The role of phase separation on Rayleigh-Plateau type instabilities in alloys

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S. Supporting Information:

S.1 Bulk construction: The 256 atom NiAg structure was created, equilibrated, and replicated following our previous work[28]. In this work, the replication led to a bulk structure of 829,440 atoms. This bulk structure was equilibrated and quenched at temperatures starting at 3000K and ending at 1400K in 200K increments. For temperatures 1800K-3000K the bulk structure was sequentially equilibrated using 400ps of NPT, 600ps of NVT, and 200ps of NVE (total of 1.2ns).

Thus, the cooling rate from 3000K to 1800K was 1.67×10^{11}K/s. At 1600K, the bulk is equilibrated for 4.8ns (400ps NPT, 2400ps NVT, 2000ps NVE) so the cooling rate from 1800K to 1600K was 0.42×10^{11}K/s. We cooled the bulk to 1400K from 1600K in 6.6ns (400ps NPT, 1800ps NVT, and 4400ps NVE), so the cooling rate was 0.30×10^{11}K/s. Note that lower temperatures required more time to equilibrate temperature and energy. The two chosen bulk structures used in this paper (2000K and 1400K) are shown in Fig.1(a) and (b). The bulk structure in Fig.1(c) was created differently.

This bulk (Fig.1(c)) was first created with 5120 atoms, equal parts Ni and Ag. The energy was minimized and both NPT/NVT ensembles were used to equilibrate pressure, volume, and temperature at 300K, 1000K, and then finally 1400K. The small bulk structure was then replicated (expanded) into one containing 921,600 atoms (this is where the stripe patterns emerge) and subsequently equilibrated with 1.2ns of NVT and 9ns of NVE, which we found to be sufficient for convergence.

The process of creating these bulk structures (Fig.1) made the lengths, L, unequal ((a): 1463.2 Å, (b): 1433.7 Å, (c) 1598.9 Å). The only bearing this has is on the initial prescribed stripe length, which is determined by L.

S.2 Lines: At the lateral ends of the lines, periodic boundary conditions are imposed. The widths of these lines are calculated so that for lines of length L, and wavelength λ, $L = N\lambda$ where N is an integer (we used N=5 or N=3).

Slices:

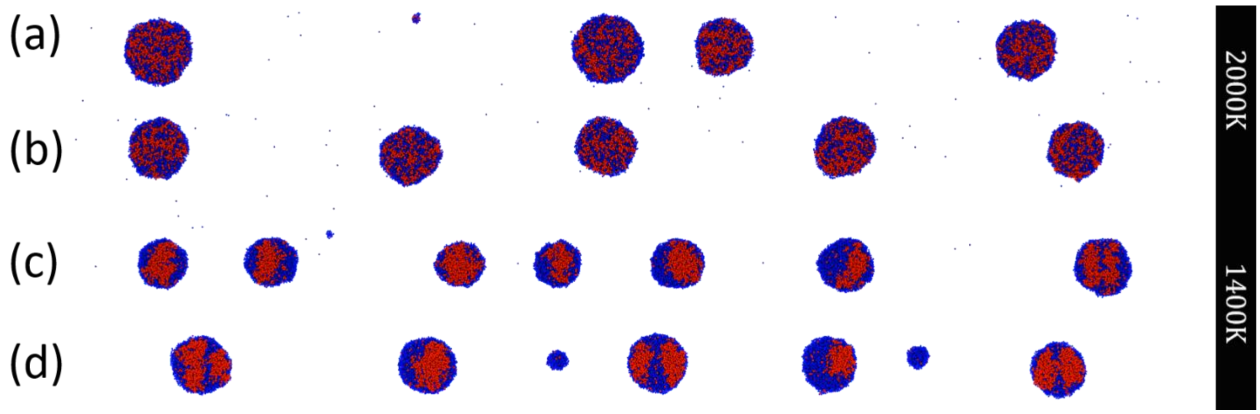

Figure S.1. Longitudinal slices of resultant nanoparticles at the final frames v) for (a) Fig.3 2000K, straight line (left), (b) Fig.3 2000K, large amplitude (right), (c) Fig.3 1400K, straight line (left), (d) Fig.3 1400K, large amplitude (right). In (d) satellite droplets of pure Ag are present.

