# X-ray radiography of granular systems – particle densities and dynamics

FRIEDRICH-ALEXANDER
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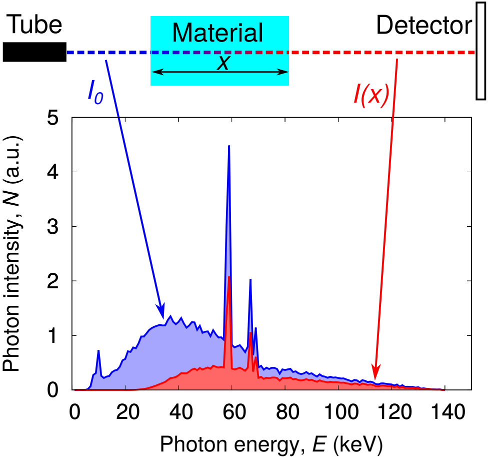
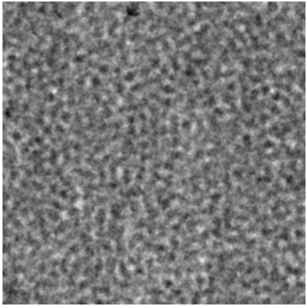
Institute for
Multiscale Simulation
MSS

PhD thesis
**Manuel Baur**



Funded by the German Federal Ministry for Economic Affairs and Energy, grant no. 50WM 1653

# Granular flows - are ubiquitous

replace text by images:

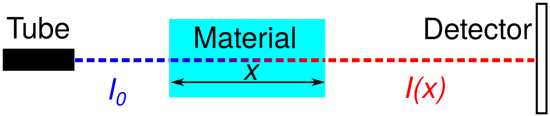
- Granular flows - fluidized bed reactors
- optically opaque
- X-ray reveal the inside
- Tomography: full 3D information - but slow (no dynamics)
- Radiography: single projections - knowledge on imaging physics
- Here two techniques to quantify: densities & dynamics

# Granular flows - are ubiquitous

Study of granular flows difficult, because opaque: optically not possible
Video of Welm Pätzold’s Master thesis - only boundary observable
here X-rays
tomogram of particles
video of radiograms - check what is interesting

# Correction of beam hardening in X-ray radiograms

# Attenuation of X-rays

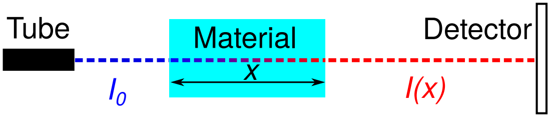



Beer-Lambert's law

$$I(x) = I_0 \exp(-\mu\, x)$$

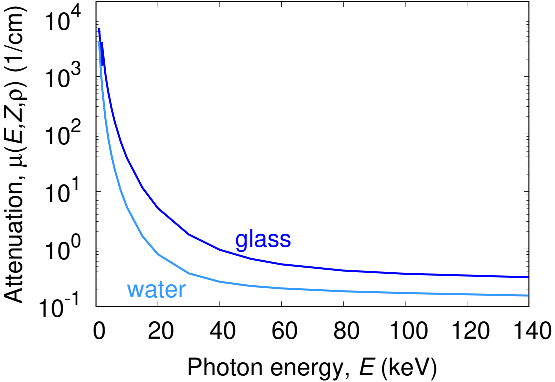
Thickness: $x = -\frac{1}{\mu} \ln \frac{I(x)}{I_0}$

# Attenuation of X-rays




Beer-Lambert's law

$$I(x) = I_0 \exp(-\mu(E, Z, \rho)\, x)$$
$$\mu \neq \text{const}, x = ?$$




`https://www.nist.gov/pml/x-ray-mass-attenuation-coefficients`

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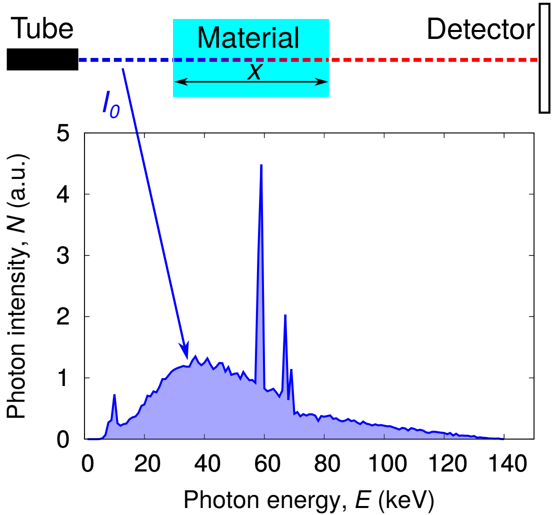


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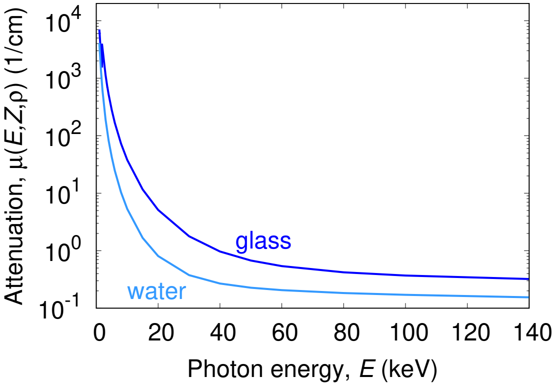


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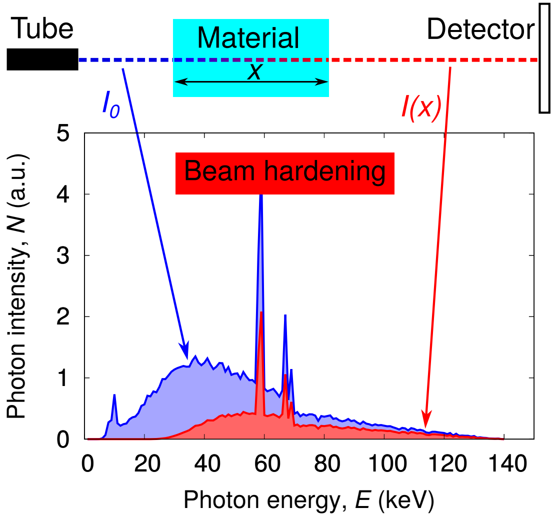


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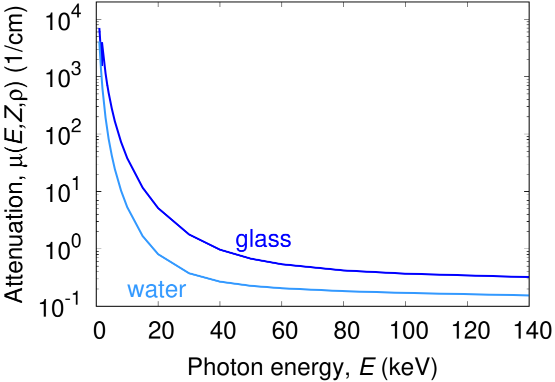


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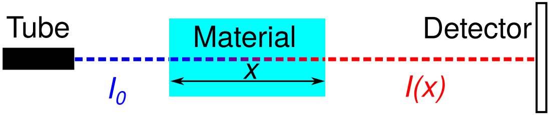
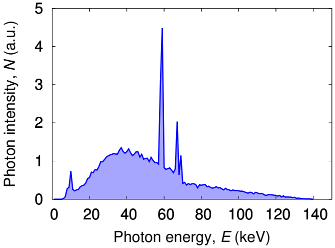
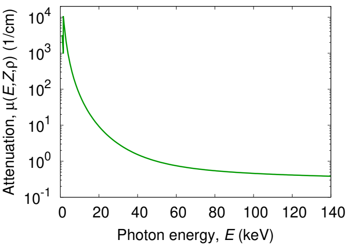
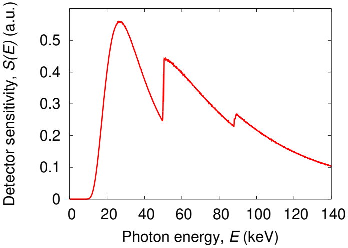
# Measurement of X-ray intensities



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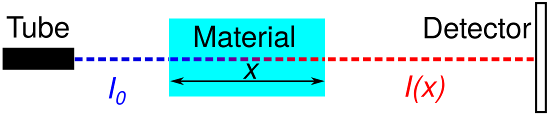
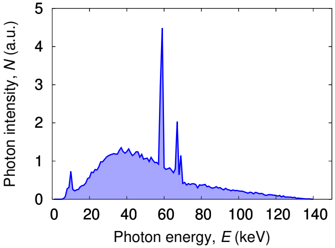
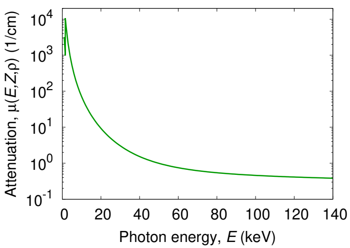
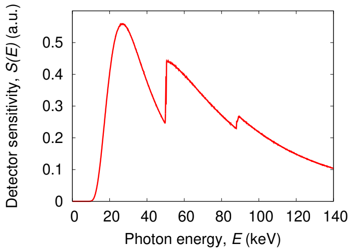


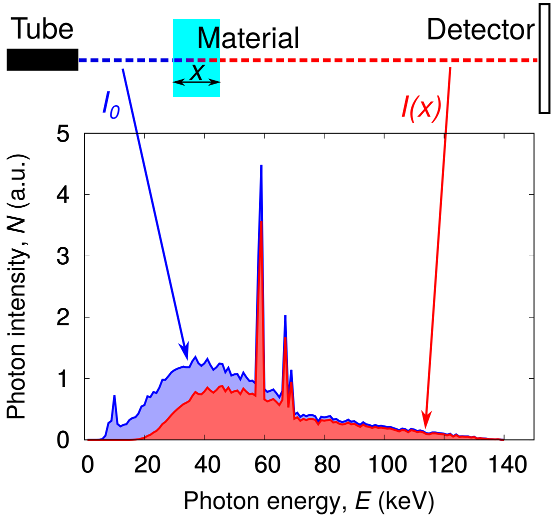



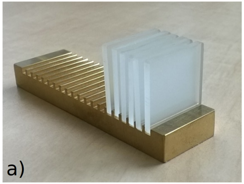



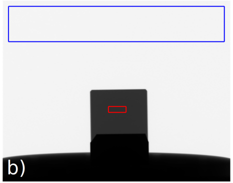




$$I(x) \propto \int N(E)\, \exp\{-\mu(E, Z, \rho)x\}\, S(E)\, \mathrm{d}E$$

# Energy averaged attenuation coefficient, $\mu_{\mathrm{eff}}$

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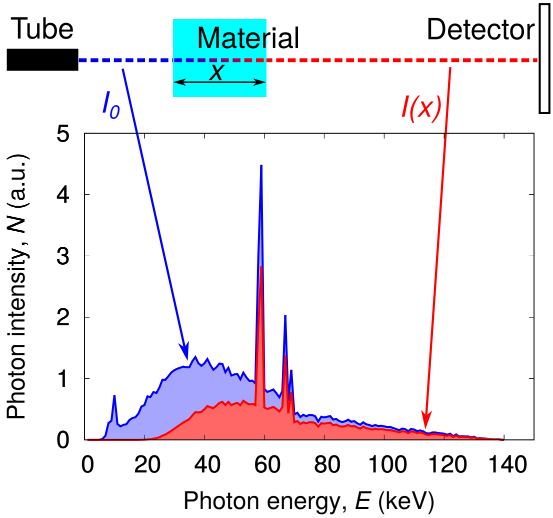


