Effect of two-particle correlations on x-ray coherent diffractive imaging studies performed with continuum models

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CFEL SCIENCE

DESY

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  and CFEL Theory Division

Outline

- Introduction
- Imaging and radiation damage modeling
- Samples exposed to XFEL beam
- The scattered intensities
- Numerical simulations
- Summary

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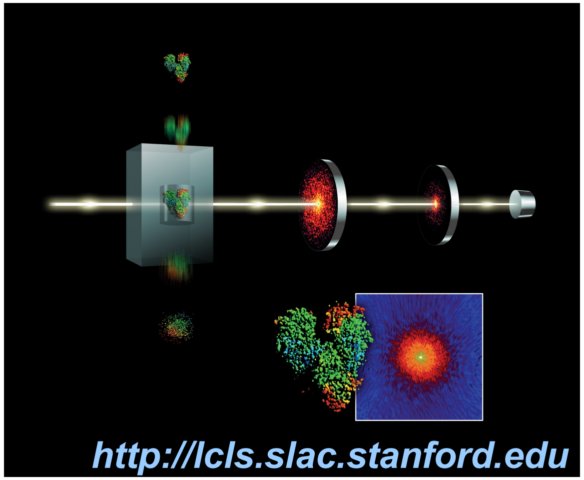
Motivation

- **Single molecule imaging**:
  high-priority project for
  X-ray Free Electron Lasers



- Scattering experiment: high resolution **patterns** are **disturbed** by the
  ionization dynamics (radiation damage) of the sample

- There is a **need for modeling** the sample dynamics;
  important role of modeling: understanding FEL experiments and
  getting useful info about the changes in the diffraction patterns

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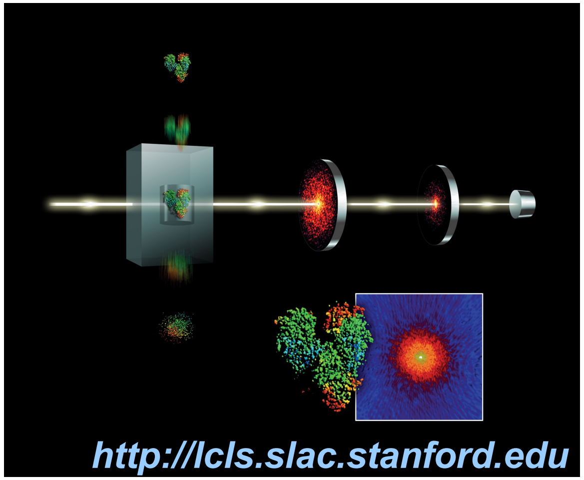

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X-ray Coherent Diffraction Imaging of small objects

- **1 shot** at one sample orientation
  ↓
  2D information

- **Many shots** at different sample orientation
  ↓
  3D information

- **Many shots** at the same sample orientation
  ↓
  averaged, better signal-to-noise ratio

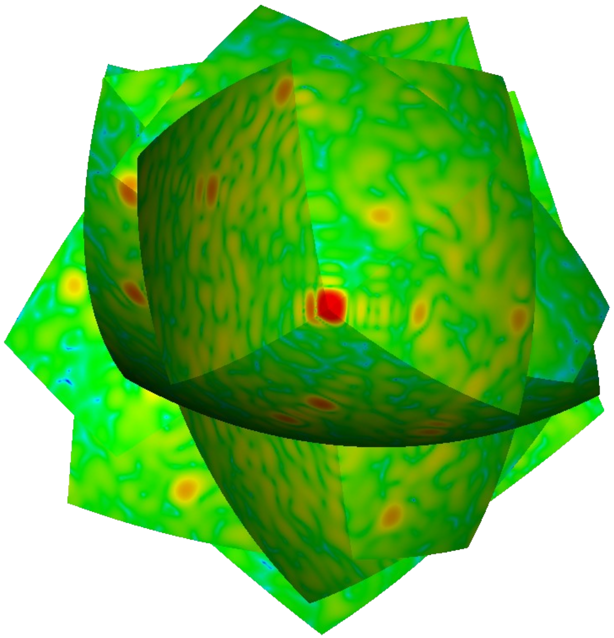


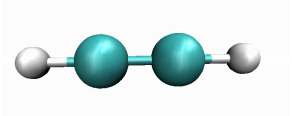
Figure from *http://xray.bmc.uu.se/spb/*

Multi-shot experiment on **many replicas** of the sample, as individual **patterns are disturbed** by the radiation damage!

