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Laue Data P

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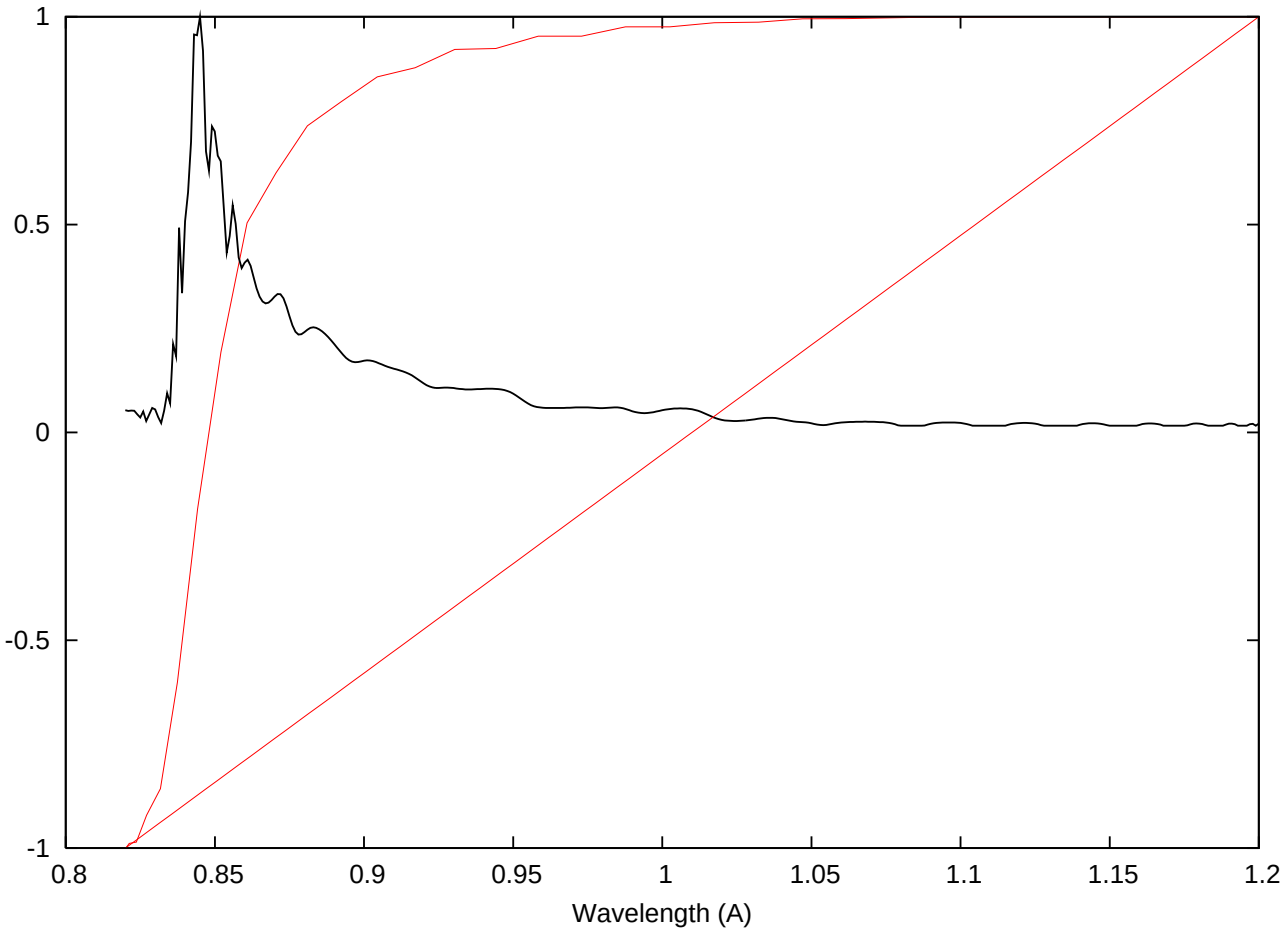
Introduction

One of the uniqueness of Laue diffraction is its use of polychromatic source, X-ray or neutron. The normalization of incident intensities at all wavelengths is therefore a critical step of Laue data processing, which is often known as wavelength normalization. The resulting wavelength-dependent function is called λ-curve. This process does not require an experimentally measured source spectrum, instead the λ-curve can be extracted from the integrated intensities of all reflections. This wavelength-dependent function can be modeled numerically or by empirical polynomials, such as Chebyshev approximation. Recent advances in this area include 1) modeling sharply-varying incident spectrum with less parameters, 2) correction of wavelength assignment errors caused in the steps prior to wavelength normalization, 3) integration over a finite mosaic spread of a protein crystal, and 4) correction of residual errors unaccounted by the spectral model λ-curve. These new features are incorporated in the commercial software Precognition™ for Laue data processing (`http://renzresearch.com/Precognition`).