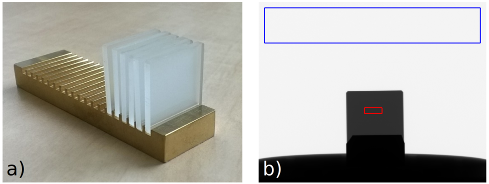


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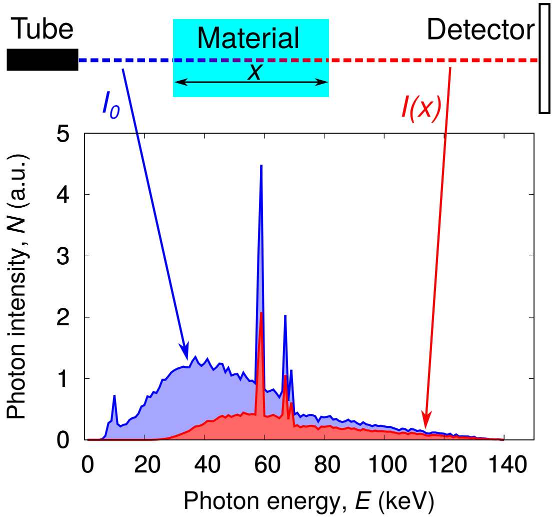


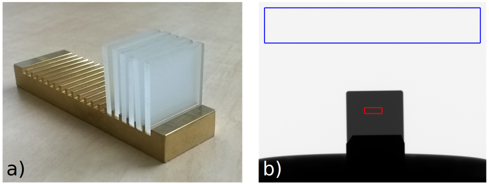


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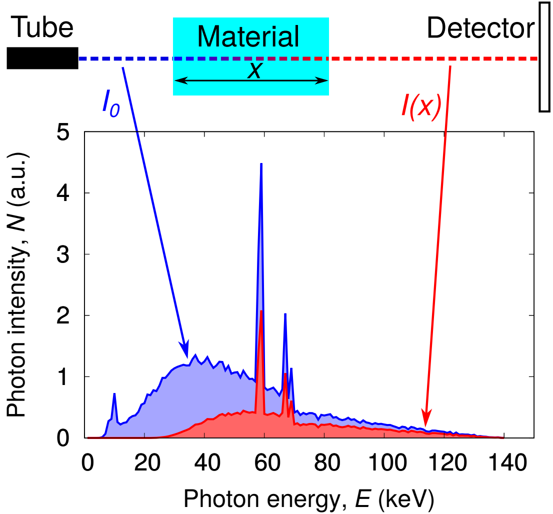


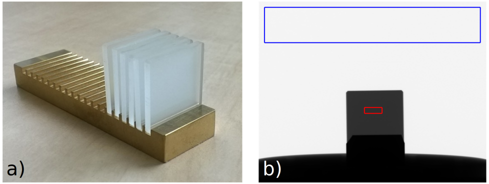


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*effective* attenuation coefficient:

$$I(x) = I_0 \exp(-\mu_{\text{eff}}(x)\, x)$$

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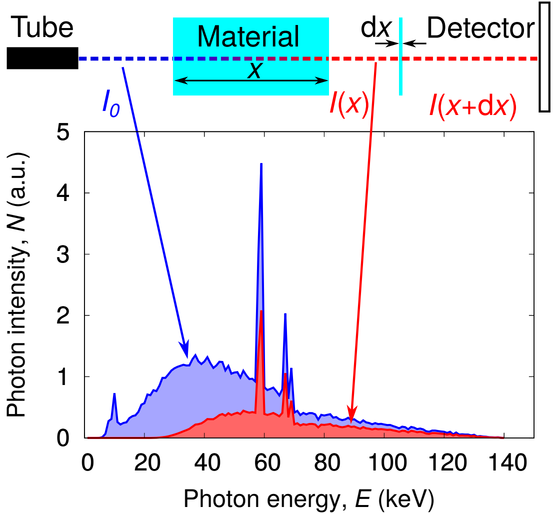


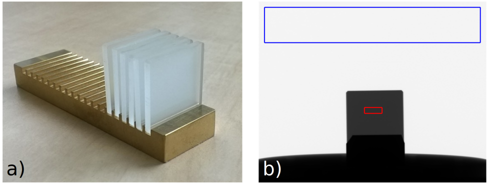


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*effective* attenuation coefficient:

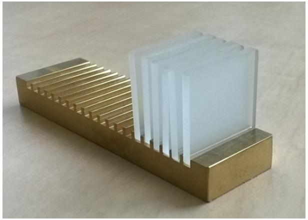
$$I(x) = I_0 \exp(-\mu_{\text{eff}}(x)\, x)$$

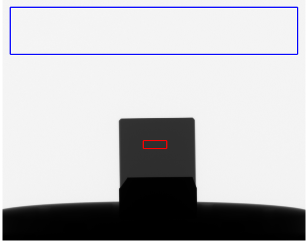
*differential* attenuation coefficient:

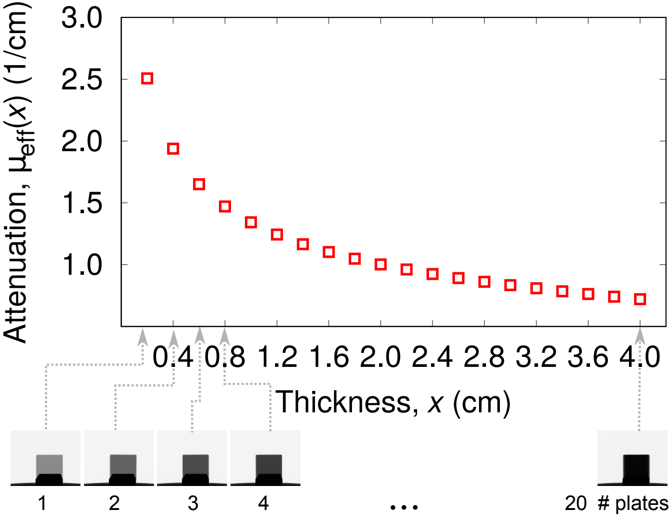
$$\frac{\mathrm{d}I(x)}{\mathrm{d}x} = \bar{\mu}(x)\, I(x)$$

# Experimental procedure

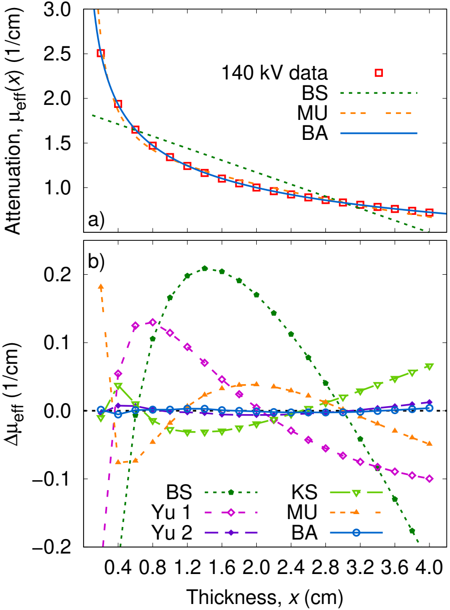
*effective* attenuation coefficient: $I(x) = I_0 \exp(-\mu_{\text{eff}}(x)\, x)$







# Heuristic model functions




$$\mu_{\text{eff}}(x) = \mu_0 - \lambda x$$

Bjärngard & Shackford (1994)

$$\mu_{\text{eff}}(x) = \frac{\mu_0}{1+\lambda x}$$

$$\mu_{\text{eff}}(x) = \frac{\mu_0}{(1+\lambda x)^\beta}$$

Yu *et al.* (1997)

$$\mu_{\text{eff}}(x) = \mu(E_{\text{max}}) + \frac{2\mu_1}{x\sqrt{-\lambda_1^2+4\lambda_2}} \times \left[\arctan\left(\frac{\lambda_1+2\lambda_2 x}{\sqrt{-\lambda_1^2+4\lambda_2}}\right) - \arctan\left(\frac{\lambda_1}{\sqrt{-\lambda_1^2+4\lambda_2}}\right)\right]$$

Kleinschmidt (1999)

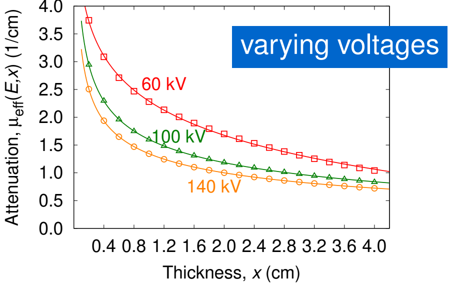
$$\mu_{\text{eff}}(x) = -\frac{1}{x}\ln\left[A + B\exp(-x/C)\right]$$

Mudde *et al.* (2008)

$$\mu_{\text{eff}}(x) = a + \frac{b}{x^\alpha}$$

Baur *et al.* (2019) (this work)

# Universality of $\mu_{\text{eff}}(x) = a + \frac{b}{x^{\alpha}}$



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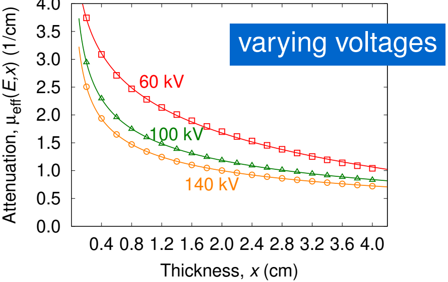


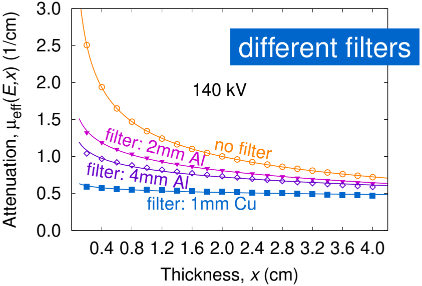

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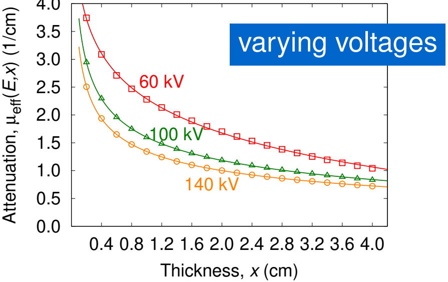