Radiation damage models

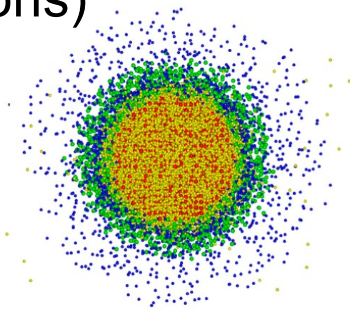
> **Ab initio methods** (electron wave functions: $\psi(\mathbf{r}_1,\mathbf{r}_2,\ldots,t)$)

- few-atom systems



> **Molecular Dynamics** (particles: $\mathbf{r}_i(t)$, $\mathbf{v}_i(t)$; random ionizations)

Z. Jurek et al., EPJD **29**, 217 (2004)

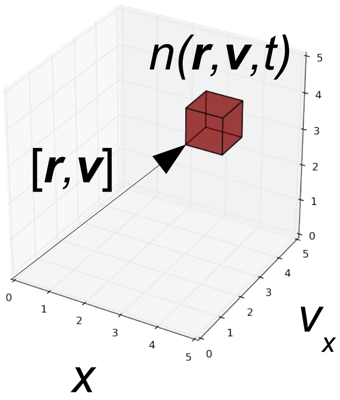


- CPU time ~ $N^2 \rightarrow N>10^5$ modeling not feasible
- one run → one realization of the time evolution

> **Continuum approach** (probability density ρ; $n(\mathbf{r},\mathbf{v},t) = N\,\rho(\mathbf{r},\mathbf{v},t)$; rate equations)

Transport (Boltzmann) Approach, B. Ziaja et al., EPJD **40**, 465 (2006)
Hydrodynamical model, S. P. Hau-Riege et al., PRE **69**, 051906 (2004)

- efficient for large systems: no direct scaling on the number of particles
- one run → average dynamics



Continuum model

↓

How to get relevant information on the imaging?

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Continuum model vs. imaging

- **Basic formulae** of diffraction – static case:

$$F(\boldsymbol{q}) = \int d^3r\, n(\boldsymbol{r})\, e^{i\boldsymbol{q}\cdot\boldsymbol{r}} \qquad I(\boldsymbol{q}) \sim |F(\boldsymbol{q})|^2 = \int d^3r\, d^3r'\, n(\boldsymbol{r})\, n(\boldsymbol{r}')\, e^{i\boldsymbol{q}\cdot(\boldsymbol{r}-\boldsymbol{r}')}$$

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- **Measurement**: integrating patterns over time in **one realization** of the sample dynamics.

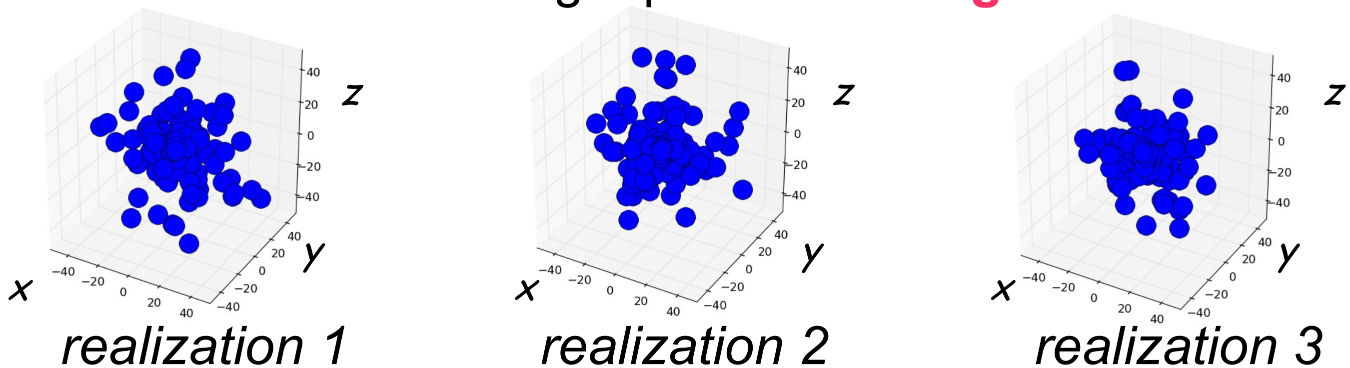
In the case of short coherence time of the beam:

