BornAgain

Software for simulating and fitting
X-ray and neutron small-angle scattering
at grazing incidence

User Manual

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Introduction

BornAgain is a free and open-source software package to simulate and fit small-angle scattering at grazing incidence (GISAS). It supports analysis of both X-ray (GISAXS) and neutron (GISANS) data. Its name, BornAgain, indicates the central role of the distorted-wave Born approximation (DWBA) in the physical description of the scattering process. The software provides a generic framework for modeling multilayer samples with smooth or rough interfaces and with various types of embedded nanoparticles.

BornAgain almost completely reproduces the functionality of the widely used program IsGISAXS by Rémi Lazzari [1]. BornAgain goes beyond IsGISAXS by supporting an unrestricted number of layers and particles, diffuse reflection from rough layer interfaces, particles with inner structures, neutron polarization and magnetic scattering. Adhering to a strict object-oriented design, BornAgain provides a solid base for future extensions in response to specific user needs.

BornAgain is a platform-independent software, with active support for Linux, MacOS and Microsoft Windows. It is a free and open source software provided under the terms of the GNU General Public License (GPL, version 3 or higher). This documentation is released under the Creative Commons license CC-BY-SA. When BornAgain is used in preparing scientific papers, please cite software and manual as follows:

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> BornAgain — Software for simulating and fitting X-ray and neutron small-angle scattering at grazing incidence, version 1.0.0,
> http://www.bornagainproject.org

This User Manual is complementary to the online documentation at http://www.bornagainproject.org. It does not duplicate information that is more conveniently read online. Therefore, Section 1 just contains a few pointers to the web site. The remainder of this User Manual mostly contains background on the sample models and on the scattering theory implemented in BornAgain, and some documentation of the corresponding Python functions.

Software and documentation are work in progress. We cannot guarantee that they are accurate and correct. Anyway, it is entirely in the responsibility of users to ensure that their data interpretation is physically meaningful. If in doubt, please contact us.

We are grateful for all kind of feedback: criticism, praise, bug reports, feature requests or contributed modules. If questions go beyond normal user support, we will be glad to discuss a scientific collaboration.

Chapter 1

Online documentation

This User Manual is complementary to the online documentation at the project web site `http://www.bornagainproject.org`. It does not duplicate information that is more conveniently read online. This dummy chapter contains no more than a few pointers to the web site.

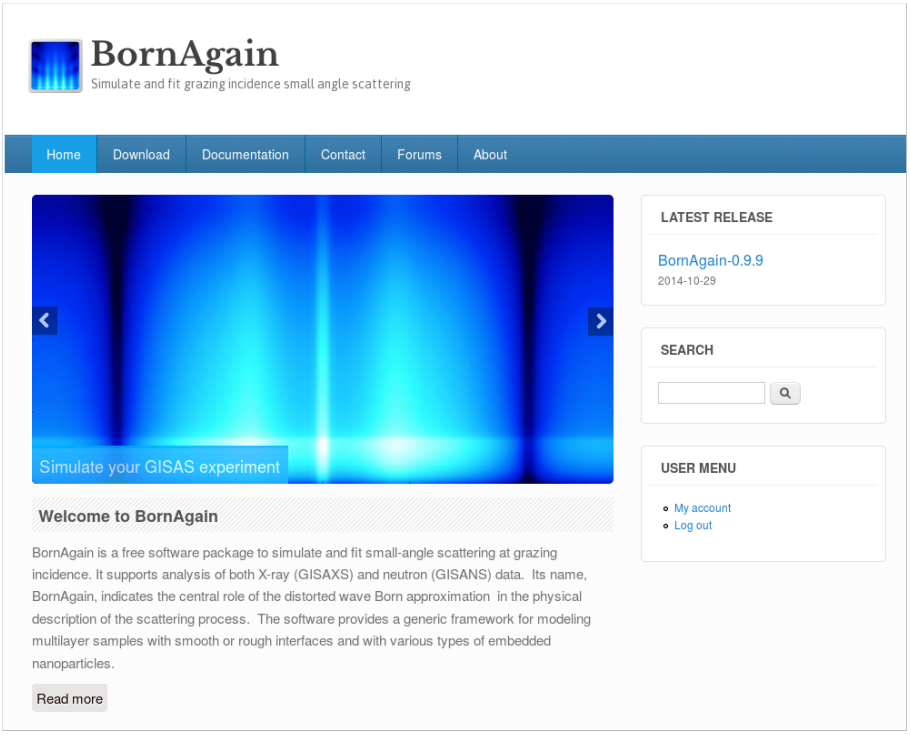

Figure 1.1: A screenshot of the home page `http://www.bornagainproject.org`.

