Detecting excessive instantaneous flux with `screen19`

Ben Williams

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The problem

Photon counting detectors are paralysable, meaning that after a photon is detected at a pixel, there is a short dead time during which the pixel is inactive. This results in some photons going undetected and the higher the frequency of incident photons, the larger the proportion that go missing. The detected count rate for a given pixel can be modelled as

$$C = C_0\, e^{-C_0 \tau},$$

where C_0 is the true rate of incident photons. Up to a point, the detector corrects for this under-counting — the total intensity for each pixel in a given exposure is multiplied by a correction factor. However, there is an implicit assumption that the flux incident on each pixel does not vary significantly during the exposure. The correction is applied to the total measured intensity, effectively correcting an unweighted time-average of the measured flux. Because the averaging takes no account of the non-linear count rate response, a large variation in the instantaneous flux can lead to undetected under-counting, as illustrated in figure A. Where this occurs, the 'corrected' measured flux, Φ_m, will fall short of the true average flux for the pixel, $\bar{\Phi}$. The recorded intensity, $I \propto \Phi_\mathrm{m}$, will therefore be too low, a phenomenon referred to as instantaneous overloading.

In a standard rotation/oscillation single-crystal x-ray diffraction experiment, this effect will be particularly significant when the angular dispersion of a reflection (the mosaic spread) is much smaller than the angle swept during the exposure, since the time-averaged flux will then be a poor estimate of the peak flux. Such small, bright reflections are often a feature of small-molecule crystallography. `screen19` serves as an aid in screening a sample, warning of excessive instantaneous flux to prevent collection of compromised data.

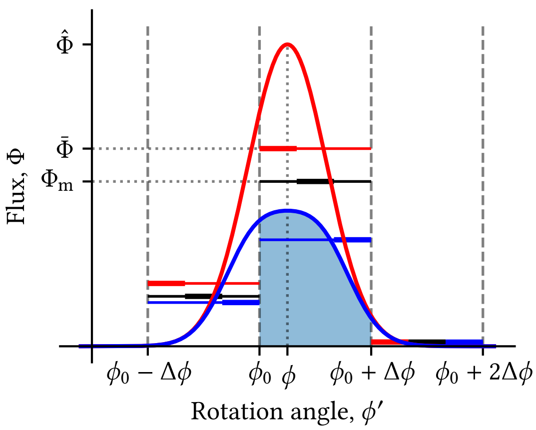

Figure A: Flux at a certain pixel, as a function of rotation angle, in the vicinity of a Bragg peak. Each *dashed vertical line* denotes the boundary between consecutive exposures. *Red trace:* the true incident flux on the pixel. *Blue trace:* the systematically under-counted measured flux. The measured flux for a single exposure (*e.g.* the *shaded blue region*) is aggregated and averaged to achieve a value represented by the *blue bar*. Applying the count rate correction to this average yields the pixel's recorded flux for the exposure, represented by the *black bar*, which systematically falls short of the pixel's true average flux for the exposure (the *red bar*).

The `screen19` solution

The notation below largely follows that of Kabsch (2009), except where otherwise stated.

In terms of the profile coordinates, the flux of scattered photons is commonly modelled as a Gaussian:

$$\Phi = \hat{\Phi}(\varepsilon_1, \varepsilon_2) \cdot \exp\left(-\frac{{\varepsilon_3}^2}{2{\sigma_\mathrm{M}}^2}\right).$$

Hereafter, we ignore the effect of the beam divergence, since σ_D is typically much larger than the pixel size and so there is relatively little variation in flux across a single pixel — instead, $\hat{\Phi}$ is taken to be independent of ε_1 and ε_2.

`screen19` attempts to warn the user of the presence of instantaneous overloading by calculating the profile of the reflections and estimating for each pixel the peak instantaneous flux $\hat{\Phi}$ from the measured flux, Φ_m. It has been empirically determined that the measured flux is unreliable (that is to say, the count correction is poor and $\hat{\Phi} \approx \Phi_\mathrm{m}$ starts to break down) when the instantaneous flux, Φ, is greater than $0.25\times$ the upper limit of the manufacturer's stated trusted range. We can use the fact that $\bar{\Phi}$ is easy to calculate in terms of $\hat{\Phi}$ and, assuming Φ_m increases monotonically with $\hat{\Phi}$, exploit $\hat{\Phi} \approx \Phi_\mathrm{m}$ to convert the upper bound on instantaneous flux into an estimated upper bound on measured flux.

