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Paganin, David M.; Sales, Morten; Kadletz, Peter M.; Kockelmann, Winfried; Beltran, Mario Alejandro; Poulsen, Henning Frilis; Schmidt, Søren

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Revisiting neutron propagation-based phase contrast imaging and tomography: Effective-brilliance amplification

David M. Paganin
School of Physics and Astronomy, Monash University, Australia

Morten Sales
Department of Physics, Technical University of Denmark, DK-2800 Kgs. Lyngby, Denmark

Peter M. Kadletz
European Spallation Source ESS ERIC, Lund, Sweden

Winfried Kockelmann
STFC-Rutherford Appleton Laboratory, ISIS Facility, Harwell OX11 0QX, UK

Mario A. Beltran, Henning F. Poulsen, Søren Schmidt
Department of Physics, Technical University of Denmark, DK-2800 Kgs. Lyngby, Denmark

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Propagation-based neutron phase-contrast tomography was demonstrated on an insect sample, using the ISIS pulsed spallation source. The tomogram with Paganin-type phase-retrieval filter applied exhibited an effective net boost of 23 ± 1 in the signal-to-noise ratio as compared to an attenuation-based tomogram, implying an effective boost in neutron brilliance of over two orders of magnitude. The phase-retrieval filter applies to monochromatic as well as poly-energetic neutron beams. Expressions are provided for the optimal phase-contrast geometry as well as conditions for the validity of the method. The underpinning theory is derived under the assumption of the sample being composed of a single material, but this can be generalized. The effective boost in brilliance may be employed to give reduced acquisition time, or may instead be used to keep exposure times fixed while improving contrast and spatial resolution.

I. INTRODUCTION

Propagation-based phase contrast has been known for millennia, e.g. in its manifestation as heat shimmer over hot sand or in the twinkling of starlight. The term “propagation-based phase contrast” arises due to the conversion of spatially-varying phase shifts in an optical wave field, namely spatial deformations in its associated wave fronts, into corresponding spatial intensity variations upon propagation [1]. In a more modern setting, one has the out-of-focus contrast that is well known in visible-light microscopy [2, 3] and electron microscopy [4], together with propagation-based phase contrast for X-rays [5, 6] and neutrons [7, 8].

Klein and Opat [7, 8] gave an early demonstration of neutron propagation-based phase contrast, via Fresnel diffraction from a ferromagnetic domain in the context of demonstrating the spinor nature of neutrons. Later examples include Allman et al. [9], who also incorporated phase–amplitude retrieval using the algorithm of Paganin and Nugent [10]. For a textbook account of neutron imaging, see e.g. Utsuro and Ignatovich [11], and for a recent review see e.g. Nelson et al. [12].

In addition to propagation-based methods for achieving neutron phase contrast, which form the main focus of the present paper, several other important methods must be mentioned. Each has their relative strengths, which are clearly delineated in the series of contributions to the book edited by Bilheux et al. [13]. For the use of a two-crystal setup to achieve neutron phase contrast, in which the first crystal serves as a monochromator and the second as an analyzer, see e.g. Freimer et al. [14]. For the use of a Bonse–Hart [15] three-blade interferometer cut from a single monolithic crystal, see e.g. Dubus et al. [16]. For grating-based neutron imaging, see Pfeiffer et al. [17].

This last-mentioned method has also been used to recover small-angle-scattering contrast (also spoken of as “dark-field contrast” within the neutron-imaging and X-ray-imaging communities [18]), that is due to unresolved micro-structure within the sample: see the paper on dark-field neutron tomography by Strobl et al. [19]. For summaries of all methods for neutron phase contrast, together with their relative strengths and differences, see e.g. Pfeiffer [20] and Kardjilov et al. [21]. While all of the listed methods for neutron phase contrast are of high importance, we restrict consideration to the propagation-based method for the remainder of the paper.

One challenge of neutron phase contrast imaging in particular, and neutron imaging more generally, is low neutron brilliance. Exposure times on the order of one hundred seconds are not unusual for neutron microscopy [11], with exposures on the order of tens of seconds per projection in neutron tomography being typical (see e.g. Zhang et al. [22]). The low brilliance requires one to
highly collimate the neutron beam to observe coherence-based effects such as propagation-based phase contrast, further increasing the necessary exposure times. All of the above leads to typical neutron-tomography exposure times on the order of hours (see e.g. [LaManna et al. [23]), with total tomographic scan times on the order of 10 s being deemed “ultra fast” and two-dimensional image-acquisition times on the order of a few seconds being described as having “high temporal resolution” [24].

Reduced acquisition time, for neutron imaging in both two and three dimensions, is a key driver for the present paper. Rather than seeking an increase in neutron source brilliance or detector efficiency, we instead consider how propagation-based phase contrast and subsequent phase retrieval may lead to a significant increase in the effective brilliance of existing sources. This is achieved in an indirect manner which may be viewed as “one step backwards followed by one larger step forwards”, in the sense that it first decreases signal-to-noise ratio (SNR) (via the flux-reducing step of significant collimation) in order to enable a net increase in SNR:

1. We first sufficiently collimate the neutron beam in order to have sufficient spatial coherence for propagation-based phase contrast to be manifest [7–9].

2. Subsequent application of a phase retrieval step, to the propagation-based phase contrast images, boosts the effective SNR of the image and thereby leads to a boost in the effective brilliance of the neutron source.

We close this introduction with a summary of the remainder of the paper. Section II outlines how propagation-based neutron phase contrast, for a single-material sample, may be employed for significant boosts in the effective brilliance of sources used for neutron imaging. We give an experimental demonstration of these ideas, for neutron imaging in both two and three dimensions, in Sec. III. Some broader implications of this work are discussed in Sec. IV together with some suggested avenues for future work. We conclude with Sec. V.