1.1 Download and installation

The DOWNLOAD section on the `BornAgain` web site points to the download location for binary and source packages. It also provides a link to our `git` server where the unstable development trunk is available for contributors or for users who want to live on the edge.

The DOCUMENTATION section contains pages with *Installation instructions*.

1.2 Further online information

The DOCUMENTATION section of the project web site contains in particular

- an overview of the software architecture,
- a list of implemented functionality,
- tutorials for "Working with `BornAgain`", using either the Graphical User Interface or `Python` scripts,
- a comprehensice collection of examples that demonstrate how to use `BornAgain` for modelling various sample structures and different experimental conditions,
- a link to the API reference for using `BornAgain` through `Python` scripts or C++ programs.

1.3 Registration, contact, discussion forum

To stay informed about the ongoing development of `BornAgain`, register on the project homepage `http://www.bornagainproject.org` ("Create new account"). You will then receive our occasional newsletters, and be authorized to post to the discussion forum

To contact the `BornAgain` development and maintenance team in the Scientific Computing Group of Heinz Maier-Leibnitz Zentrum (MLZ) Garching, write a mail to `contact@bornagainproject.org`, or fill the form in the CONTACT section of the project web site.

For questions that might be of wider interest, please consider posting to the discussion forum, accessible through the FORUMS tab of the project web site.

Chapter 2

Foundations of GISAS

2.1 Wave propagation

2.1.1 X-ray propagation

...to come

2.1.2 Neutron propagation

In a GISANS experiment, we are only interested in small scattering vectors $\mathbf{q}$, and therefore do not register ordinary Bragg scattering by crystal lattices or other correlations at atomic level; at most, Bragg scattering must be accounted for as a loss channels. We therefore describe the neutron-sample interaction in continuum approximation by a refractive index n with

$$n^2 = 1 - \frac{4\pi}{K^2}\rho_s \tag{2.1}$$

where ρ_s is an effective scattering length density [2, 3].

2.1.3 Helmholtz equation

X-ray and neutron propagation are both governed by the same stationary wave equation, the *Helmholtz equation*

$$\left(\boldsymbol{\nabla}^2 + K^2 n(\mathbf{r})^2\right)\Psi(\mathbf{r}), \tag{2.2}$$

where $K = 2\pi/\lambda_0$ is the vacuum wave number, and n is the refractive index. In this equation, the time dependence $\mathrm{e}^{i\omega t}$ of the wave field does not appear explicitly. In GISAS, we are only interested in elastic scattering. Therefore, the wave frequency ω is considered constant.

Implicitly, K depends on ω through a *dispersion relation*. This relation is fundamentally different for X-rays (where it is linear, $\omega \propto K$) and for thermal neutrons (where it is quadratic, $\omega \propto K^2$). However, for stationary problems this frequency dependence of K does not matter. Therefore we can use K instead of ω as a constant that

characterizes the incoming radiation. In this way, scalar X-ray and neutron scattering can be described by exactly the same wave equation (2.2).

2.2 Scalar scattering

For both X-rays and neutrons the refractive index n is close to 1. In the following, we shall use this fact to solve the Helmholtz equation in a perturbation expansion that is known as the Born approximation though it goes back to Lord Rayleigh who devised it for the scattering of sound, and later also applied it to electromagnetic waves.

2.2.1 Born approximation

We rewrite the scalar Helmholtz equation (2.2) as

$$\left(\boldsymbol{\nabla}^2 + K^2\right)\Psi(\mathbf{r}) = \frac{\chi(\mathbf{r})}{4\pi}\Psi(\mathbf{r}) \tag{2.3}$$

with

$$\chi(\mathbf{r}) := \frac{4\pi}{K^2}\left(1 - n^2(\mathbf{r})\right). \tag{2.4}$$

For neutrons, this definition just compensates (2.1) so that $\chi = \rho_s$. Equation (2.2) looks like an inhomogenous differential equation — provided we neglect for a moment that unknown function Ψ reappears on the right side. The homogeneous equation

$$\left(\boldsymbol{\nabla}^2 + K^2\right)\Psi_0(\mathbf{r}) = 0 \tag{2.5}$$

is solved by plane waves and superpositions thereof. For an isolated inhomogeneity,

$$\left(\boldsymbol{\nabla}^2 + K^2\right)G(\mathbf{r}) = \delta(\mathbf{r}) \tag{2.6}$$

is solved by the Green's function[1]

$$G(\mathbf{r}) = \frac{\mathrm{e}^{iKr}}{4\pi r}, \tag{2.7}$$

which is an outgoing spherical wave originating from $\mathbf{r} = 0$. Convoluting this function with the given inhomogeneity $(\chi/4\pi)\Psi$, we obtain the formal solution of the full inhomogeneous equation (2.3)

$$\Psi(\mathbf{r}) = \Psi_0(\mathbf{r}) + \int \mathrm{d}^3 r' G(\mathbf{r} - \mathbf{r}')\frac{\chi(\mathbf{r}')}{4\pi}\Psi(\mathbf{r}'). \tag{2.8}$$

