Glass Transition Seen through Asymptotic Expansions

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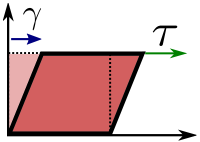
October 20^{th}, 2011

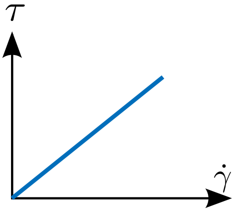
Joint work with M. Renardy (VirginiaTech)

Outline

Simple shear

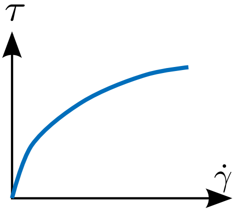


- τ: stress
- γ: shear
- $\dot{\gamma}$: shear rate



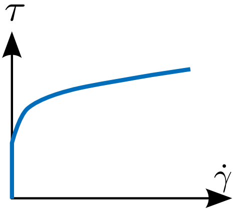
Newton

$\tau = \eta\dot{\gamma}$



Power law

$\tau = A\dot{\gamma}^n$



Herschell-Bulkley

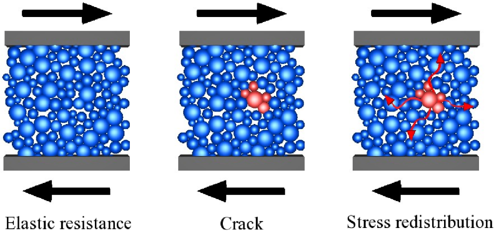
$\tau = \tau_0 + A\dot{\gamma}^n$

Solid or fluid?

Pitch drop experiment currently at University of Queensland.

Zoom to the mesoscopic scale

- Several type of response to a same external forcing



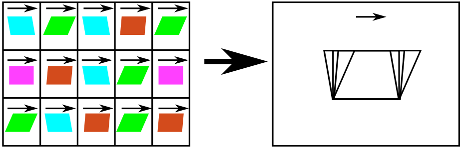
- Glassy transition

$$A\dot{\gamma}^n \quad \Phi_c \quad \tau_0 + B\dot{\gamma}^p$$

Principles of the model

Generic glassy material.

- Replace spatial heterogeneities of local stresses → global statistics p :



- Write the evolution equation of p directly
- Recover the macroscopic stress by averaging :

$$\tau = \int_{\sigma \in \mathbf{R}} \sigma p(t, \sigma) \mathrm{d}\sigma$$

Hébraud -Lequeux equation

Elasticity

$$\partial_t p = -G_0\dot{\gamma}(t)\partial_\sigma p - \frac{1}{T_0}H(|\sigma|-\sigma_c)p + \overline{\Gamma}(p(t))\delta_0(\sigma) + D(p(t))\partial_\sigma^2 p$$

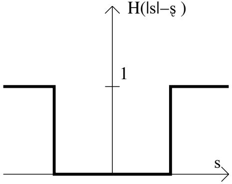
- G_0: shear modulus
- $\dot{\gamma}$: shear rate

Hébraud -Lequeux equation

Relaxation

$$\partial_t p = -G_0\dot{\gamma}(t)\partial_\sigma p - \frac{1}{T_0}H(|\sigma| - \sigma_c)p + \overline{\Gamma}(p(t))\delta_0(\sigma) + D(p(t))\partial_\sigma^2 p$$

- T_0: relaxation time
- H: Heaviside step function



Hébraud -Lequeux equation

Relaxation

$$\partial_t p = -G_0\dot{\gamma}(t)\partial_\sigma p - \frac{1}{T_0}H(|\sigma| - \sigma_c)p + \overline{\Gamma}(p(t))\delta_0(\sigma) + D(p(t))\partial_\sigma^2 p$$

- $\overline{\Gamma}(p(t)) = \frac{1}{T_0}\int_{|\sigma|>\sigma_c} p(t,\sigma)\mathrm{d}\sigma$: fluidity
- δ_0 Dirac measure at 0

Hébraud -Lequeux equation

Plasticity

$$\partial_t p = -G_0\dot{\gamma}(t)\partial_\sigma p - \frac{1}{T_0}H(|\sigma| - \sigma_c)p + \overline{\Gamma}(p(t))\delta_0(\sigma) + D(p(t))\partial_\sigma^2 p$$