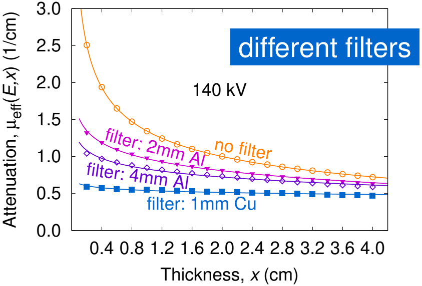


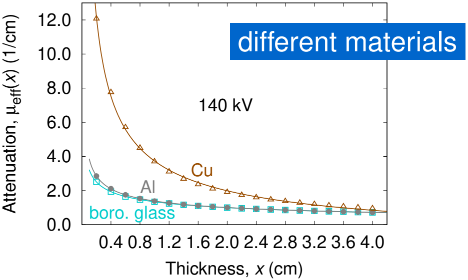

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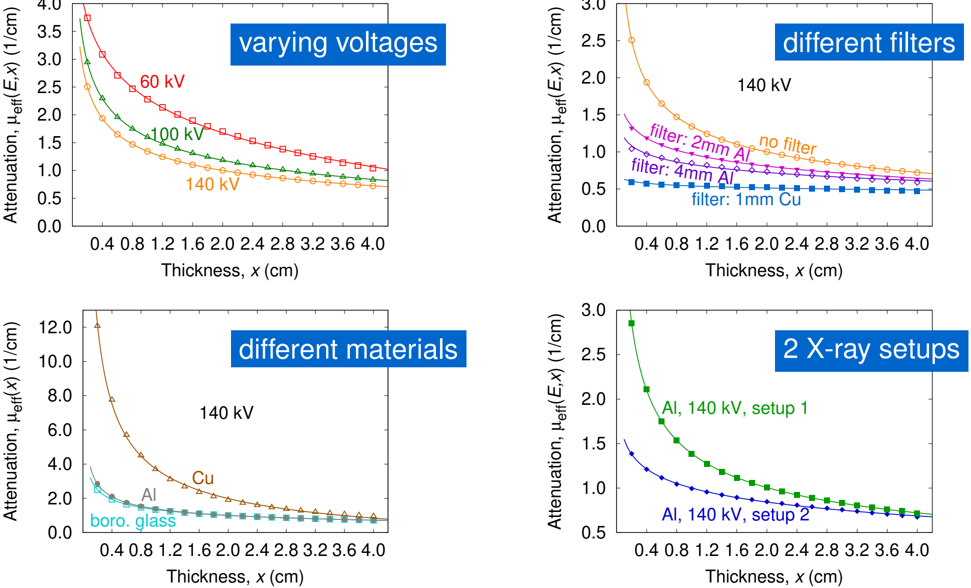


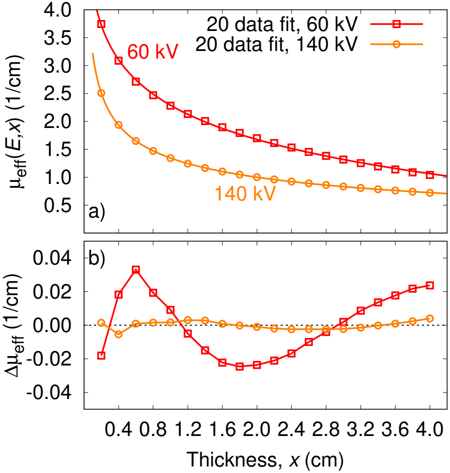
# Determining the material thickness $x$

Generalized Beer-Lambert

$$I(x) = I_0 \exp(-\mu_{\text{eff}}(x)\, x)$$

Model function

$$\mu_{\text{eff}}(x) = a + \frac{b}{x^{\alpha}}$$



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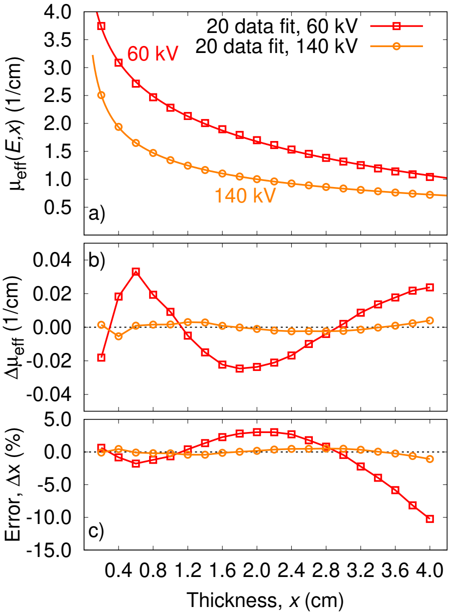
Solve

$$ax + bx^{1-\alpha} + \ln\left(\frac{I(x)}{I_0}\right) = 0$$

e.g. Newtons method or look-up table



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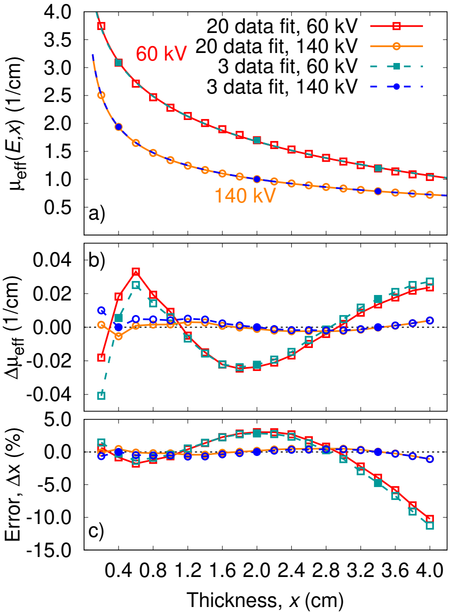
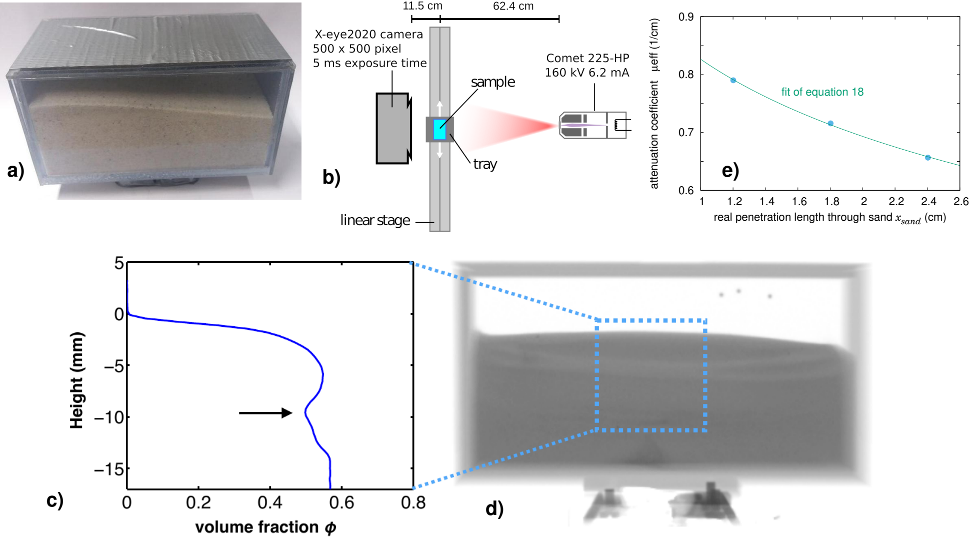
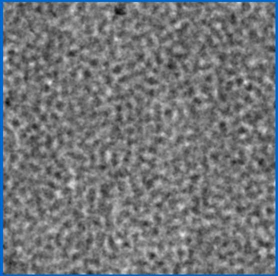


# Applications

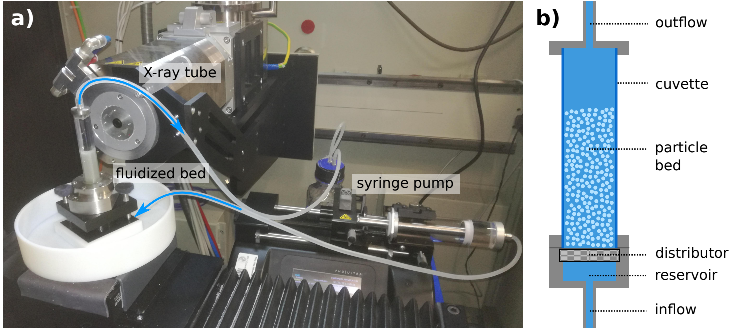


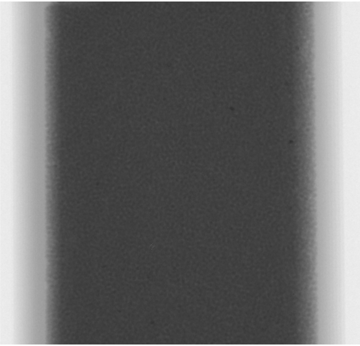
# X-ray Digital Fourier Analysis (X-DFA)

## A technique to measure granular dynamics



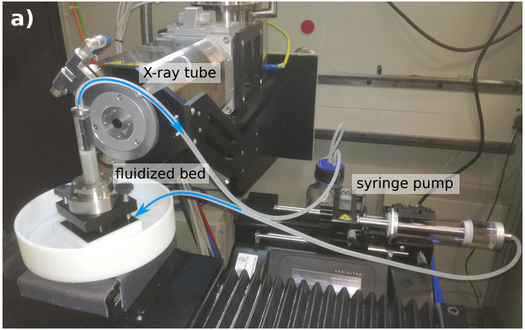
# Experiments

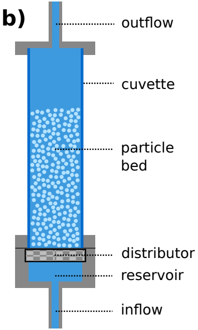


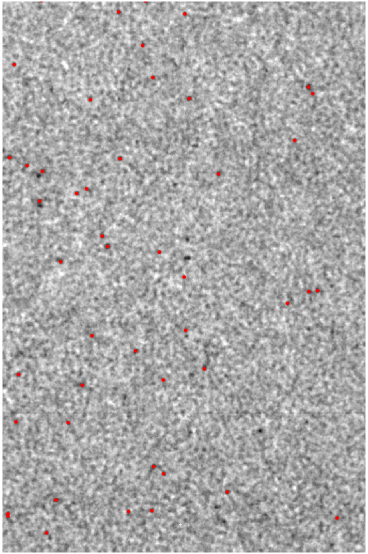



- volume fraction: $0.45 < \Phi < 0.56$
- control dynamics and $\Phi$ via pump rate

# Experiments



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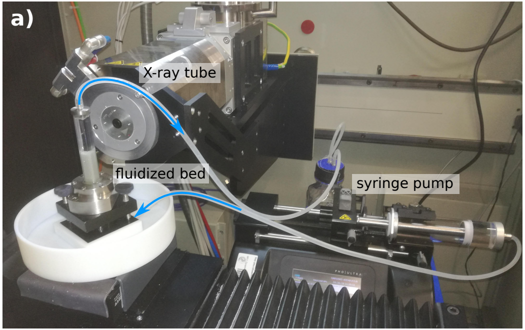