II. THEORY OF EFFECTIVE-BRILLIANCE AMPLIFICATION FOR NEUTRON IMAGING

A. Qualitative explanation of the method

Before presenting the theory underpinning the method, we describe its key aspects in qualitative terms. Consider Fig. 1 which shows a static non-magnetic sample $A$ composed of a single material. This sample is, by assumption, sufficiently weakly attenuating that a contact image, registered over the plane $B$ at the exit surface of the sample, will display little contrast. However if this sample is illuminated by a parallel neutron beam that has been sufficiently highly collimated, the angular divergence $\Theta$ of the neutron beam will be sufficiently small for the refractive properties of the sample to be visible when a propagated image is measured over the plane $C$ at some distance $\Delta$ downstream of the sample. More precisely, we need the penumbral blur width

$$W = \Theta \Delta$$

over the plane $z = \Delta$, to not be so large as to wash out the subtle refractive features due to the sample, in the image that is registered in the plane $z = \Delta$. These refractive features are analogous to the pattern of bright lines seen at the bottom of a swimming pool on a hot sunny day. Rather than the surface of the water in the pool serving to refract sunlight, here local spatial variations in the refractive index of the sample serve to refract neutrons. Thus e.g. if we restrict ourselves to sample materials having a refractive index that is less than unity, as is often but not always the case (see e.g. Table 8.1 in [Pfeiffer [20] for some exceptions), the concave feature at $D$ will act as a converging neutron lens with focal length that is much larger than $\Delta$; thus the neutron intensity measured at $D'$ will be slightly larger due to the converging effect of neutrons passing through point $D$ in the sample. Similarly, convex points on the sample such as $E$ act as locally-diverging neutron lenses, causing the intensity at $E'$ to be slightly less than it would have been in the absence of the sample. This mechanism of using free-space propagation to convert the refractive effects of a sample into contrast in a measured neutron image [8, 9], is known as propagation-based phase contrast. This use of the term “phase” refers to the phase of the neutron wavefunction $\Psi$, since surfaces of constant phase $\varphi \equiv \arg \Psi = \text{constant}$ define neutron wave fronts, and the refractive effects we have described may be considered as arising from distortions in the initially-planar neutron wave front arising from its passage through the sample en route to the detector plane $z = \Delta$. One key aspect, of this form of contrast, is that it emphasizes very fine features that are present in the sample. This image-sharpening behavior is a consequence of the fact that “a smaller lens is a stronger lens”, thus e.g. the smaller the radius of curvature of the concavity at $D$ or the convexity at $E$ in Fig. 1, the greater the consequent intensity increase at $D'$, and the greater the consequent intensity decrease at $E'$. Again, we note that in order to see this effect, the degree of collimation of the neutron beam must be sufficiently high that the penumbral-blur width $\Theta \Delta$ is smaller than the transverse length scale of the fine-level spatial structures that are amplified in visibility in the process of propagation-based neutron phase contrast.

Propagation-based neutron phase contrast images, unlike their attenuation-contrast counterparts, bear only an indirect relationship to the sample being imaged. Stated differently, a decoding step is required in order to obtain quantitative information regarding a sample. This gives the inverse problem [23] of processing the measured propagation-based phase contrast image over the plane $z = \Delta$, to give the projected density of the sam-
FIG. 1. Generic setup for effective-brilliance amplification in neutron phase contrast imaging and tomography. Neutrons are collimated using a pinhole with diameter $d$, before propagating a distance $L$ so as to illuminate a thin single-material object $A$ with number density of nuclei $\rho(x, y, z)$ that is confined to the region $-T < z < 0$. The transmitted neutrons propagate from the exit surface $B$ to a pixellated energy-resolving detector $C$, at distance $\Delta$ downstream of $B$.

B. Mathematical basis of the method

Figure 1 shows a static non-magnetic sample $A$ composed of a single material. Both the number density of nuclei and the number density of atoms are assumed to be equal to one another, being denoted by $\rho(r_\perp, z)$, where $r_\perp = (x, y)$ denote transverse spatial coordinates in planes perpendicular to the optic axis $z$. Neutrons propagating in the $z$ direction are transmitted through the sample, before traveling a distance $\Delta$ and having their resulting spatial intensity distribution $I(r_\perp, z = \Delta)$ recorded by a pixellated planar energy-resolving detector $C$. Mono-energetic neutrons are assumed here, but we note that the theory will later be generalized to the case of broad-band poly-energetic illumination. The neutron radiation is considered to have a divergence

$$\Theta = d/L,$$

(2)

which is sufficiently small that the associated penumbral blur width $\Theta \Delta$, over the plane $z = \Delta$, is less than the transverse shift of neutrons, in the plane $z = \Delta$, that is due to the refractive effects of the sample. Here, $d$ is the diameter of the collimating pinhole, and $L$ is the distance from the collimating pinhole to the sample.

Assume that the degree of collimation is sufficiently high that the image, that is measured over the plane $z = \Delta$, indeed exhibits propagation-based neutron phase contrast (see next sub-section for further detail on this point). We can then turn attention to the corresponding inverse problem of recovering the object projected density, given the propagation-based phase contrast image $I(r_\perp, z = \Delta)$ as input data. If the object is rotated step-wise around the axis $F$ through a variety of angles $\Phi$ (see Fig. 1), and an image over plane $C$ recorded for each $\Phi$ step, the projected density for each angular orientation of the object may subsequently be used for a tomographic reconstruction of the density.
The sample’s complex refractive index is \[ n(r_\perp, z) = 1 - \delta(r_\perp, z) + \frac{i\lambda}{4\pi} \mu(r_\perp, z), \] (3)

where \( \lambda \) is the wavelength and \( \mu \) is the linear attenuation coefficient. Here, the strong assumption of a single-material sample enables us to use the Fermi thin-slab formula \[ \delta(r_\perp, z) = 2r_\perp^2/\lambda^2 \] (4)

for the refractive index decrement, and

\[ \mu(r_\perp, z) = \sigma \rho(r_\perp, z) \] (5)

for the linear attenuation coefficient. Here, \( b \) is the bound neutron scattering length and \( \sigma \) is the total neutron cross section. \[ \rho(r_\perp, z) \]

The projection approximation \[ \chi \] is assumed to hold, hence the intensity \( I \) and phase \( \varphi \) of the neutron beam, at a specified energy \( E \) over plane \( B \), are given by:

\[ I(r_\perp, z = 0) = I_0 \exp[-\sigma \rho_\perp(r_\perp)], \] (6)

\[ \varphi(r_\perp, z = 0) = -b \lambda \rho_\perp(r_\perp). \] (7)

Here, \( I_0 \) is the intensity of the illuminating neutron beam, \( \rho(r_\perp, z) \) is assumed to be non-zero in the volume \( -T \leq z \leq 0 \), and \( \rho_\perp(r_\perp) \) is the projected density:

\[ \rho_\perp(r_\perp) \equiv \int_{-T}^{0} \rho(r_\perp, z)dz. \] (8)

Assuming all streamlines of the neutron current density in the slab \( -T \leq z \leq 0 \) to be almost parallel with the \( z \) axis, we can invoke the paraxial form of the continuity equation implied by the time-independent Klein–Gordon equation. Known as the transport-of-intensity equation \[ \chi \] in a phase-retrieval context, this is:

\[ -\frac{\lambda}{2\pi} \nabla_\perp \cdot [I(r_\perp, z = 0) \nabla_\perp \varphi(r_\perp, z = 0)] = \left. \frac{\partial I(r_\perp, z)}{\partial z} \right|_{z=0}, \] (9)

where \( \nabla_\perp \equiv (\partial/\partial x, \partial/\partial y) \) denotes the gradient operator in the \( xy \) plane.