- $D(p(t)) = \alpha\overline{\Gamma}(p(t)) = \frac{\alpha}{T_0}\int_{|\sigma|>\sigma_c} p(t,\sigma)\mathrm{d}\sigma$
- α: Transition parameter

Initial question

Stationary dimensionless Hébraud -Lequeux equation

$$\begin{cases} -\alpha\overline{\Gamma}(p)\partial_\sigma^2 p + \dot{\gamma}\partial_\sigma p + h(\sigma)p = \overline{\Gamma}(p)\delta_0(\sigma) \\ p \geq 0, \ \int_{\mathbf{R}} p(\sigma)\mathrm{d}\sigma = 1 \end{cases}$$

Does this model accounts for a glass transition?

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Does this model accounts for a glass transition?
[HÉBRAUD, LEQUEUX,1998] : Yes, without formal proof

Results of [HL, 98]

If $\alpha > 1/2$, Newtonian behaviour,

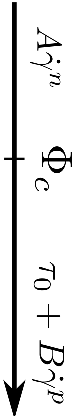
$$\tau \approx \eta\dot{\gamma} \qquad \eta \approx (\alpha - 1/2)^{-2}$$

If $\alpha = 1/2$, power law behaviour,

$$\tau \approx A\dot{\gamma}^{1/5}$$

If $\alpha < 1/2$, threshold behaviour,

$$\tau \approx \tau_0 \qquad \tau_0 \approx (1/2 - \alpha)^{1/2}$$



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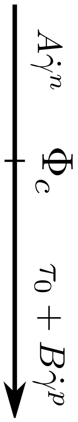

Goal : Prove mathematically this result.

Direct approach

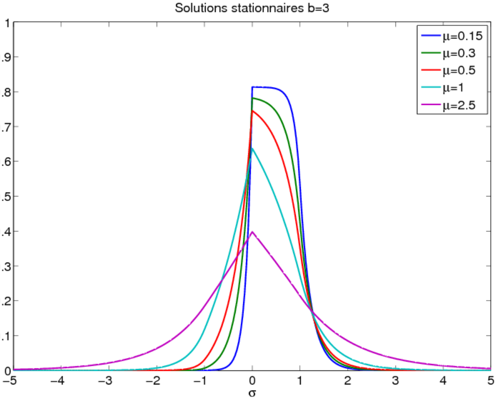

Figure: Examples of solution to the HL model

Direct approach

- Self consistency : $f_\alpha(\alpha\Gamma, \dot{\gamma}) = 0$

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$$+\left(\frac{3\alpha\Gamma^2}{\dot\gamma} + \alpha\Gamma\dot\gamma\right)\mathrm{sh}\left(\frac{\dot\gamma}{\alpha\Gamma}\right) + \sqrt{\dot\gamma^2 + 4\alpha\Gamma}\left(\frac{\alpha\Gamma^2}{\dot\gamma^2}\left(\mathrm{ch}\left(\frac{\dot\gamma}{\alpha\Gamma}\right) - 1\right)\right.$$
$$\left. + \frac{\alpha\Gamma}{\dot\gamma}\mathrm{sh}\left(\frac{\dot\gamma}{\alpha\Gamma}\right) + \alpha\Gamma\mathrm{ch}\left(\frac{\dot\gamma}{\alpha\Gamma}\right)\right)\Bigg]$$
$$- \sqrt{\dot\gamma^2 + 4\alpha\Gamma}\mathrm{ch}\left(\frac{\dot\gamma}{\alpha\Gamma}\right) - \left(\frac{2\alpha\Gamma}{\dot\gamma} + \dot\gamma\right)\mathrm{sh}\left(\frac{\dot\gamma}{\alpha\Gamma}\right)$$

Direct approach

- Self consistency : $f_\alpha(\alpha\Gamma, \dot\gamma) = 0$

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$$-\sqrt{\dot\gamma^2+4\alpha\Gamma}\,\mathrm{ch}\left(\frac{\dot\gamma}{\alpha\Gamma}\right)-\left(\frac{2\alpha\Gamma}{\dot\gamma}+\dot\gamma\right)\mathrm{sh}\left(\frac{\dot\gamma}{\alpha\Gamma}\right)$$