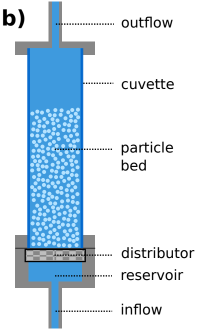


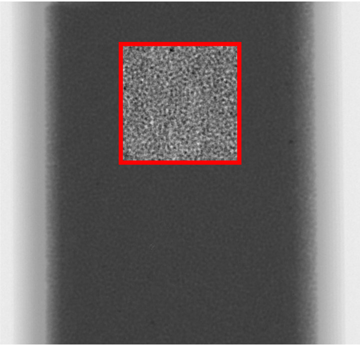

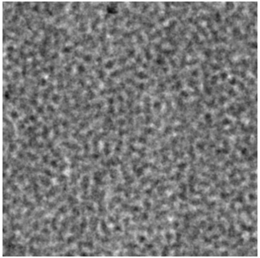

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# Differential Dynamic Microscopy (DDM)

| | **up to now** | **this work** |
|---|---|---|
| **system** | dispersion, gels | fluidized bed |
| **particles** | colloids | granulate |
| **part. diameter** | $< 1$ µm | $\approx 200$ µm |
| **volume fraction** | $\Phi \leq 0.33$ | $0.45 < \Phi < 0.56$ |
| **imaging** | light microscope | x-ray radiography |
| **dynamics** | Brownian motion, caging, glassy, collective motion | |

Giavazzi *et al*, PRE **80**, 031403 (2009)

# Extending Differential Dynamic Microscopy (DDM) to X-ray imaging

| | up to now | this work |
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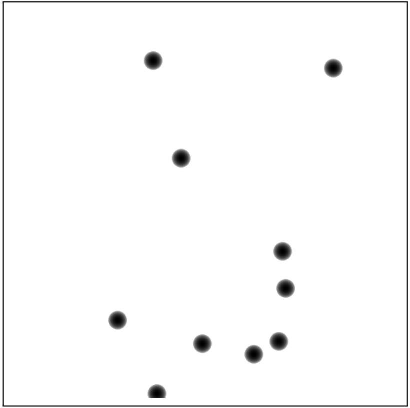
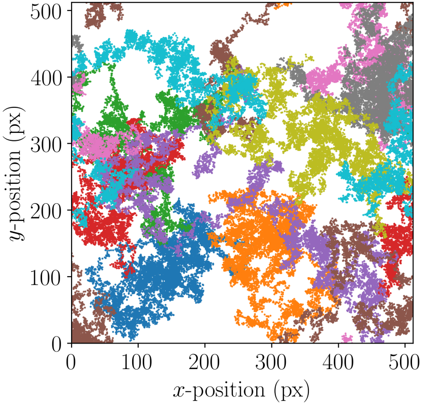
## Digital Fourier Analysis of X-Ray Radiograms (X-DFA)

Giavazzi *et al*, PRE **80**, 031403 (2009)

# Synthetic radiograms







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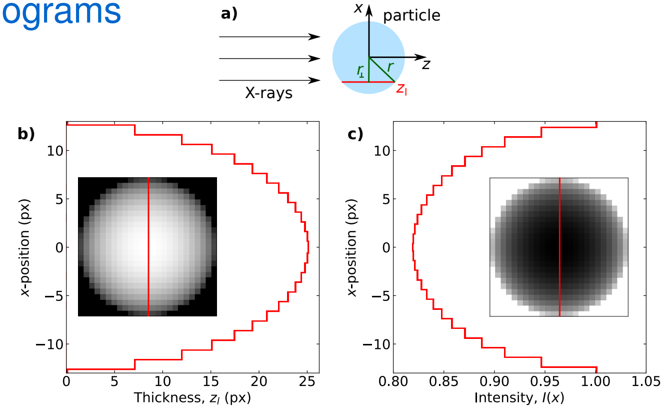
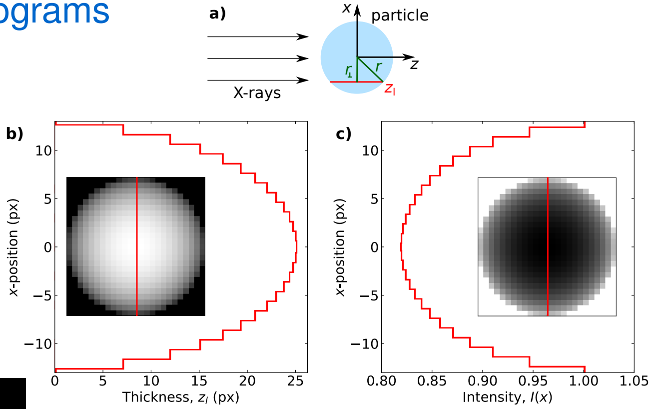
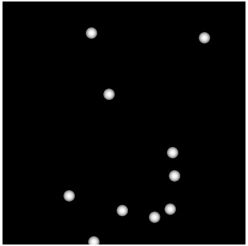



Beer-Lambert

$$I(z_l) = I_0 \exp(-\mu z)$$

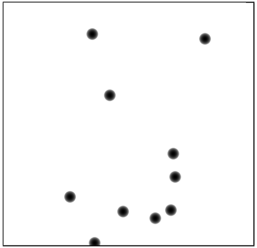
# Synthetic radiograms



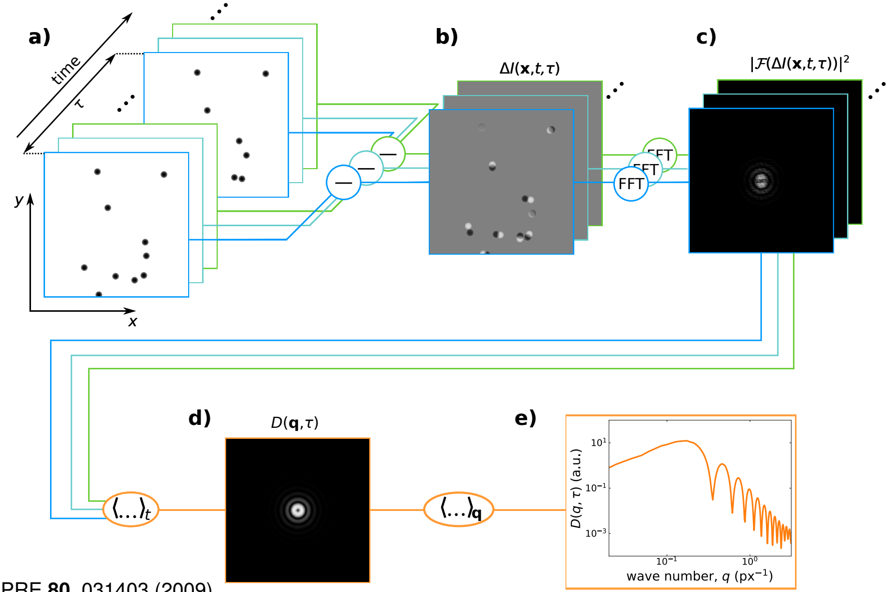



Beer-Lambert

$$I(z_l) = I_0 \exp(-\mu z)$$



# The image structure function $D(\mathbf{q}, \tau)$




Giavazzi *et al*, PRE **80**, 031403 (2009)

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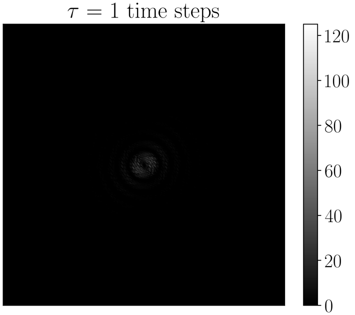
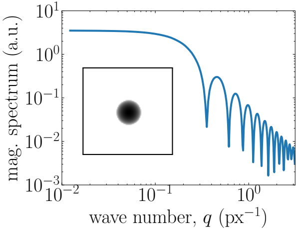
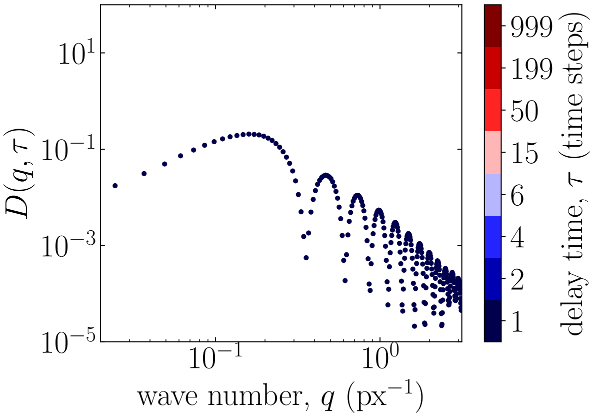



Time averaging: $D(\mathbf{q}, \tau) = \langle |\mathscr{F}(\Delta I)|^2 \rangle_t$

Azimuthal averaging: $D(\mathbf{q}, \tau) \rightarrow D(q, \tau)$ ...



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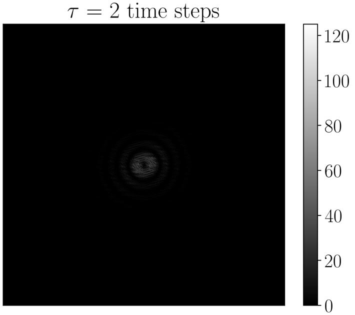


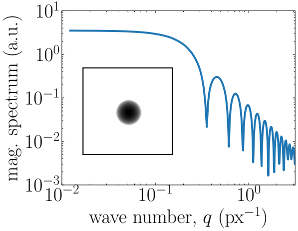


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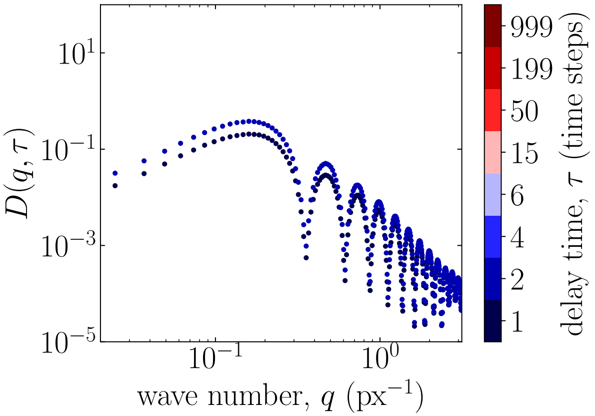

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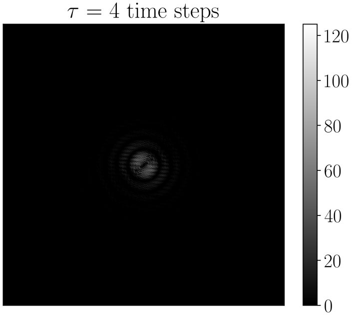


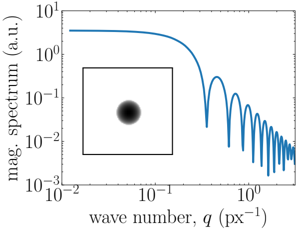


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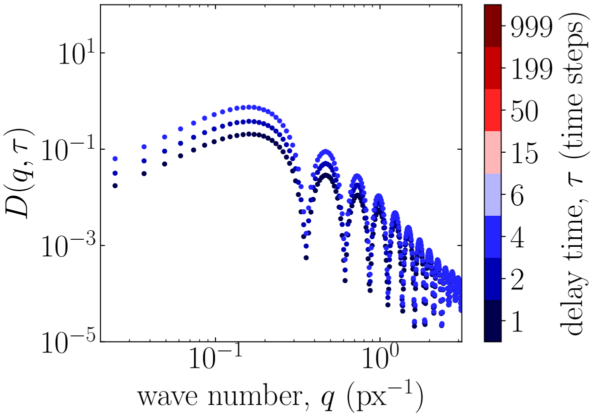
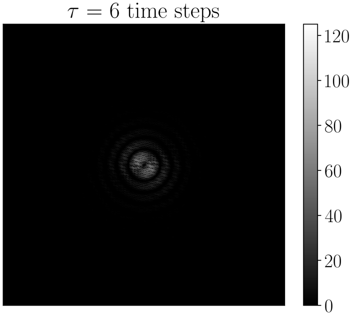
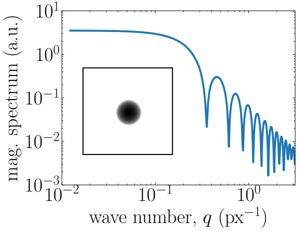