Nonlinear Wavelength Mapping

Some X-ray sources like undulators generate very sharp-edged spectra, which require more parameters to be modeled accurately compared to a smooth one. New, nonlinear wavelength mapping technique effectively reduces the number of required parameters. For a relatively smooth neutron spectrum, very small number of parameters, say 8 or less, would be enough to describe.

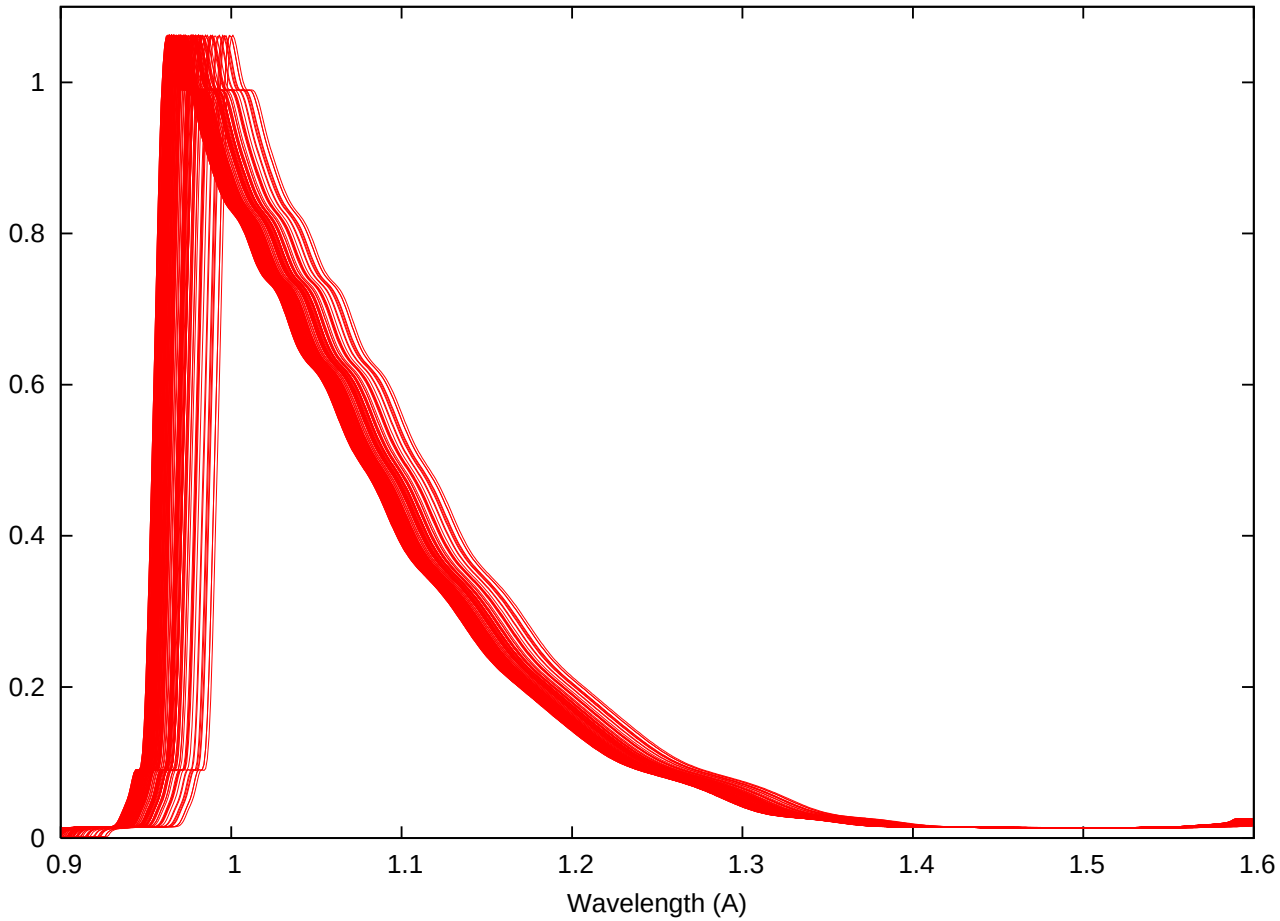
The black λ-curve below models an undulator spectrum of 1.6% bandwidth. This curve is a 40-term Chebyshev approximation with a nonlinear wavelength mapping function shown as the red curve. In comparison, a regular mapping would simply map the wavelength range 0.82 - 1.2 Å to the interval [-1, 1] linearly (straight line in red). A strategically designed nonlinear function maps large intervals on the vertical axis to those wavelength regions where the λ-curve varies sharply or has strong intensity, and only small intervals to two wings of the λ-curve.


