Universidade do Minho
Escola de Ciências

Fourier Analysis: Phonons in hBN

Alexandre Silva

João Henriques

Tiago Rodrigues

Laboratórios Avançados de Física

Braga, Março de 2020.

Objectives

- Introduce the basics of Fourier analysis and the case of study (probing phonons in hBN using EELS);
- Apply Fourier analysis to the experimental and theoretical data;
- Understand what processes might have caused the differences between the experimental and simulated data;
- ***Ultimate goal:*** Understand how Fourier analysis is used to validate theoretical models with experimental data;

Introduction…

Fourier Analysis

The Fourier transforms are defined as:

$$\underbrace{A_k = \sum_{m=0}^{n-1} a_m e^{-2\pi i \frac{mk}{n}},}_{\text{Direct}} \quad \underbrace{a_m = \frac{1}{n} \sum_{k=0}^{n-1} A_k e^{2\pi i \frac{mk}{n}}}_{\text{Inverse}}$$

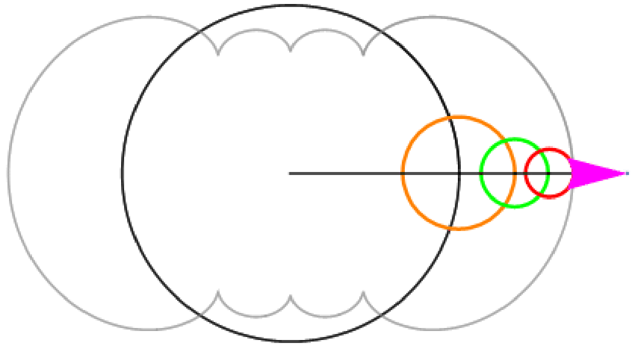
Fourier Analysis

The Fourier transforms are defined as:

$$A_k = \sum_{m=0}^{n-1} a_m e^{-2\pi i \frac{mk}{n}}, \quad a_m = \frac{1}{n}\sum_{k=0}^{n-1} A_k e^{2\pi i \frac{mk}{n}}$$

Direct

Inverse



Case of Study: What & Why?

Case of study: Probing phonons in hexagonal boron nitride (hBN) using electron energy loss spectroscopy (EELS) - R. J. Nicholls *et. al*. PRB **99** 2019.

Phonons: Crystal lattice excitations. **Why?** To understand lattice Dynamics.

hBN: Most stable form of BN polymorphs, widely used in its 2D configuration.

EELS: Beam of electrons impinges on a sample and the transmitted electrons' energy is measured. **Why?** Allows us to study momentum depedence and nanometer spatial resolution.

DFT: Density functional theory is a powerful, widely spread method at the heart of today's computational physics research… and it is like a black box to us.

Case of Study: Details

Theory:

1. Scattering formalism is used to compute cross sections;

2. To simplify the problem some approximations are introduced;

3. Density functional perturbation theory was used to compute the phonon eigenvalues.

Case of Study: Details

Theory:

1. Scattering formalism is used to compute cross sections;

2. To simplify the problem some approximations are introduced;

3. Density functional perturbation theory was used to compute the phonon eigenvalues.

Experiment:

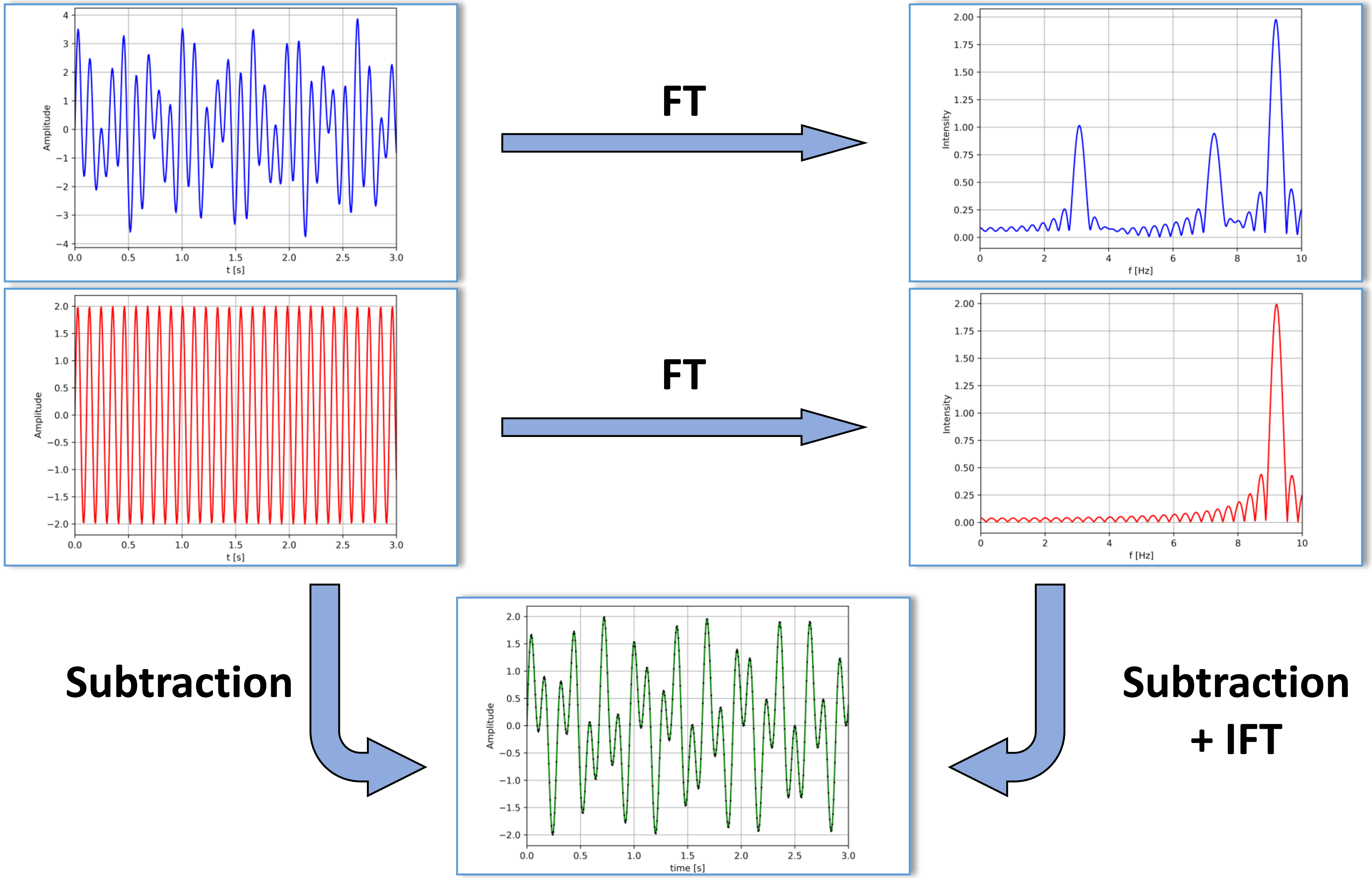
1. A transmission electron microscope (STEM) was used to perform EELS;

