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Computer simulation tests of optimized neutron powder diffractometer configurations

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ABSTRACT

Recent work has developed a new mathematical approach to optimally choose beam elements for constant wavelength neutron powder diffractometers. This article compares Monte Carlo computer simulations of existing instruments with simulations of instruments using configurations chosen using the new approach. The simulations show that large performance improvements over current best practice are possible. The tests here are limited to instruments optimized for samples with a cubic structure which differs from the optimization for triclinic structure samples. A novel primary spectrometer design is discussed and simulation tests show that it performs as expected and allows a single instrument to operate flexibly over a wide range of measurement resolution.

1. Introduction

Neutron powder diffraction is a valuable technique in studies of condensed matter. In comparison with other techniques, notably X-Ray diffraction, neutron diffraction has advantages in locating light atoms in crystal lattices and elucidating magnetic structures. The intensities in neutron scattering work are often very low and so measurements are usually quite slow and are usually conducted with relatively poor resolution. In this context, it would be useful to improve the performance of neutron powder diffractometers (PDs).

Neutron powder diffractometers must distinguish the Bragg peaks scattered from a sample and this may be done using time-of-flight (TOF) methods (usually using a spallation neutron source) or using a crystal monochromator to produce a constant wavelength (CW) beam (usually using a reactor source).

For CW PDs, one common instrumental arrangement is to have a primary spectrometer, which delivers a CW beam to the sample, followed by a sample and a collimator-detector pair which is stepped through a range of scattering angles, $2\theta_S$, to produce a map of scattered intensity as a function of $2\theta_S$. It is usual now to use a bank of many collimator-detector pairs to speed data collection. The primary spectrometer consists of the source and a crystal monochromator with beam collimators between source and monochromator and also between monochromator and sample. A second common arrangement is to use an open geometry where the detector is a continuous multi-wire position sensitive detector (PSD or "banana" detector). Collimated geometries have the advantages that noise tends to be low and that because sample size does not affect the resolution, larger samples can be used although, in practice, large samples are often simply unavailable. In any case, multiple scattering from the sample reduces the beam fraction scattered usefully and is a major contributor to background, so samples are usually chosen to scatter a maximum beam fraction of 1/e. Open geometries have the advantage of greatly increased count rates due to the larger effective detector solid angle but are sensitive to exact sample position and tend to be more susceptible to background noise. Radial oscillating collimators between sample and detector are often used to reduce background coming from air scattering or sample environment at a modest cost in count rate (of order 10%).

Assuming that the scattering plane is horizontal (as is usual), the monochromator is often vertically curved or "focussed" to increase vertical beam divergence and hence intensity at the sample. Many hope that horizontally curved monochromators may be exploited to further increase count rates by transforming beam spatial spread to angular spread thus increasing the flux at the sample position. Monochromator mosaic is needed in the scattering plane to deliver sufficient wavelength spread. Vertical mosaic has the effect of diffusing the beam and this can significantly reduce sample flux in some cases, notably for vertically focussed

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monochromators using small segments. Crystals with smaller vertical than horizontal mosaic can be produced, although with difficulty, and can improve performance in some cases. Ref. [1] shows that it is possible to get sufficient wavelength spread even with small mosaic if the monochromator uses a relatively small Bragg angle and is curved in the scattering plane. Then good performance becomes possible without the added complexity of anisotropic monochromator mosaic. All models here use crystals with homogeneous mosaic of a reasonable value, though the mathematics in [1] show that it is straightforward to describe different horizontal and vertical mosaicity.

Recent work [1,2] presents a new "Acceptance Diagram" approach to describing beams from primary spectrometers and an analytic approach to optimize the choice of beam elements for CW PDs. The "optimization" minimizes the RMS value of R_P, the "peak separation ability", at fixed instrument transmission. R_P is the ratio of the Bragg peak angular widths ($A_{1/2}$, the peak FWHM) to the expected separation of neighbouring peaks calculated from the peak density in reciprocal space for the sample crystal class considered. For samples with cubic structures this is

$$R_P = A_{1/2} a_0^2 \lambda^{-2} \sin 2\theta_S \quad (1)$$

where a_0 is the cubic unit cell side length. The optimization [2] shows that at constant wavelength the line intensity L is proportional to

$$L \propto u \cdot a_{in} \cdot a_3 \cdot \varphi_{in} \cdot \varphi_{out} \quad (2)$$

Here u ($=\sqrt{U}$ where U is the first of the well-known parameter set U, V,W used to describe PD resolution) is proportional to $\Delta\lambda/\lambda$ at the sample. α_{in} is the effective incident beam collimation at the sample and α_3 is the detector collimation. The optimization result is that $\alpha_{in}=\alpha_3$ and $u \propto \alpha_3$. The vertical beam divergence should be equal before and after the sample: $\varphi_2=\varphi_3=\varphi$ and optimally, $\varphi \propto \sqrt{\alpha_3}$. Any set of optimised parameters can be scaled to adjust the intensity resolution trade-off and then the transmission (intensity) scales as the 4-th power of the in-plane divergence and the 8-th power of vertical divergence or monochromator height (since the optimal vertical divergence is proportional to the square root of the in plane divergence) if the parameters are chosen to deliver an optimal configuration.