1
0.5
0
-0.5
-1
0.8
0.85
0.9
0.95
1
1.05
1.1
1.15
1.2
Wavelength (A)

Correction for Wavelength Assignment Error

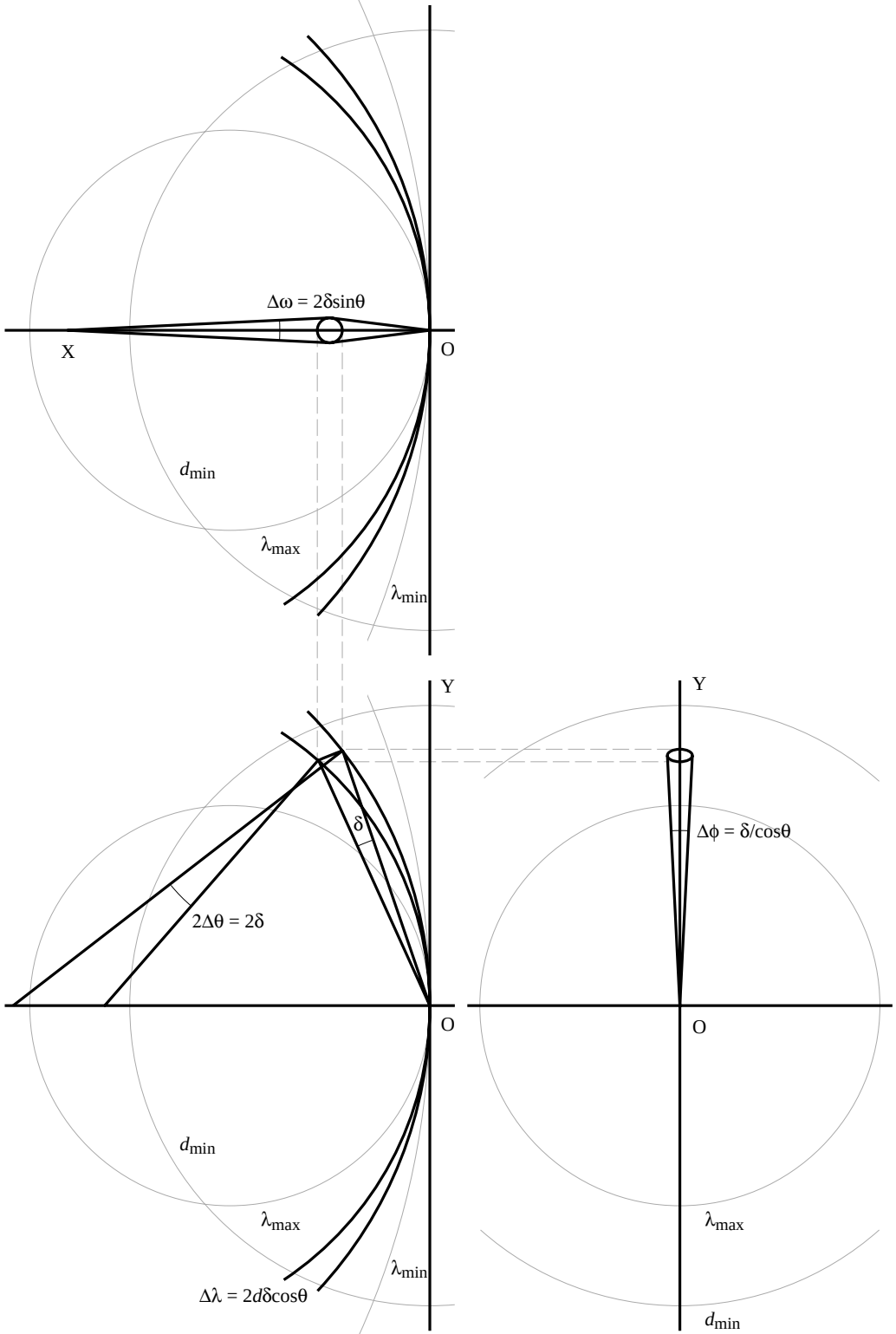
Laue diffraction experiment from X-rays often lacks a characteristic wavelength marker. As a result, interactions between several linear variables (cell lengths, crystal-to-detector distance, detector pixel size, and wavelength) in a Laue experiment does not have a fixed reference scale, which is prone to the possibility of a wavelength assignment error. Since this error is highly systematic, and therefore correctable during the modeling of source spectrum. Time-of-flight neutron diffraction experiment provides a gridlock in wavelength assignment, so that effectively limits the possible error. However, the regular neutron Laue diffraction shares the same problem with X-ray Laue diffraction.

Specific to each exposure, the wavelength assignment error can be modeled by a constant shift and a linear term as if the crystal cell is isotropically expanded. The constant term corrects most of the errors, and the linear one does the rest. Corrections for higher order distortion do not seem to be necessary. As a result of these corrections, λ-curves of all frames are shifted and stretched differently to overcome wavelength assignment error.