# The image structure function $D(q,\tau)$

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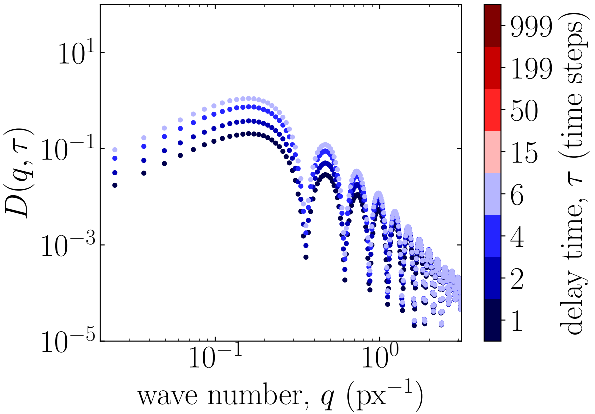

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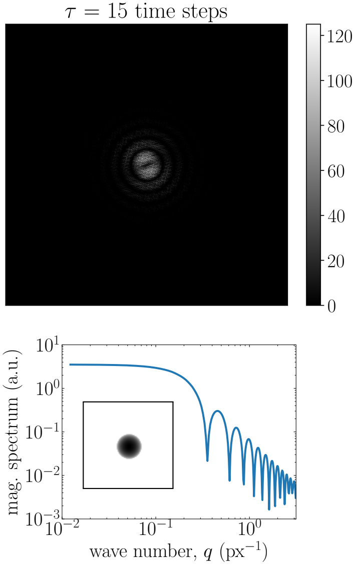


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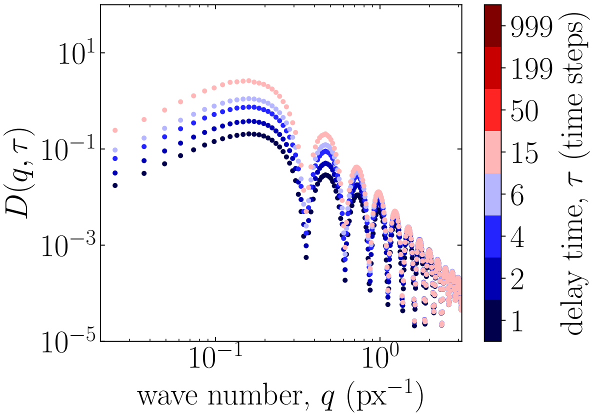

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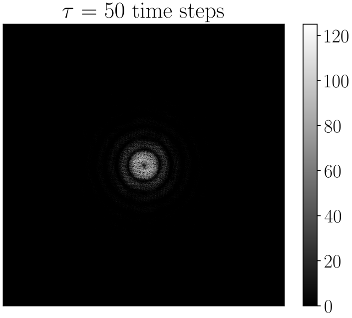


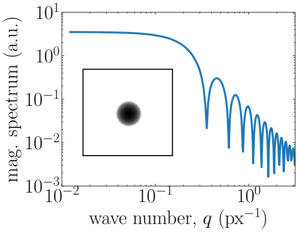


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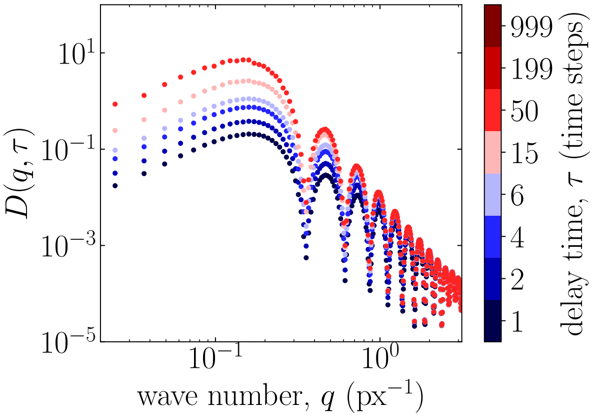

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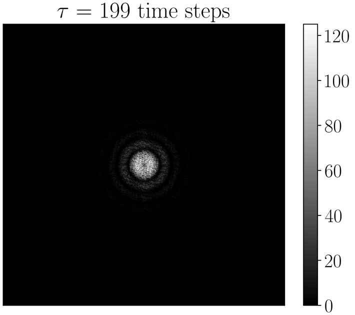


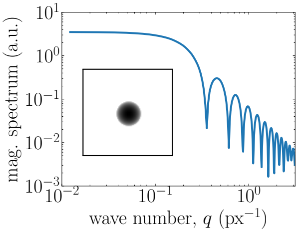


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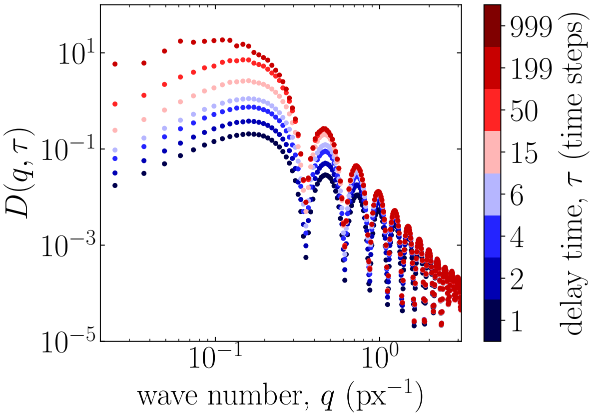

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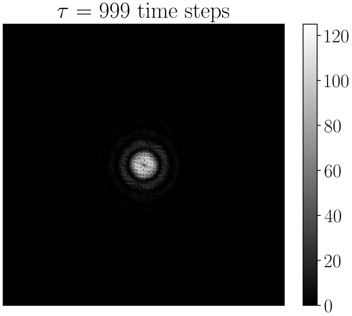


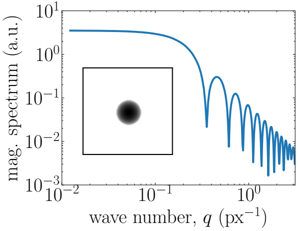


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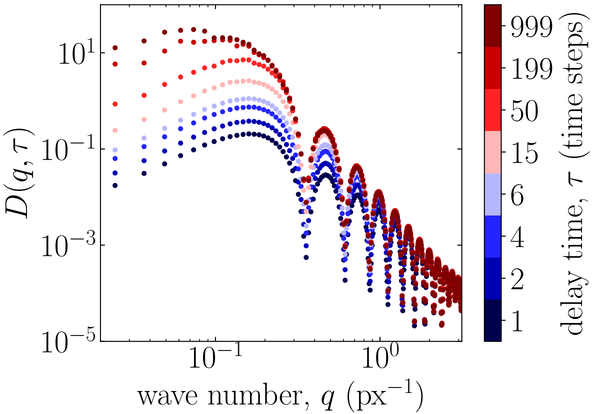

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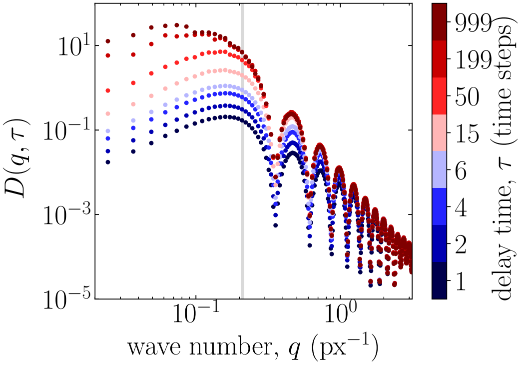


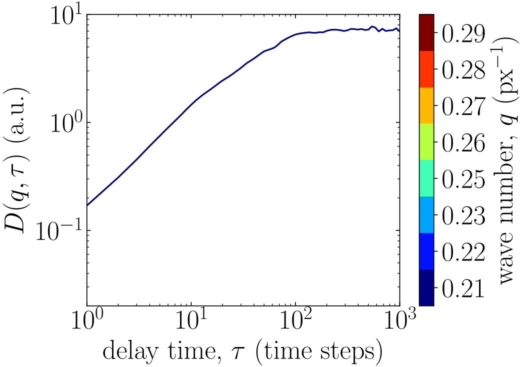

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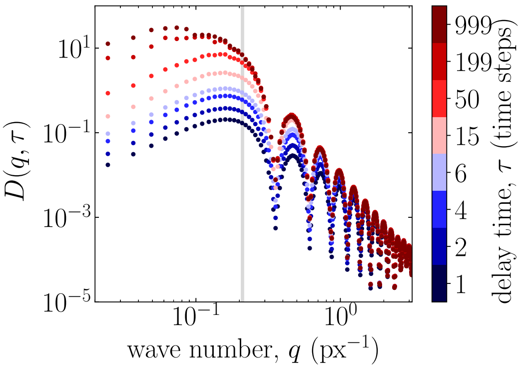


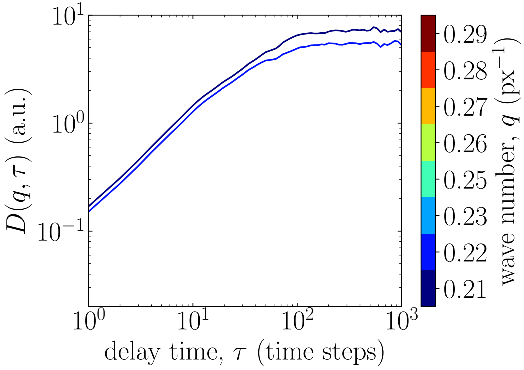

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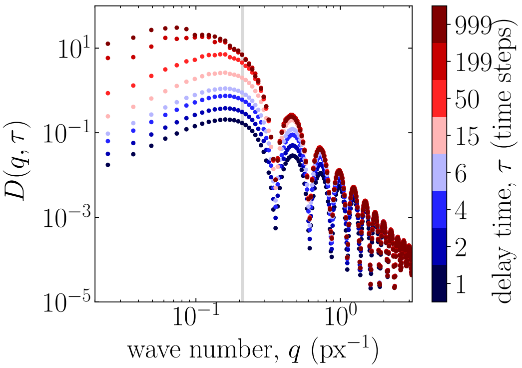