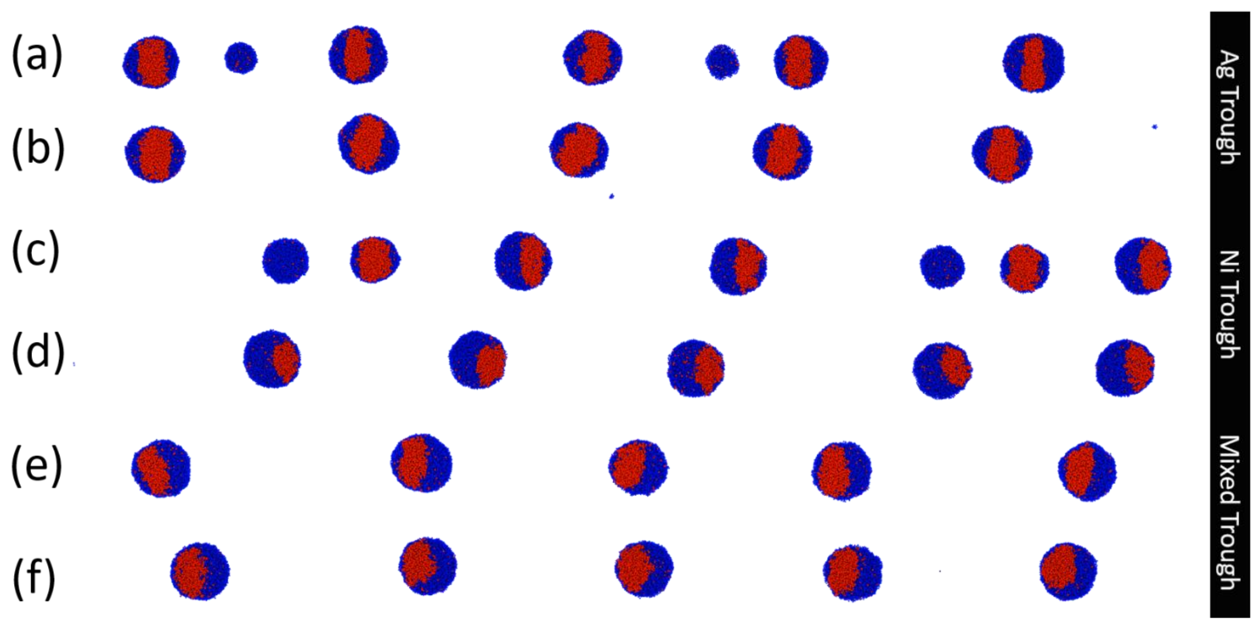

Figure S.2. Longitudinal slices of resultant nanoparticles at the final frames v) for (a) Fig.4 Ag trough, small amplitude (left), (b) Fig.4 Ag trough, large amplitude (right), (c) Fig.4 Ni trough, small amplitude (left), (d) Fig.4 Ni trough, large amplitude (right), (e) Fig.4 mixed trough, small amplitude (left), and (f) Fig.4 mixed trough, large amplitude (right). In (a) satellite droplets of pure Ag are present.

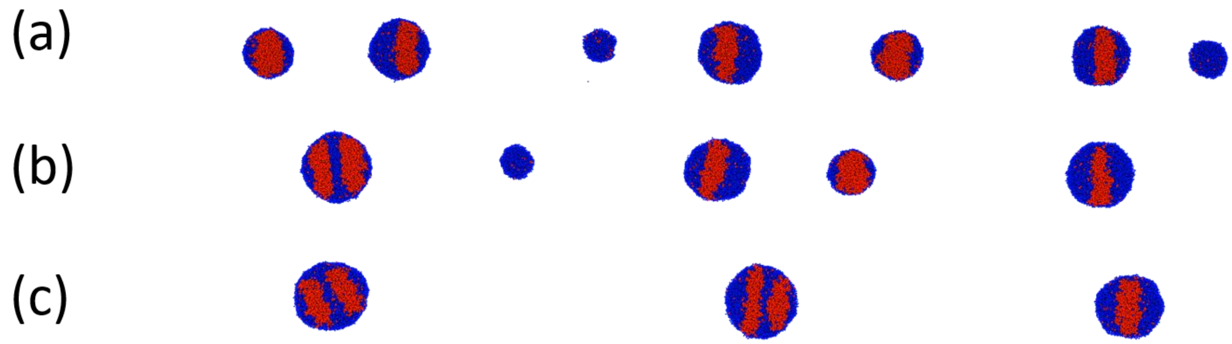

Figure S.3. Longitudinal slices of resultant nanoparticles at the final frames v) for (a) Fig.5 A=3.2Å (b) Fig.5 A=5Å, and (c) Fig.5 A=6Å. In (a) and (b) satellite droplets of pure Ag are present.

