A microscopic view of the yielding transition in concentrated emulsions

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Yielding of concentrated emulsions

\[ \tan \left( \frac{G''}{G'} \right) \]

\( \sigma \) (dynes/cm\(^2\))
\( \delta \) (rad)
\( G' \), \( G'' \) (dynes/cm\(^2\))

strain \( \gamma \)

Mason et al 1996.
Questions

• Microscopic picture of yielding: crystalline solids OK (defects), what about amorphous solids?

• Oscillatory drive: very few simulations and experiments! BUT relevant to rheology and fatigue tests, easier to detect irreversible rearrangements

• Relation to reversible-irreversible transition in complex fluids?
Oscillatory drive: DWS echo

- the **same**, small fraction of drops undergo **irreversible** rearrangements at each cycle
- the fraction of rearranged drops increases with $\gamma$
- at yielding, just a few % of drops undergo an irreversible rearrangement

- need to make (strong) assumptions on the nature of motion, no spatial/temporal resolution…

*Courtesy of D.J. Pine, Hebraud et al., PRL 1997*
The reversible-irreversible transition: revisiting Taylor’s experiment

wind…

unwind…

New Mexico University – http://youtu.be/p08_KITKP50
Suspension of (non-Brownian) particles

Pine’s group

\( g_0 = 3 \)

\( g_0 = 2 \)

Corte et al, Nature Physics 2008
Suspension of (non-Brownian) particles

2nd order dynamical transition

Corte et al, Nature Physics 2008
Dense/strongly interacting systems

Yielding and microstructure in a 2D jammed material under shear deformation†

*Soft Matter, 2013, 9, 6222

Nathan C. Keim and Paulo E. Arratia*

Oscillatory athermal quasistatic deformation of a model glass

PHYSICAL REVIEW E 88, 020301(R) (2013)

Davide Fiocco,1, Giuseppe Foffi,1,2,† and Srikanth Sastry3,4,†

Same behavior as in Pine’s experiments??
Confined colloids w/ hydrodynamic interactions: 1st order transition??

Back to our drops…

Emulsion: PDMS oil + TMN-10 in H₂O + glycerol
\[ 2r = 2.4 \, \mu m, \text{ polydispersity} = 20\% \]
\[ \varphi = 65 – 88\% \]

Microscopy: 100x DIC, gap ~100 \( \mu m \)
Visualizing the emulsion

Motion analysis (over one cycle): Image Correlation Velocimetry (PIV-like)
coarse graining ~ 3.8 µm, resolution ~ 10 nm
Rheology: strain sweep ($\phi = 0.83$)

Yielding transition: very smooth!
Microscopy: rms displacement

Yielding transition: quite sharp (especially at high $\varphi$)
Microscopic yielding corresponds to macroscopic onset of non-linearity.
Motion is heterogeneous

\[ \varphi = 0.65, \quad \gamma = 4.33\% \]

\[ \varphi = 0.74, \quad \gamma = 7.21\% \]

**Fig. 4** Difference between two successive strobed images: regions where motion occurred show up as features brighter or dimmer than the average background, while immobile regions are featureless. For each sample, the applied strain is just above the yield strain \( \gamma_y \).
Probability distribution of drop displacements

Abrupt change at the yielding transition (compare $\gamma = 6.25\%$ to $\gamma = 7.21\%$)

$\phi = 0.74$ (same at higher $\phi$)
Probability distribution of drop displacements

Above $\gamma_y$: non-Gaussian pdf: $\sim$ double exponential tails
Closer to the jamming transition…

\[ \phi = 0.65 \]

Smooother transition, but again ‘mobile’ and ‘supermobile’ drops.
A Lindemann’s criterion for yielding?

Jump size for ‘supermobile’ drops: \( \sim 11\% \) of drop size (irrespective of \( \varphi \) and \( \gamma \))

Lindemann’s criterion for melting a crystal: particles ‘jiggle’ over \( \sim 15\% \) of their size

Yielding transition when all particles become ‘supermobile’??

![Graph showing the relationship between \( f_{sm} \) and \( \gamma \) for different values of \( \varphi \).]
Dynamics: spatial and temporal organization
Temporal correlation: bursts of motion

\[ c(\tau) = \langle \langle |\Delta y(t, R)| \Delta y(t + \tau, R) | \rangle \rangle_R \]

- Histogram of temporal correlation function \[ c(\tau) \] with bins for different \( \gamma_y \) values: 4.81%, 6.25%, 9.62%.

\[ \varphi = 0.74 (\gamma_y = 6.5\%) \]

- Bursts of motion may last hundreds of cycles, but eventually mobile/quiescent drops do exchange.

- No clear indication of divergence around \( \gamma_y \): transition not so 2nd order…
Spatial correlation

\[ g_4(\Delta r) \sim \langle \langle \Delta y(R, t) \Delta y(R + \Delta r, t) \rangle \rangle_R \]

- Spatial correlations extend up to \(~10-15\) drops
- \(\xi_y\) larger at higher \(\varphi\): stress transmission important?
- Behavior around \(\gamma_y\): varies with \(\varphi\)…
Conclusions

• Yield transition at a microscopic level quite sharp, as opposed to rheology

• Motion is heterogeneous: ‘quiescent’, mobile and supermobile particles coexist

• extended spatio-temporal correlations of dynamics, but quiescent/mobile populations eventually do exchange

• A Lindemann’s criterion for fluidization of amorphous systems?

• Nature of the transition unclear:
  - Close to $\phi_j$: smoother $\Delta y(\gamma)$, but no diverging length/time scales around $\gamma_y$
  - At higher $\phi$: sharp transition, but length/time scales grow significantly around $\gamma_y$
Thanks to…

NYU
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You all!