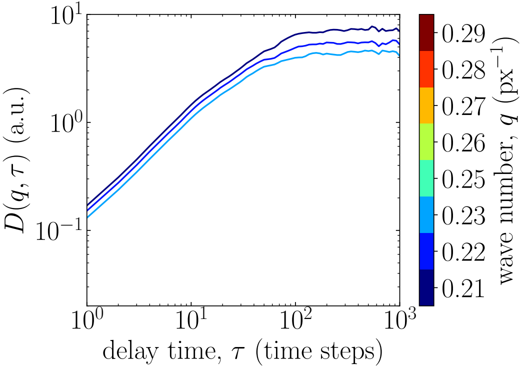

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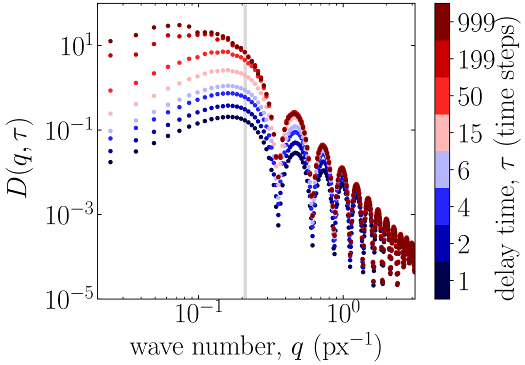
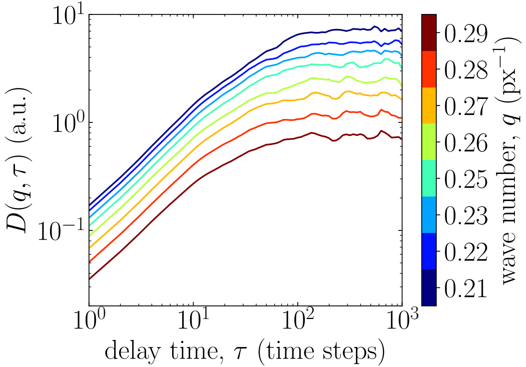
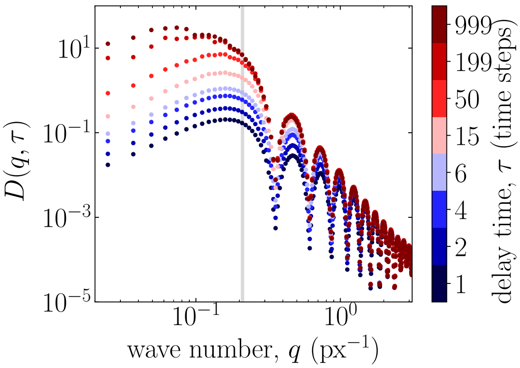


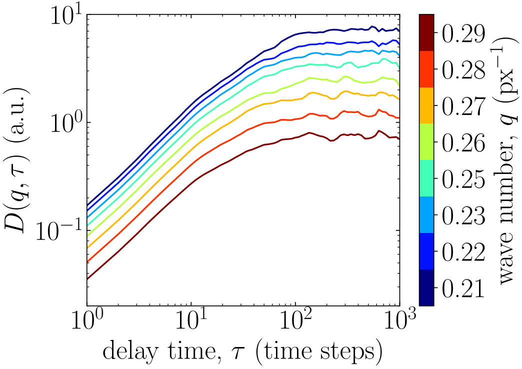




$$D(q, \tau) = \left\langle \left| I(q, t + \tau) - I(q, t) \right|^2 \right\rangle_t$$

$$= A(q) \left[ 1 - \frac{\left\langle I^*(q, t) I(q, t + \tau) \right\rangle_t}{\left\langle |I(q, t)|^2 \right\rangle_t} \right] + B(q)$$

# The image structure function $D(q, \tau)$




$$D(q,\tau) = \left\langle \left| I(q, t+\tau) - I(q,t) \right|^2 \right\rangle_t$$

$$= A(q)\left[1 - \underbrace{\frac{\langle I^*(q,t) I(q,t+\tau)\rangle_t}{\langle |I(q,t)|^2\rangle_t}}_{\text{image correlation function}}\right] + B(q)$$

# Linear space invariant imaging

image correlation function

$$g(\mathbf{q}, \tau) = \frac{\langle I^*(\mathbf{q}, t) I(\mathbf{q}, t + \tau) \rangle_t}{\langle |I(\mathbf{q}, t)|^2 \rangle_t}$$

# Linear space invariant imaging

image correlation function

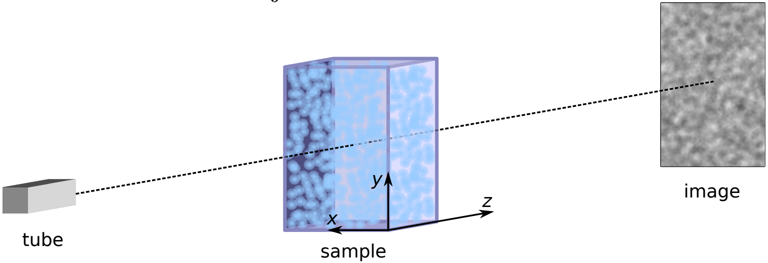
$$g(\mathbf{q},\tau) = \frac{\langle I^*(\mathbf{q},t) I(\mathbf{q},t+\tau)\rangle_t}{\langle |I(\mathbf{q},t)|^2\rangle_t}$$

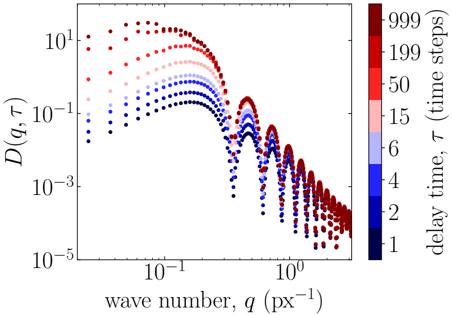
intermediate scattering function

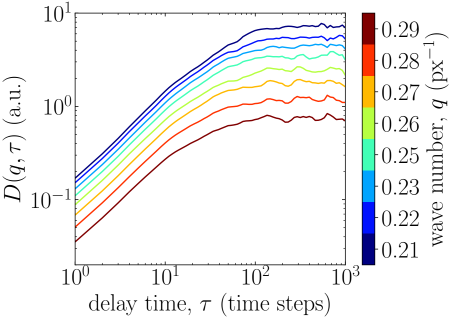
$$f(\mathbf{q},\tau) = \frac{\langle \rho^*(\mathbf{q},t) \rho(\mathbf{q},t+\tau)\rangle_t}{\langle |\rho(\mathbf{q},t)|^2\rangle_t}$$

Linear space-invariant imaging:

$$I(\mathbf{r},t) = I_0 + \int \mathrm{d}\mathbf{r}'\, \mathrm{d}z'\, T(\mathbf{r}-\mathbf{r}', -z') c(\mathbf{r}', z', t)$$

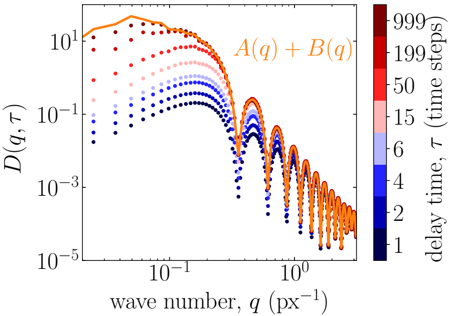


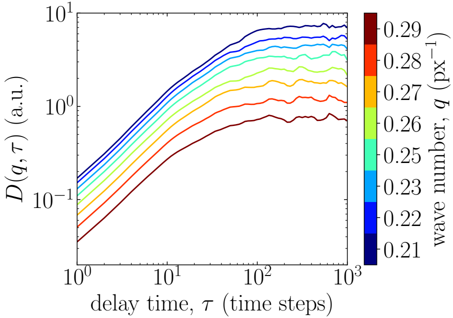



$$D(q,\tau) = \left\langle |I(q,t+\tau) - I(q,t)|^2 \right\rangle_t$$

$$= A(q)\left[1 - \underbrace{\frac{\langle I^*(q,t)I(q,t+\tau)\rangle_t}{\langle |I(q,t)|^2\rangle_t}}_{\text{image correlation function}}\right] + B(q)$$



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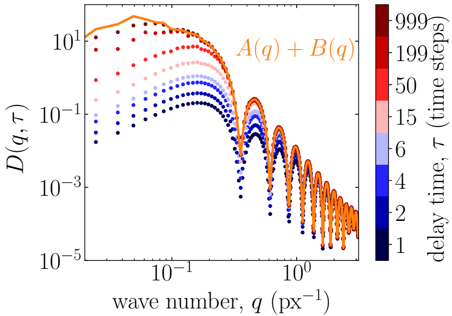
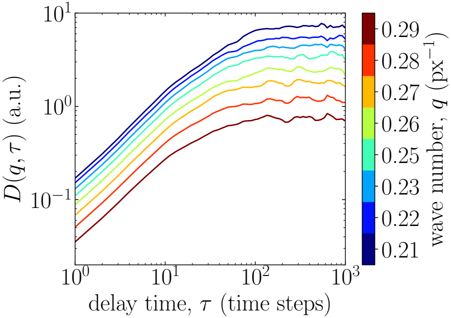
$$= A(q)\left[1 - \frac{\langle I^*(q,t)I(q,t+\tau)\rangle_t}{\langle |I(q,t)|^2\rangle_t}\right] + B(q)$$

- $d(q,\tau\to 0) = B(q) = 0$
- $d(q,\tau\to 0) = A(q) + B(q)$

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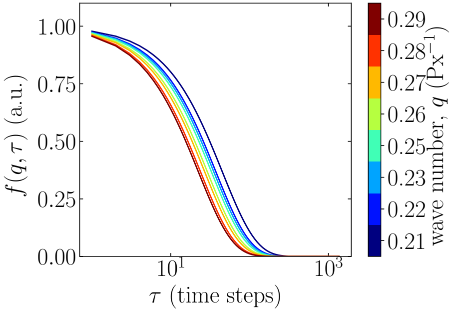
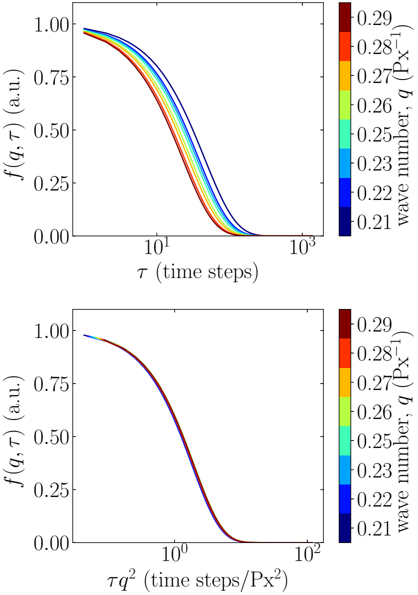


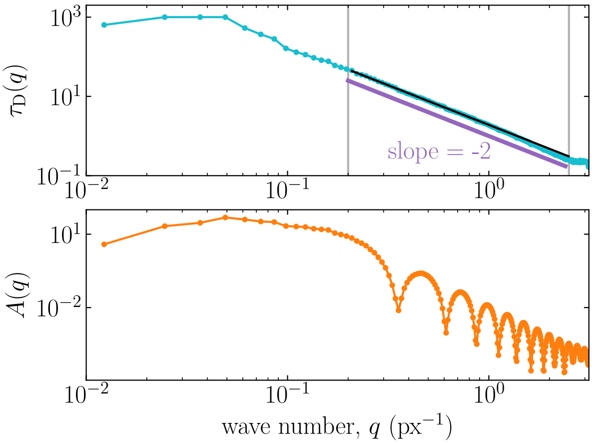
# Intermediate scattering function $f(q,\tau)$