2. The apparatus was callibrated no minimize electron damage;

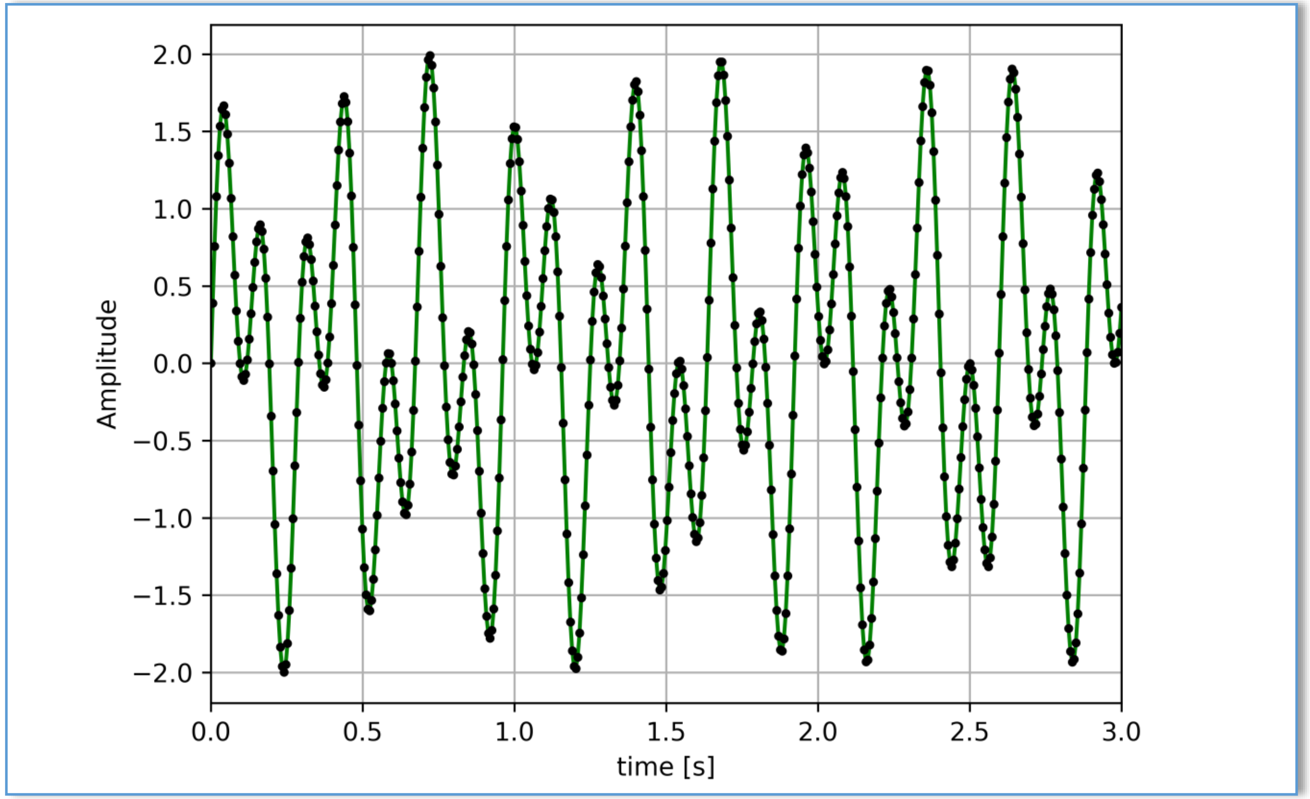
3. No denoising or deconvolution routines were used.