S.3 Maximum RP wavelength case:

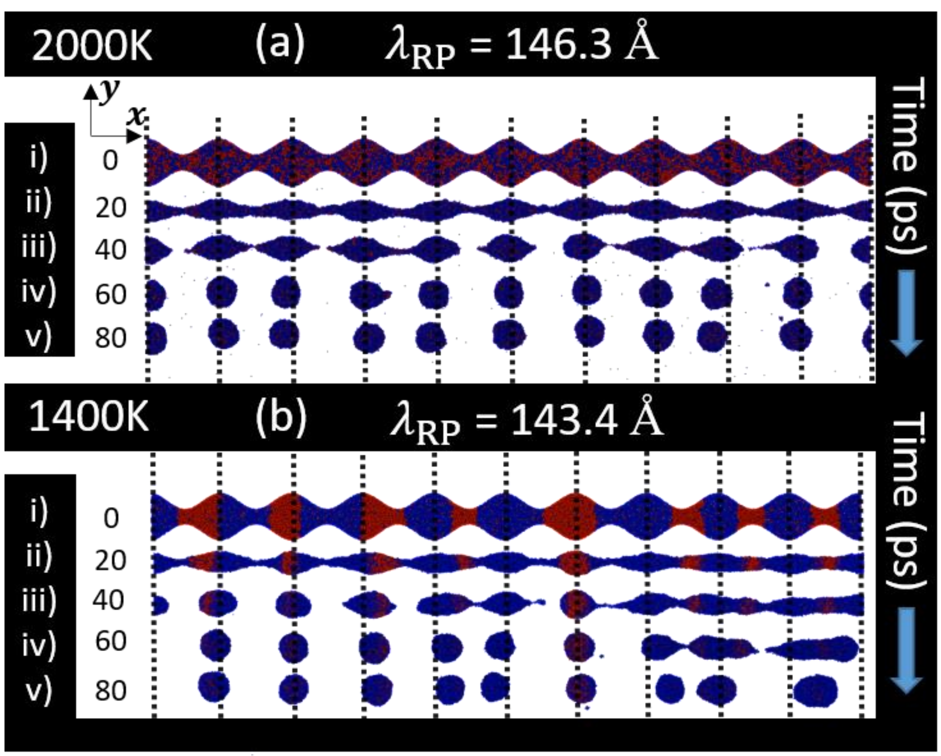

Figure S.4. Time evolution of synthetic lines patterned with amplitude A=16 Å and maximum RP wavelength λ_{RP}=146.3Å (a) and λ_{RP}=143.4Å (b). Time labels (in ps) are placed next to the corresponding lines. Guides (dotted lines) are placed at the location of the original peak in i)**.**

Figure S.4 shows an example of synthetic lines at (a) 2000K and (b) 1400K patterned with the maximum RP wavelength, $\lambda_{\mathrm{RP}} = \lambda_{\mathrm{m}}$. The time evolution is marked i)-v) and is shown in 20ps increments. In Fig.S.4(a), at 2000K, where the phase separation lengthscale is very small, the line breaks up into 10 equally spaced nanoparticles according to the RP mechanism. In Fig. S.4(b), the PS length scale is comparable in size to the prescribed maximum wavelength, (λ_{PS}=170.7Å). Similar to what was shown in for longer λ_{RP}, when the trough is aligned with the Ag/Ni interface or in a Ag region, the soluto-capillary flow cooperates with the RP mechanism and droplets form slightly faster and aligned with the peak positions (see the center and left side of S.4(b)). When the troughs are mostly Ni, however, the soluto-capillary flow competes with the RP mechanism as early stages of the line are observed to coarsen as Ag atoms migrate to this Ni-rich region. Ultimately, where the troughs are Ni-rich, the breakup is slower and the particle positions are not aligned with the original synthetic perturbation peak positions, consistent with the soluto-capillary effects competing with the RP.