[1] Verification under the condition $\mathbf{r} \neq 0$ is a straightforward exercice in vector analysis. For the special case $\mathbf{r} = 0$, one encloses the origin in a small sphere and integrates by means of the Gauß-Ostrogadsky divergence theorem. This explains the appearance of the factor 4π.

However, the integral kernel still contains the full solution Ψ. If we replace this occurence of Ψ by the full right-hand side of (2.8) we obtain

$$\Psi(\mathbf{r}) = \Psi_0(\mathbf{r}) + \int \mathrm{d}^3 r' G(\mathbf{r}-\mathbf{r}') \frac{\chi(\mathbf{r}')}{4\pi} \left(\Psi_0(\mathbf{r}') + \int \mathrm{d}^3 r'' G(\mathbf{r}'-\mathbf{r}'') \frac{\chi(\mathbf{r}'')}{4\pi} \Psi(\mathbf{r}'') \right). \tag{2.9}$$

This can be iterated to obtain an infinite series representation of Ψ. Successive terms in this series contain rising powers of χ. Since χ is assumed to be small, the series is likely to converge. In first Born approximation, only the linear order in χ is retained,

$$\Psi(\mathbf{r}) \simeq \Psi_0(\mathbf{r}) + \int \mathrm{d}^3 r' G(\mathbf{r}-\mathbf{r}') \frac{\chi(\mathbf{r}')}{4\pi} \Psi_0(\mathbf{r}'). \tag{2.10}$$

This is practically always adequate for material investigations with X-rays or neutrons, where the aim is to deduce $\chi(\mathbf{r}')$ from the scattered intensity $|\Psi(\mathbf{r})|^2$.

2.2.2 Far-field expansion

In a scattering experiment, the intensity $|\Psi(\mathbf{r})|^2$ is measured at a detector location $\mathbf{r}$ far away from the sample volume. We choose the coordinate origin at the nominal center of the sample so that the integral in (2.10) runs over $\mathbf{r}'$ with $r' \ll r$. This allows us to expand

$$\left|\mathbf{r}-\mathbf{r}'\right| \simeq \sqrt{r^2 - 2\mathbf{r}\,\mathbf{r}'} \simeq r - \frac{\mathbf{r}\,\mathbf{r}'}{r} \equiv r - \frac{\mathbf{k}_f \mathbf{r}'}{K}, \tag{2.11}$$

where we have introduced the outgoing wave vector

$$\mathbf{k}_f := K \frac{\mathbf{r}}{r}. \tag{2.12}$$

We apply this to the Green's function (2.7), retaining the first order in $\mathbf{r}'$ in the exponent, but not in the denominator:

$$G(\mathbf{r}-\mathbf{r}') \simeq \frac{\mathrm{e}^{iKr - i\mathbf{k}_f\mathbf{r}'}}{4\pi r}. \tag{2.13}$$

This is an outgoing spherical wave with respect to $\mathbf{r}$, but a plane wave with respect to $\mathbf{r}'$. If the incident radiation is also a plane wave,

$$\Psi_0(\mathbf{r}) = \mathrm{e}^{i\mathbf{k}_i\mathbf{r}}, \tag{2.14}$$

then the Born approximation (2.10) becomes essentially a Fourier transform of χ,

$$\Psi(\mathbf{r}) \simeq \Psi_0(\mathbf{r}) + \frac{\mathrm{e}^{iKr}}{r} \int \mathrm{d}^3 r' \mathrm{e}^{i\mathbf{q}\mathbf{r}'} \chi(\mathbf{r}') \tag{2.15}$$

with the *scattering vector*

$$\mathbf{q} := \mathbf{k}_i - \mathbf{k}_f. \tag{2.16}$$

2.2.3 Differential cross section

Above, we said somewhat sloppily that a scattering experiment measures intensities $|\Psi(\mathbf{r}|^2$. We shall now make this more rigorous. In the case of neutron scattering, one actually measures the probability flux. We define it in arbitrary relative units as

$$\mathbf{J} := \Psi^* \frac{\boldsymbol{\nabla}}{2i} \Psi - \Psi \frac{\boldsymbol{\nabla}}{2i} \Psi^*. \tag{2.17}$$