1
0.8
0.6
0.4
0.2
0
0.9
1
1.1
1.2
1.3
1.4
1.5
1.6
Wavelength (A)

Effect from Crystal Mosaicity

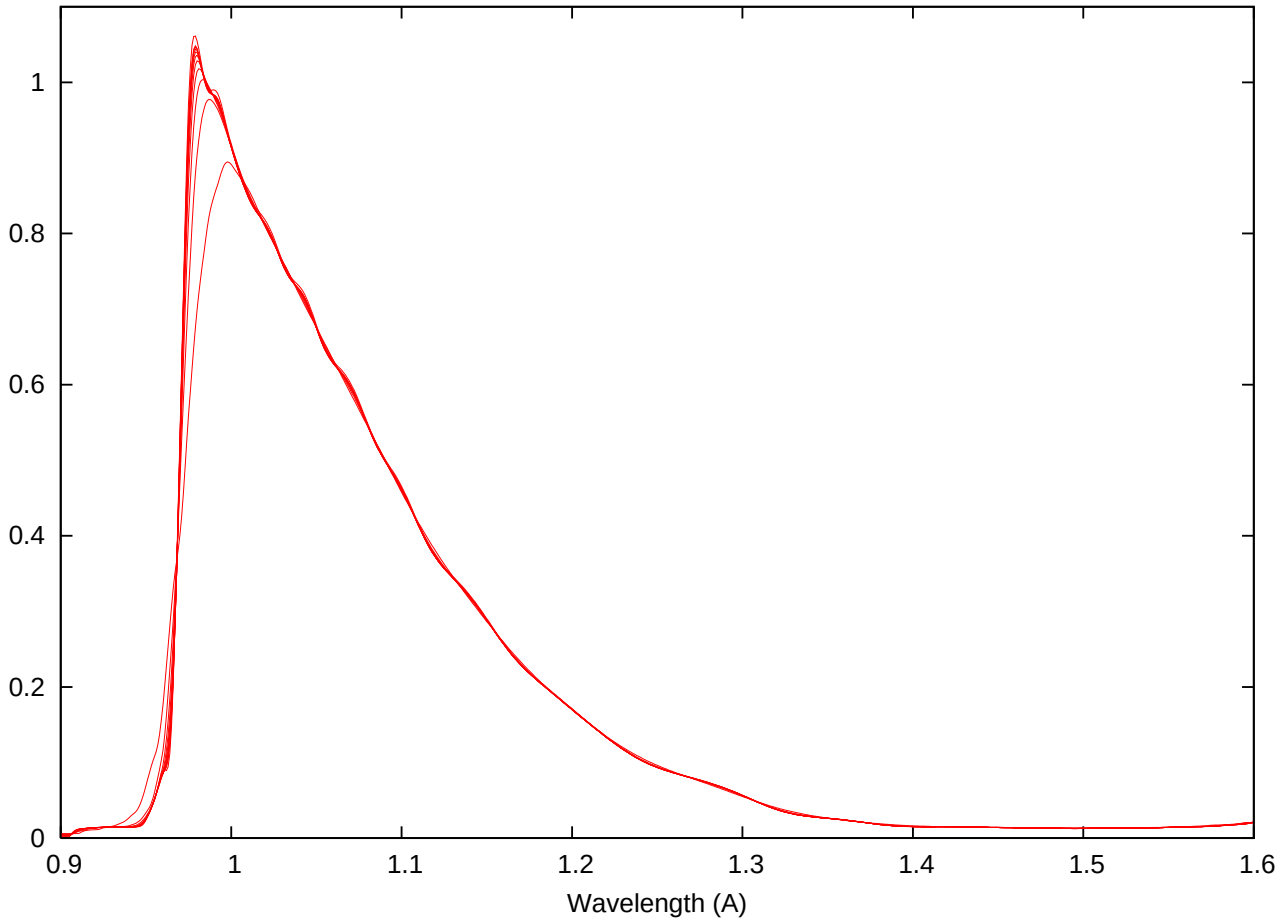
Mosaicity of protein crystals plays an important role in Laue diffraction, but the wavelength span of a reflection due to a finite mosaic spread is often oversimplified to none. This strongly wavelength- and resolution-dependent variable makes some reflections, especially those at lower resolution, partial, and virtually all partial in a time-of-flight neutron diffraction. These energy partials, respect to those angular partials in monochromatic diffraction, can be fully reduced to useful observations if the effect of crystal mosaicity is properly modeled. The graph below shows an Ewald construction with a reciprocal lattice 'cap' instead of a 'point' in order to deduce relationships under the assumption of a crystal mosaic spread of δ.


Δω = 2δsinθ
X
O
d_{min}
λ_{max}
λ_{min}
Y
δ
2Δθ = 2δ
O
d_{min}
λ_{max}
λ_{min}
Δλ = 2dδcosθ
Y
Δϕ = δ/cosθ
O
λ_{max}
d_{min}

It can be shown that relative bandwidth of a Laue reflection is proportional to crystal mosaic spread δ and inversely proportional to tangent of Bragg angle, and approximately the angle itself in nearly all protein cases.

