CATCH-U-DNA
UAM group
Theory and simulations

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Modelling of Quartz Micro Balance (QCM)

1. **Reproduce experiments** (results/trends)
   - Frequency and decay rate shifts, ΔF, ΔD
   - Relation between acoustic ratio (dissipation/frequency) and molecule intrinsice viscosity.
2. **Insights**
   - How the mechanic energy dissipates into the fluid-macromolecule system?
   - Role of the protein linker in dissipation.
   - Origin of the intercept "a" in $\Delta D/\Delta F = a + m\,[\eta]$ (related with the linker stiffness).
   - Plateau regime in the relation $\Delta D/\Delta F$ versus $[\eta]$ (viscous limit?)

Modelling of Quartz Micro Balance

Figure 2 shows the relation between experimental $\Delta D/\Delta F$ values and the corresponding intrinsic viscosities for linear

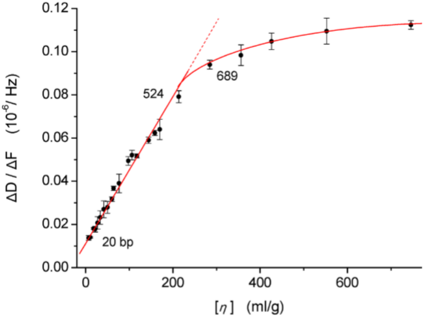

Figure 2. Acoustic ratio/intrinsic viscosity relationship for linear dsDNA for the 35 MHz frequency of the QCM device. The $[\eta]$ values are from the experimentally obtained eq 5. The numbers adjacent to the line indicate the number of base pairs giving an approximate value of 600 bp at the "break" point.

- **Quarzt wall**

$$\ddot{x} + \omega_0^2 x - \frac{\eta}{\sigma} \left. \frac{\partial v_x(z,t)}{\partial z} \right|_{z=0}$$

- **Fluid** (Unsteady base flow)

$$\rho_f \frac{\partial v_x}{\partial t} = \eta \frac{\partial^2 v_x}{\partial z^2}$$

- $\omega_0 = (k_w/m)^{1/2}$ fundamental QCM-frequency
- $\sigma = m/A$ surface mass density of the wall
- Solution (normal modes) $x(t) \sim \exp[-\lambda t]$, $v_x(z,t) \sim f(z) \exp[-\lambda t]$
- Complex frequency

$$\lambda = \omega + i\Gamma$$

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$$\lambda = \omega + i\Gamma$$

- Define $\Lambda = \lambda/\omega_0$
- Characteristic equation

$$\left(\Lambda^2 + 1\right)^2 = C\,\Lambda^3$$

- The C parameter:

$$C = \frac{\eta\rho_f}{\omega_0\sigma^2}. \quad (1)$$

- Experimental value: $C \simeq 10^{-8}$

$$C = \frac{\eta \rho_f}{\omega_0 \sigma^2}. \tag{2}$$

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Meaning of C

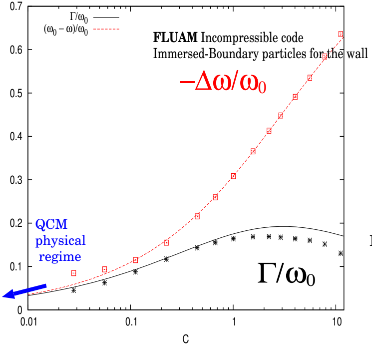
- QCM without DNA (solvent)
  - $\Delta w = \omega - \omega_0 < 0 \quad \text{Oscillation}$
  - $\Delta \Gamma = \Gamma - 0 > 0 \quad \text{Dissipation rate}$
- For very small C (i.e. $\omega \simeq \omega_0$)
  - $\Delta w / \omega_0 = 2^{-3/2} C^{1/2}$
  - $\Gamma / \omega_0 = 2^{-3/2} C^{1/2}$
- Number of oscillations before decay

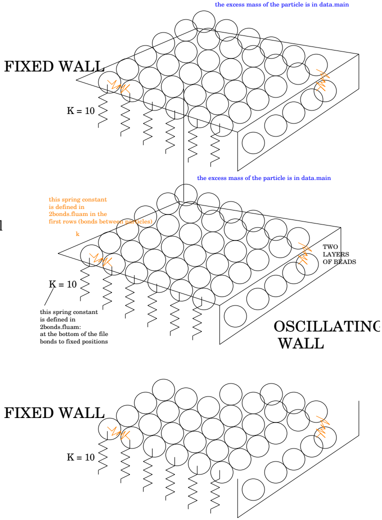
$$\frac{\tau_{osc}}{\tau_{decay}} = 2\pi \frac{\Gamma}{\omega_0} \simeq 10^5$$

$$C = \eta\, \rho_f / (\omega_0\, \sigma^2)$$

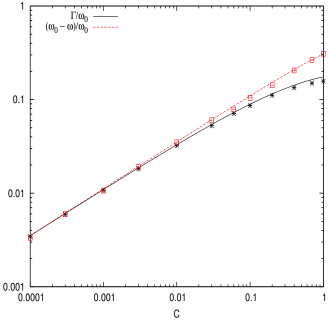
FLUAM Incompressible code
Immersed-Boundary particles for the wall