The ζ-approximation allows us to express the reflection profile in terms of the sample rotation angle, $\varepsilon_3 \simeq \zeta \cdot \left(\phi' - \phi\right)$. The parameter ζ is a function of reciprocal lattice vector and experiment geometry. Though the distribution of ζ values for all observed reflections is not trivial to determine, replacing ζ with a single average value $\bar{\zeta}$ allows us to express Φ in terms of ϕ'. The determination of an appropriate value for $\bar{\zeta}$ is left as a point for discussion. For a pixel containing the peak of a reflection, we can then find $\bar{\Phi}$ in terms of $\hat{\Phi}$ by taking the mean over ϕ' from ϕ_0 to $\phi_0 + \Delta\phi$,

$$\begin{aligned}
\bar{\Phi} &= \frac{1}{\Delta\phi} \int_{\phi_0}^{\phi_0+\Delta\phi} \Phi \,\mathrm{d}\phi' \\
&= \hat{\Phi} \cdot \frac{\sqrt{2\pi}\,\sigma_\mathrm{M}}{\bar{\zeta}\,\Delta\phi} \cdot \frac{1}{2}\left(\operatorname{erf}\left(z\left(\phi_0 + \Delta\phi\right)\right) - \operatorname{erf}\left(z\left(\phi_0\right)\right)\right) \\
&= \hat{\Phi} \cdot \frac{\sqrt{\pi}}{2\,\Delta z} \cdot \left(\operatorname{erf}\left(\frac{\bar{\zeta}\cdot\left(\Delta\phi - x\right)}{\sqrt{2}\,\sigma_\mathrm{M}}\right) + \operatorname{erf}\left(\frac{\bar{\zeta}\,x}{\sqrt{2}\,\sigma_\mathrm{M}}\right)\right),
\end{aligned}$$

where $z\left(\phi'\right) = \frac{\bar{\zeta}\cdot(\phi' - \phi)}{\sqrt{2}\,\sigma_\mathrm{M}}$, $x = \phi - \phi_0$ and

$$\Delta z = z\left(\phi_0 + \Delta\phi\right) - z\left(\phi_0\right) = \frac{\bar{\zeta}\,\Delta\phi}{\sqrt{2}\,\sigma_\mathrm{M}}\,.$$

From this, we can see that $\hat{\Phi}/\bar{\Phi}$ lies in the range

$$\frac{\Delta z}{\sqrt{\pi}\,\operatorname{erf}\left(\frac{\Delta z}{2}\right)} \leq \hat{\Phi}/\bar{\Phi} \leq \frac{2\,\Delta z}{\sqrt{\pi}\,\operatorname{erf}\left(\Delta z\right)}$$

according to the value of ϕ (i.e. the position of the flux peak within an oscillation). The lower bound of $\hat{\Phi}/\bar{\Phi}$ corresponds to $\phi = \phi_0 + \Delta\phi/2$ and the upper bound corresponds to $\phi = 0$.

Finding all pixels containing reflection centroids and calculating the value of $\hat{\Phi}/\bar{\Phi}$ in each case is expensive. Rather, we would like to have an indicative measure of the expected ratio of $\hat{\Phi}/\bar{\Phi}$ for pixels containing a peak. $\phi \in \left[\phi_0, \phi_0 + \Delta\phi\right)$ is evenly distributed, but because there is no analytical solution to $\int \hat{\Phi}\,\mathrm{d}\phi$, we cannot calculate $\langle\hat{\Phi}\rangle/\bar{\Phi}$ and must instead use an approximate average, such as $\langle\hat{\Phi}^{-1}\rangle^{-1}/\bar{\Phi}$. We can exploit the symmetry of $\hat{\Phi}$ about $x = \Delta\phi/2$ to simplify the sum:

$$\begin{aligned}
\frac{\left\langle\hat{\Phi}^{-1}\right\rangle^{-1}}{\bar{\Phi}} &= \frac{\Delta\phi}{\bar{\Phi}}\left(\int_{\phi_0}^{\phi_0+\Delta\phi} \hat{\Phi}^{-1}\,\mathrm{d}\phi\right)^{-1} \\
&= \frac{\Delta z\,\Delta\phi}{\sqrt{\pi}}\left(\int_0^{\Delta\phi} \operatorname{erf}\left(\frac{\bar{\zeta}x}{\sqrt{2}\,\sigma_\mathrm{M}}\right)\mathrm{d}x\right)^{-1} \\
= \Delta z^2 &\left(\left[\frac{\sqrt{\pi}\,\bar{\zeta}x}{\sqrt{2}\,\sigma_\mathrm{M}} \operatorname{erf}\left(\frac{\bar{\zeta}\,x}{\sqrt{2}\,\sigma_\mathrm{M}}\right) + \exp\left(-\left(\frac{\bar{\zeta}\,x}{\sqrt{2}\,\sigma_\mathrm{M}}\right)^2\right)\right]_0^{\Delta\phi}\right)^{-1} \\
&= \frac{\Delta z^2}{\sqrt{\pi}\,\Delta z \operatorname{erf}(\Delta z) + e^{-\Delta z^2} - 1}\,. \qquad (1)
\end{aligned}$$