With (2.14), the incident flux is

$$\mathbf{J}_0 = \mathbf{k}_i. \tag{2.18}$$

The scattered wavefield (2.15) is observed at a detector position $\mathbf{r}$ that is not illuminated by the incident beam, hence $\Psi_0(\mathbf{r}) = 0$. This leaves us with

$$\mathbf{J}(\mathbf{r}) = \left| \frac{1}{r} \int \mathrm{d}^3 r' \mathrm{e}^{i\mathbf{q}\mathbf{r}'} \chi(\mathbf{r}') \right|^2 K \frac{\mathbf{r}}{r}. \tag{2.19}$$

The ratio of the scattered current hitting an infinitesimal detector area $r^2 \mathrm{d}\Omega$ to the incident flux is expressed as a *differential cross section*

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} := \frac{r^2 J(\mathbf{r})}{J_0}. \tag{2.20}$$

Using $k_i = K$ and changing notation $\mathbf{r}' \to \mathbf{r}$, we find

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = \left| \int \mathrm{d}^3 r \mathrm{e}^{i\mathbf{q}\mathbf{r}} \chi(\mathbf{r}) \right|^2 =: \left| \chi(\mathbf{q}) \right|^2 \tag{2.21}$$

The differential cross section is just the squared modulus of the Fourier transform $\chi(\mathbf{q})$ of the scattering-length distribution (2.4).

2.3 Distorted Wave Born Approximation

As we have seen the Born approximation relies on plane waves, which basically means that incoming and scattered radiation are assumed to propagate along straight lines, with directions expressed by $\mathbf{k}_i$ and $\mathbf{k}_f$. This is adequate for most experiments with weakly interacting radiation, and especially for most X-ray and neutron scattering techniques, but not for grazing-incidence small-angle scattering where radiation is refracted and reflected by interfaces and therefore does not propagate along straight lines. The Born approximation also fails for collisions of charged particles. For them, Massey and Mott devised in the early 1930s the distorted wave Born approximation (DWBA). A particularly simple variant of this approximation is routinely used to account for refraction and reflection in grazing-incidence scattering.

Refraction and reflection are caused by vertical variations of the average refractive index $n_0(z)$.

2.3.1 Multilayer systems

In multilayer systems, the first term of (**??**) denotes the specular part of the reflection, while the second term is responsible for the off-specular scattering. This off-specular part is caused by deviations from the perfectly smooth layered system, as e.g. interface roughnesses or included nanoparticles. In here only the case of nanoparticles will be treated.

In the conventions where $H = -\Delta + V$, the potential splits into two parts V_1 and V_2, where only the second part is treated as a perturbation:

$$V_1 = K^2\left(1 - n_0^2(\mathbf{r})\right)$$
$$V_2 = \sum_i K^2\left(n_0^2(\mathbf{R}^i) - n_i^2\right) S^i(\mathbf{r}) \otimes \delta(\mathbf{r} - \mathbf{R}^i),$$

where $n_0(\mathbf{r})$ denotes the refractive index of the unperturbed system (which, in case of a multilayer system, will only depend on its z-coordinate) and n_i is the refractive index of the nanoparticle with shape function S^i and position $\mathbf{R}^i$.

For nanoparticles in a specific layer j, i.e. $V_2 \neq 0$ only in layer j, one only needs the unperturbed solutions in layer j:

$$\left\langle \mathbf{r} | \Psi^+_{1k_i} \right\rangle = (2\pi)^{-3/2}\left[R_j(\mathbf{k}_i)e^{i\mathbf{k}_{j,R}(\mathbf{k}_i)\cdot\mathbf{r}} + T_j(\mathbf{k}_i)e^{i\mathbf{k}_{j,T}(\mathbf{k}_i)\cdot\mathbf{r}}\right]$$
$$\left\langle \Psi^-_{1k_f} | \mathbf{r} \right\rangle = (2\pi)^{-3/2}\left[R_j(-\mathbf{k}_f)e^{i\mathbf{k}_{j,R}(-\mathbf{k}_f)\cdot\mathbf{r}} + T_j(-\mathbf{k}_f)e^{i\mathbf{k}_{j,T}(-\mathbf{k}_f)\cdot\mathbf{r}}\right].$$