- Singularity removal : $X = \alpha\Gamma$, $Y = \dot\gamma/(\alpha\Gamma)$

Stress

Assuming $\Gamma = \phi_\alpha(\dot\gamma) \sim \phi_{\text{app}}(\dot\gamma)$, how to prove the theorem?

$$\begin{aligned}\tau(\alpha\Gamma,\dot\gamma) &= \dot\gamma + \frac{\alpha\Gamma}{2\alpha\dot\gamma} - \frac{(\alpha\Gamma)^3}{\alpha\dot\gamma^3} \\ &\quad + \frac{(\alpha\Gamma)^2}{\dot\gamma^3\Delta(\alpha\Gamma,\dot\gamma)}\left(\frac{\dot\gamma^2}{\alpha\Gamma} - 2\left(\mathrm{ch}\left(\frac{\dot\gamma}{\alpha\Gamma}\right) - 1\right)\right) \\ &\quad + \frac{(\alpha\Gamma)^3}{\dot\gamma^4\Delta(\alpha\Gamma,\dot\gamma)}\left(\dot\gamma\left(\left(\frac{\dot\gamma}{\alpha\Gamma}\right)^2 + \frac{4}{\alpha\Gamma}\right)^{1/2} + 2\mathrm{sh}\left(\frac{\dot\gamma}{\alpha\Gamma}\right)\right)\end{aligned}$$

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Theorem

Glass transition is as announced by HÉBRAUD *and* LEQUEUX.

Link to the penalization

$$\begin{cases} -\alpha\overline{\Gamma}(p)\partial_\sigma^2 p + \dot{\gamma}\partial_\sigma p + h(\sigma)p = \overline{\Gamma}(p)\delta_0(\sigma) \\ p \geq 0, \ \int_{\mathbf{R}} p(\sigma)\mathrm{d}\sigma = 1 \end{cases}$$

What is the behaviour of this system when $\dot{\gamma} \to 0^+$?

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What is the behaviour of this system when $\dot{\gamma} \to 0^+$?

- From previous proof : $\Gamma \sim c\dot{\gamma}$ when $\alpha < 1/2$.
- Replace Γ by its equivalent :

$$-\alpha c\partial_\sigma^2 p + \partial_\sigma p + \frac{1}{\dot{\gamma}}h(\sigma)p = c\delta_0(\sigma)$$

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$$-\eta\Delta u + V\cdot\nabla u + \frac{1}{\varepsilon}\mathbf{1}_\omega u + \nabla\pi = f \qquad \mathrm{div}\, u = 0$$

Boundary layers

By analogy with the penalization : boundary layers.
Example, for $\sigma > 1$:

$$p(\sigma) \approx \sum_k \dot{\gamma}^{k/2}\overline{R}^k(\sigma) + \dot{\gamma}^{k/2}R^k\left(\frac{\sigma - 1}{\dot{\gamma}^{1/2}}\right).$$

Goal :

1. find the expansion of p without *a priori* assumptions
2. justification of the expansion in an appropriate space

Rewriting

Note $q = p_{[-1,1]}$ and $r = p_{[-1,1]^c}$.

$$\begin{cases} -\alpha\Gamma\partial_\sigma^2 q + \dot\gamma\partial_\sigma q = \Gamma\delta_0 & \text{in }]-1,1[, \\ -\mu\Gamma\partial_\sigma^2 r + \dot\gamma\partial_\sigma r + r = 0 & \text{in } [-1,1]^c, \\ r(\pm 1) = q(\pm 1), \\ \partial_\sigma r(\pm 1) = \partial_\sigma q(\pm 1), \\ \int_{|\sigma|>1} r(\sigma)\mathrm{d}\sigma + \int_{-1}^{1} q(\sigma)\mathrm{d}\sigma = 1. \end{cases}$$

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Ansaetze for q and r

We use ansaetze for q and r but of unknown scale and boundary layer size.

$$q(\sigma) = \sum_{k=0}^{+\infty} (\dot{\gamma}^{1/s})^k \overline{Q}^k(\sigma),$$

$$r(\sigma) = \sum_{k=0}^{+\infty} (\dot{\gamma}^{1/s})^k \overline{R}^k(\sigma) + (\dot{\gamma}^{1/s})^k R^k \left(\operatorname{sign}(\sigma), \frac{|\sigma| - 1}{\dot{\gamma}^{l/s}} \right).$$

