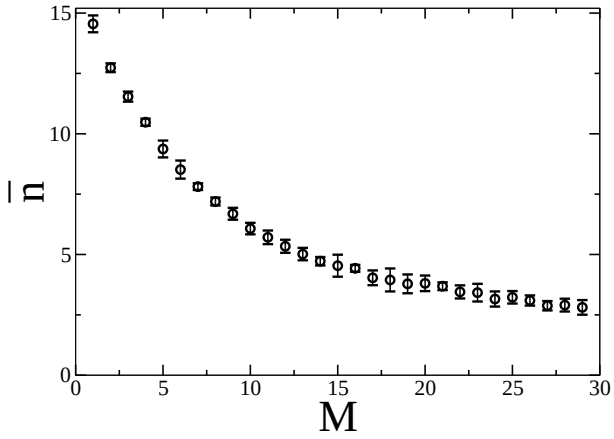

FIG. 1: Dependence of mean finger number $\bar{n}$ on mobility M, i.e., on the ratio of the time scales of solvent evaporation and nanoparticle diffusion, for a straight evaporative dewetting front. The parameters are: lattice size 1200 $\times$ 1200, nanoparticle coverage $\phi = 0.1$, kT=0.2, $-\mu = 2.2$, $\varepsilon_l = 1, \varepsilon_n = 2, \varepsilon_{nl} = 1.5$. A sufficient number of Monte Carlo steps were simulated such that the entire field is finally dry (about 5000-7000). Finger numbers are counted at the final structure.