The off-specular contribution to the scattering amplitude then becomes:

$$f(\theta,\phi) = -\int d^3\mathbf{r}\frac{V_2(\mathbf{r})}{4\pi}\Big[T_i T_f e^{i(\mathbf{k}_{j,i} - \mathbf{k}_{j,f})\cdot\mathbf{r}} + R_i T_f e^{i(\widetilde{\mathbf{k}}_{j,i} - \mathbf{k}_{j,f})\cdot\mathbf{r}}$$
$$+ T_i R_f e^{i(\mathbf{k}_{j,i} - \widetilde{\mathbf{k}}_{j,f})\cdot\mathbf{r}} + R_i R_f e^{i(\widetilde{\mathbf{k}}_{j,i} - \widetilde{\mathbf{k}}_{j,f})\cdot\mathbf{r}}\Big],$$

where the following shorthand notations were used:

$$T_i \equiv T_j(\mathbf{k}_i) \qquad R_i \equiv R_j(\mathbf{k}_i)$$
$$T_f \equiv T_j(-\mathbf{k}_f) \qquad R_f \equiv R_j(-\mathbf{k}_f)$$
$$\mathbf{k}_{j,i} \equiv \mathbf{k}_{j,T}(\mathbf{k}_i) \qquad \widetilde{\mathbf{k}}_{j,i} \equiv \mathbf{k}_{j,R}(\mathbf{k}_i)$$
$$\mathbf{k}_{j,f} \equiv -\mathbf{k}_{j,T}(-\mathbf{k}_f) \qquad \widetilde{\mathbf{k}}_{j,f} \equiv -\mathbf{k}_{j,R}(-\mathbf{k}_f).$$

From this expression, one sees that the scattering amplitude consists of a weighted sum of Fourier transforms of the potential V_2. Using

$$V_2(\mathbf{r}) = \sum_i 4\pi\rho_{s,rel,i} S^i(\mathbf{r}) \otimes \delta(\mathbf{r} - \mathbf{R}^i),$$

with $\rho_{s,rel,i} \equiv K^2\left(n_0^2(\mathbf{R}^i) - n_i^2\right)/4\pi$, the scattering amplitude becomes

$$f(\theta,\phi) = -\sum_i \rho_{s,rel,i}\mathscr{F}^i_{\mathrm{DWBA}}(\mathbf{k}_{j,i}, \mathbf{k}_{j,f}, \mathbf{R}^i_z)e^{i(\mathbf{k}_{j,i\parallel} - \mathbf{k}_{j,f\parallel})\cdot\mathbf{R}^{i\parallel}},$$

with

$$\mathscr{F}^{i}_{\mathrm{DWBA}}(\mathbf{k}_i,\mathbf{k}_f,R_z) \equiv T_i T_f F^i(\mathbf{k}_i-\mathbf{k}_f)e^{i(k_{iz}-k_{fz})R_z} + R_i T_f F^i(\widetilde{\mathbf{k}}_i-\mathbf{k}_f)e^{i(-k_{iz}-k_{fz})R_z} + T_i R_f F^i(\mathbf{k}_i-\widetilde{\mathbf{k}}_f)e^{i(k_{iz}+k_{fz})R_z} + R_i R_f F^i(\widetilde{\mathbf{k}}_i-\widetilde{\mathbf{k}}_f)e^{i(-k_{iz}+k_{fz})R_z},$$

TO MERGE IN: In the DWBA, the form factor of a particle in a multilayer system is given by

$$F_{\mathrm{DWBA}}(\mathbf{k}_i,\mathbf{k}_f,r_z) = T_i T_f F_{\mathrm{BA}}(\mathbf{k}_i-\mathbf{k}_f)e^{i(k_{i,z}-k_{f,z})r_z} + R_i T_f F_{\mathrm{BA}}(\widetilde{\mathbf{k}}_i-\mathbf{k}_f)e^{i(-k_{i,z}-k_{f,z})r_z} + T_i R_f F_{\mathrm{BA}}(\mathbf{k}_i-\widetilde{\mathbf{k}}_f)e^{i(k_{i,z}+k_{f,z})r_z} + R_i R_f F_{\mathrm{BA}}(\widetilde{\mathbf{k}}_i-\widetilde{\mathbf{k}}_f)e^{i(-k_{i,z}+k_{f,z})r_z}, \tag{2.22}$$

where F_{BA} is the expression of the form factor in the Born approximation, r_z is the z-coordinate of the particle's position (measured from the bottom of the particle), $\mathbf{k}_i = (k_{i,x}, k_{i,y}, k_{i,z})$ $\mathbf{k}_f = (k_{f,x}, k_{f,y}, k_{f,z})$ are the incident and scattered wave vectors in air, respectively [4]. With a tilde ($\widetilde{\ }$), these wavevectors components are evaluated in the multilayer system (the refractive indices of the different constituting materials have to be taken into account). T_i, T_f, R_i, R_f are the transmission and reflection coefficients for the incident wave (index i) or the scattered one (index f). These coefficients can be calculated using the Parratt formalism [5] or the matrix method [6]. $\mathbf{k}_i - \mathbf{k}_f$ is equal to the scattering vector $\mathbf{q}$ and the z-axis is pointing upwards.