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Ansaetze for Γ

From $\Gamma = \int_{|\sigma|>1} r(\sigma)\mathrm{d}\sigma$ we also have

$$\Gamma = \sum_{k=0}^{+\infty} \tilde{c}_k y^{k/s}$$

with

$$\tilde{c}_k = \begin{cases} \overline{c}_k & \text{if } 0 \le k \le l-1, \\ \overline{c}_k + c_{k-l} & \text{if } k \ge l, \end{cases}$$

$$\overline{c}_k = \int_{|\sigma|>1} \overline{R}^k(\sigma)\mathrm{d}\sigma,$$

$$c_k = \int_0^{+\infty} [R^k(-1,z) + R^k(1,z)]\mathrm{d}z.$$

Scales and sizes

Goal : find the expansion of p without *a priori* assumptions

Theorem (J. O., M. RENARDY, 2011)

$\alpha > 1/2$: $s = 1$, *all* R^k *vanish,* $\Gamma(\dot{\gamma}) \to \Gamma(0) > 0$

$\alpha < 1/2$: $s = 2$, $l = 1$, *all* $\overline{R}^k$ *vanish* , $\Gamma(\dot{\gamma}) \sim c\dot{\gamma}$

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$\alpha < 1/2$: $s = 2$, $l = 1$, *all* $\overline{R}^k$ *vanish* , $\Gamma(\dot\gamma) \sim c\dot\gamma$

$\alpha = 1/2$: $s = 5$, $l = 2$, *all* $\overline{R}^k$ *vanish*, $\Gamma(\dot\gamma) \sim c\dot\gamma^{4/5}$

Proof

- First relate the shape of the expansion to the first non zero coefficient $\tilde{c}_k$.
  - $\tilde{c}_0 \neq 0$: no boundary layer terms
  - $\tilde{c}_0 = 0$: no non boundary layer terms. First non zero term necessarily $\tilde{c}_{2l} = c_l$.
- Next relate the fundamental problem to the range of α that gives a solution.
- Finally find the next non trivial profile and deduce the minimal s and the corresponding l.

Example: the case $\tilde{c}_0 = 0$ (I)

Assume $2l \geq s$.

- $2l \geq s \implies 2l = s$.
- Fundamental problem

$$\begin{cases} -\alpha c_l \partial_\sigma^2 \overline{Q}^0 + \partial_\sigma \overline{Q}^0 = c_l \delta_0, \\ \overline{Q}^0(\pm 1) = 0, \\ \int_{-1}^{1} \overline{Q}^0(\sigma) \mathrm{d}\sigma = 1, \\ c_l \neq 0. \end{cases}$$

- $c_l \neq 0$ satisfies $\alpha = \alpha c_l \tanh\left(1/(2\alpha c_l)\right)$. Solvable iff $\mu < 1/2$.
- Next non trivial interior problem for $\overline{Q}^l$: $l = 1$ and $s = 2$.

Example: The case $\tilde{c}_0 = 0$ (II)

Assume $2l + 1 \leq s$.

- $2l \leq s - 1$.
- Fundamental problem

$$\begin{cases} -\alpha c_l \partial_\sigma^2 \overline{Q}^0 = c_l \delta_0, \\ \overline{Q}^0(\pm 1) = 0, \\ \displaystyle\int_{-1}^{1} \overline{Q}^0(\sigma)\mathrm{d}\sigma = 1, \\ c_l \neq 0. \end{cases}$$

- Only fixes $\alpha = 1/2$.

Example: The case $\tilde{c}_0 = 0$ (II)

- Non trivial following interior problem on $\overline{Q}^{s-2l}$.
  Incompatibility with $c_l \neq 0$ if $l \leq s - 2l$ so $s \leq 3l - 1$ and $l \geq 2$.
- c_l still not defined!
- Next problem is for $\overline{Q}^{2s-4l}$.
  Incompatibility with $c_l \neq 0$ except if $l = 2s - 4l$ that is $2s = 5l$.
- $c_l = 1/(3\sqrt{2})^{2/5}$.
- Minimize the number of profile to compute by taking $l = 2$ and $s = 5$.

Convergence Theorem

Theorem (J. O., M. RENARDY (2010))

For all $\alpha > 0$ the described ansaetze converge (that is the series converge on small intervals $[0, \varepsilon(\alpha)[$) in weighted Sobolev spaces.