Let us further assume the object-to-detector distance \( \Delta \) to be sufficiently small that the resulting Fresnel diffraction pattern at \( z = \Delta \) is in the near field of the object. We may now adapt the logic in \[ \chi \] from X-rays to neutrons, as follows: (i) Make a first-order finite-difference approximation to the right-hand side of Eq. (6), using the intensity distributions over the planes \( z = 0 \) and \( z = \Delta \); (ii) use Eqs. (6) and (7); (iii) isolate the intensity over the plane \( z = \Delta \) in the resulting expression. This gives (cf. Eq. (7) in \[ \chi \]):

\[ \frac{I(r_\perp, z = \Delta)}{I_0} = \left( 1 - \frac{b\lambda^2 \Delta}{2\pi\sigma} \nabla_\perp^2 \right) \exp[-\sigma \rho_\perp(r_\perp)], \] (10)

where \( \nabla_\perp^2 \) denotes the Laplacian in the \( xy \) plane. The propagation-based phase contrast in the recorded intensity distribution \( I(r_\perp, z = \Delta) \) is manifest in the Laplacian term on the right side, with its multiplier proportional to the object-to-detector distance \( \Delta \). The right-hand side of Eq. (10) is mathematically identical to the application of a Laplacian-type unsharp-mask image sharpening operator \[ \chi \] to the attenuation-contrast image \( I_0 \exp[-\sigma \rho_\perp(r_\perp)] \), an observation which renders precise our earlier qualitative statements that propagation-based phase contrast serves to sharpen an image. As we show later, this observation is key to the brilliance-boosting nature of the method considered in the present paper.

To proceed further, Fourier transform Eq. (10) with respect to \( x \) and \( y \), utilize the Fourier derivative theorem, solve the resulting algebraic equation for the Fourier transform of \( \exp[-\sigma \rho_\perp(r_\perp)] \), then inverse Fourier transform and solve for the projected density \( \rho_\perp(r_\perp) \). This gives the neutron-optics form of an algorithm previously published for X-rays \[ \chi \] :

\[ \rho_\perp(r_\perp) = -\frac{1}{\sigma} \log_e \left( F^{-1} \left\{ \frac{F[I(r_\perp, z = \Delta)/I_0]}{1 + \frac{b\lambda^2 \Delta}{2\pi\sigma} (k_x^2 + k_y^2)} \right\} \right). \] (11)

Here, \( (k_x, k_y) \) are Fourier-space spatial frequencies corresponding to \((x, y)\), \( F \) denotes Fourier transformation with respect to \( x \) and \( y \), and \( F^{-1} \) is the corresponding inverse transform with respect to \( k_x \) and \( k_y \). We have used a convention in which Fourier transformation converts \( \partial/\partial x \) to \( ik_x \) and \( \partial/\partial y \) to \( ik_y \), according to the Fourier derivative theorem. Note that we require \( b \geq 0 \) to avoid a division-by-zero singularity in the denominator of Eq. (11). This is indeed the case for most but not all materials at thermal and cold neutron energies. Manganese, titanium, vanadium, lithium, and the \(^{1}H\) isotope of hydrogen (together with certain compounds thereof) are important exceptions \[ \chi \].

Since blurring due to finite size is important for typical neutron sources, we follow \[ \chi \] in noting that a stable partial deconvolution for the effective source-blurring area

\[ A = W^2 = (\Theta \Delta)^2 \] (12)

(referred to the imaging plane \( z = \Delta \)) can be achieved using the following logic. Blurring of a two-dimensional image, over the transverse length scale \( W \), can be achieved by applying the blurring operator \( 1 + \frac{b\lambda^2 \Delta}{2\pi\sigma} \nabla_\perp^2 \) to that image \[ \chi \]. Hence the effects of divergence-induced blurring may be taken into account by acting on the right side of Eq. (10) with the operator \( 1 + \frac{b\lambda^2 \Delta}{2\pi\sigma} \nabla_\perp^2 \approx 1 + \frac{1}{8} A \nabla_\perp^2 \), to give an equation of the Fokker–Planck type...
where the algorithm is thus rather simple, being the low-pass \( \tau > \sigma / \sigma \) depends on the single parameter \( \sqrt{\pi \sigma} \). Thus, to account for divergence-induced blurring, we need to make the following replacement in Eq. (11) \([46]\):

\[
\frac{b\lambda^2}{2\pi\sigma} \rightarrow \frac{b\lambda^2}{2\pi\sigma} - \frac{A}{8}.
\]  

This replacement extends Eq. (11) into the main result of the present paper (cf. Eq. (8) of Beltran et al. \([46]\)):

\[
\rho_\perp(r_\perp) = \frac{1}{\sigma} \log_e \left( F^{-1} \left\{ \frac{F[I(r_\perp, z = \Delta)/I_0]}{1 + \tau(k_x^2 + k_y^2)} \right\} \right),
\]  

where

\[
\tau = \frac{\lambda^2 b\lambda}{2\pi\sigma} - \frac{A}{8} = \frac{1}{2} \left[ \frac{\lambda^2 b\lambda}{\pi\sigma} - \frac{(\Theta\Delta)^2}{4} \right].
\]

The above expression permits the projected density \( \rho_\perp(r_\perp) \) of the single-material sample, to be obtained from a single propagation-based phase contrast image \( I(r_\perp, z = \Delta) \). With the exception of the numerically trivial multiplicative constant \( 1/\sigma \), this reconstruction depends on the single parameter \( \tau > 0 \). The core of the algorithm is thus rather simple, being the low-pass Lorentzian Fourier-space filter \( 1/(1 + \tau(k_x^2 + k_y^2)) \).