Validation…

Learning The Tools

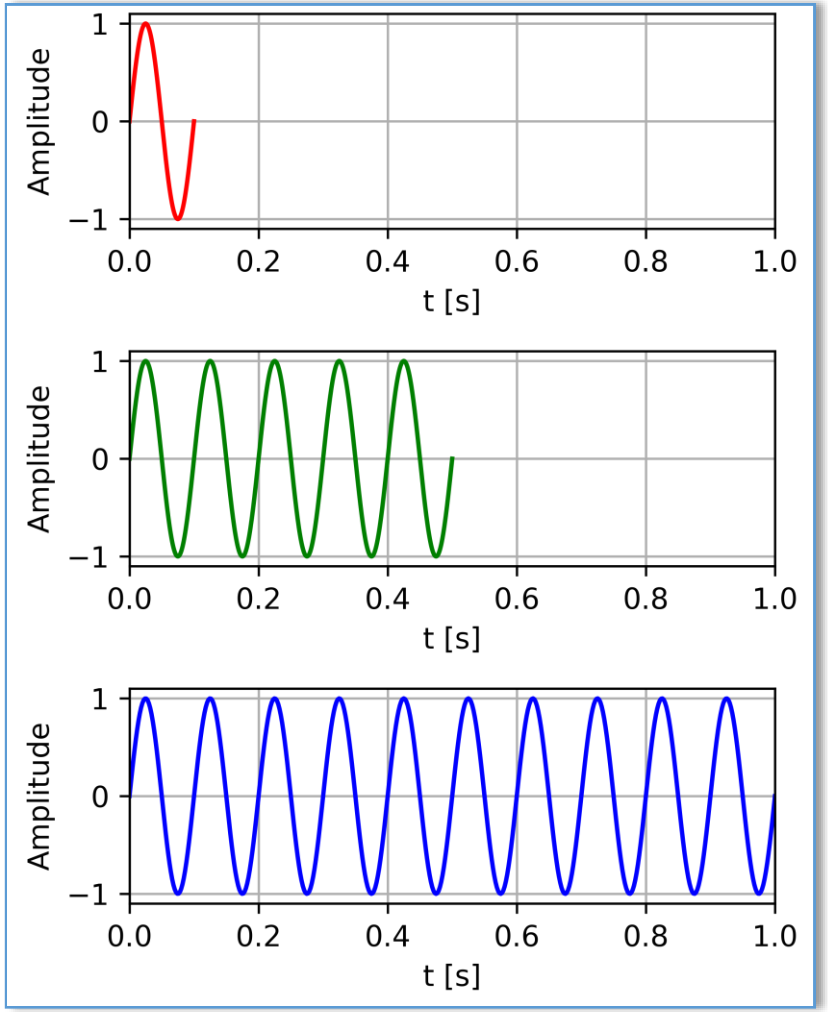


Learning The Tools

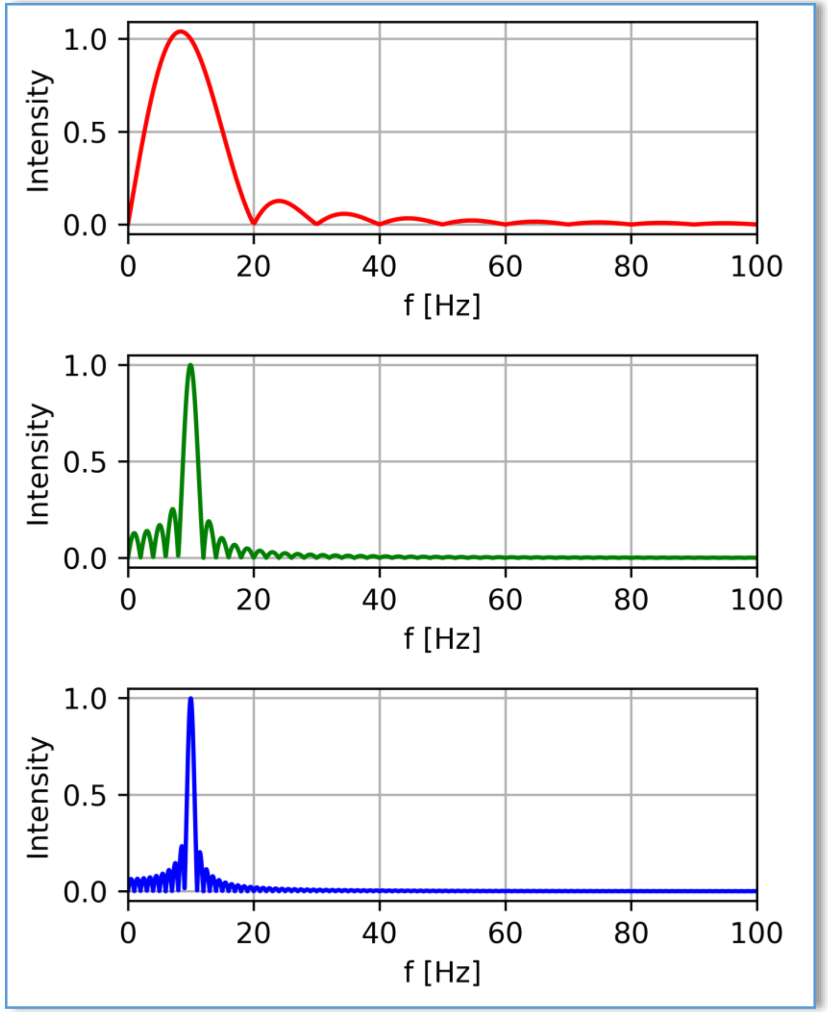


It Works!