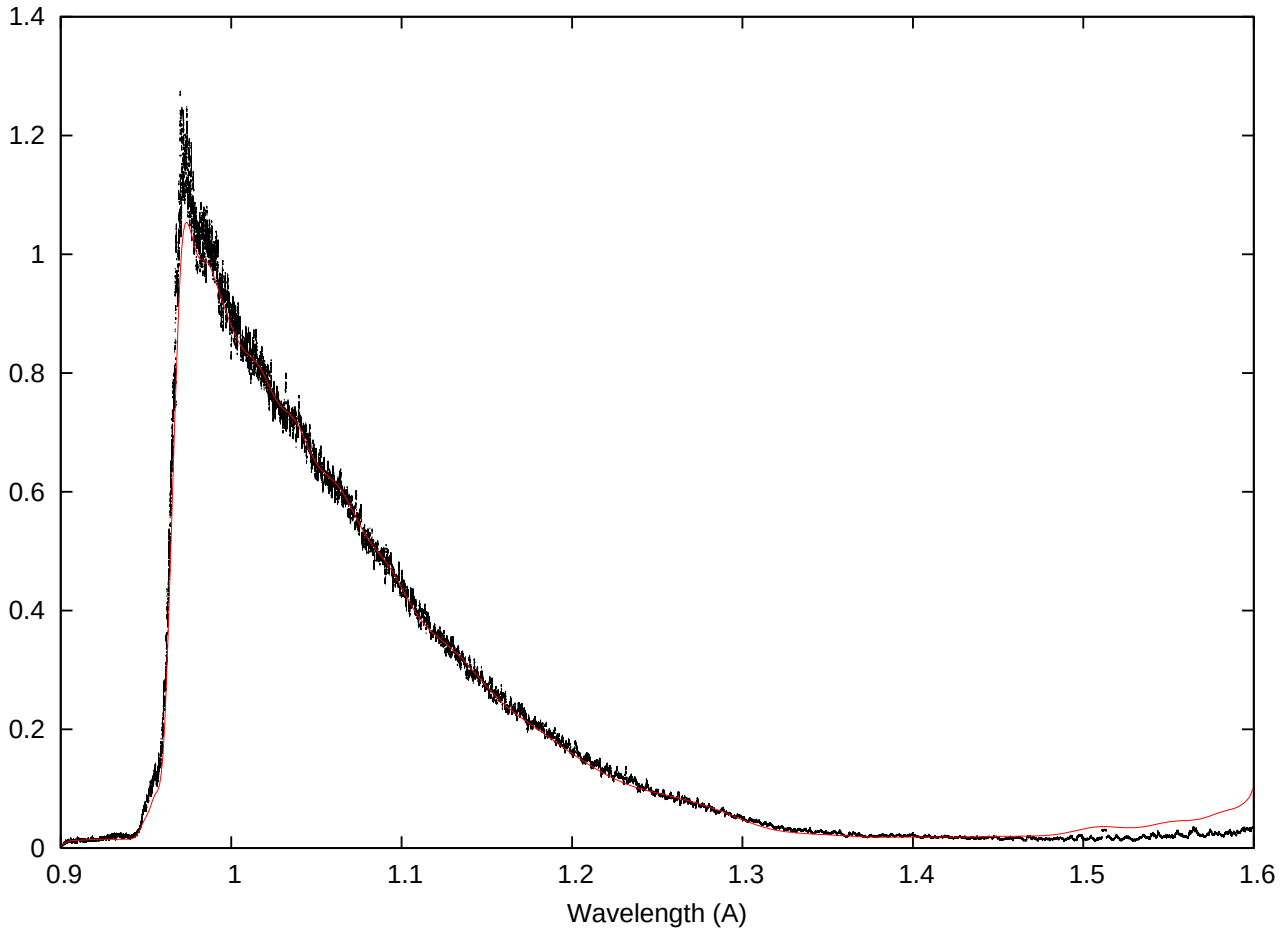
$$\frac{\Delta\lambda}{\lambda} = \frac{\delta}{\tan\theta} \approx \frac{\delta}{\theta}.$$

Integration over the bandwidth $\Delta\lambda$ caused by mosaic spread δ at wavelength λ and Bragg angle θ results in a λ-surface instead of λ-curve. The top curve below stands for wavelength-dependent correction equivalent to no effect of crystal mosaicity. The other curves are at Bragg angles of 1, 2, …, and 10 degrees, respectively, from bottom to top. The mosaic spread is refined to 0.034 degree in full-width at half-maximum.