The analysis process, given in Eqs. (15) and (16), is equivalent to the following algorithm:

1. Take a single propagation-based phase contrast neutron image with measured intensity distribution \( I(r_\perp, z = \Delta) \), as a function of 2D position coordinates \( r_\perp \) in the detector plane, and then normalise (or, more generally, flat-field correct) via division by the background intensity \( I_0 \).

2. Apply a fast Fourier transform \( F \) to the normalised image, thereby generating a complex image that is a function of the Fourier-space coordinates \( (k_x, k_y) \). Each pixel in this Fourier-space image will have height (width) equal to \( 1/W_x \) or \( 1/W_y \), where \( W_x \) (\( W_y \)) is the physical width (height) of the original image input into Step #1 above.

3. Multiply the result of Step #2 by the low-pass Fourier filter (Lorentzian filter) \( 1/(1 + \tau(k_x^2 + k_y^2)) \), where the numerical value of \( \tau \) is given by Eq. (16).

Optional: As an alternative to using Eq. (16) to calculate \( \tau \), for non-quantitative studies we may simply tune this parameter according to the criterion that \( \tau \) should be sufficiently small to eliminate phase-contrast fringes from the data, but not so large as to introduce excessive blurring.

4. Apply an inverse fast Fourier transform \( F^{-1} \).

5. Take the natural logarithm of the resulting image, and then divide by \( -\sigma \). The resulting image is a map of the projected density \( \rho_\perp(r_\perp) \). For non-quantitative studies this step may be omitted.

In a tomographic setting, the above process may be applied to each projection, with the resulting processed images subsequently being input into a conventional tomography reconstruction process such as filtered-backprojection or algebraic reconstruction methods \([39]\).

C. Choice of degree of neutron collimation

A condition that the denominator in Eq. (15) never vanishes is that \( \tau > 0 \). Making use of the second definition for \( \tau \) in Eq. (16), we obtain the collimation condition:

\[
\Theta < \Theta_{\text{critical}} = 2\lambda \sqrt{\frac{b}{\pi\sigma\Delta}} = \sqrt{\frac{8\delta}{\Delta\mu}}.
\]  

This condition may be thought of as quantifying the need for sufficiently high spatial coherence, for the propagation-induced neutron phase contrast to be non-negligible \([50]\). It may therefore be considered as a rule of thumb required to be in a phase-contrast imaging regime, which ensures that the penumbral blurring due to non-zero divergence does not entirely wash out propagation-based phase contrast (cf. Gureyev et al. \([51]\)). Note that the various functional dependencies in Eq. (17) make intuitive physical sense. Thus, the larger the object-to-detector distance \( \Delta \), the greater the effect of penumbral blurring and hence the more stringent is the required collimation condition. Conversely, the smaller the value of \( b/\sigma \) or \( \delta/\mu \), the weaker the propagation-based phase contrast signal and hence the more stringent the collimation condition needs to be.

The collimation condition in Eq. (17) immediately implies the following trade-off. Greater beam divergence \( \Theta \) gives more neutrons and therefore better statistics, but worse phase contrast on account of the associated blurring leading to suppressed propagation-based phase contrast. Conversely, smaller beam divergence gives improved phase contrast on account of the improved spatial coherence, at the expense of higher noise in the measurement statistics. What is an optimum degree of collimation, given this trade-off?

The term in large round brackets on the final line of Eq. (13), namely \( \tau \) as given in Eq. (16), is a measure of the visibility of the propagation-based phase contrast.
features that we would observe in the high-neutron-flux limit (zero noise limit). See the Appendix for a justification of this claim. This noise-free-case visibility decreases with increasing divergence $\Theta$, since $\tau$ gets smaller as $\Theta$ gets bigger. Conversely, the SNR associated with a finite neutron flux, will scale proportionally to the square root of the solid angle $\Theta^2$ associated with the divergence, so that SNR $\propto \Theta$. Therefore the quantity we need to extremize with respect to $\Theta$ is $\tau$ SNR (cf. Rule #3 in the cited paper by Gureyev et al. [51]):

$$\tau \text{SNR} \propto \left[ \frac{\lambda^2 b \Delta}{\pi \sigma} - \frac{(\Theta \Delta)^2}{4} \right] \Theta.$$  

(18)

Differentiating the above equation with respect to $\Theta$, and setting the result to zero, gives an optimal divergence $\Theta_{\text{optimum}}$ that is $1/\sqrt{3} \approx 60\%$ of the maximum value given in the collimation condition (Eq. (17)). Thus we have the optimum divergence

$$\Theta_{\text{optimum}} = (1/\sqrt{3}) \Theta_{\text{critical}}.$$  

(19)

This gives rise to another simple rule of thumb: Reduce the divergence to approximately 60% of the value at which phase contrast is first observed in the image, to give a near-optimal experiment in light of the trade-off mentioned in the previous paragraph. Further convenient rules of thumb, for optimized propagation-based phase contrast imaging, can be found in the previously cited paper by Gureyev et al. [51]. Note also, that in line with the well-known trade-off between noise and resolution [52], one may opt to reduce the divergence below the “optimum effective brilliance” value given above, which will improve spatial resolution in the phase-retrieval reconstruction at the expense of increasing either the noise or the data-acquisition time [53].

D. Effective-brilliance amplification

The single-parameter reconstruction in Eq. (15) is mathematically identical to the so-called Paganin method [20]. This latter algorithm, which has been widely applied in an X-ray setting, exhibits extreme stability with respect to noise in the input phase-contrast image [54]. Indeed, in an X-ray setting, the algorithm has been seen to boost SNR by two orders of magnitude or more, enabling acquisition times to be reduced by four orders of magnitude or more [27,51]. However for neutrons the situation is less favorable, since, as has already been mentioned, this SNR boost must be traded off against the SNR reduction that results from collimating the divergence to a sufficient degree that the collimation condition in Eq. (17) is satisfied. We consider this trade-off in more detail below, with a view to determining the conditions under which the loss in flux, due to increased collimation, is sufficiently compensated by the effective-brilliance boost of the subsequent phase-retrieval step.

