Dr Helio Tolentino,

JSR, editor

Grenoble, September 15th 2024

Dear Helio,

We thank you for your email on July 31st. regarding our paper "Formulation of perfect-crystal diffraction from Takagi-Taupin equations. Numerical implementation in the *crystalpy* library". We also thank the referees for the time invested, and helpful questions, comments and recommendations. We have considered all of them in our new manuscript (we address below the individual points answered in red). We appreciated the positive comments and every single remark, which contributed to improving our manuscript.

We have submitted our modified manuscript, and we are looking forward to seeing it published soon in JSR. We thank you in advance for all your help.

Sincerely yours,

Manuel Sanchez del Rio and Jean-Pierre Guigay

Referee 1 (our answers and comments in red)

This paper is focused on developing a dynamical theory of X-ray diffraction library for the crystalpy software by using the simplest form of the Takagi-Taupin equations with transfer matrices. The manuscript is very well and detailed written. The presented results and software library is extremely useful for the X-ray optics community.

Therefore, I have only few comments and questions that, maybe, can help to improve the final manuscript version.

1. In section “1. Introduction”:

a) What the authors meant by “polychromators”? The authors could add some references.

b) The authors mentioned: “single crystal Laue monochromators”. Should not be “bendable single

crystal Laue monochromators”?

Polychromator is a name used to describe monochromators that provide a very large energy bandwidth and sometimes are dispersive.

We have reformulated the sentence to clarify this and we include references of the mentioned crystal systems.

c) 2nd paragraph: There are few references missing when mentioning ray tracing programs, e.g., RAY and Xray Tracer. They should be briefly described and mentioned. Then, please, add the following references:

Klementiev, K., & Chernikov, R. (2023). New Features of xrt: Bent Crystals, Coherent Modes, Waves with OAM. Synchrotron Radiation News, 36(5), 23-27. https://doi.org/10.1080/08940886.2023.2274735

Chernikov, R., Klementiev, K. (2017). Recent progress of the XRT: ray tracing and wave propagation toolkit (Conference Presentation), Proc. SPIE 10388, Advances in Computational Methods for X-Ray Optics IV, 1038806. https://doi.org/10.1117/12.2273430

Schafers, F. (2008). Modern Development in X-ray and Neutron Optics, Springer Series on Optical Sciences, Vol. 137, edited by A. Erko, M. Idir, Th. Krist and A. G. Michette. Berlin: Springer.

We now mention the RAY and XRT codes and added these references.

d) Please, remove “equations” after “Taupin, 1967”

Done

2. In sub-section “3.4. Reflection and transmission amplitudes in transmission geometry (Laue case)”:

a) Since we have reflection and transmission for both, Bragg and Laue cases, to avoid confusion, please change the sub-section title to: “3.4. Reflection and transmission amplitudes in Laue case”.

3. In sub-section “3.5. Reflection and transmission amplitudes in reflection geometry (Bragg case)”:

a) Again, since we have reflection and transmission for both, Bragg and Laue cases, to avoid confusion, please change the sub-section title to: “3.5. Reflection and transmission amplitudes in Bragg case”.

Done (both titles).

b) With the equations presented here, would you the authors be able to handle with transmissions (forward diffracted o-beam) for the case of enhanced anomalous transmission in Bragg-case asymmetrically-cut crystals? What about multiple beam diffraction and X-ray back diffraction with multiple beam diffraction? Is there any plan to implement it? What are the difficulties? Please, clarify.

The equations of the Dynamical Theory implemented in *crystalpy* permit to compute correctly the diffracted and transmitted intensities. This reveals the “anomalous transmission”. To verify it, we used the example shown in Fig. 5.44 of the book by Yu. Shvyd’ko “X-ray optics: high-energy-resolution applications”. Our code reproduced exactly these results (see appendix at the end of this document).

Concerning the multiple beam diffraction, we understand that the referee is asking about simultaneous diffraction/reflections of multiple sets of Miller indices. *Crystalpy* does not treat it, as all the theoretical framework is based on the two-waves approximation, therefore a single set of Miller indices or, in other words, a single vector ***h***. Multiple excited waves in a crystal, as discussed, for example in https://doi.org/10.1364/OE.505208 cannot be treated with our approach, and we have no plans to develop it in a near future. X-ray backdiffraction can certainly be calculated in the two-beams approximation, which is sufficient for many cases, but multiple-beam corrections are not considered.

c) Please change “3.4.1 Reflection amplitude ...” to “3.5.1 Reflection amplitude ...”

d) Please change “3.4.2 Reflection amplitude ...” to “3.5.2 Reflection amplitude ““

Done (both)

4.In sub-section “3.6. Calculation of reflection and transmission amplitudes using the transfer matrix”:

a) If the extinction depth varies from few microns to tens of microns (Fig. 4), a 1 micron half-layer (Fig. 5). in the transfer matrix method, would be enough? Why 1 micron? Is it based in some fraction of the Pendellosung distance in the Bragg geometry? Please, clarify.

