Effect of SASE Pulse Properties on Diffuse X-ray Scattering Measurements

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Abstract

In diffuse X-ray scattering experiments at X-ray Free-Electron Laser (XFEL) facilities, the measured scattering signal depends critically on the properties of the Self-Amplified Spontaneous Emission (SASE) pulses used for the measurements. This document examines how the spectral properties of SASE pulses affect the diffuse scattering patterns recorded by a pixel detector, with particular attention to shot-to-shot variability and finite bandwidth effects. We also propose a differentiable approach using 3D Gaussians with the reparameterization trick to model these effects in analysis workflows.

1 Introduction

In diffuse X-ray scattering experiments at X-ray Free-Electron Laser (XFEL) facilities, the measured scattering signal depends critically on the properties of the Self-Amplified Spontaneous Emission (SASE) pulses used for the measurements. This document examines how the spectral properties of SASE pulses affect the diffuse scattering patterns recorded by a pixel detector.

2 Physical Framework

2.1 Diffuse Scattering Model

The diffuse scattering intensity as a function of momentum transfer vector $\mathbf{q}$ can be represented as $I_d(\mathbf{q})$. This is the physical quantity we aim to measure, which represents correlations in electron density fluctuations within the sample.

2.2 SASE Pulse Properties

SASE pulses are characterized by:

- A spectral intensity distribution $I_{\text{SASE}}(\omega)$ or equivalently $I_{\text{SASE}}(q)$
- Shot-to-shot variations in both central energy ω_0 and spectral width σ_ω
- Finite bandwidth even within a single shot

2.3 Measurement Process

The intensity detected at pixel position (x, y) on the detector can be expressed as:

$$I_{\text{det}}(x, y) = \int I_d(\mathbf{q}(x, y, \omega)) \cdot I_{\text{SASE}}(\omega) \cdot G(x, y, \omega)\, d\omega \tag{1}$$

where:

- $\mathbf{q}(x, y, \omega)$ maps the pixel position and photon energy to momentum transfer
- $G(x, y, \omega)$ represents geometric correction factors

3 Key Considerations

3.1 Shot-to-Shot Variability (Jitter)

The SASE central energy typically fluctuates by $\Delta E/E \approx 10^{-3}$ to 10^{-2} at current XFEL facilities. This means that for a 10 keV X-ray beam, the central energy may jitter by 10-100 eV between shots.

The impact on measured intensity can be estimated by:

$$\frac{\Delta I_{\text{det}}}{I_{\text{det}}} \approx \left| \frac{\partial \ln I_d}{\partial q} \right| \cdot \frac{\Delta E}{E} \cdot |\mathbf{q}| \tag{2}$$

For typical diffuse scattering with significant features where $\left| \frac{\partial \ln I_d}{\partial q} \right| \approx 1 - 10$ Å and $|\mathbf{q}| \approx 0.1 - 1$ Å^{-1}, this yields:

$$\frac{\Delta I_{\text{det}}}{I_{\text{det}}} \approx 10^{-4} - 10^{-1} \tag{3}$$

Conclusion on jitter: The shot-to-shot jitter can indeed introduce significant stochasticity in the measured intensity, especially at higher resolution (larger $|\mathbf{q}|$) and for diffuse features with sharp gradients. This effect becomes particularly relevant when analyzing small differences in diffuse scattering patterns.

3.2 Finite Bandwidth of Single Pulses

SASE pulses typically have a relative bandwidth of $\Delta E/E \approx 10^{-3}$ for a single shot. This leads to an integration over a range of $\mathbf{q}$ values for each pixel:

$$\delta q \approx \frac{\Delta E}{E} \cdot |\mathbf{q}| \tag{4}$$

For $|\mathbf{q}| \approx 1$ Å^{-1}, this gives $\delta q \approx 10^{-3}$ Å^{-1}.

Conclusion on bandwidth: The finite bandwidth means that each detector pixel measures an integral over a small region in reciprocal space rather than a point. However, this effect is generally small compared to other experimental factors unless measuring extremely sharp features in reciprocal space.