Let $\Theta_0 > \Theta_{\text{critical}}$ denote the beam divergence that would be utilized in the context of attenuation-contrast neutron imaging. If we collimate sufficiently hard that the divergence is now reduced to $\Theta_{\text{critical}}$, the corresponding flux will scale by the multiplicative factor $(\Theta_{\text{critical}}/\Theta_0)^2$. This leads to a loss in SNR corresponding to the multiplicative factor

$$f = \frac{\Theta_{\text{critical}}}{\Theta_0} = \frac{2\lambda}{\Theta_0 \sqrt{\frac{b}{\pi \sigma \Delta}}}.$$  

(20)

Next, we apply the formulae of Nesterets and Gureyev [29] and Gureyev et al. [30]. There, the maximum SNR gain (due to the phase-retrieval step) $G_{\text{max}}$ in a tomographic setting [55] is:

$$G_{\text{max}} \approx \frac{0.3 \delta}{\beta} \approx \frac{\pi \delta}{\lambda \mu},$$  

(21)

where (i) $\beta = \text{Im}(n) = \lambda \mu/(4\pi)$; (ii) we have made the approximation that $0.3 \times 4 \approx 1$. Using Eqs. (4) and (5),

$$G_{\text{max}} \approx \frac{b \lambda}{2 \sigma}.$$  

(22)

To take the effects of penumbral blurring into account, Eq. (14) implies that we must make the following replacement in Eq. (22):

$$\frac{b \lambda}{2 \sigma} \rightarrow \frac{b \lambda}{2 \sigma} - \frac{\pi A}{8 \lambda \Delta},$$  

(23)

leading to:

$$G_{\text{max}} \rightarrow \frac{b \lambda}{2 \sigma} - \frac{\pi A}{8 \lambda \Delta}.$$  

(24)

The net corresponding boost in effective brilliance, when both the collimation SNR loss and the phase retrieval SNR boost are taken into account, is:

$$B_{\text{max}} = f^2 G_{\text{max}}^2 = \frac{b \lambda^2}{\pi \sigma \Delta \Theta_0^2} \left( \frac{b \lambda}{\sigma} - \frac{\pi A}{4 \lambda \Delta} \right)^2 > 1.$$  

(25)

If we choose the optimal collimation given by Eq. (19), then we may take $A = \Theta_{\text{optimum}} \Delta^2$ and so Eq. (25) becomes:

$$B_{\text{max}} = \frac{4 b^3 \lambda^4}{9 \pi \sigma^3 \Delta \Theta_0^2} = \frac{\lambda (\delta/\beta)^3}{18 \pi \Delta \Theta_0^2} > 1.$$  

(26)

It is only when the above inequality is satisfied, i.e. when $B_{\text{max}} > 1$, that there is a net boost in effective brilliance on account of the combined effects of increased collimation followed by phase retrieval. If the above inequality is not satisfied, there is no net benefit to be obtained using a phase-contrast approach. Note also that the maximal boost is strongly material dependent as it is proportional to $(\delta/\beta)^3$. 


E. Application to poly-energetic neutron beams

Poly-energetic neutron beams are able to yield propagation-based phase contrast [56, 57] (cf. analogous work in the X-ray regime [58]). We now use a similar argument to that of several X-ray papers on phase retrieval using poly-energetic radiation [52, 51], to show how the method may be applied to poly-energetic neutron beams, for the case of weakly-attenuating samples that are composed of a single material. The assumption of weak attenuation allows us to make the approximation

\[ \exp[-\sigma_\perp(r_\perp)] \approx 1 - \sigma_\perp(r_\perp) \]

in the final line of Eq. (13), leaving us with:

\[
\frac{I_E(r_\perp, z = \Delta)}{I_{0,E}} = [1 - \tau_E \nabla^2] [1 - \sigma_\perp \rho_\perp(r_\perp)] \\
= 1 - \sigma_\perp \rho_\perp(r_\perp) + \sigma_\perp \tau_E \nabla^2 \rho_\perp(r_\perp).
\]

In the above equation, we have (i) made use of the definition for \( \tau \) in Eq. (16), and (ii) explicitly indicated the energy dependence of various quantities via a subscript \( E \). If we multiply through by the neutron energy spectrum \( I_{0,E} \) and then average over energies, as indicated via an overline, we obtain:

\[
I_E(r_\perp, z = \Delta) = I_{\bar{0},E} - (I_{\bar{0},E}\sigma_\bar{E} - I_{\bar{0},E}\tau_E \nabla^2 \rho_\perp(r_\perp)).
\]

Upon introducing the spectrally-averaged quantities defined via

\[
I_{av}(r_\perp, z = \Delta) \equiv \frac{I_E(r_\perp, z = \Delta)}{I_{0,E}}, \quad (\sigma_\av)_{av} \equiv \frac{\sigma_\bar{E}I_{\bar{0},E}/I_{0,E}}, \quad (\sigma\tau)_{av} \equiv \frac{\sigma_\bar{E}\tau_E I_{\bar{0},E}/I_{0,E}},
\]

and applying the weak-attenuation approximation in reverse (this is analogous to passing from the second line to the first line of Eq. (28)), we obtain the following poly-energetic variant of Eq. (13):

\[
I_{av}(r_\perp, z = \Delta) = \left[ 1 - \frac{(\sigma\tau)_{av}}{\sigma_\av} \nabla^2 \right] \exp[-(\sigma_\av)(r_\perp)].
\]

Solving for the projected density, we obtain the main result of this sub-section, namely a poly-energetic version of Eq. (15) that is valid for weakly-attenuating samples:

\[
\rho_\perp(r_\perp) \equiv \frac{1}{\sigma_\av} \log_e \left( \mathcal{F}^{-1} \left\{ \mathcal{F}[I_{av}(r_\perp, z = \Delta)]/ \left[ 1 + \frac{(\sigma\tau)_{av}}{\sigma_\av} (k_x^2 + k_y^2) \right] \right\} \right).
\]

This is identical to the mono-energetic form of the algorithm in Eq. (15) aside from the replacements:

\[
\sigma \rightarrow \sigma_\av, \quad \tau \rightarrow (\sigma\tau)_{av},
\]

together with the use of a poly-energetic neutron phase contrast image rather than an energy-filtered image. This is likely to be advantageous, since there will be a significant boost in utilisable neutron flux on account of there being no need to energy-filter broad-band poly-energetic neutron phase-contrast images.