Brownian motion:

$$f(q,\tau) = \exp(-q^2\tau/\tau_\mathrm{D})$$

Accuracy: 2% - 6%

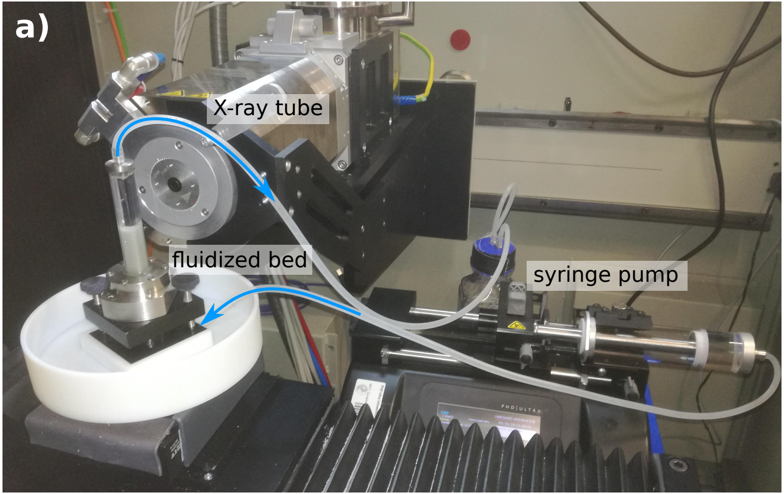


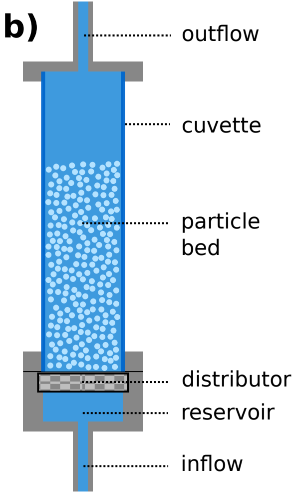
# Validating X-DFA on a sedimenting suspension

## Measurements on a fluidized bed

# Experimental validation: Sedimenting suspension

track sedimentation front $\leftrightarrow$ DFA



# Experimental validation: Sedimenting suspension

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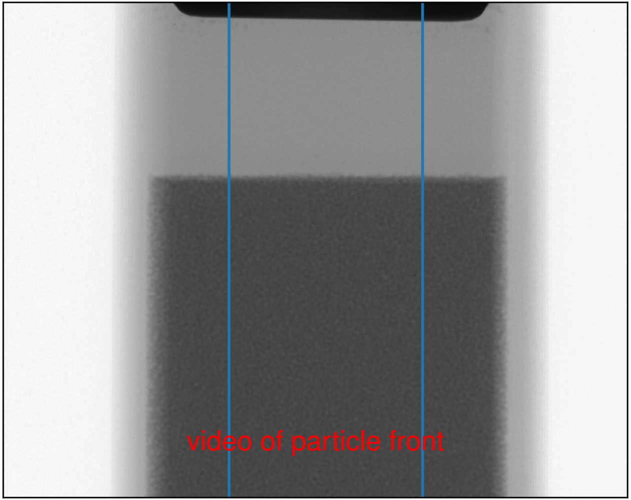
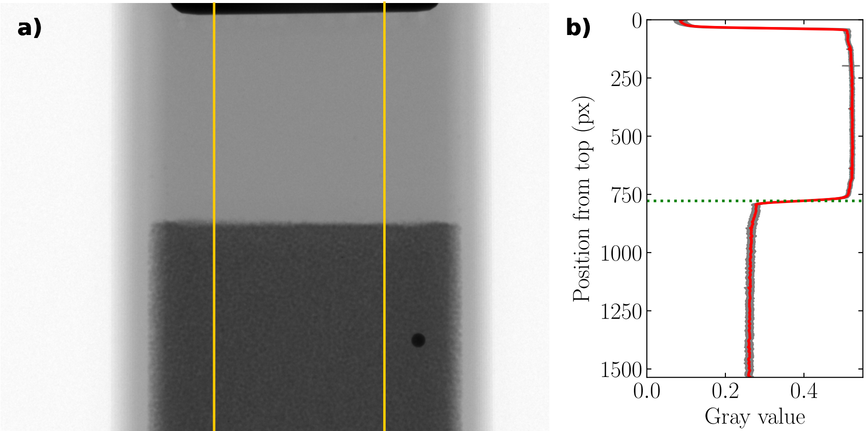


# Tracking of particle front



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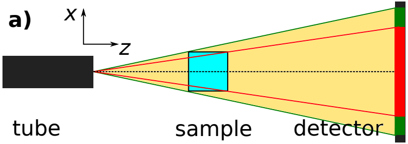


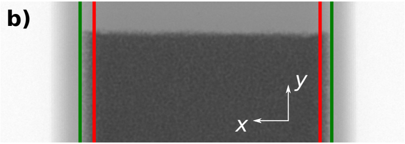


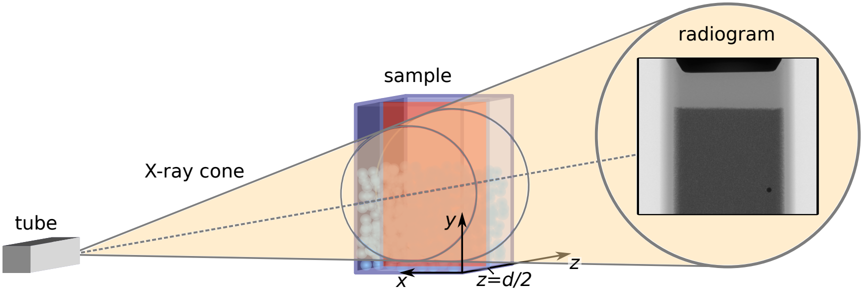

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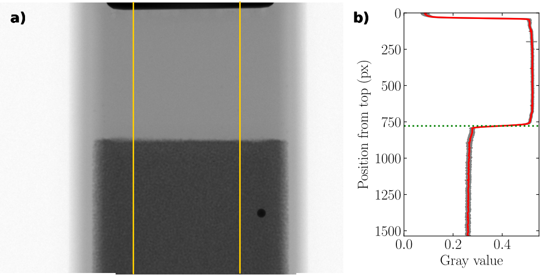
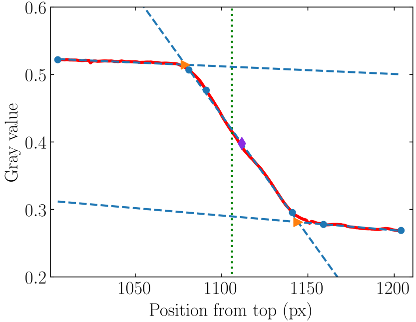
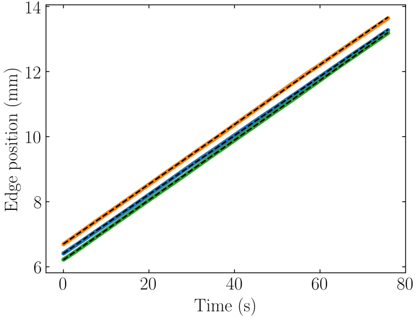
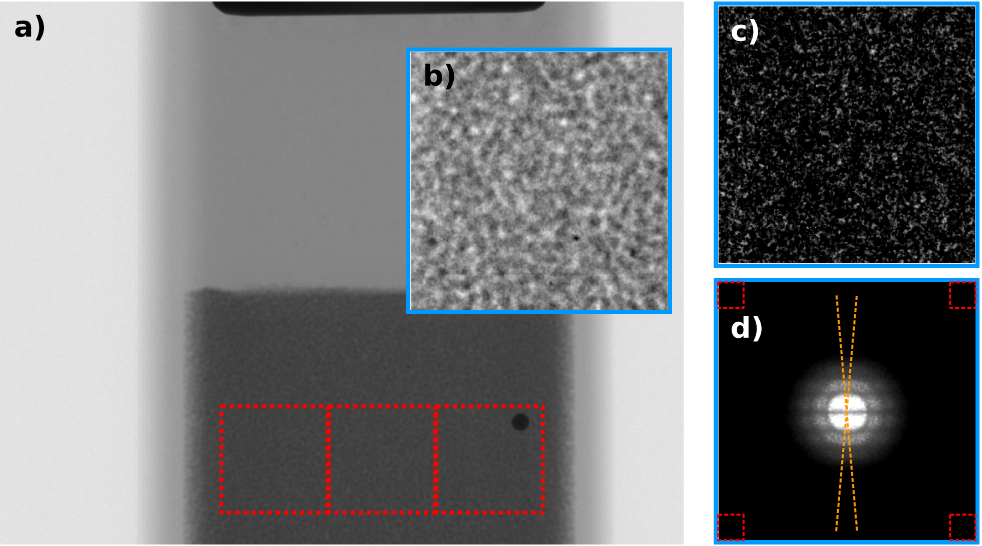


# X-DFA for sedimenting particles



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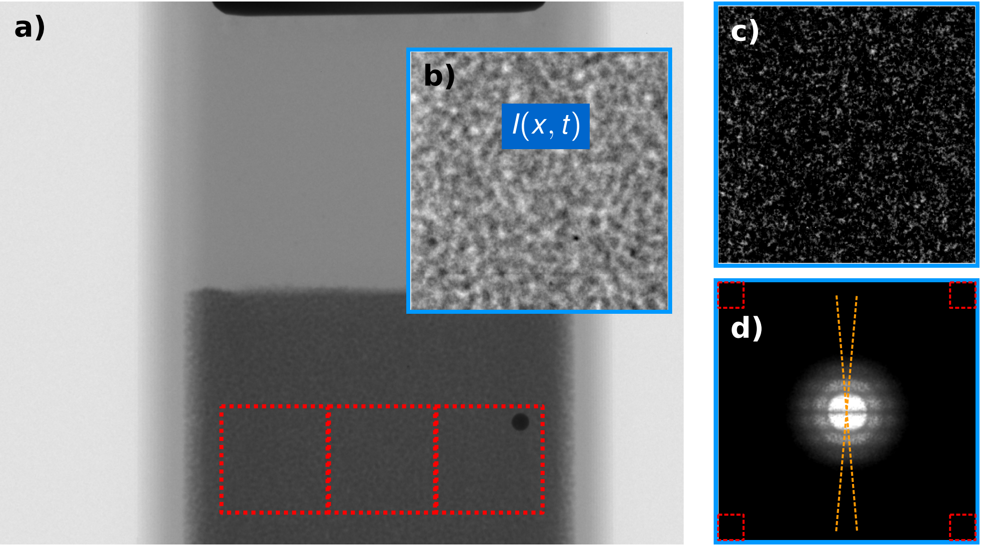