Uncertainty Principle

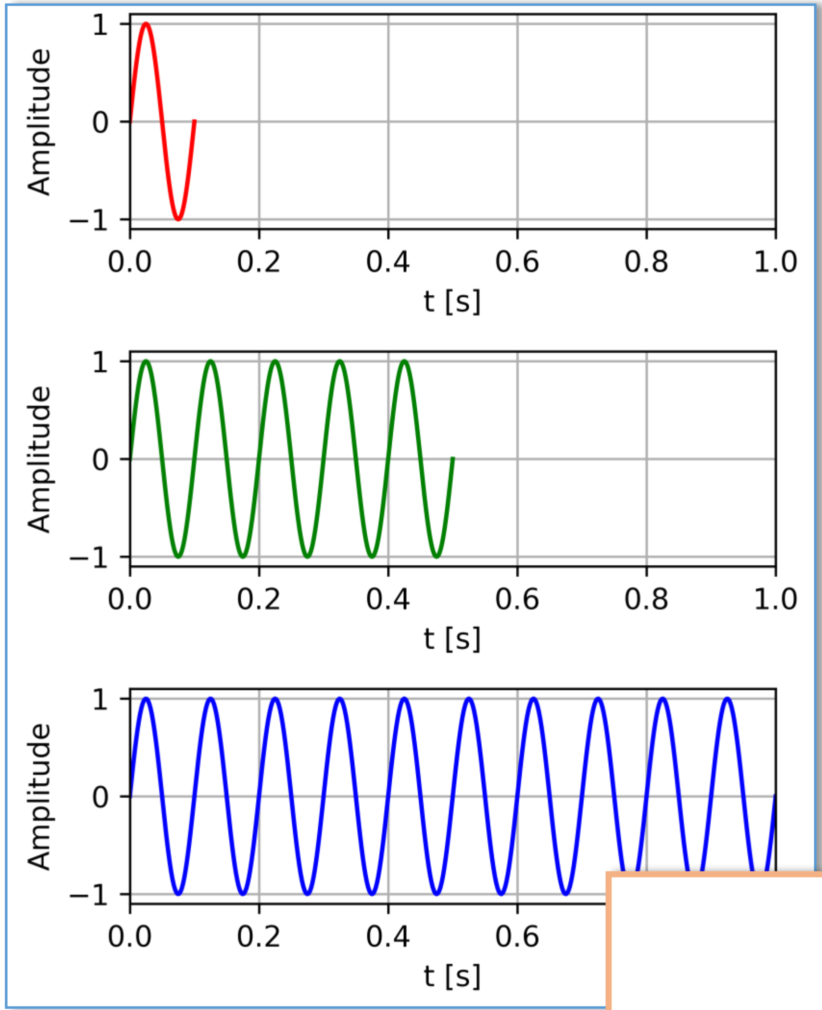


FT

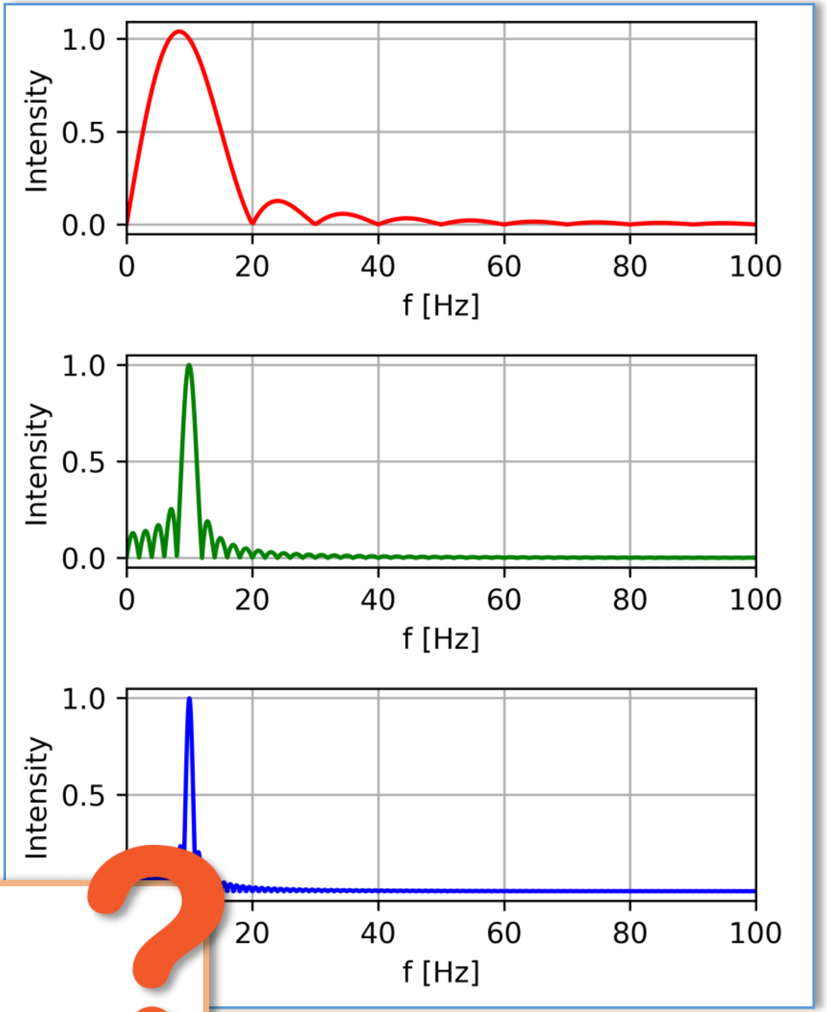


$$\text{Smaller } \Delta t \Longrightarrow \text{Bigger } \Delta\omega$$

Uncertainty Principle



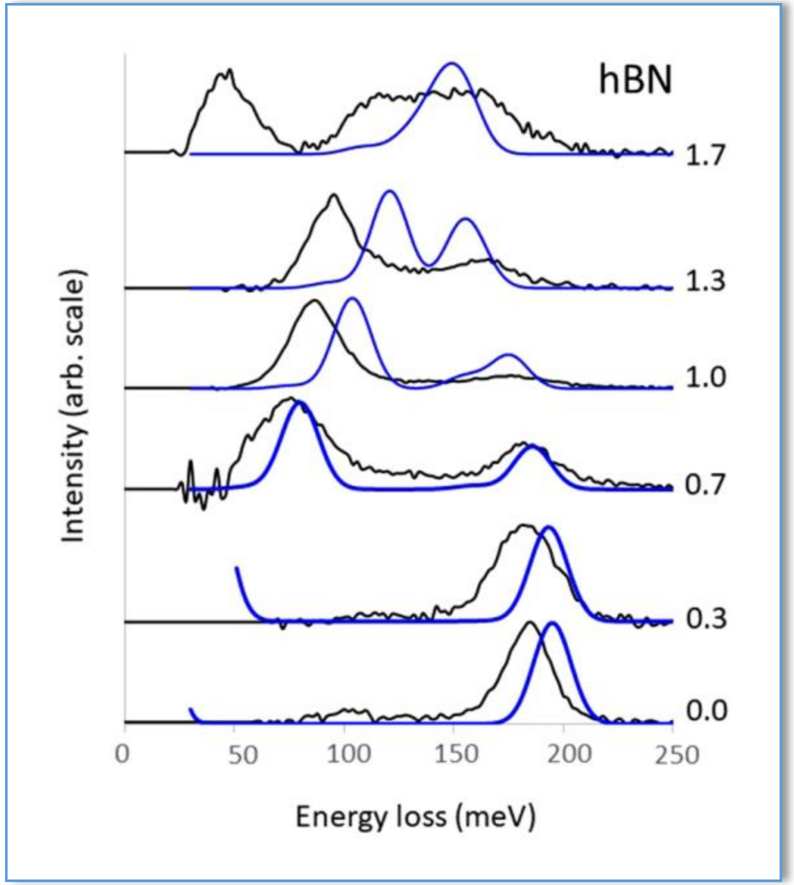
FT

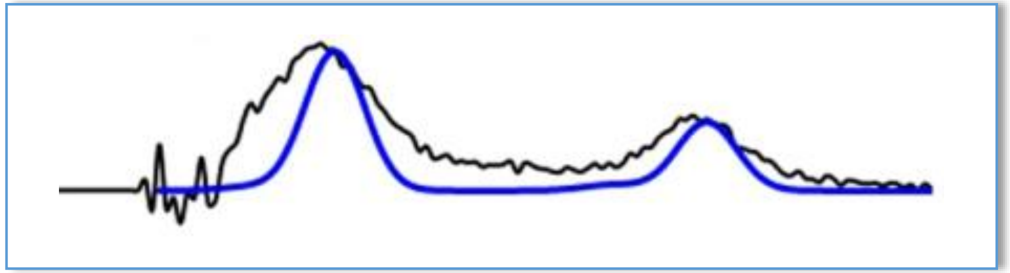