III. EXPERIMENT

The measurement was performed at IMAT (Imaging and Materials Science & Engineering) neutron imaging and diffraction instrument at ISIS, Oxfordshire, United Kingdom [62-64]. This pulsed spallation source yields both thermal and cold neutrons, with 50 kW power at 10 Hz [62]. Downstream of both source and moderator lie a neutron guide, wavelength-band choppers, beam-shaping elements (pinhole plus slits), sample stage and position-sensitive Multi-Channel-Plate (MCP) Timepix 2 detector [63]. Energy resolution is obtained using time of flight analysis [62, 63]. Exposure times were \( \sim 6 \) minutes per tomographic projection, binned into 524 equal-width wavelength bins ranging from 0.70 Å to 6.76 Å. The pinhole-to-sample distance was \( L = 10 \) m, and the pinhole diameter was \( d = 40 \) mm. The divergence was \( \Theta = \tan^{-1}(d/L) = 0.004 \) radians. 2D tomographic projection angles were used, ranging between \( 0^\circ \) and \( 360^\circ \) in equal angular steps of \( 1.8^\circ \). 2D projection images were taken on a 256 × 256 pixel array, corresponding to one quarter of the available detector surface. The pixel size was \( 55 \times 55 \) \( \mu \)m\(^2\), and the sample-to-detector distance was \( \Delta \approx 30 \) mm. The sample was assumed to be composed of amorphous \(^{12}\)C, giving a scattering length \( b = 6.65 \times 10^{-15} \) m and total neutron cross section \( \sigma = 5.55 \times 10^{-28} \) m\(^2\).

The sample was a common honey bee, shown in the upper panel of Fig. 2. The middle panel of the same figure shows a sample 2D projection image, obtained using the experimental parameters listed above. Here, the selected neutron wavelength was \( \lambda = 5.919 \) Å \( \pm 0.006 \) Å, corresponding to one of the 524 wavelength bins. The event rate recorded by the utilized part of the detector in the selected wavelength bin was 110 neutrons/s/cm\(^2\), this being 0.07% of events in the full wavelength range. This event rate, together with the stated exposure time and pixel dimensions, corresponds to an average of 1.2 neutrons per pixel, hence the SNR of the detected tomographic projection is on the order of unity. Before proceeding, we note that (i) this detected event rate of one neutron per pixel in each tomographic projection has been chosen to demonstrate the ultimate limits of the method; (ii) Paganin-type phase retrieval has previously been successfully applied to quantitative analyses using transmission electron microscope images with on the order of one detected electron per pixel [32]. Application of the phase-retrieval process based on Eqs. (15) and (16), to the tomographic projection in the middle panel of Fig. 2 yielded the image in the lower panel of...
FIG. 2. Top: Image of bee used for measurements. Middle: Neutron propagation-based phase-contrast image data, for one projection, without phase contrast filtering. Bottom: Corresponding neutron attenuation data with phase contrast filter applied.

Fig. 2. A significant boost in SNR is evident in passing from the middle panel to the lower panel of Fig. 2. This SNR boost is consistent with similar observations made in studies using both X-rays [27–31] and electrons [32].

The results of the subsequent tomographic-reconstruction step, based on filtered back-projection [49], are shown in Fig. 3. The top-left (isosurface rendering) and top-middle (tomographic slice of recovered density) panels correspond to filtered-backprojection tomographic reconstruction [49] being applied to the raw projection data. The corresponding post-phase retrieval data yields the iso-surface rendering and tomographic-slice images shown in the top-right and middle-right of Fig. 3, respectively. The bottom row of Fig. 3 shows line profiles of the reconstructed tomographic density, with the blue gray-level profile corresponding to the diagonal trace in the middle-left panel (i.e. without the phase-retrieval step) and the orange gray-level profile corresponding to the diagonal trace in the middle-right panel (i.e. with the phase retrieval step).

Validity condition for the transport-of-intensity equation: Using similar triangles applied to the geometry in Fig. 1, together with the numerical values listed above, the resolution $R$ associated with penumbral blurring can
be estimated as:
\[ R = \frac{d\Delta}{L} \approx 0.1 \text{ mm.} \]  \hspace{1cm} (34)
This resolution of 100 \( \mu \text{m} \) is well-matched to the pixel size of 55 \( \mu \text{m} \). It corresponds to Fresnel number \( N_F \) of
\[ N_F = \frac{R^2}{\lambda \Delta} \approx 800. \]  \hspace{1cm} (35)
This Fresnel number easily satisfies the validity condition
\[ N_F \gg 1 \]  \hspace{1cm} (36)
for the transport-of-intensity equation \([11]\), upon which our analysis is based (see Eq. \([9]\)).

**Collimation condition:** With the specified values of \( b, \lambda, \sigma, \Delta \) as listed above, the collimation condition in Eq. \([17]\) gives the critical divergence (minimum degree of collimation) as \( \Theta_{\text{critical}} = 75^{-1} = 0.013 \) radians. The actual divergence of \( \Theta = 250^{-1} = 0.004 \) radians therefore meets the collimation condition in Eq. \([17]\). Also, from Eq. \([19]\), the optimum divergence is \( \Theta_{\text{optimum}} = 125^{-1} = 0.008 \) radians. Note that the utilized divergence is double that which would be routinely used for attenuation-based neutron imaging (i.e. \( \Theta_0 \approx 125^{-1} \) radians \([62, 63]\)), hence there is a four-fold reduction in flux that is implied by collimating the beam in the present experiment, to improve the spatial coherence for the purpose of increasing the propagation-based phase contrast.

**Effective-brilliance-boosting condition:** With the specified numerical values for \( b, \lambda, \sigma, \Delta \), the condition in Eq. \([26]\) for effective-brilliance boosting becomes \( \Theta_0 < 32 \) radians, which is always true; recall that \( \Theta_0 \approx 125^{-1} \) radians. This establishes the experiment to indeed be in an SNR-boosting regime, a fact which is otherwise clear from the experimental reconstructions in Figs.\([2]\) and \([3]\). To estimate the actual SNR boost obtained in the present experiment, we use the purple and green boxed regions in the left-middle row of Fig. \([3]\) to estimate the pre-phase-retrieval SNR, and use the purple and green boxed regions in the right-middle row of Fig. \([3]\) to estimate the post-phase-retrieval SNR. The ratio of these SNRs gives an SNR boost, due to the phase-retrieval step, of 45 \pm 1 (error bars estimated using similar regions of interest). If we instead use the yellow and green boxed regions, corresponding to a part of the object that has significantly smaller density, the estimated SNR boost (for the phase-retrieval step) is also 45 \pm 1. The SNR boost is relative to the previously-mentioned collimation-related flux reduction of a factor of 4, hence the effective SNR boost needs to be divided by \( \sqrt{4} = 2 \) in order to properly quantify the net effect of (i) SNR reduction due to sufficiently-hard collimation needed to achieve propagation-based phase contrast, followed by (ii) SNR increase due to the subsequent phase-retrieval step. Thus the measured net SNR boost is 23 \pm 1, corresponding to a net effective-brilliance boost of 530 \pm 50. Our experiments are therefore consistent with a boost in effective neutron brilliance of over two orders of magnitude. This may be compared to: (i) the current state-of-the-art in X-ray experiments, which have reported SNR boosts well in excess of four orders of magnitude using Paganin-type phase retrieval \([27, 31]\); (ii) the upper limit of Eq. \([26]\), which indicates that increases in effective brilliance of up to seven orders of magnitude are theoretically possible, for the neutron case considered in the present paper.

