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Design of a neutron diffractometer on biological macromolecules

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Abstract

In order to develop a diffractometer dedicated to neutron protein crystallography, a new special detector which has a large position-sensitive area with a high positional and time resolution is eagerly required. To determine the detector specifications a simulation software is needed, which can predict both the positions and times of scattered intensities from macromolecular crystals. Its development is still under progress. If an ideal detector becomes available, the data-taking efficiency will be better by a factor of 100, compared to that of BIX-3 at JRR-3M reactor in Japan, one of the best diffractometers for biology in the world. Measurement time of less than half a day for a complete data set can be achieved. Ultimately it is intended to cover up to 200 Å in unit cell dimensions and to get reliable data from crystals of less than 1 mm cube in volume.

1. Introduction

Neutron crystallography on biological macromolecules is expected to understand the mechanism of their various functions, which are deeply related to the behaivior of hydrogen bonds and hydration at the surface and inside of the molecule, after positions of all atoms on backbone are determined by X-rays and NMR except functional hydrogens. When the Japanese Joint Project (JJP) is realized, the experimental opportunity will be widely opened to many people and applications including profit companies. In this report consideration is concentrated on the problem of spots-separation related to wavelength, especially to λ_{min}.

2. Requirements

When we design a Time-of-Flight (TOF) diffractometer for protein crystallography (PXD), there are four reqiurements to be realized; (1) intense neutrons to detect spots at $d_{sp} > 0.7$ Å from crystals less than $0.5^3 mm^3$ in volume; (2) separation of spots from crystals up to 200 Å in cell constant; (3) one week measurement time per a sample in average (one month as the max); (4) normal environment. Off course these reqirements will be realized only when the

very intense neutron source like SNS, ESS and JJP etc. will be constructed. On the other hand we should know that these are the relatively low level requirements from biochemists and pharmaceutists to neutron crystallography in biology.

3. Design parameters

When collecting diffraction data on a TOF-PXD, the reciprocal space should be covered by Ewald spheres corresponded to each wavelength as much as possible like by quasi-Laue method. For a given detector maximum angle $2\theta_{max}$ the value of λ_{min} to use in TOF method is very important because it fixes the resolution d_{min} and the time-intervals and spatial-distances between diffracted spots as follows. Fig.1 shows a reciprocal space covered by a TOF-PXD. Shaded region is detectable scan area (low angle region is neglected).

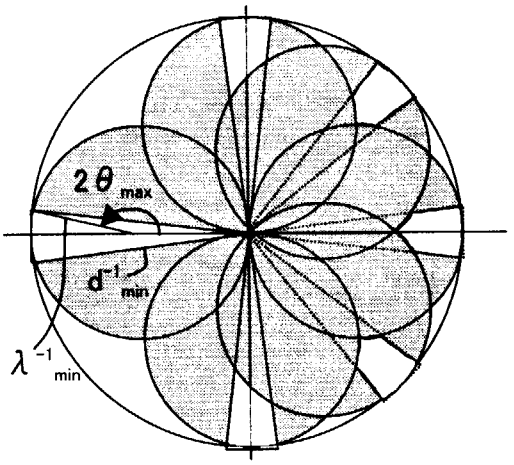

Fig. 1 Detectable reciprocal space on the equator.

One of the largest problems to design a TOF-PXD is detector because there are no suitable detectors for it. In this section we consider the separation of spots on a detector for various conditions if a certain specification detector is available.

3-1. Spots separation in time

Fig.2 shows the concept of time separation condition; ΔT : time difference between peaks in spots, St : time to pass through the sample, and ΔTd : pulse width of λ along time, respectively. When the ratio of $\Delta T/(St+\Delta Td)$ is more than 1, two spots will be separated on the detector along time axis. Considering the shortest time difference of spots diffracting to the same direction, the time separation condition can be written as $[\lambda^2(L1+L2)/(8a)]/[St+k\lambda] \sim \lambda^2(L1+L2)/(8ak) > 1$, if $St/\Delta Td \ll 1$. Here L1 : moderator-sample distance, L2 : sample-detector distance [mm], a : cell constant [Å], k : coefficient related to pulse width ; k=10 for $\lambda < 1.3$ Å, $10+30x(\lambda-1.3)/(6.4-1.3)$ for $1.3 < \lambda < 6.4$ Å which are deduced from calculated results by JJP moderator working group.

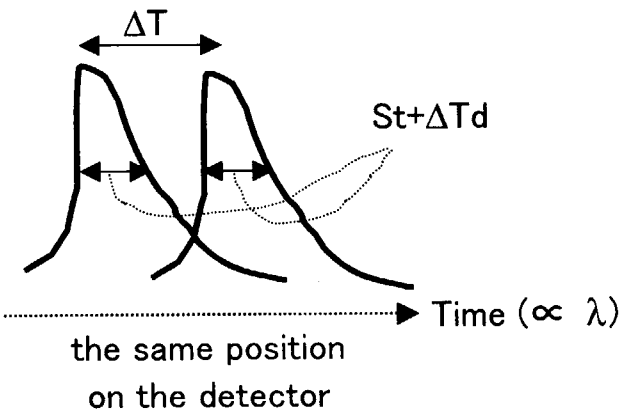

Fig. 2 Time separation condition.