With this last expression, the same techniques as demonstrated in section **??** can be applied, leading to the following expression for the expectation value of the scattering cross-section:

$$\begin{aligned}&\left\langle \frac{d\sigma}{d\Omega}(\mathbf{k}_i,\mathbf{k}_f)\right\rangle_{\text{Off-specular}} \\ &= \sum_\alpha p_\alpha \left|\mathscr{F}_\alpha(\mathbf{k}_{j,i},\mathbf{k}_{j,f},R_{\alpha,z})\right|^2 + \frac{\rho_S}{S}\sum_{\alpha,\beta} p_\alpha p_\beta \mathscr{F}_\alpha(\mathbf{k}_{j,i},\mathbf{k}_{j,f},R_{\alpha,z})\mathscr{F}^*_\beta(\mathbf{k}_{j,i},\mathbf{k}_{j,f},R_{\beta,z}) \\ &\times \iint_S d^2\mathbf{R}^{\parallel}_\alpha d^2\mathbf{R}^{\parallel}_\beta \mathscr{G}_{\alpha,\beta}\left(\mathbf{R}^{\parallel}{}_\alpha,\mathbf{R}^{\parallel}{}_\beta\right)\exp\left[i\mathbf{q}_{j\parallel}\cdot(\mathbf{R}^{\parallel}_\alpha-\mathbf{R}^{\parallel}_\beta)\right].\end{aligned}$$

The main differences with respect to the cross-section in the Born approximation are:

1. The particle form factor now consists of a more complex expression and now depends on both incoming and outgoing wavevectors and also on the z-coordinate of the particle;

2. Since the z-coordinate of the particles is implicitly included in its formfactor, the position integrals only run over x- and y-coordinates and the volume and density gets replaced with the surface area and surface density respectively.

2.4 Polarization

ome GISAS experiments are performed with polarized radiation or/and involve polarization-dependent interactions. Therefore we need also to consider the propagation of polarized X-rays and neutrons. Electromagnetic field vectors and neutron spinors are fundamentally different mathematical objects Nevertheless, for our purpose they can be mapped onto a uniform .... <to elaborate> .... so that one and the same formalism can be applied to polarized GISAXS and GISANS. This shall be derived in the following.

2.4.1 Polarized X-rays

2.4.2 Polarized neutrons

2.5 TO RECYCLE

Consider a scattering volume V, containing N scattering centers with shape functions $S^i(\mathbf{r})$, positions $\mathbf{R}^i$ and scattering length density ρ_s (relative to the ambient material).

The differential cross-section (per scattering center) is then given by:

$$\frac{d\sigma}{d\Omega}(\mathbf{q}) = \frac{1}{N}\left|\int_V \rho_s(\mathbf{r}) e^{i\mathbf{q}\cdot\mathbf{r}} d^3\mathbf{r}\right|^2. \tag{2.23}$$

Chapter 3

Wave propagation through multilayers

This chapter describes how to calculate the coherent wave solution for neutrons incident on a multilayered sample. In the first section, only scalar interactions will be considered. The second part uses a spinor and matrix formalism to account for neutron polarization.

⚠ Remark: In `BornAgain` there is no limitation to the number of layers composing the sample.

3.1 Geometry of the sample

The geometry used to describe the sample is shown in figure 3.2. The z-axis is perpendicular to the sample's surface and pointing upwards. The x-axis is perpendicular to the detector plane. The input and the scattered output beams are each characterized by two angles α_i, ϕ_i and α_f, ϕ_f, respectively. Our choice of orientation for the angles α_i and α_f is so that they are positive as shown in figure 3.2.

The layers are defined by their thicknesses (parallel to the z-direction), their possible roughnesses (equal to 0 by default) and the materials they are made of. They have an infinite extension in the x and y directions. And, except for roughness, their interfaces are plane and perpendicular to the z-axis. There is also no limitation to the number of layers that could be defined in `BornAgain`. Note that the thickness of the top and bottom layer are not defined.