1
0.8
0.6
0.4
0.2
0
0.9
1
1.1
1.2
1.3
1.4
1.5
1.6
Wavelength (A)

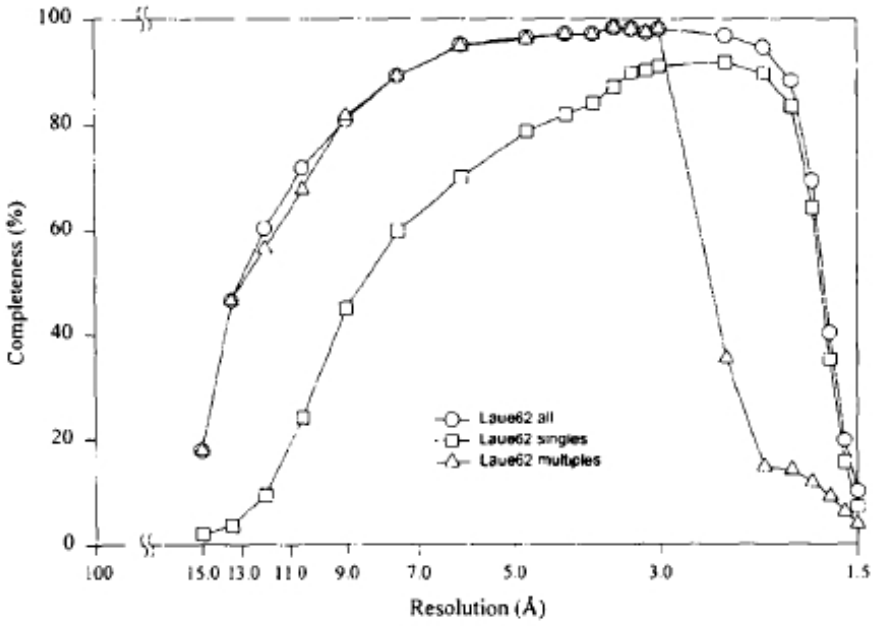
Local Scaling

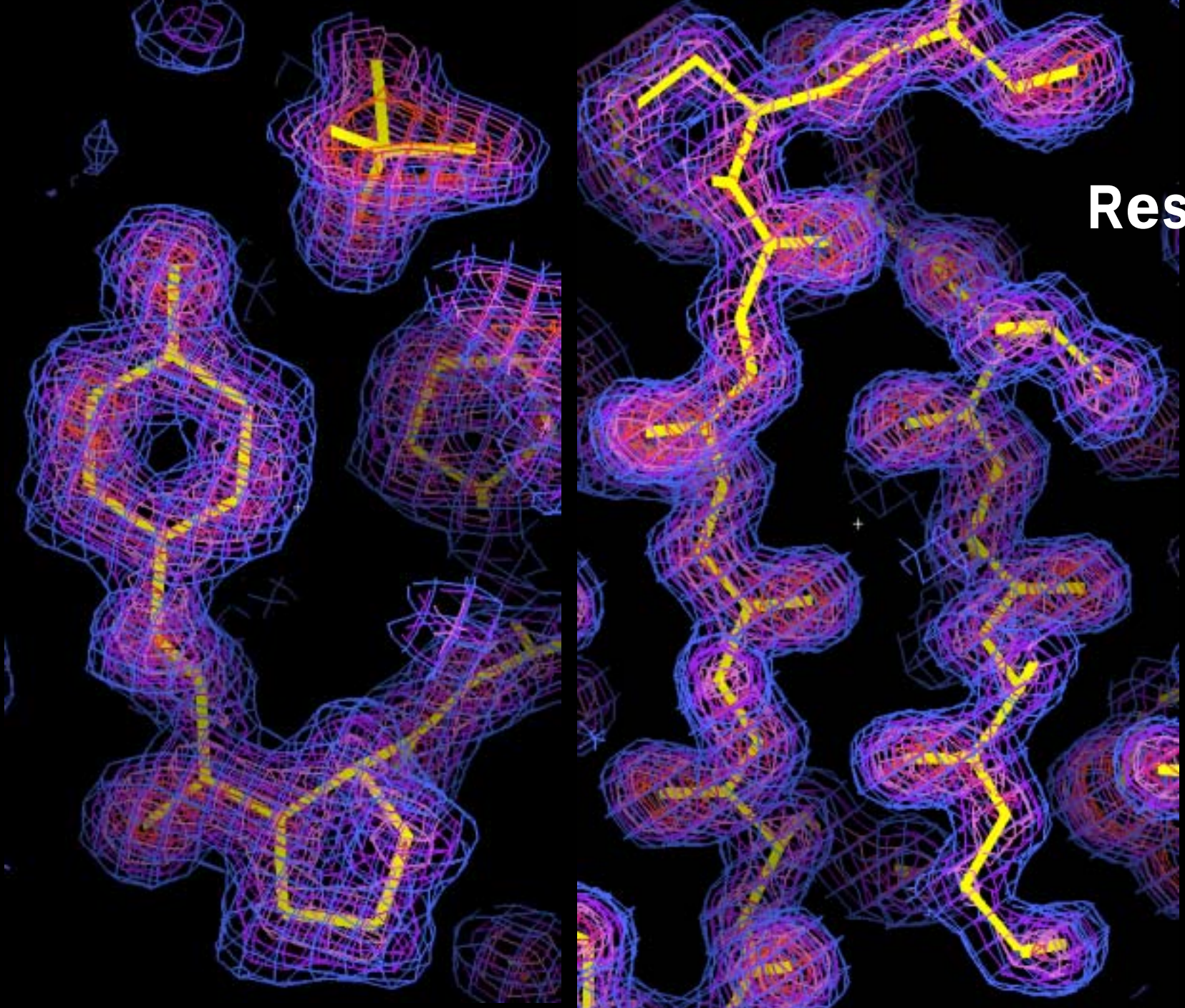
After all, a local scaling procedure corrects residual errors when a rational spectral model has been obtained. Local scaling targets various types of unexplained errors as function of wavelength, integrated intensity, location on detector, Bragg angle, and resolution. The following example shows two λ-curves before (in red) and after (in black) local scaling. The latter becomes even sharper.