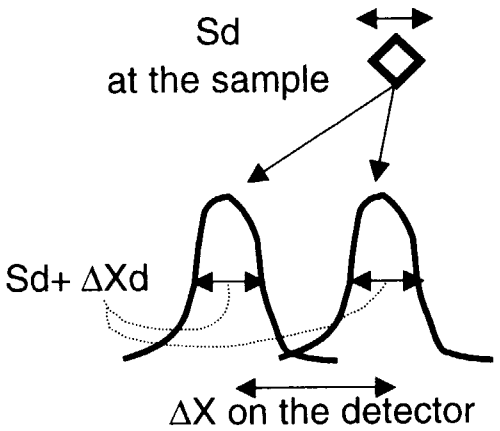

Fig. 3 Spatial separation condition.

The separation check was done for two

Table 1. Time separation of spots on $\lambda_{min.}$

cell const. a	L1=10m	L1=20m
50Å	0.39Å	0.20Å
100Å	0.78Å	0.39Å
200Å	>2.0Å	0.79Å

geometrical conditions of L1=10m and 20m against a=50, 100, 200Å of cell const. of samples, respectively. L2 was fixed as 300mm.

Table 1 shows the crritical wavelength $\lambda_{min.}$ All spots are separable when the range of λ is more than this $\lambda_{min.}$

3-2. Spots separation in space

The concept of spatial separation condition is drawn in Fig. 3 ; ΔX : distance between peaks in spots on the detector, Sd : sample dimension, and ΔXd : spot divergence on the detector from the sample, respectively. When the ratio of $\Delta X/(Sd+\Delta Xd) > 1$, then two spots will be separated on the detector spatially. Considering the nearest spatial distance of spots detected simultaneously, the spatial separation condition can be written as $(L2\lambda/a)/(Sd+\sqrt{[(L2\Delta\theta)^2+(L2\Delta\eta_M)^2+(\Delta xy)^2]}) \sim (\lambda/a)/\sqrt{[(\Delta\theta)^2+(\Delta\eta_M)^2]} > 1$, if $Sd/L2$ & $\Delta xy/L2 << 1$. Here L2 : sample-detector distance [mm], Sd : sample dimension [mm], λ : wavelength [Å], a : cell constant [Å], $\Delta\theta$: beam divergence [rad], $\Delta\eta_M$: sample mosaicity [rad], Δxy : detector spatial resolution [mm], respectively.

Table 2. Spatial separation of spots on $\lambda_{min.}$

cell const. a	$\Delta\theta$=0.29 deg[1)]	$\Delta\theta$=0.15 deg[2)]	$\Delta\theta$=0.29 deg[1)]	$\Delta\theta$=0.15 deg[2)]
	L2 infinity		L2=300mm, , Δxy=1mm, Sd=0.5mm	
50Å	0.30Å	0.20Å	0.43 Å	0.36 Å
100Å	0.61Å	0.43Å	0.86 Å	0.71 Å
200Å	1.22Å	0.86Å	1.72 Å	1.42 Å

[1)]Lm=50mm, L1=10m, [2)]Lm=50mm, L1=20m

The separation conditions were checked for two beam divergences of $\Delta\theta$=0.29 deg and 0.15 deg under infinity of L2 against a=50, 100, 200Å of cell const. of samples, respectively. After that the condition of L2=300mm, Δxy=1mm and Sd=0.5mm was examined as a realistic case. In Table 2, Lm : moderator size [mm] and $\Delta\eta_M$: sample mosaicity [deg].

4. Summary

Up to now the TOF-PXD specification is summarized in Table 3. Concerning to moderator choice, coupled liquid H_2 is better because it has more intensity spectrum over the necessary λ region from calculations by JJP moderator group. In

Table 3. PXD specification in JJP.

items	values
Moderator(M.)	Coupled Liq. H_2
M. surface size	5 X 5cm^2
L1	10m
L2	300 [1000][*)] mm
Sample dimension	0.5mm cube
Q resolution	0.35% (d_{min}=0.7Å, a=200 Å)
λ range	0.3 - 6 Å
2θ range	5 - 170deg
Acc. freq.	25Hz
Detector	0.5 X 0.5 [3 X 3][*)] mm^2

*)Temporal use

case of spots-overlapping despite of proper λ_{min} a data reduction software will be required so that it can deconvolute close spots by profile fitting in pulse shapes and spatial shapes. Finally a new powerful detector is strongly expected by neutron crystallographers in biology.

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