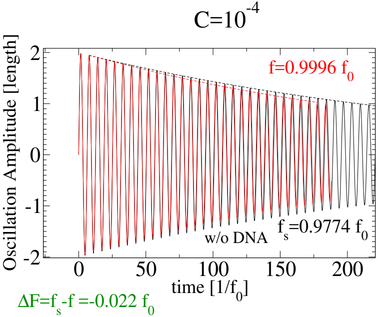
Is not able to resolve C<0.01





Solution: Coupling wall equations and base flow





C=10^{-4}

- Quarzt wall

$$m\ddot{x} + k_w x - \eta A \left. \frac{\partial v_x(z,t)}{\partial z} \right|_{z=0} - K_0 \left(x - X_0 \right) = 0.$$

- Fluid (Unsteady base flow)

$$\rho_f \frac{\partial v_x}{\partial t} = \eta \frac{\partial^2 v_x}{\partial z^2}$$

- Polymer (Brownian dynamics)

$$dR_i = v_x(Z_i;t)dt + \boldsymbol{\mu} F_{\text{springs}}(\mathbf{R})\, dt + \mu K_0 \left(x - X_0 \right) \delta_{Kr}(i,0)\, dt + d\tilde{R}_i$$

UAMMD GPU-code by Raul Pérez Peláez under supervision of R. Delgado-Buscalioni.

- Brownian dynamics without hydrodynamic couplings between DNA beads: $\mu = D_0/k_B T$ **scalar mobility**
- Brownian dynamics **with** hydrodynamic coupling between DNA beads:

$$\boldsymbol{\mu} = \text{Blake Mobility tensor}$$

- Total velocity field is sum of BASE flow plus PARTICLE perturbations

$$\mathbf{v}(\mathbf{r}, t) = v_b(z, t)\hat{\mathbf{x}} + \mathbf{v}_p(\mathbf{r}(t))$$

- The perturbative velocity given by

$$\mathbf{v}_p(\mathbf{r}, t) = \int \boldsymbol{\mu}(\mathbf{r} - \mathbf{r}')\mathbf{F}(\mathbf{r}')d\mathbf{r}'$$

Why it is possible to use Brownian dynamics?

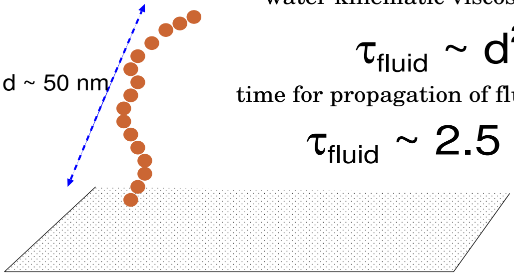
We are in the limit of validity of Brownian hydrodynamics

water kinematic viscosity $\nu = 10^{-6}\ m^2/s$

$$\tau_{fluid} \sim d^2/\nu$$

time for propagation of fluid momentum

$$\tau_{fluid} \sim 2.5\ 10^{-9}\ s$$



$$\tau_{osc} \sim 1/f_0 \sim 3\ 10^{-8}\ s$$

Period of oscillation for 35 MHz

$$\tau_{fluid}/\tau_{osc} \sim 10^{-1}$$

Transients (delays) in fluid propagation are not very important

But, what shall we do if f_0=1 GHz ?

FLUAM with compressible fluid (ongoing work)

$$\begin{aligned}
\text{Fluid} \quad &: \quad \rho \left[\frac{\partial \mathbf{v}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{v} \right] = -\nabla p(\rho) + \eta \nabla^2 \mathbf{v} - \sum_i \mathbf{F}_i^{(fl)} \, \mathrm{S}(\mathbf{q}_i - \mathbf{r}) \\
\text{DNA beads} \quad &: \quad m_e^{(i)} \frac{d^2 \mathbf{q}_i}{dt^2} = \mathbf{F}_i^{(fl)} + \mathbf{F}_{springs} \\
\text{Quarzt wall} \quad &: \quad m \ddot{x} = k_w x - \eta A \left. \frac{\partial v_x(z,t)}{\partial z} \right|_{z=0} - K_0 \left(x - X_0 \right)
\end{aligned} \tag{3}$$