$$\Delta t \Delta \omega \geq \frac{1}{2}$$

Cleaning the Signal…

Raw Data



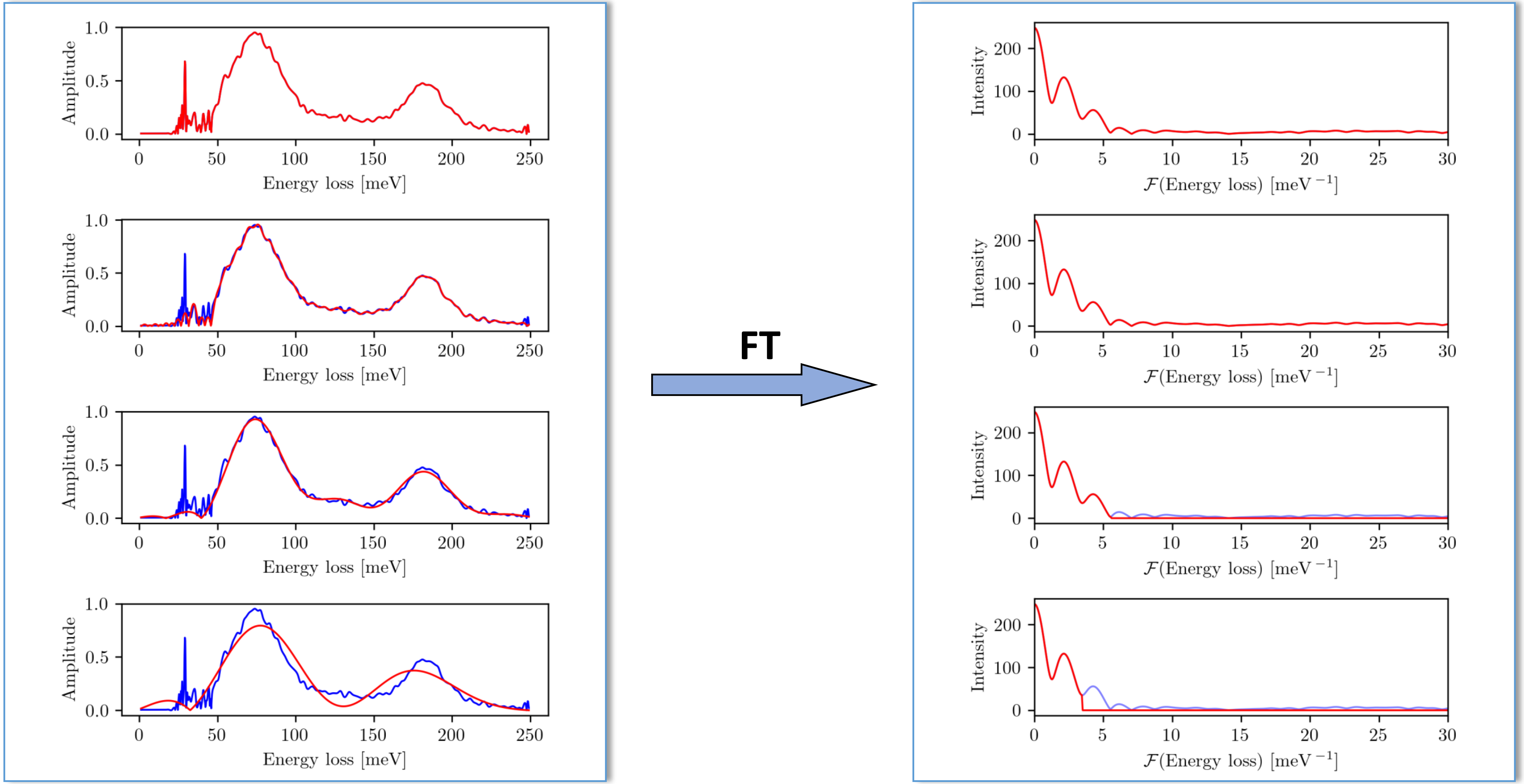


General features of the data

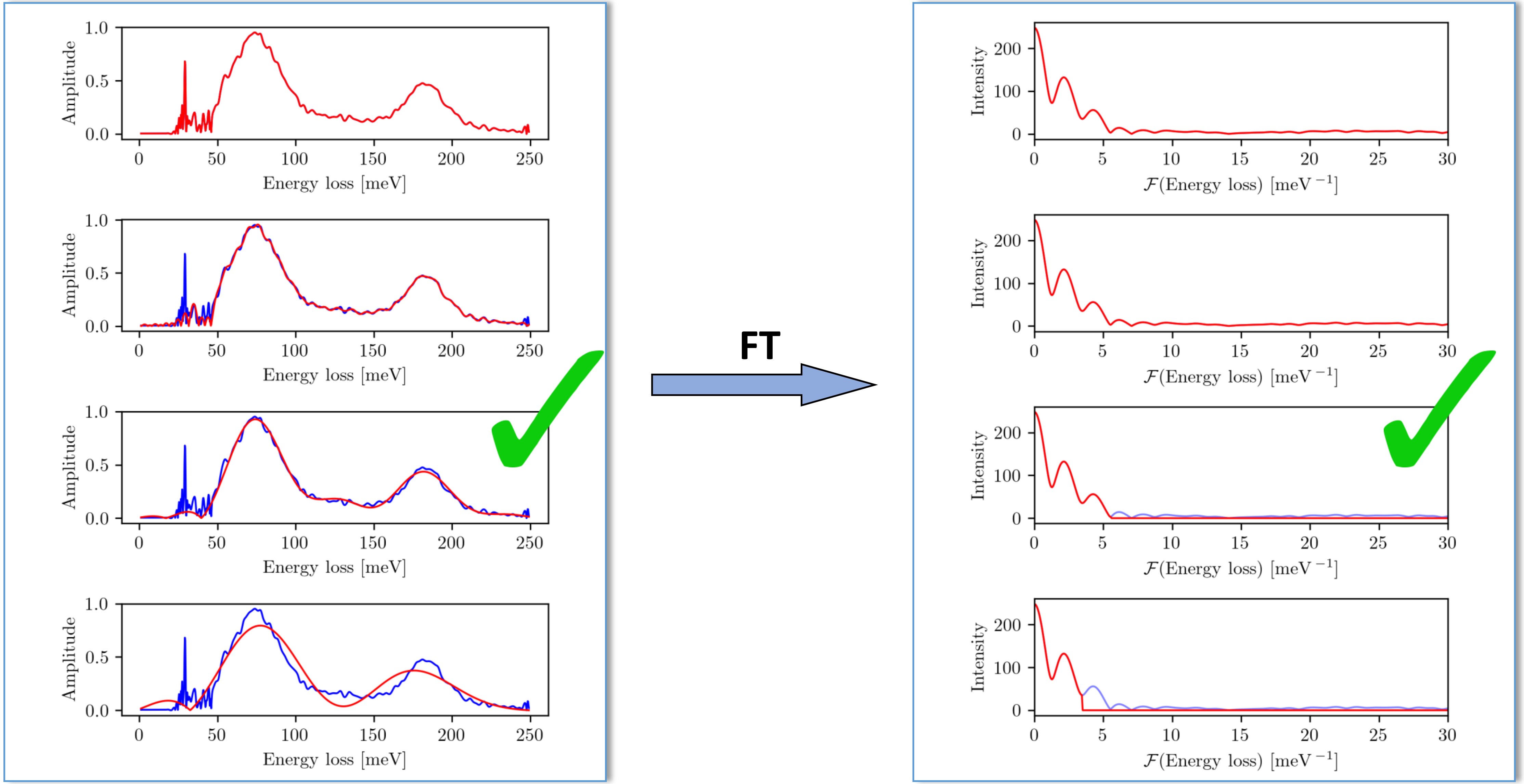
(R. J. Nicholls *et. al*. PRB **99** 2019):

- Two independent peaks;
- Different broadening;
- Initial noise.