$$\mathcal{I}(\boldsymbol{q}) \equiv \int dt\, h(t)\, I(\boldsymbol{q},t) = \int dt\, h(t) \int d^3r\, d^3r'\, n(\boldsymbol{r},t)\, n(\boldsymbol{r}',t)\, e^{i\boldsymbol{q}\cdot(\boldsymbol{r}-\boldsymbol{r}')}$$

- **Continuum model**: ensemble single-particle **average over realizations**

e.g.

at time t:



etc.

Continuum model vs. imaging

- **Imaging**: **averaging patterns**:

$$\langle \mathcal{I}(\boldsymbol{q}) \rangle_R = \int dt\, h(t)\, \langle I(\boldsymbol{q},t) \rangle_R = \int dt\, h(t) \int d^3r\, d^3r' \boxed{\langle n(\boldsymbol{r},t)\, n(\boldsymbol{r}',t) \rangle_R} e^{i\boldsymbol{q}\cdot(\boldsymbol{r}-\boldsymbol{r}')}$$

- **Continuum model**: **averages single-particle density** over realizations:

$$\mathcal{I}^C(\boldsymbol{q}) = \int dt\, h(t) \int d^3r\, d^3r' \boxed{\langle n(\boldsymbol{r},t) \rangle_R\, \langle n(\boldsymbol{r}',t) \rangle_R} e^{\boldsymbol{q}\cdot(\boldsymbol{r}-\boldsymbol{r}')}$$

Continuum model vs. imaging

> **Imaging**: **averaging patterns**:

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> **Difference**: depends on the correlation between realizations:

$$\langle n(\boldsymbol{r},t)\, n(\boldsymbol{r}\,',t) \rangle_R - \langle n(\boldsymbol{r},t) \rangle_R\, \langle n(\boldsymbol{r}\,',t) \rangle_R$$

Question

- Can we use $\langle n(\boldsymbol{r})\rangle_{\mathrm{R}}$ obtained from the continuum model for imaging studies?

Question

- Can we use $\langle n(\boldsymbol{r}) \rangle_R$ obtained from the continuum model for imaging studies?
- Generally not, as the average intensity also depends on the correlations between the realizations
- But for specific systems?

$\rightarrow$ investigations needed

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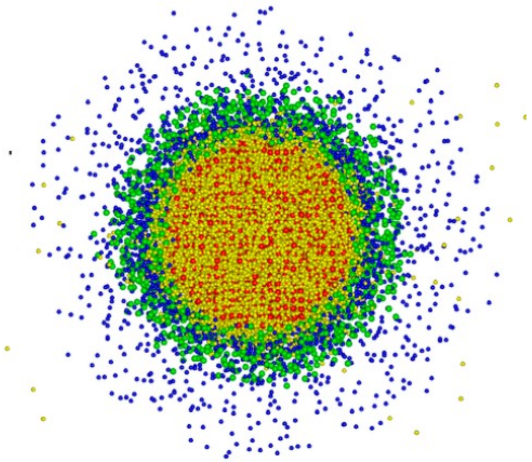
Sample dynamics

- Modeling of radiation damage within a sample built of light elements

  - fast electrons escaping

  - ions: electronic damage; possible movement

  - hot trapped electrons (T_{el} ~ 20eV–100eV, n_{el} ~1–4 x ion density)



Damage components

- Electronic damage – form factors of ions are decreasing
- Scattering from free and quasi-free electrons
- Ion movement