3.3 3D Visualization

The finite bandwidth effect can be visualized as measuring a thin "slab" through reciprocal space rather than an exact 2D Ewald sphere. The thickness of this slab is:

$$\text{Slab thickness} \approx \frac{\Delta E}{E} \cdot |\mathbf{q}| \tag{5}$$

For typical SASE parameters and $|\mathbf{q}| \approx 1$ Å^{-1}, this gives a thickness on the order of 10^{-3} Å^{-1}.

4 Practical Implications

1. **Data Processing Strategy**: Multiple shots should be normalized by their spectral distribution and intensity before averaging to reduce the impact of jitter.
2. **Feature Resolution**: Features in diffuse scattering with characteristic length scales smaller than the slab thickness will be averaged out.
3. **Simulation Comparison**: When comparing experimental results to theoretical predictions, the experimental data should be understood as representing a weighted average over a small volume in reciprocal space.
4. **Monochromator Consideration**: Using a monochromator would reduce these effects but at significant cost to intensity.

5 Differentiable Modeling of Slab Thickness with 3D Gaussians

If the slab thickness effect proves significant enough to require explicit modeling, a differentiable approach using 3D Gaussians can be implemented. This approach is particularly valuable when fitting theoretical models to experimental data using gradient-based optimization.

5.1 Gaussian Representation of SASE Spectral Distribution

The SASE pulse's spectral distribution can be modeled as a 3D Gaussian in reciprocal space:

$$I_{\text{SASE}}(\mathbf{q}) \approx \exp\left(-\frac{1}{2}(\mathbf{q}-\mathbf{q}_0)^T\Sigma^{-1}(\mathbf{q}-\mathbf{q}_0)\right) \tag{6}$$

where:

- $\mathbf{q}_0$ is the center of the distribution (determined by the central energy)
- Σ is the covariance matrix that defines the shape and orientation of the slab

The covariance matrix Σ can be parametrized to reflect the anisotropic nature of the slab (thin in one direction, extended along the Ewald sphere).

5.2 Reparameterization Trick for Differentiability

To make this model differentiable for gradient-based optimization, we can use the reparameterization trick from variational inference:

$$\mathbf{q} = \mathbf{q}_0 + L\epsilon \tag{7}$$

where:

- L is the Cholesky decomposition of Σ (i.e., $\Sigma = LL^T$)
- ϵ is a standard normal random variable

This allows gradients to flow through the parameters $\mathbf{q}_0$ and L during optimization.

5.3 Implementation in Diffuse Scattering Model

The measured intensity at pixel (x, y) can then be approximated as:

$$I_{\text{det}}(x, y) \approx \frac{1}{N} \sum_{i=1}^{N} I_d(\mathbf{q}_0(x, y) + L\epsilon_i) \cdot G(x, y) \tag{8}$$

where N is the number of Monte Carlo samples used to estimate the integral. This Monte Carlo integration is differentiable with respect to all parameters.

5.4 Advantages of This Approach

1. **Differentiability**: Enables the use of efficient gradient-based optimization algorithms.
2. **Flexibility**: The covariance matrix Σ can be adapted to model different SASE beam properties and geometries.
3. **Computational Efficiency**: The reparameterization approach allows backpropagation through the model while maintaining computational tractability.
4. **Uncertainty Quantification**: Natural extension to modeling shot-to-shot variations by treating the parameters as random variables with priors.

5.5 Practical Implementation

In a PyTorch or JAX implementation, this might look like:

```
def diffuse_model(q_0, L, I_d_func, num_samples=100):
    # q_0: central q vector for a pixel
    # L: Cholesky factor of covariance matrix
    # I_d_func: function that computes diffuse intensity at a given q

    # Sample from standard normal
    epsilon = torch.randn(num_samples, 3)

    # Apply reparameterization trick
```

```
    q_samples = q_0.unsqueeze(0) + torch.matmul(epsilon, L.T)

    # Compute intensity at each sampled q point
    I_samples = torch.stack([I_d_func(q) for q in q_samples])

    # Average the intensities (Monte Carlo integration)
    return I_samples.mean()
```

This framework allows for efficient, differentiable modeling of the finite bandwidth effects in SASE-based diffuse scattering experiments.