The transfer matrices discussed in our paper act on the amplitudes, not on the intensities. Therefore, the thickness of the layers is not important. The total thickness (2 um) has been selected as “thin” (i.e. showing Pendellosung fringes) to make evident the differences with a crystal of 1 um. We just verify that the result is equal to two layers of 1 um. Similarly, we could reproduce the results of 2um with 4 layers of 0.5um. This is an exact calculation.

Another different thing, that perhaps the referee has in mind, is the “multilamellar method” first introduced by White [1] and implemented in our software tools [2]. This algorithm is used to compute the crystal reflectivity of a curved crystal by adding the intensity profiles of the layers in which the total thickness is divided. Here, we add intensities, and the number of layers (or the layer thickness) must be carefully selected following a given recipe to obtain reasonable results. This is an approximated method.

[1] J. E. White “X-Ray Diffraction by Elastically Deformed Crystals” J. Appl. Phys. 21, 855–859 (1950)

https://doi.org/10.1063/1.1699774

[2] M Sanchez del Rio, N Perez-Bocanegra, X Shi, V Honkimaki, L Zhang (2015) Simulation of X-ray diffraction profiles for bent anisotropic crystals Journal of Applied Crystallography 48: 2. 477-491. https://doi.org/10.1107/S1600576715002782

Appendix: Verification of anomalous transmission.

The Fig. 5.44 in Shvyd'Ko, Y. (2004) "X-ray optics: high-energy-resolution applications" shows a simple example of "anomalous transmission": the beam transmitted through a 100 um plate of Si220 (asymmetric, incident photon 19.7 keV) presents a high transmission value (~0.8) at angle ~-6urad. This figure is reproduced here.

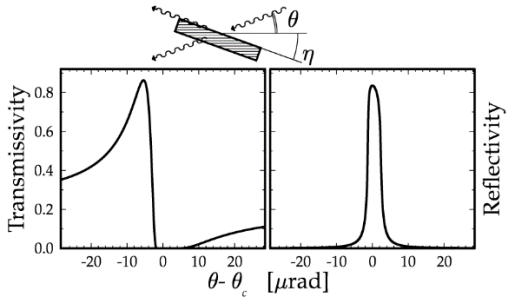
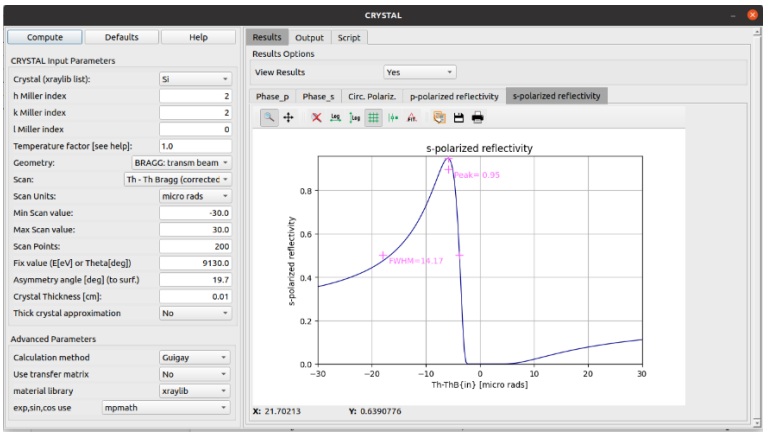
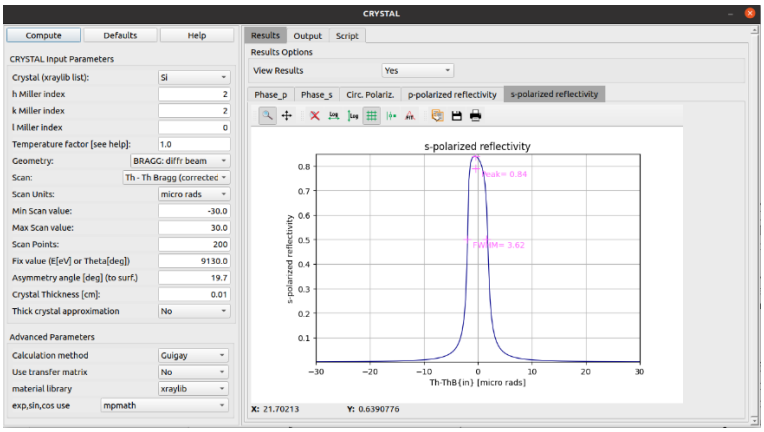


Fig. 5.44. Angular dependence of transmission (left), and reflection (right) of x rays with $E = 9.13$ keV photon energy in Bragg diffraction by a silicon crystal, the (220) Bragg reflection. Crystal plate thickness $d = 100$ μm, $\theta_c = 20.7°$, asymmetry angle $\eta = 19.7°$, asymmetry factor $b = -37$ (the same parameters as the collimator crystal of the monochromators presented in Table 5.6).
At $\theta - \theta_c \simeq -6$ μrad the crystal is highly transparent. Such transparency will have the collimator crystal (C) in the $(+,+,\pm)$ monochromator shown in Fig. 5.38(a) set in this angular position with respect to x rays propagating from the dispersing element (DE) to the wavelength selector (W).