Damage components

- Electronic damage – form factors of ions are decreasing
- Scattering from free and quasi-free electrons

always present

- Ion movement $\rightarrow$ can be avoided by using pulses short enough

Sample dynamics

- Modeling of radiation damage (T_{pulse} ≤ 5 - 10 fs)

  - fast electrons escaping

  - ions: electronic damage; no movement

  - hot trapped electrons (T_{el} ~ 20eV–100eV, n_{el} ~1–4 x ion density)

$$\Gamma = \frac{E_{Coulomb}}{k_B T} = \frac{1}{4\pi\epsilon_0}\frac{e^2}{k_B T}\left(\frac{4\pi n}{3}\right)^{\frac{1}{3}} \approx 0.5$$

$$\Theta = \frac{k_B T}{E_F} = \frac{k_B T}{\frac{\hbar^2}{2m_e}(3\pi^2 n)^{2/3}} \approx 2.5$$

→ **classical, ideal plasma**: classical treatment is adequate; also small effect of electron correlations

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Time integrated scattered intensity

- Average intensity:

$$\langle \mathcal{I}(\boldsymbol{q}) \rangle_R = \int dt\, h(t)\, \underline{\langle I(\boldsymbol{q},t)\rangle_R} = \int dt\, h(t) \int d^3r\, d^3r'\, \underline{\langle n(\boldsymbol{r},t)\, n(\boldsymbol{r}',t)\rangle_R}\, e^{i\boldsymbol{q}\cdot(\boldsymbol{r}-\boldsymbol{r}')}$$

- Intensity obtained directly from the continuum model:

$$\mathcal{I}^C(\boldsymbol{q}) = \int dt\, h(t)\, \underline{I^C(\boldsymbol{q},t)} = \int dt\, h(t) \int d^3r\, d^3r'\, \underline{\langle n(\boldsymbol{r},t)\rangle_R\, \langle n(\boldsymbol{r}',t)\rangle_R}\, e^{\boldsymbol{q}\cdot(\boldsymbol{r}-\boldsymbol{r}')}$$

Contributions to the scattered intensity

$$\langle I(\boldsymbol{q},t)\rangle_R = \left\langle \left| \int (n_b(\boldsymbol{r},t) + n_t(\boldsymbol{r},t) + n_e(\boldsymbol{r},t))\; e^{i\boldsymbol{q}\cdot\boldsymbol{r}}\, d^3r \right|^2 \right\rangle_R$$

bound electrons — trapped electrons — escaped electrons

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bound electrons — trapped electrons — escaped electrons

$$= \left\langle |n_b(\boldsymbol{q},t)|^2 \right\rangle_R + \left\langle |n_t(\boldsymbol{q},t)|^2 \right\rangle_R + \left\langle |n_e(\boldsymbol{q},t)|^2 \right\rangle_R$$

$$+2\,Re\,[\,\langle n_b(\boldsymbol{q},t)\, n_t^\star(\boldsymbol{q},t)\rangle_R + \langle n_b(\boldsymbol{q},t)\, n_e^\star(\boldsymbol{q},t)\rangle_R + \langle n_t(\boldsymbol{q},t)\, n_e^\star(\boldsymbol{q},t)\rangle_R\,]$$

Modulus square terms

Cross terms

Scattered intensity averaged over realizations – uncorrelated case

$$\mathcal{J}^{uncorr}(\boldsymbol{q}) = \int dt\, h(t)\, \langle I^{uncorr}(\boldsymbol{q},t)\rangle_R$$

$$= \int dt\, h(t) \left[I^C(\boldsymbol{q},t) + \sum_{j=1}^{N_a} \left(\left\langle |f_j(\boldsymbol{q},t)|^2 \right\rangle_R - \left| \langle f_j(\boldsymbol{q},t)\rangle_R \right|^2 \right) \right.$$

$$\left. - \frac{1}{N_t} I_t^C(\boldsymbol{q},t) + N_t(t) + N_e(t) \right]$$

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Simulations

- We test the formula on a simple system in the typical parameter range:

  - 100 C-atoms within a sphere, n_{atom}=15Å^{-3}, random, but static positions, average charge of ions increases to +2
  - 200 electrons, T=20eV, n_{el}=2 n_{atom}, Coulomb interactions
  - 100 realizations, T = 10fs