Cleaning the Experimental Data

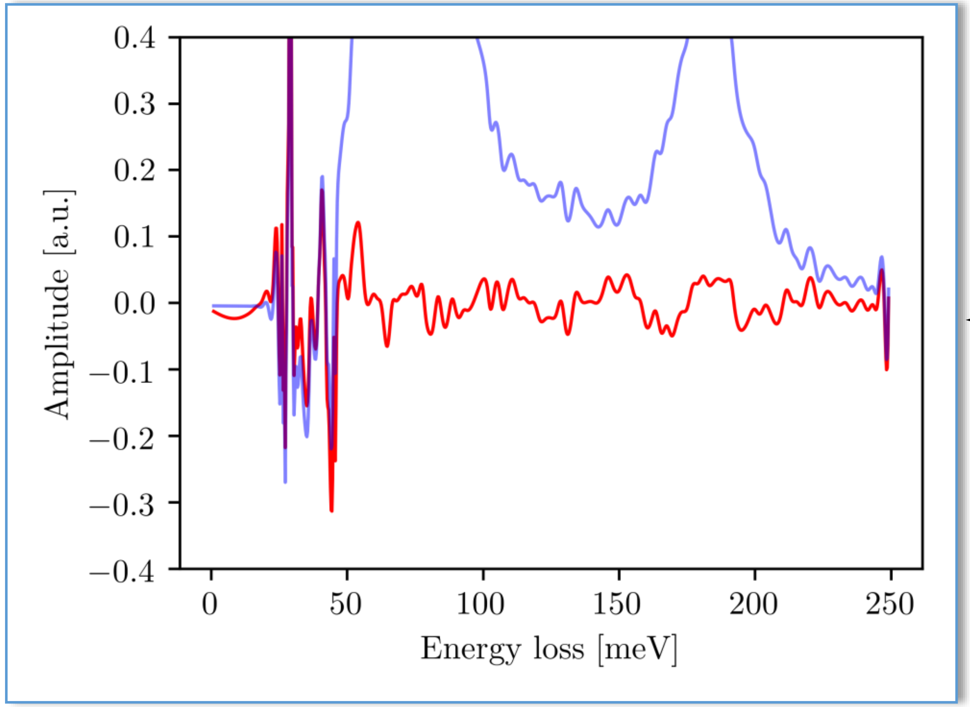


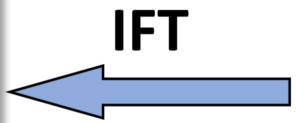
Cleaning the Experimental Data

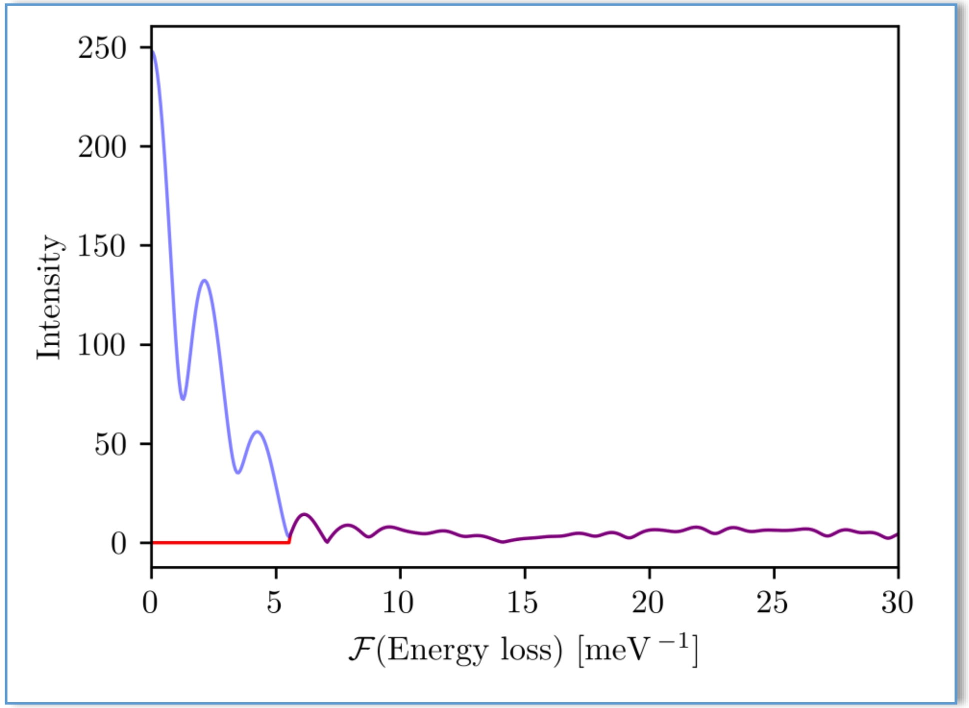


Cleaning the Experimental Data

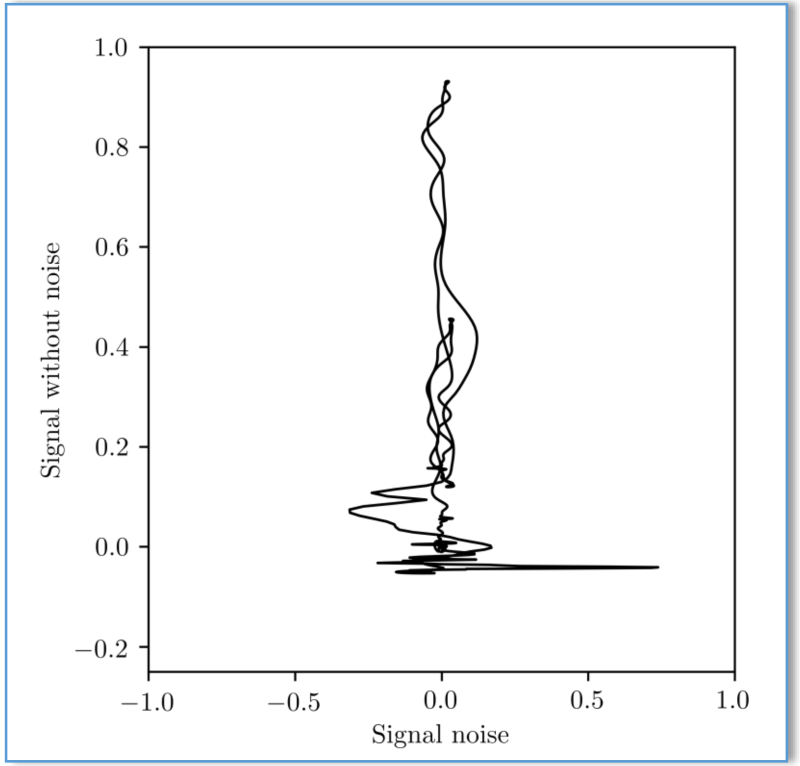
Looking at the noise:

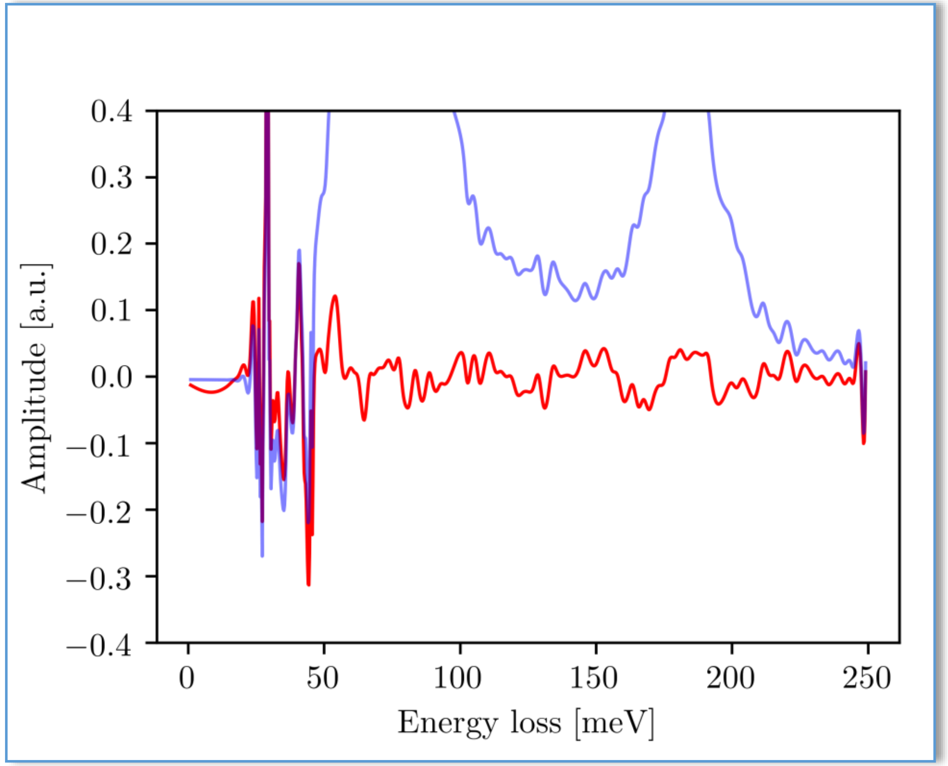






Cleaning the Experimental Data

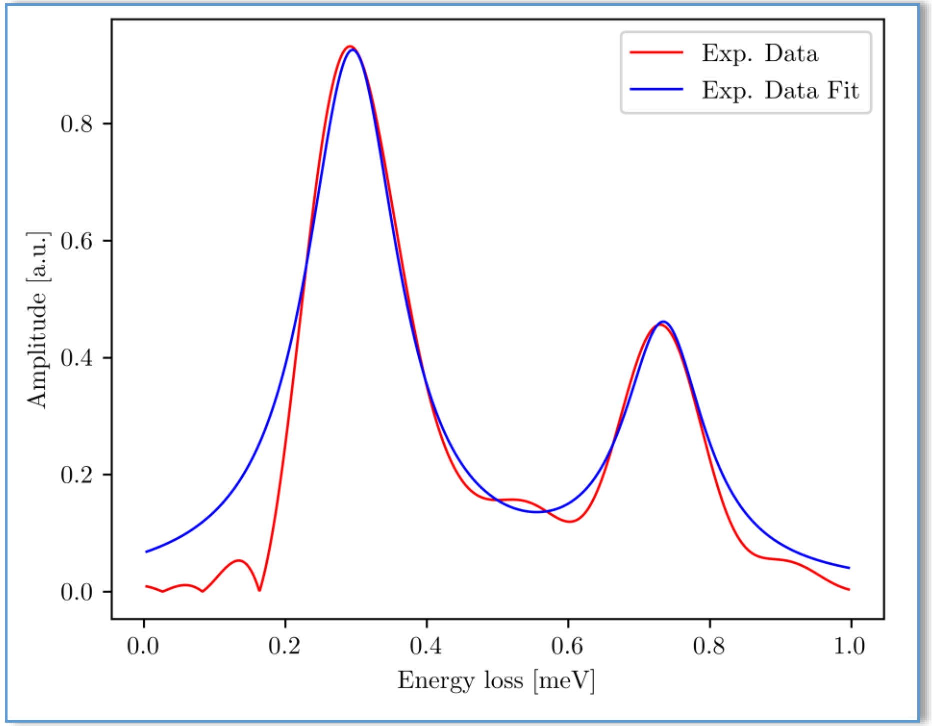




It is noise...