The nanoparticles are characterized by their form factors (*i.e.* the Fourier transform of the shape function - see Appendix **??** for a list of form factors implemented in `BornAgain`) and the composing material. The number of input parameters for the form factor depends on the particle symmetry; it ranges from one parameter for a sphere (its radius) to three for an ellipsoid (its three main axis lengths).

By placing the particles inside or on top of a layer, we impose their vertical positions, whose values correspond to the bottoms of the particles. The in-plane distri-

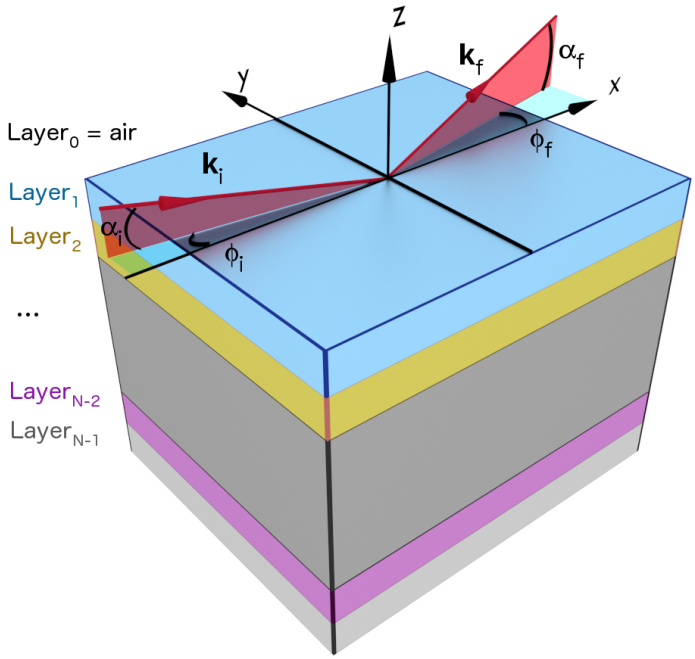

Figure 3.1: Representation of the scattering geometry. n_j is the refractive index of layer j and α_i and ϕ_i are the incident angles of the wave propagating. α_f is the exit angle with respect to the sample's surface and ϕ_f is the scattering angle with respect to the scattering plane.

bution of particles is linked with the way the particles interfere with each other. It is therefore implemented when dealing with the interference function.

The complex refractive index associated with a layer or a particle is written as $n = 1 - \delta + i\beta$, with $\delta, \beta \in \mathbb{R}$. In our program, we input δ and β directly.
The input beam is assumed to be monochromatic without any spatial divergence.

3.2 Basic formalism for scalar index of refraction

In this section, we investigate refraction by a multilayer system with flat interfaces and with scalar refraction indices. We first develop the basic formalism without absorption, and far from total reflection conditions. Absorption, total reflection and evanescent waves will be discussed in subsection 3.2.5. In section 3.3, we generalize the formalism to include polarization-dependent interaction.

Polarized neutron propagation at glancing incidence in multilayer systems has been studied by Spiering and Deak [...], using Abeles' matrix formalism and matrix exponentials. In Abeles' formalism [], plane waves are parametrized by ψ and $\partial_z \psi$. We prefer a parametrization (3.4) by the amplitudes of forward and backward propagat-

ing plane waves, which is mathematically simpler, and allows a more direct physical interpretation.

3.2.1 Wave equation for a basic multilayer model

The geometry for reflection by a multilayer sample is shown in figure 3.2. Here we only consider specular reflection, hence the azimuthal angle $\phi_f = 0$.