1.4
1.2
1
0.8
0.6
0.4
0.2
0
0.9
1
1.1
1.2
1.3
1.4
1.5
1.6
Wavelength (A)

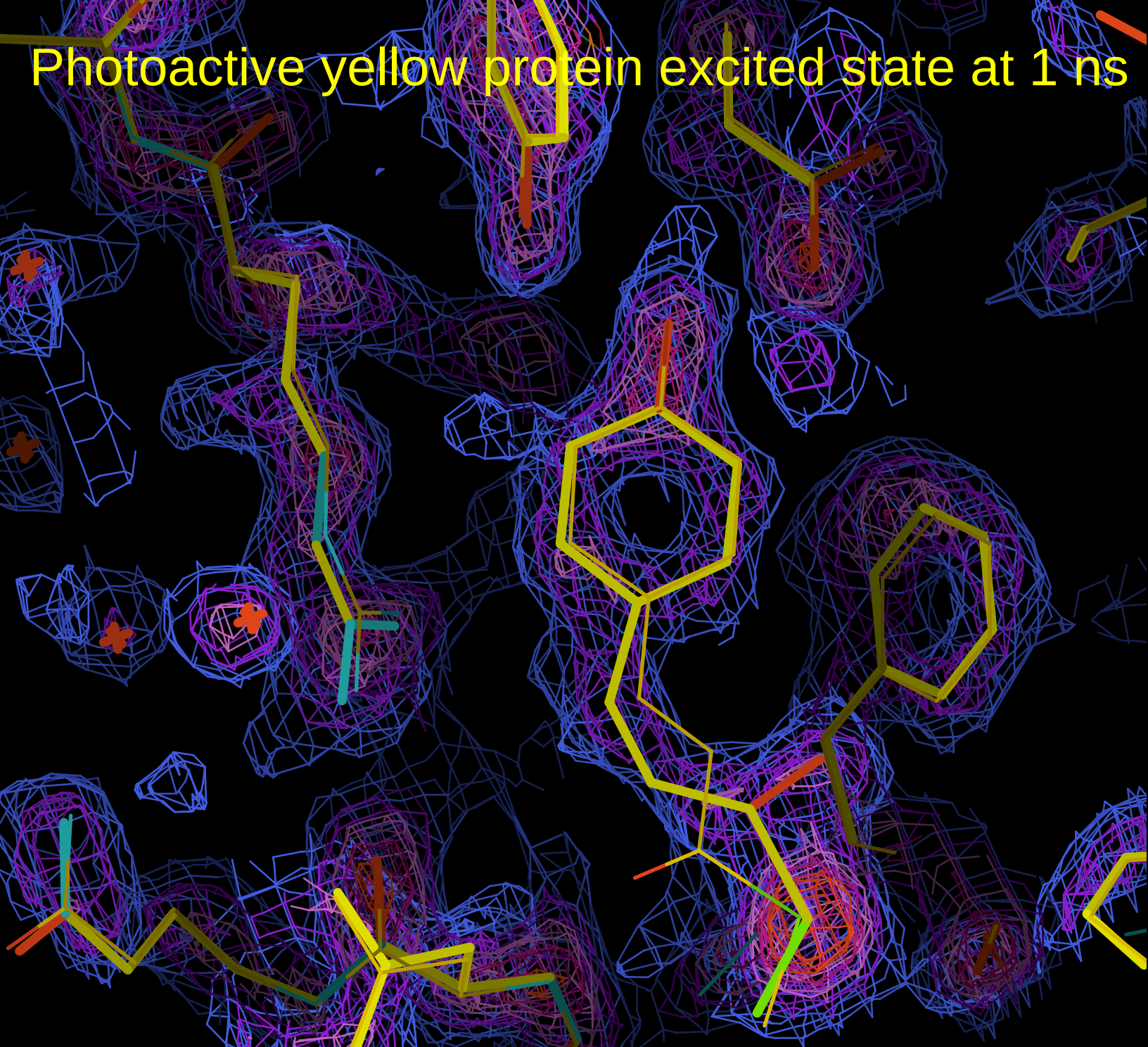
Completeness

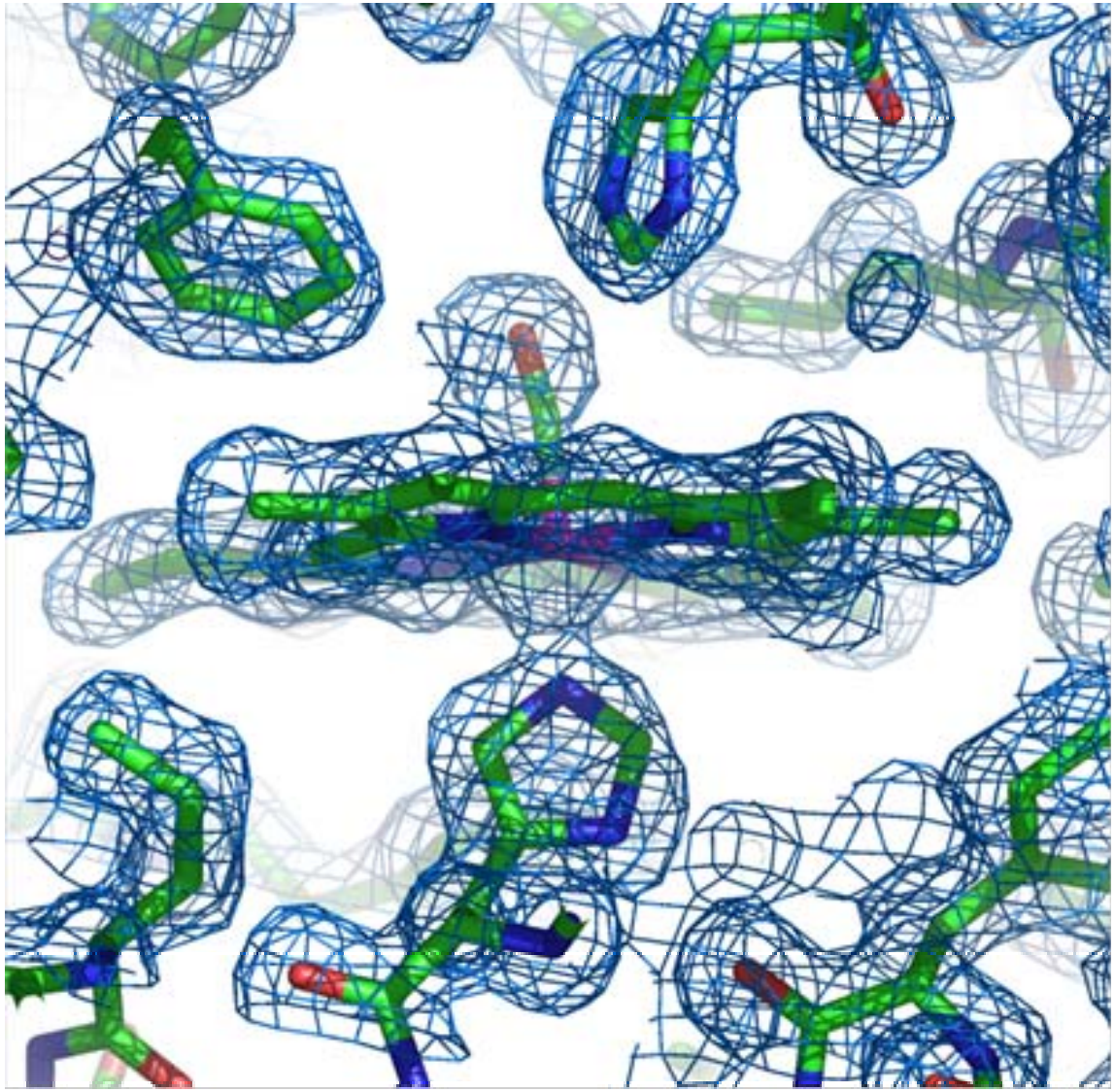
The quality of Laue data critically depends how they are collected and processed. In general, it is very easy to obtain a modestly complete data set in the first a few pictures, if they are wisely spaced. However, it is very hard to absolutely complete a data set at the very low and high resolution ranges, if it is already over 90% complete. The additional exposures will largely measure the data redundantly. This figure shows a typical completeness curve as function of resolution. The accuracy of a Laue data set benefits greatly from its naturally high redundancy. The next several pages show that Laue data can be of superb quality for structural analysis.





Restrictocin
$2F_o-F_c$ maps at 1.7 A


Photoactive yellow protein excited state at 1 ns



$2F_o$-F_c map of dimeric clam hemoglobin at 1.6 Å resolution

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