- We calculated

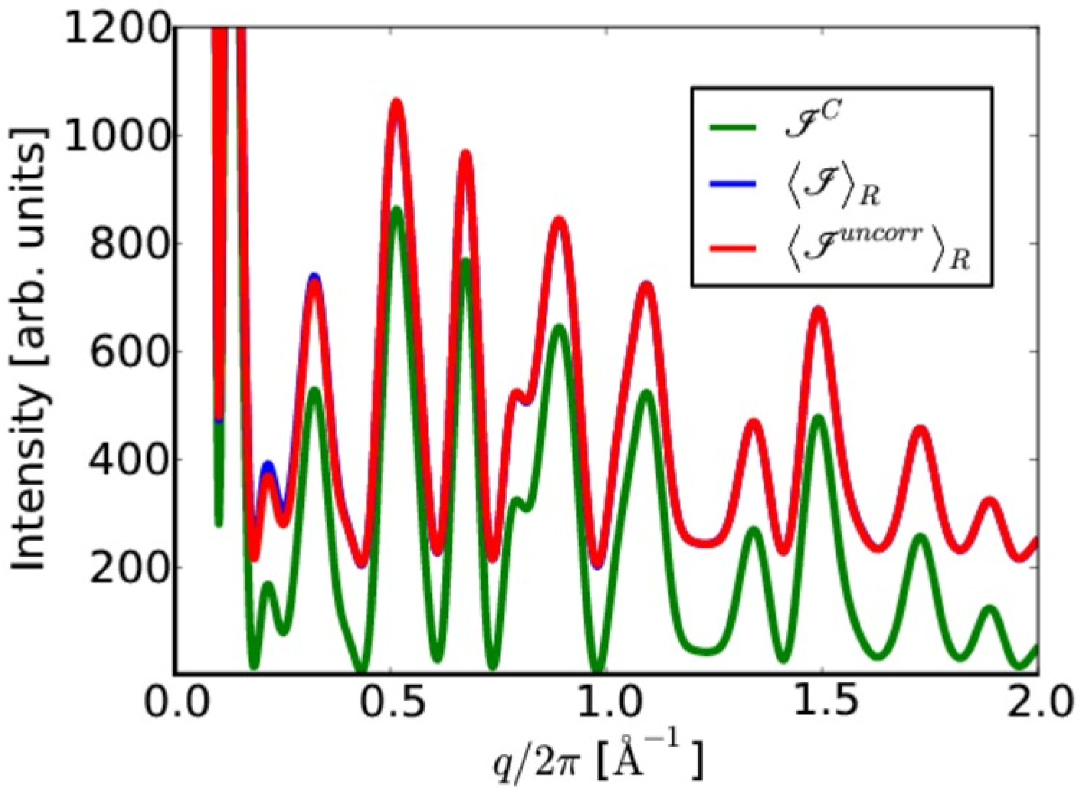
$$\langle \mathcal{I}(\boldsymbol{q}) \rangle_R = \int dt \, \langle I(\boldsymbol{q}, t) \rangle_R$$

$$\langle \mathcal{I}^{uncorr}(\boldsymbol{q}) \rangle_R = \int dt \, \langle I^{uncorr}(\boldsymbol{q}, t) \rangle_R$$

$$\mathcal{I}^C(\boldsymbol{q})$$

Simulations – ions and trapped electrons

- System: 200 electrons and
  100 carbon ions in fixed positions, increasing charge to +2



- The dominant correction to $\mathcal{I}^C(\boldsymbol{q})$ is the constant, q-independent **shift by the** average **number of free electrons**.

$$\langle I^{uncorr}(\boldsymbol{q},t)\rangle_R = I^C(\boldsymbol{q},t) - \frac{1}{N_t} I_t^C(\boldsymbol{q},t) + N_t(t) + N_e(t)$$

Summary

- Average intensity depends on the correlations between realizations

- For systems in a few-fs, short coherence-time XFEL beam, an uncorrelated estimate for the average scattered intensity can be derived from the continuum model.

- The dominant correction to the intensity (constructed from average density) is a **q-independent shift by the number of free electrons** in the beam.