Hence

$$\hat{\Phi} \approx \left\langle\hat{\Phi}^{-1}\right\rangle^{-1} \approx \frac{\left\langle\hat{\Phi}^{-1}\right\rangle^{-1}}{\bar{\Phi}}\,\Phi_\mathrm{m} = \frac{\Delta z^2}{\sqrt{\pi}\,\Delta z \operatorname{erf}(\Delta z) + e^{-\Delta z^2} - 1}\,\Phi_\mathrm{m}\,.$$

`screen19` multiplies the intensity of every pixel by this approximate factor of $\hat{\Phi}/\Phi_\mathrm{m}$. Pixels with a deduced intensity greater than $0.25\times$ the upper limit of the trusted range of the detector are flagged as overloaded. Naturally, since this intensity correction has been optimised for pixels containing the peak of a reflection, it does not produce an accurate estimate of the maximum instantaneous flux for pixels that do not contain a peak. In fact, it still underestimates the maximum instantaneous flux for non-peak pixels. Since the peak-containing pixels are necessarily the brightest however, correctly identifying these is a sufficient guide to the presence and location of instantaneous overloads.

Sanity checks

It is not clear by inspection how equation 1 behaves in the limit $\Delta z \longrightarrow 0$. We expect in such a case, where the mosaicity, σ_M, far exceeds the oscillation, $\Delta\phi$, that the instantaneous flux should not vary significantly across a single exposure. The pixel count correction should therefore give an accurate representation of the true peak flux — we would expect $\hat{\Phi} \approx \Phi_m$ to be a good approximation. Sure enough, a double application of l'Hôpital's rule to equation 1 yields

$$\lim_{\Delta z \to 0} \frac{\langle \hat{\Phi}^{-1} \rangle^{-1}}{\bar{\Phi}} \overset{\left[\frac{0}{0}\right]}{\underset{\mathrm{H}}{=}} \lim_{\Delta z \to 0} \frac{2\,\Delta z}{\sqrt{\pi}\,\mathrm{erf}\,(\Delta z)} \overset{\left[\frac{0}{0}\right]}{\underset{\mathrm{H}}{=}} \lim_{\Delta z \to 0} e^{\Delta z^2} = 1\,.$$

For higher values of Δz, we are less interested in the exact behaviour of $\langle \hat{\Phi}^{-1} \rangle^{-1} / \bar{\Phi}$, since the approximation $\Phi_m \approx \hat{\Phi}$ becomes very unreliable. We need only be satisfied that $\langle \hat{\Phi}^{-1} \rangle^{-1}$ increases monotonically with Δz, such that instantaneous overloads never go undetected.

Choice of value for $\bar{\zeta}$

In a typical goniometer geometry, the sample is rotated around an axis that is normal to the plane in which the detector moves on the 2θ arm. A point detector in this geometry would only be able to measure reflections for which the incident and scattered ray vectors are coplanar with the plane in which the detector moves. An area detector allows for measurement of reflections where the scattering plane (the plane of the incident and scattered ray vectors) may be at a small angle to the plane of the 2θ arm. ζ takes account in the Lorentz correction of this deviation of the scattering plane from the plane of the 2θ arm and theoretically has a range $0 \leq \zeta \leq 1$. In practice, unless the detector is infinitely wide, $\zeta \neq 0$ and in most cases $\zeta \approx 1$.

In the estimation above of expected peak instantaneous flux, it was necessary to designate an average value $\bar{\zeta}$. A proper calculation of this value would require averaging the expected distribution of reflections across the width of the detector. However, since there would be little deviation from $\zeta = 1$ for most geometries, and since little can be lost by making a more conservative estimate, namely the value of $\bar{\zeta}$ that maximises Δz, `screen19` uses the value $\bar{\zeta} = 1$.