The instrument transmission, τ, or the peak intensity, L, is proportional to ${A_{1/2}}^4$ or ${R_P}^4$ for an optimised machine. Any quality factor, Q_{PD}, for the instruments must then include a $\tau/{R_P}^4$ term. For reference, a $\sqrt[4]{10} \approx 1.8$ fold improvement in resolution at constant peak intensity is equivalent to a 10 fold increase in count rate at constant resolution. The optimization is less clear on the effect of wavelength but numerical tests show that to maximize count rate in measuring some desired range of sample d-spacings, the wavelength should be made almost as long as is possible. The results of the optimization [2] suggest that a proper choice of elements can deliver large performance gains over current best practice, better described as reduced losses.

The optimization mathematics is self-consistent and has been checked in several ways but some further independent verification would be useful, especially given the large performance gains predicted. Some believe that the only true test of a prediction of instrument improvement is to build a new instrument and compare the data with that from existing machines. Such an exercise would cost many million Euros and permit testing only a single configuration. In the absence of a widely accepted expression for Q_{PD} for these measurements, it is surprisingly difficult to compare different instrument configurations unless the performance differences are truly dramatic. Significant work has been devoted to developing, testing, comparing and benchmarking "Monte Carlo" (MC) computer simulation packages for neutron scattering instruments, notably "McStas", "RESTRAX" and "VITESS" [3–5]. These programs have proved useful and cost effective in designing neutron scattering instruments. They provide a relatively cheap and straightforward way to accurately compare different instrument configurations.

This article presents data from MC simulations of existing best practice CW PDs and of instrument configurations optimized using the new methods. The simulations were conducted using McStas (by LDC) and independently using VITESS (by KL) and then compared for consistency to provide an additional check of their validity. All figures here (except Fig. 5) display the McStas data. The reference instruments here are taken to be the instruments D2B and D20 at the Institut Laue-Langevin, highly regarded examples of so-called High Resolution and High Intensity PDs (HRPD & HIPD). As far as possible, the work avoids questions of component performance and technical development which is peripheral to the topic at hand. Thus, for example, monochromator mosaic is set to the same value wherever this can be accommodated by the flexibility in the optimization and all detectors are assumed to be 100% efficient. This removes from consideration any effect that changes to these parameters might have.

Computer simulations, such as those presented here, cannot prove that the proposed new configurations are optimal, so this article aims only to show that the optimization procedure delivers significant performance improvements. There are many equivalent optimal configurations for neutron CW PDs. There are many, many more non optimal configurations. This work should be regarded as a preliminary and limited illustration of the performance improvements possible over current best practice. The optimization used to design the machines tested is mathematical and shows the upper limits to possible performance. It is hoped that application of the new optimization method will lead to better instruments, better use of existing technology for instrument components, to better measurements, to new types of measurements and to other unexpected improvements.

The Monte Carlo simulations are independent of the mathematical optimization and can be believed independently. The only limit to that independence is where equivalent assumptions have been made in the different approaches; an example is that both the mathematics and all simulations here assume that monochromators have zero thickness. Even if the improved instrument configurations described here had been discovered by accident or guesswork they would still represent a significant and useful advance on current best practice. That the improvements have been found using a rational approach makes them more believable and useful.

2. A baseline – simulations of existing instruments

A list of symbols used for instrument parameters is presented in Appendix A. Appendix B presents details of the parameters used in each of the simulations which should allow readers to repeat the simulations to verify the results presented here.

D2B is a conventional collimated high resolution CW PD using a bank of 128 detectors and 5′ FWHM collimators separated by 1.25°, thus spanning 160° in $2\theta_S$. The instrument uses a 0.30 m high, vertically focussed monochromator (VFM). There is some freedom in choosing the in-plane collimation between source and monochromator, ($\alpha_1=5'$, 10′ or open ($\approx 22'$) FWHM) and an electronically variable detector height, $2H_D$. The wavelength, λ, is usually chosen to be either 1.594 Å or 2.4 Å. The two most commonly used configurations (E. Suard 2009 *Private Communication*) use $\alpha_1=5'$, a germanium (Ge533) monochromator at Bragg angle $\theta_M=67.5°$ giving $\lambda=1.594$ Å and $2H_D=0.30$ m or 0.10 m. These are reportedly used for almost all measurements.