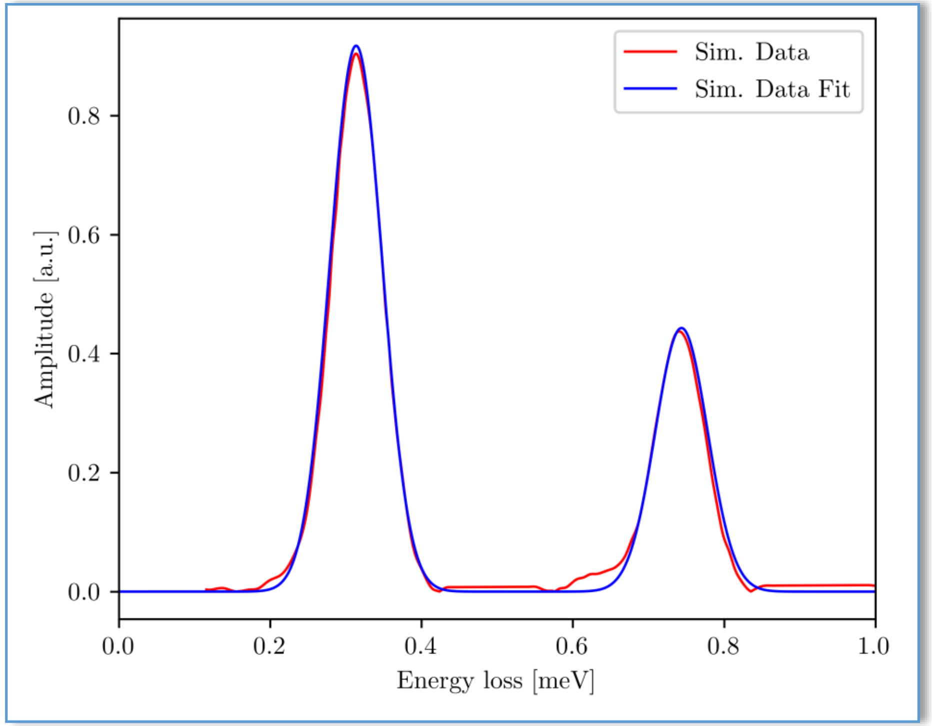
Data Fits

Experimental



Lorentzian

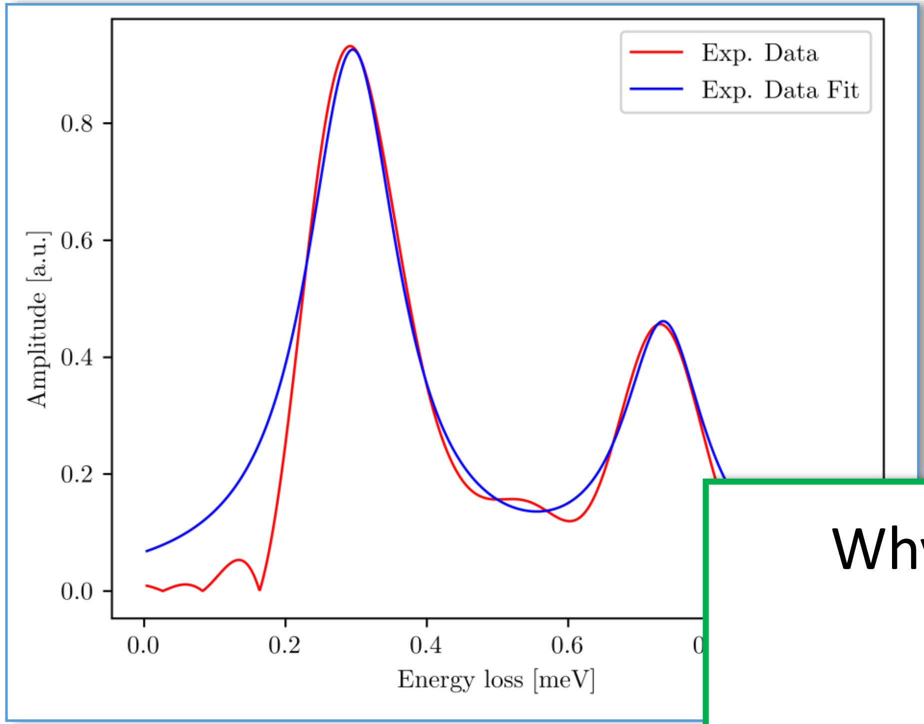
Simulated



Gaussian

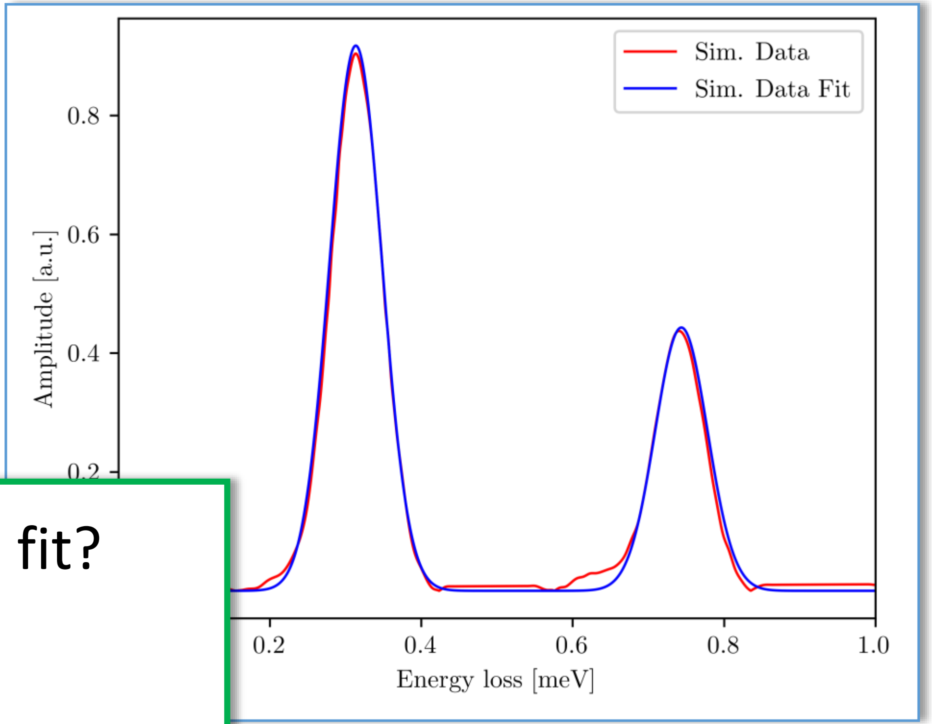
Data Fits

Experimental



Lorentzian

Simulated



Gaussian

Why the different fit?