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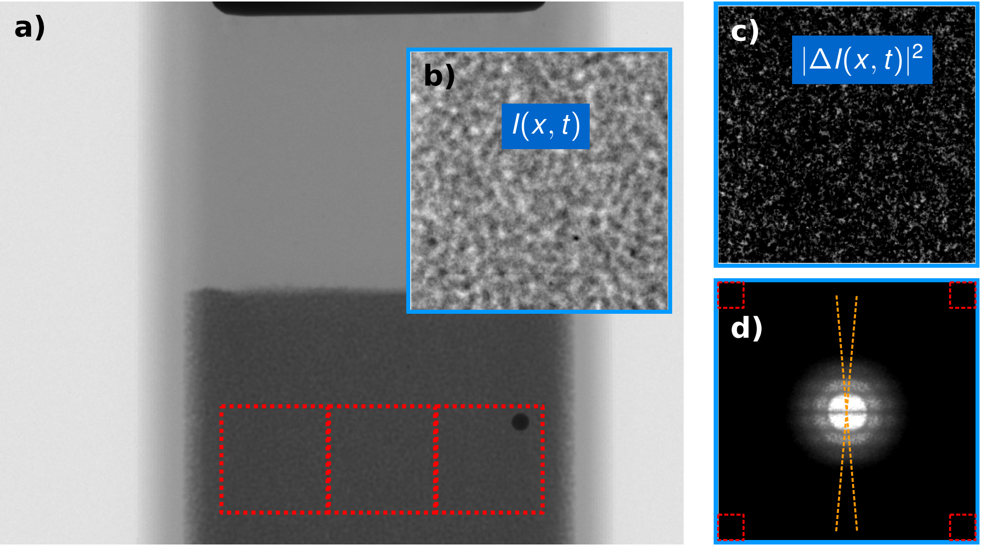

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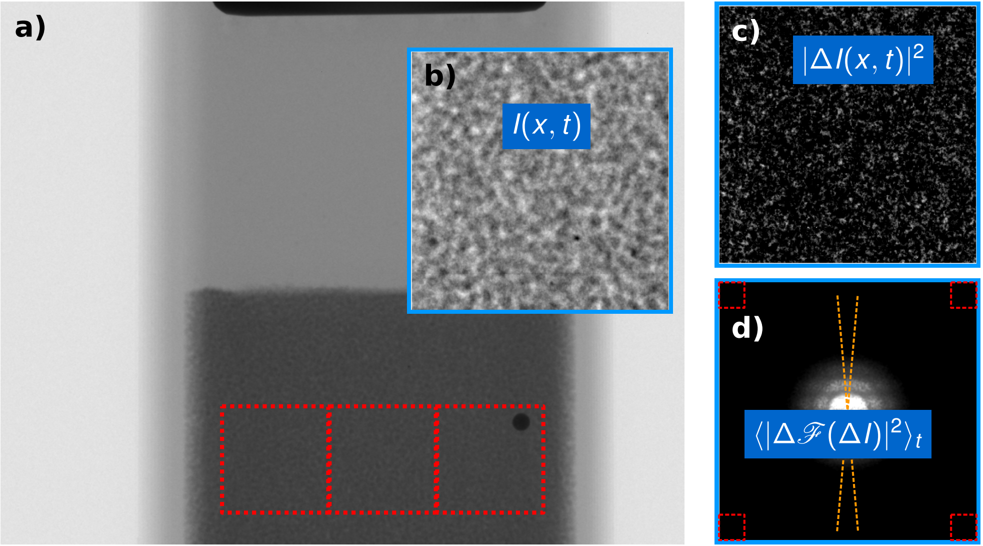
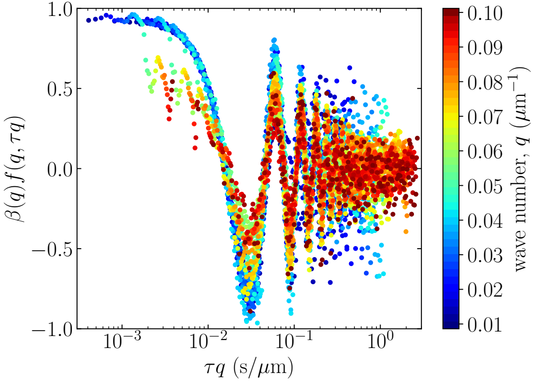
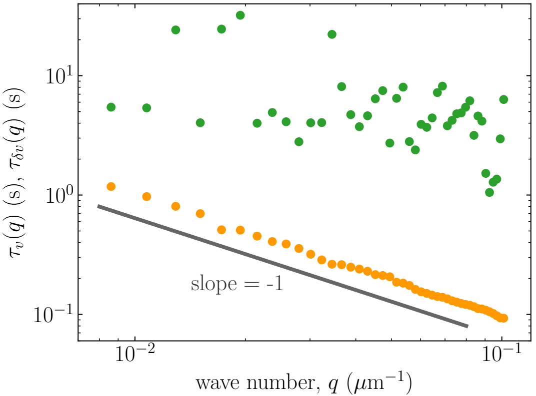


# X-DFA for sedimenting particles

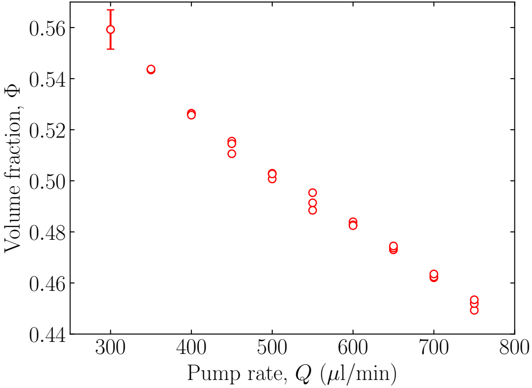
$$f(q,\tau) = \cos(q\langle v_s\rangle\tau)\exp\left(-\frac{1}{2}q^2\delta v^2\tau^2\right)$$

$$\langle v_s\rangle = \langle \Delta r\rangle/\tau_\nu, \langle \delta v\rangle = \langle \delta r\rangle/\tau_{\delta\nu}$$

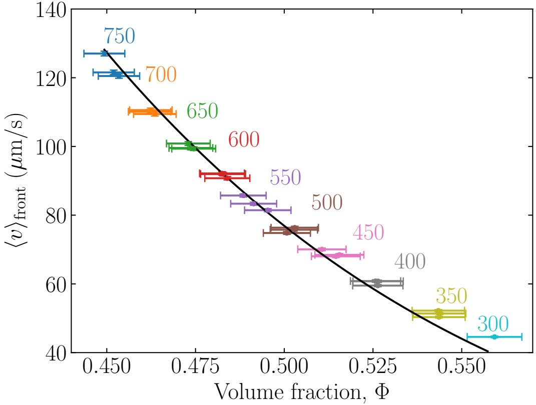


# Richardson-Zaki law

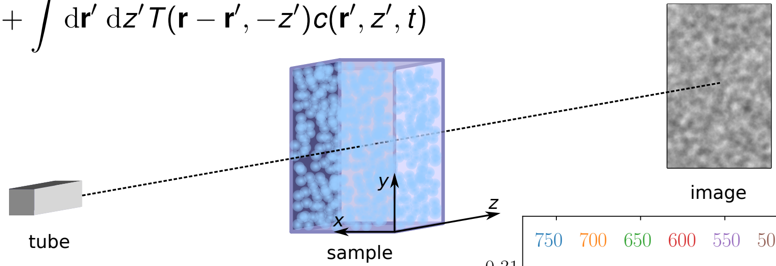


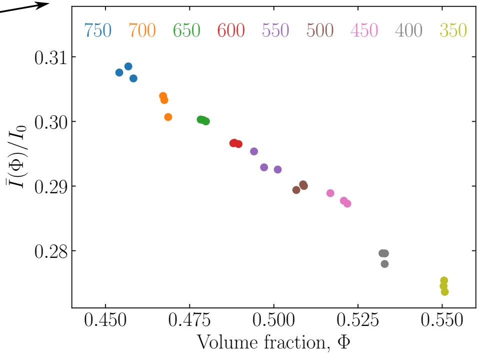

$$\frac{\langle v \rangle_{\text{front}}}{v_{\text{Stokes}}} = (1 - \Phi)^n$$



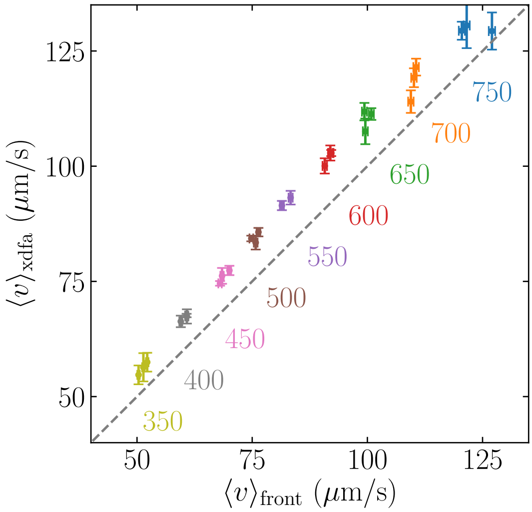
# X-DFA: Requirement of linear space invariant imaging

$$I(\mathbf{r}, t) = I_0 + \int \mathrm{d}\mathbf{r}' \, \mathrm{d}z' \, T(\mathbf{r} - \mathbf{r}', -z') c(\mathbf{r}', z', t)$$

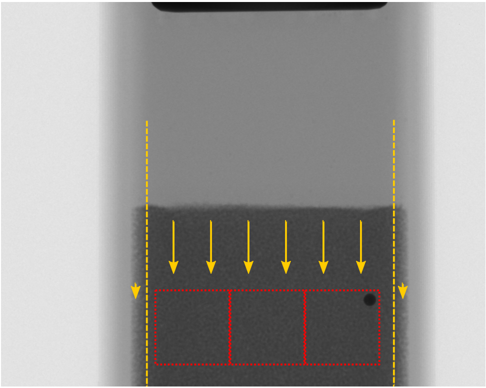


# Front tracking vs. X-DFA

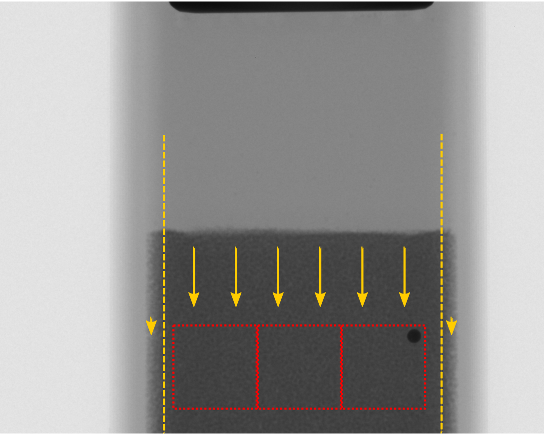



$\langle v \rangle_{\mathrm{xdfa}} > \langle v \rangle_{\mathrm{front}}$ by 9.4%



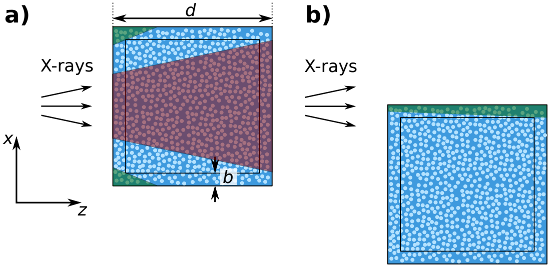
sample video

# Estimate width of boundary layer



$\langle v \rangle_{\text{xdfa}} > \langle v \rangle_{\text{front}}$ by 9.4%

$\langle v \rangle_{\text{xdfa}}$ takes two layers into account
$\langle v \rangle_{\text{front}}$ takes four layers into account




**Estimation:**
Boundary velocity = 0
Else = const.
$\rightarrow b \approx 3$ particle diameters


# Thank you for your attention!