Corollary

The macroscopic stress $\tau(\dot\gamma) = \int_{\mathbf{R}} \sigma p^{\dot\gamma}(\sigma) \mathrm{d}\sigma$ has the following expansion:

$$\alpha > 1/2:\ \tau(\dot\gamma) = \eta_0\dot\gamma + \eta_1\dot\gamma^2 + \eta_2\dot\gamma^3 + \dots$$

$$\alpha < 1/2:\ \tau(\dot\gamma) = \tau_0 + \tau_1\sqrt{\dot\gamma} + \tau_2\dot\gamma + \dots$$

$$\alpha = 1/2:\ \tau(\dot\gamma) = A_0\dot\gamma^{1/5} + A_1\dot\gamma^{2/5} + A_2\dot\gamma^{3/5} + \dots$$

Proof

- Rewrite as a two parameter model in $a = \dot{\gamma}/\Gamma$ and $b = \sqrt{\Gamma}$ (plus one for the exponential decay).
- Use analytic perturbation “à la Kato” + analytic local inversion theorem.

Multidimensional flows

Some rheological properties cannot be accessed in simple shear flows.

Example : normal differences.

Extension from simple shear models to general flow models : not systematic.

Example : Maxwell 1d $\rightarrow$ Upper/under Convected Maxwell.

Typical rheological flows

- Shear flows:

$$\nabla u = \dot{\gamma} \begin{pmatrix} 0 & 1 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

- 3d elongational flow

$$\nabla u = \dot{\gamma} \begin{pmatrix} 2 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & -1 \end{pmatrix}$$

- 2d axisymmetric elongational flow

$$\nabla u = \dot{\gamma} \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

Frame indifference

Principle of frame indifference

The constitutive law of a material must be the same in all frames.

- If a block of material at point x has stress tensor Σ then in the frame $x^* = x_0(t) + Q(t)x$ it must have the stress:

$$\Sigma^* = Q\Sigma Q^{-1}.$$

- Consequence: a model based on $p(t, x, \Sigma)$ must be invariant by the change $p^*(t, x^*, \Sigma^*)$

A possible extension

- Works only for incompressible flows
- Σ: generic point of the space of symmetric tensors (not traceless)

$$
\begin{aligned}
&\partial_t p(t,x,\Sigma) + u\cdot\nabla_x p - g_a(\Sigma,\nabla u) : \nabla_\Sigma p = \\
&-2\lambda D(\nabla u) : \nabla_\Sigma p(t,x,\Sigma) \\
&\qquad - \frac{\mathbf{1}_{|\Sigma|>1}(\Sigma)}{We} p(t,x,\Sigma) + \frac{1}{We}\Gamma(p)(t,x)\rho(\Sigma) \\
&\qquad\qquad + \frac{\alpha}{We}\Gamma(p)(t,x)\Delta_\Sigma p(t,x,\Sigma)
\end{aligned}
$$

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$$\partial_t p(t,x,\Sigma) + u\cdot\nabla_x p - g_a(\Sigma,\nabla u) : \nabla_\Sigma p = \\ -2\lambda D(\nabla u) : \nabla_\Sigma p(t,x,\Sigma) \\ - \frac{\mathbf{1}_{|\Sigma|>1}(\Sigma)}{We} p(t,x,\Sigma) + \frac{1}{We}\Gamma(p)(t,x)\rho(\Sigma) \\ + \frac{\alpha}{We}\Gamma(p)(t,x)\Delta_\Sigma p(t,x,\Sigma)$$

Glass transition in this model?

Theorem

1. *Same formal asymptotic expansion as in the $1d$ case.*
2. *There exists $\alpha_c(\rho)$ so that*

   if $\alpha > \alpha_c$, *"Newtonian" or "threshold"*
   if $\alpha = \alpha_c$, *"power-law" or "threshold"*
   if $\alpha < \alpha_c$, *only "threshold"*

Some of the difficulties

- If one wants $\rho = \delta_0$: non variationnal elliptic theory.
- Unable to compute any of the profiles.
- Needs some properties on the characteristics of $\Sigma \mapsto g_a(\nabla u, \Sigma) - 2\lambda D(\nabla u)$.
- Careful study of the symmetry of the profile equations.
- ...