The simulations with crystalpy reproduce very well these results





Referee 2 (our answers and comments in red)

This paper concentrates very heavily on deriving formulas for the diffraction of plane waves from perfect crystals by using the Takagi-Taupin equations. The calculations take up well over half the paper. This scarcely seems necessary because the required reflectivity and transmittivity are already well known from Authier, Zachariasen and others.

We agree with the idea that no new physics is presented in the paper, and what we present has been obtained in several works, many of them very old. However, it is difficult to find a comprehensive deduction of the main and basic results of Dynamical Theory in a short text. The books mentioned go through many pages of methodology before arriving to the results. We used an approach in which i) all results, including the Takagi-Taupin equations are directly deduced from basic concepts, ii) all results are expressed as a few (complex) parameters (a, omega, T), and iii) this allows reduced expressions, well adapted to be programmed, instead of much longer expressions found in typical textbooks that are more interested in discussing the physical meaning thus requiring real parameters.

In order to make clear these points to the reader, we have added a new paragraph in the introduction.

It is unsurprising that, for plane-wave diffraction from perfect crystals, the Takagi-Taupin equations reduce to Zachariasen's formulas as shown in Appendix C.

Yes, it is not surprising, but it is neither evident to see. Therefore, for completeness, and also for double-checking our formulation, the appendix C is included.

I therefore don't clearly see what is novel about this treatment. It is true that the authors develop a matrix formalism that is convenient for ray tracing, especially when diffraction from layers of crystals must be treated, but could this not also have been developed equally well from Authier's or Zachariasen's work?

The matrix formalist has the benefit of reducing all the physics to four simple equations (eq 31). The known results in the textbooks cited are simply deduced from them, but the other way around is not always possible: from the equations shown in these texts (in many cases regarding the intensity and not treat in depth the phase, see e.g. our footnote in page 30) is not possible to construct the transfer and scattering matrices. We believe this is a novelty in our development and an added value to our paper. Again, in the paragraph added in the introduction we make clear that we do not claim new physical results, only a new way to deduce them, perhaps more adapted for coding.

If too much attention is paid to the Takagi-Taupin equations, too little is paid to other topics that would naturally be connected with this paper:

Section 3.6 discusses the use of the previously derived matrices to calculate the reflection and transmission amplitudes of a crystal composed of layers of varying orientations. It would seem natural to cite (Sanchez del Rio et al, 2015) here. The authors could also apply both their matrix formalism and the multilamellar model of XOP to the same bent crystal, and then compare the results. Is this what the authors are proposing in their last sentence of Section 3.6?

We mentioned that the curved crystals are left for a future work. However, is worth to mention that the transfer matrix method presented here and the "multilamellar" method in XOP are different things. The transfer matrix method is exact and allows the same result for a crystal made by 1, 2 (or many layers) as shown in Fig. 5. In the multilamellar method in XOP (described in the 2015 reference, that reused the methodology already existing) the crystal reflectivity of a curved crystal is calculated by adding the *intensity* profiles of the layers in which the total thickness is divided. Here, we add intensities, we neglect phases, and the layer thickness must be carefully selected following a given recipe to obtain reasonable results. This is an approximated method.

Very little detail about the crystalpy library is given. Is any documentation provided elsewhere?

The web site of *crystalpy* (https://github.com/oasys-kit/crystalpy) contains all the code, examples, and also programmer documentation (available at https://crystalpy.readthedocs.io)

As a side note, on Page 2, Lines 5-7, the authors state,

"… the calculation of the crystal structure factor, which is an essential ingredient to calculating diffracted amplitudes and intensities, is obtained from tabulated scattering functions of multiple origins."

The determination of crystal structure factors is indeed not trivial, and it becomes even more complex once noncubic crystals are treated. Alpha quartz (also called "low quartz") illustrates the complexities especially well because of its chirality, its strong anisotropy, its non-linear temperature dependence, and the many different conventions used by different authors to describe its crystal structure. The correct treatment and interpretation of X-ray diffraction from alpha quartz has been the topic of many papers published in the last decade. The authors may wish to consult them, see what solutions have been found, and apply them to other non-cubic crystals.

This is true. We certainly need to calculate the structure. We mention in the paper (pag 22) that crystalpy "delegate" the work of computing the structure factors to other libraries, either xraylib and dabax. Both libraries use similar methods that are described in more detail in the paper [1]. The possibility of computing noncubic and complex crystals is included, once the structure is known. The dabax library offers the flexibility to modify structural data of the crystals database and to include new ones. Very long crystal lists are available.

We added a paragraph to explain how and where we calculate the structure factors, pointing to the reference [1], and mentioning the difficulties with some parameters in some crystals, with references to quartz and YB66.

[1] X J Yu, X Chi, T Smulders, A T S Wee, A Rusydi, M Sanchez del Rio, M B H Breese (2022) Beamline simulations using monochromators with high d-spacing crystals Journal of Synchrotron Radiation 29: 5. https://doi.org/10.1107/S160057752200707X