**IV. DISCUSSION**

Numerous public-domain software implementations exist, for the X-ray version of Eqs. \([15]\) and \([32]\) \([28]\). These include ANKAphase \([66]\), X-TRACT \([67]\), pyHST2 \([68]\), TomoPy \([69]\) and the SYRMEP TomoProject \([70]\). Since the X-ray and neutron versions are mathematically identical single-parameter reconstructions, such X-ray software (many of which also enable tomographic processing) may be utilized for propagation-based phase contrast neutron data without any modification.

Parallel \( z \)-directed neutrons have been assumed throughout the development of the present paper. However, all of the results readily carry over to the case of divergent illumination from a small neutron source, on account of the Fresnel scaling theorem \([26, 40, 71]\). Here, we simply replace the sample-to-detector propagation distance \( \Delta \) by the scaled propagation distance \( \Delta/M \), where \( M \) is the geometric magnification associated with the point-projection geometry. Thus cone-beam tomographic reconstructions may be performed, in both mono-energetic \([35]\) and poly-energetic settings \([60]\).

The assumption of a single material is less restrictive than it might seem, particularly in three spatial dimensions (i.e. in a tomographic context), where many samples of interest may be locally described as composed of a single material. As shown by Beltran \textit{et al.} \([27, 28]\), we can typically choose the parameter \( \tau \) corresponding to a material of interest, which will only locally corrupt the tomographic reconstruction of features composed of other materials. This locality arises from the fact that the real-space version of the phase-retrieval filter \([72, 73]\), which may be applied after rather than before tomographic reconstruction if the sample is weakly attenuating, has the same functional form as the manifestly-local Yukawa potential \([74]\). Separate tomographic reconstructions may thus be performed for each material of interest, before splicing the resulting reconstructions together using the method of Beltran \textit{et al.} \([27, 28]\). What is the physical reason underpinning the significant SNR-boosting properties of Eq. \([15]\) and Eq. \([32]\) ? We refer the reader to Gureyev \textit{et al.} \([54]\) for the theoretical details, and here give a heuristic explanation to motivate the rigorous results developed there. As pointed out earlier, the propagation-based phase contrast image in Eq. \([13]\) may be viewed as creating a sharpened version of the attenuation-based contact neutron image \( I_0 \exp[-\sigma p_L(r_x)] \). This sharpening follows from
the fact that this equation corresponds exactly to approximate deconvolution (sharpening); this has an associated transverse length scale \( \ell \) given by the square root of the coefficient of the transverse Laplacian \[42\] in Eq. (13):

\[
\ell = \sqrt{\frac{\hbar^2 \Delta}{2 \pi \sigma} - \frac{A}{8}}.
\] (37)

Indeed, from another albeit closely related perspective, Eq. (13) is mathematically identical in form to unsharp-mask image sharpening using a Laplacian kernel \[42\] \[43\] \[44\]. This image sharpening is evident from the edge enhancement due to Fresnel diffraction. Regardless of how we understand the image sharpening, the crucial point to note is that this sharpening occurs before the addition of noise in the detection process. Moreover, if the aforementioned noise is white then it will be evenly spread through Fourier space, in contrast to the object which will typically have a Fourier-space power spectrum that decreases rapidly with increasing radial spatial frequency. The Lorentzian Fourier-space filters in Eqs. (15) and (32), which are low pass filters since their function is to negate the previously mentioned sharpening, will therefore suppress noise much more strongly than they suppress signal due to the object. The result is a strong increase in SNR, corresponding to a boost in effective brilliance.

As previously mentioned, the boost in effective brilliance may be used to give significantly reduced exposure times, for propagation-based neutron phase-contrast tomography and radiography, relative to their attenuation-based counterparts. Alternatively, the SNR boost associated with the effective-brilliance increase may be traded off against increased contrast-to-noise ratio (CNR) and/or resolution, while keeping acquisition times relatively fixed. Increased CNR follows directly from increased SNR. Increased resolution follows from the usual trade-off between noise and resolution \[52\], so that e.g. (i) an increase in SNR by a factor of \( K \) but with no change in spatial resolution may be exchanged for (ii) no SNR increase but an increase in spatial resolution by a factor of \( K \) (by using a smaller pinhole, whose area is \( K \times K = K^2 \) times smaller, than the pinhole that was previously used).

Dark-field imaging, namely the imaging of scattering contrast due to unresolved micro-structure within a sample, is an important topic that has been only cursorily mentioned in the present paper. Indeed, the development of dark-field neutron tomography using a grating-based setup, by \[59\] \[60\] \[61\], is a particularly significant advance that is attracting much attention within the neutron-imaging community. The propagation-based method for neutron phase-contrast imaging, which we consider in the present paper, can be generalized to include dark-field/scattering contrast. This can be achieved using a formalism for paraxial imaging \[48\] \[75\] based on the Fokker–Planck equation \[47\] \[76\]. This generalizes the transport-of-intensity equation \[41\], upon which the present paper is based, enabling it to take into account local small-angle scattering and its associated diffusive transport, in addition to the coherent transport associated with attenuation and refraction. In particular, the single-image single-material propagation-based phase-retrieval method of \[26\] may be generalized to a two-image method that is able to recover both the projected density and the projected small-angle-scattering signal of a single-material object \[48\], given two images taken at two different propagation distances. Further exploration of this extension, in the present neutron-optics context, is a topic for future work.