↓

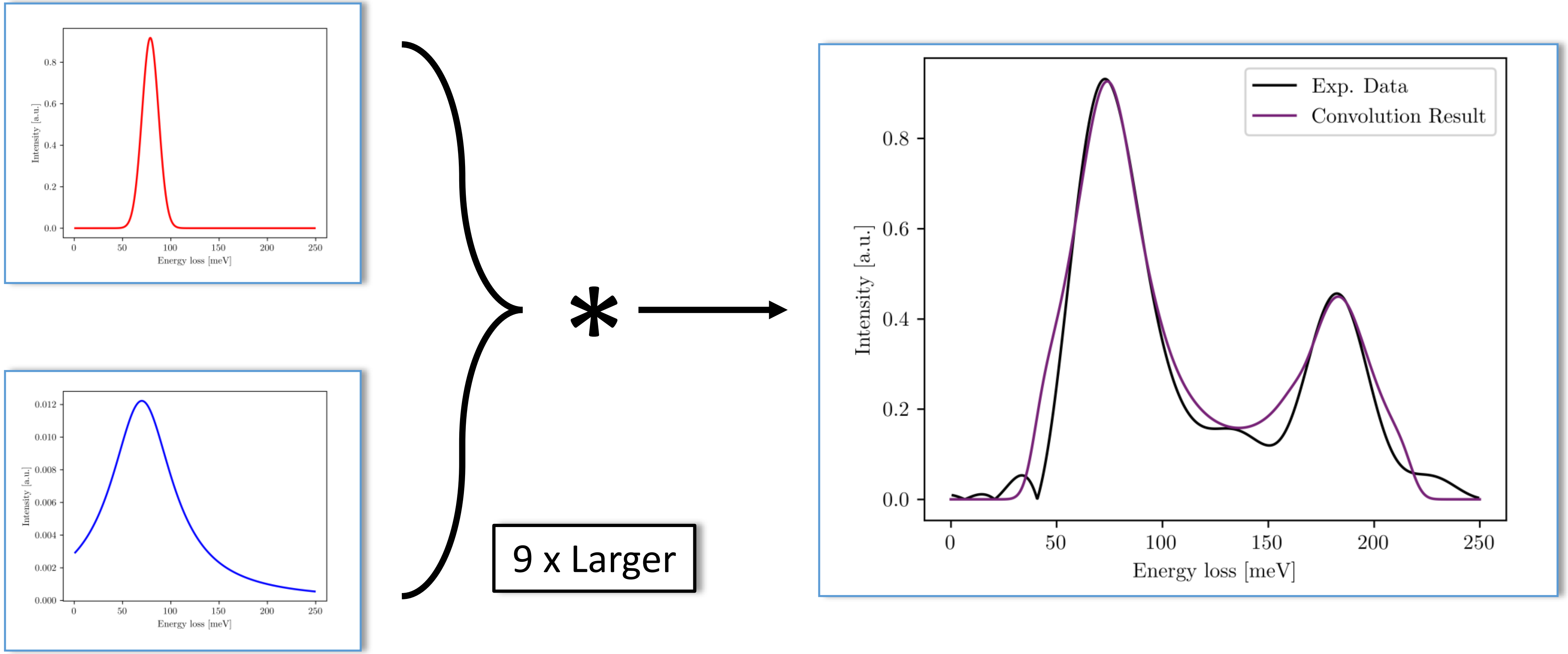
Different couplings lead to different line widths.

Smearing the Data…

Convolution

We start by convoluting the first peak with a Lorentzian. Why?

To broaden the peak. ⇒ Produce a better fit to experimental data.



Overview

- We started by validating the Fourier analysis method with controllable examples;
- A frequency filter was applied to the experimental data, removing the noise and smoothing the results;
- Experimental and simulated data were fitted with different distributions indicating different line width regimes;
- The simulated data was broadened by means of a convolution with a Lorentzian presenting good agreement with experimental data.

Backup Slides…

Let us define the discrete Fourier transform as:

$$f(t) = \sum_{n=-\infty}^{\infty} \frac{1}{2\pi} C_n e^{i\omega nt}$$

The spectral coefficients C_n are easily obtained using:

$$\int_{-\infty}^{\infty} f(t) e^{-i\omega mt} dt = \sum_{n=-\infty}^{\infty} \frac{1}{2\pi} \int_{-\infty}^{\infty} C_n e^{i\omega(n-m)t} dt$$

$$\int_{-\infty}^{\infty} f(t) e^{-i\omega mt} dt = \sum_{n=-\infty}^{\infty} C_n \delta_{m,n}$$

$$\int_{-\infty}^{\infty} f(t) e^{-i\omega mt} dt = C_m$$

Let us once again use the definition:

$$2\pi f(t) = \sum_{n=-\infty}^{\infty} C_n e^{i\omega nt}$$

which may be re-written as:

$$2\pi f(t) = \sum_{n=1}^{\infty} C_n e^{i\omega nt} + \sum_{n=1}^{\infty} C_{-n} e^{-i\omega nt} + C_0$$

we now note that $e^{i\omega t} = cos(\omega t) + i\sin(\omega t)$. Thus:

$$2\pi f(t) = \sum_{n=1}^{\infty} C_n[cos(\omega t) + i\sin(\omega t)]$$
$$+ \sum_{n=1}^{\infty} C_{-n}[cos(n\omega t) - i\sin(n\omega t)] + C_0$$

From here it is simple to show that $f(t)$ can be put in the form:

$$f(t) = \frac{1}{2\pi}\left[C_0 + \sum_{n=1}^{\infty} A_n \cos(n\omega t) + B_n \sin(n\omega t)\right]$$

where $A_n = C_n + C_{-n}$ and $B_n = i(C_n - C_{-n})$.
Some remarks:

- If $f(t)$ is an even function of t, then all B_n must vanish and thus $C_n = C_{-n}$;
- If $f(t)$ is an odd function of t, then all A_n (and C_0) must vanish and thus $C_n = -C_{-n}$;
- The parity is conserved between the dual spaces;
- C_0 gives the baseline.

Yet another way we could handle this expression is the following:

$$f(t) = \frac{1}{2\pi}\left[C_0 + \sum_{n=1}^{\infty} A_n \cos(n\omega t) + B_n \sin(n\omega t)\right]$$

$$= \frac{1}{2\pi}\left[C_0 + \sum_{n=1}^{\infty} D_n \cos(\phi_n)\cos(n\omega t) + D_n \sin(\phi_n)\sin(n\omega t)\right]$$

$$= \frac{1}{2\pi}\left[C_0 + \sum_{n=1}^{\infty} D_n \cos(n\omega t - \phi_n)\right]$$

An analogous procedure could be applied for a sin instead of a cos.