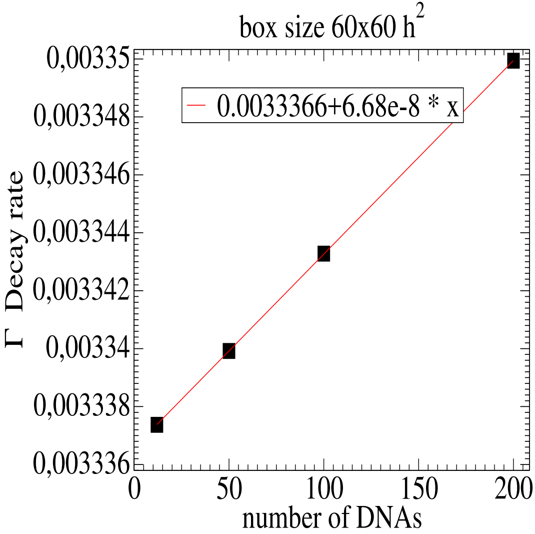
- Explicit solution of wall dynamics: $x(t)$
- Rigid boundary to the fluid with velocity $v_x(z = 0, t) = dx/dt$
- Drag evaluated numerically

- Brownian dynamics without hydrodynamic couplings
- RELEVANT non-dimensional parameters
  - C-parameter $C = 10^{-4}$
  - Penetration lenght $\delta = (\nu/2\omega_0)^{1/2} \rightarrow 2\, L_{DNA}$
  - DNA-bead radius $h \simeq 2\mathrm{nm}$
  - DNA-contour-lenght $L_{DNA} = (N-1)\, h = 50\mathrm{nm}$
  - DNA-persistence-lenght $\mathcal{L}_p = k_{bend}/k_B T$

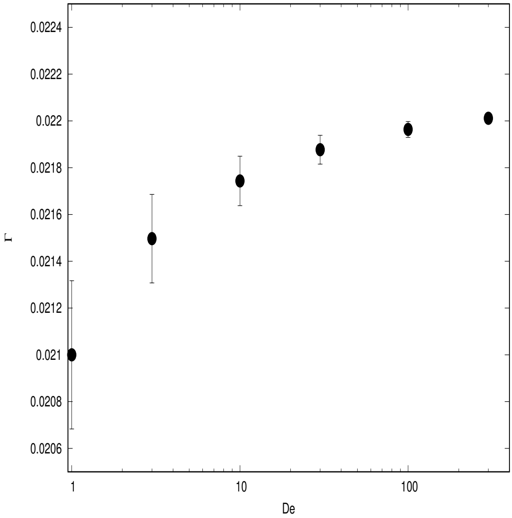
$$U_{bend} = -(k_{bend}/2)\left(\sin(\theta/2) - \sin(\theta_0/2)\right)^2$$

  - Deborah number $\mathrm{De} = \tau_{DNA} f_0$

Dissipation Γ VERSUS Surface coverage



Dissipation Γ VERSUS Deborah number (Deboray increases with chain lenght and stiffness)



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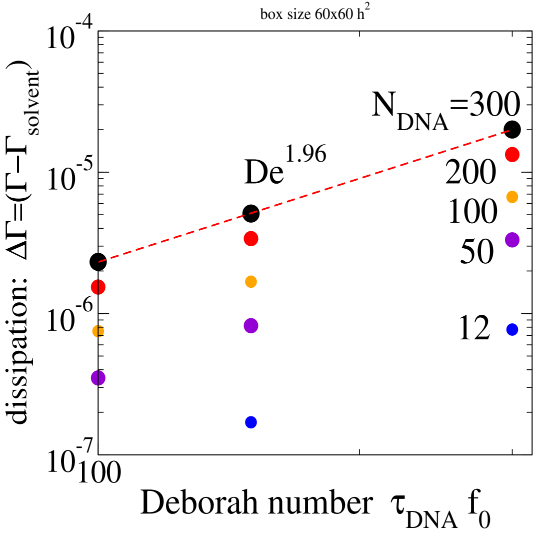

$$(L_p = 20,\ L_{DNA} = 50)$$

DNA-danze with BIG HEAD)

$(L_p = 20,\ L_{DNA} = 50$

Conclusions: Very Preliminary Work

- FLUAM-incompressible with beads-as-wall cannot be used for the physical regime of QCM
- UAMMD Brownian dynamics is consistent with the separation of time scales for $f_0 \sim 10MHz$.
- FLUAM-compressible resolves transient fluid-momentum transport should be valid for any f_0, but longer simulations (under implementation).
- Dissipative particles
  - **Easy-model for dissipation particles can be implemented using slow-diffusion bead at the end of the chain (video)**
  - It seems that the "two-fixed-ends" of the chain leads to stronger dissipation.