Another interesting avenue for future work would be to apply Eq. (32) to non-energy-binned broad-band polychromatic neutron propagation-based phase contrast images obtained with smaller pinhole sizes. The idea of phase retrieval using broad-band polychromatic neutron phase contrast images has been considered previously, using a phase-retrieval method that amplifies rather than suppresses noise \[50\]. However, the idea warrants revisiting in the context of the present paper, due to the SNR-boosting properties of Eqs. (32) and (33) \[27\]–\[31\]. For example, in the present study, time-of-flight monochromatization was achieved using 524 equally-spaced energy bins over the wavelength range from 0.70 Å to 6.76 Å. If the pinhole diameter were to have been reduced from \( d = 40 \) mm to \( d = 10 \) mm, the resulting net-flux reduction of \( d^2 = 16 \) could easily have been compensated for by a net flux-boost by a factor of 1400 \[77\], due to not needing to monochromatize and instead having a polychromatic image energy-integrating over all 524 energy bins. Indeed, by using fully poly-energetic neutrons, there would be a net increase in the utilized flux by a multiplicative factor of 1400/16=88, even with the smaller pinhole indicated above; this would boost the SNR of the raw phase-contrast data by a factor of \( \sqrt{88} \approx 9 \), relative to that utilized in the present paper, with an additional boost in SNR to be expected on account of the significantly improved spatial coherence that would be due to the use of a smaller pinhole (cf. \[58\] for an early example of this last-mentioned fact, in the X-ray literature). Also, the corresponding Fresnel number would have been reduced from 800 (see Eq. (35)) to \( N_F \approx 800/A^2 = 50 \), which is still well within the validity condition in Eq. (36). Moreover, the spatial resolution in Eq. (34) would become 25 μm, which remains well-matched to the pixel size of 55 μm.

The present study has been restricted to positive defocus \( \Delta \), since free-space propagation was utilized as the mechanism for phase contrast. However, negative defoci will be accessible if an imaging system is interposed between the sample and the detector, e.g. in possible future applications of the methods of this paper to neutron microscopy \[78\]–\[79\] using compound refractive lenses \[80\]–\[81\]. For such systems, the requirement for \( d \Delta \) to be positive, so as to avoid a division-by-zero divergence in the Fourier filter given in Eq. (15), implies that either (i) the scattering length \( b \) for the material in the single-material sample, and the defocus \( \Delta \), should both be posi-
tive; (ii) $b$ and $\Delta$ should both be negative. In the context of neutron imaging systems, for which residual optical aberrations may be present, we note that [Liu et al.] have developed a form of the algorithm which takes both defocus and spherical aberration into account [82, 83].

V. CONCLUSION

In light of the effective-brilliance boosting possibilities available for propagation-based phase-contrast neutron tomography, it is timely that this modality for three-dimensional imaging be revisited. Theoretical and experimental evidence was presented to support this, for the limited class of samples that can be locally approximated as being composed of a single material. Within its domain of applicability, the method was able to achieve effective-brilliance boosts of greater than two orders of magnitude. This may be used to significantly reduce exposure times and harness the effective-brilliance boost to improve contrast and/or spatial resolution.

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APPENDIX

Here we justify the statement, made in Sec. II C, that the term in large round brackets in the final line of Eq. (13), namely $\tau$ as given in Eq. (16), is a measure of the visibility of the propagation-based phase contrast features that we would observe in the high-neutron-flux limit (zero noise limit).

Work with one transverse dimension $x$, for simplicity. Consider a weakly attenuating single-material sample whose projected number density has the form of a sinusoidal grating:

$$\rho_\perp(x) = \rho_0[\sin(2\pi x/p) + 1].$$

Here, $\rho_0$ is a positive constant, and $p$ denotes the transverse period of the grating.

The attenuation-contrast image $I_{\text{abs}}(x, z = 0)$ will have the intensity profile given by Eq. (6) as

$$I_{\text{abs}}(x, z = 0) = I_0 \exp\{-\sigma_\rho_0[\sin(2\pi x/p) + 1]\}.$$  

The corresponding Michelson visibility [84] is

$$V_{\text{abs}} = \frac{\max[I_{\text{abs}}(x, z = 0)] - \min[I_{\text{abs}}(x, z = 0)]}{\max[I_{\text{abs}}(x, z = 0)] + \min[I_{\text{abs}}(x, z = 0)]}.$$  

where max/min respectively denotes the maximum or minimum value of the corresponding argument, and the assumption of weak attenuation has been used to make the approximation $\exp(-2\sigma_\rho_0) \approx 1 - 2\sigma_\rho_0$. For the propagation-based phase contrast image $I_{\text{prop}}(x, z = \Delta)$, Eqs. (13), (16) and (38) give, in the weak-attenuation limit,

$$I_{\text{prop}}(x, z = \Delta) \approx I_0 - \frac{I_0 - I_0 \exp(-2\sigma_\rho_0)}{I_0 + I_0 \exp(-2\sigma_\rho_0)} \approx \sigma_\rho_0,$$  

where $\sigma_\rho_0$ is a positive constant, and the approximation $\exp(-2\sigma_\rho_0) \approx 1 - 2\sigma_\rho_0$. This displays the linear proportionality with $\tau$ that we set out to demonstrate.

We close this appendix by noting that the boost in visibility, obtained in using propagation-based phase contrast in comparison to attenuation contrast, is also directly proportional to $\tau$:

$$\frac{V_{\text{prop}}}{V_{\text{abs}}} = 1 + 4\pi^2\tau \rho^2.$$  

This visibility boost underpins the effective-brilliance boost that is the main topic of the present paper.

Note that this usage of the term “dark field” differs from the 1920 review by Gage [85].
duces from a number that is much greater than unity to a number on the order of unity.

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[55] The maximum SNR gain is different for two-dimensional imaging (radiography) versus three-dimensional imaging (tomography). The gain is somewhat larger for the tomographic case [29, 30].


[76] For the experiment using energy-filtered neutrons reported in Sec. III, the neutron flux recorded by the detector without the sample in the beam, over the utilized part of the detector at the selected wavelength bin of 5.9 Å, was 110 neutrons/s/cm$^2$ (averaged over the acquisition time of 6 minutes). For the entire wavelength range measured, the flux was about $1.56 \times 10^5$ neutrons/s/cm$^2$ measured by the detector. Hence the net boost in utilized flux, that is to be obtained by not needing any energy filter, is $1.56 \times 10^5 /110 \approx 1400$.