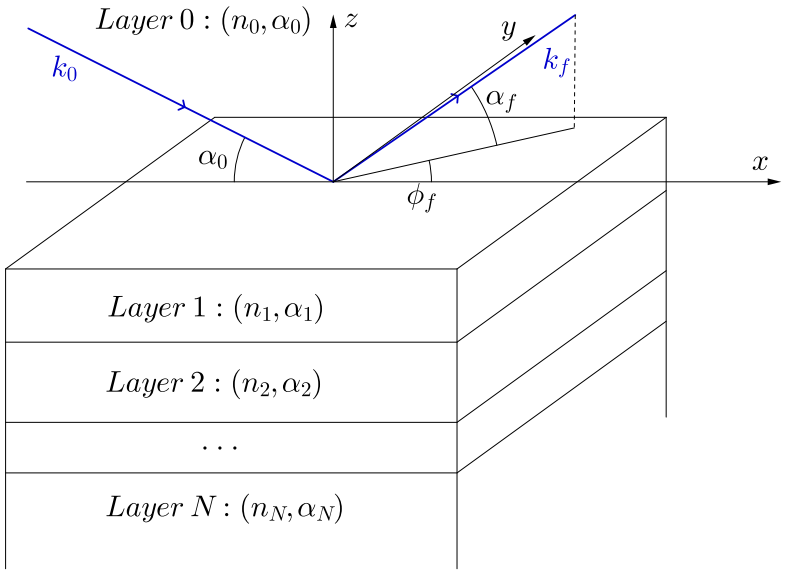

Figure 3.2: Representation of the scattering geometry for multilayer specular reflectivity. n_i is the refractive index of layer i and α_i is the incident angle of the wave propagating in layer i and incident on layer $i+1$. α_f is the exit angle with respect to the sample's surface and ϕ_f is the scattering angle with respect to the scattering plane.

All layer interfaces are assumed to be perfectly smooth. All layers are homogenous; layer i has the complex refractive index n_i. Layer 0 is the air or vacuum on top of the sample; layer N is the substrate; these two layers are assumed semi-inifinite. We first consider scalar interactions. Polarization-dependent reflection will be discussed in section 3.3.

Within one layer, stationary wave propagation with given frequency, hence with fixed vacuum wavenumber K, obeys the differential equation

$$\left(\Delta + K^2 n_i^2\right)\psi(\mathbf{r}) = 0. \tag{3.1}$$

At interfaces between layers, the wave function $\psi(\mathbf{r})$ and its first derivative $\nabla\psi(\mathbf{r})$ must evolve continuously.

3.2.2 Wave propagation within one layer

Since our multilayer model has infinite extension in x and y–directions, we can factorize $\psi(\mathbf{r})$, and assume plane-wave propagation for the in-plane component $\mathbf{r}_\parallel$:

$$\psi(\mathbf{r}) = \psi(z)\mathrm{e}^{i\mathbf{k}_\parallel \mathbf{r}_\parallel}. \tag{3.2}$$

From the continuity conditions we infer that $\mathbf{k}_\parallel$ is constant across layers. The wave equation (3.1) reduces to a one-dimensional equation in z,

$$\left(\partial_z^2 + K^2 n_i^2 - k_\parallel^2\right)\psi(z) = 0, \tag{3.3}$$

which is solved by

$$\psi_i(z) = a_i \mathrm{e}^{ik_{\perp i}(z-z_i)} + b_i \mathrm{e}^{-ik_{\perp i}(z-z_i)}, \tag{3.4}$$

where z_i is the coordinate of the *bottom* interface of layer i, introduced here as a constant offset for later convenience. In the case of the semi-infinite bottom layer N, z_N can be chosen arbitrarily.

Inserting (3.4) into (3.3), we obtain the dispersion relation

$$k_\parallel^2 + k_{\perp i}^2 = K^2 n_i^2. \tag{3.5}$$

If n_i is real and $k_\parallel < K n_i$, this is just the Pythagorean equation for the perpendicular components of either of the two wavevectors: vector $\mathbf{k}_\pm = \mathbf{k}_\parallel \pm k_{\perp i}\hat{\mathbf{z}}$. They correspond to the two summands in (3.4), and describe plane waves propagating upwards or downwards. These waves have glancing angles

$$\alpha_i := \arctan(k_{\perp i}/k_\parallel). \tag{3.6}$$

Equivalently,

$$k_\parallel = K n_i \cos\alpha_i. \tag{3.7}$$

Since $k_\parallel$ is constant across layers, we have

$$n_i \cos\alpha_i = \text{the same for all } i, \tag{3.8}$$

which is Snell's refraction law.

For later convenience we abbreviate

$$f_i := \sqrt{n_i^2 - n_0^2 \cos^2\alpha_0}, \tag{3.9}$$

which is a combination of material parameters (n_0, n_i) and a geometric parameter (α_0) that describes the incident beam. If n_i is real, and the argument of the square root nonnegative, we simply have $f_i = n_i \sin\alpha_i$. However, using f_i allows for later generalizations to cope with absorbing media or with total reflection and evanescent waves. The wavenumber in (3.4) can now be written as

$$k_{\perp i} = K f_i. \tag{3.10}$$

3.2.3 Wave propagation across layers

In this section, the continuity of $\psi(z)$ and $\partial_z\psi(z)$ at layer interfaces is used to derive a recursion rule for the coefficients a_i and b_i of (3.4). At the *bottom* interface of layer $i = 0,\ldots,N-1$, continuity requires

$$\begin{aligned} \psi_i(z_i) &= \psi_{i+1}(z_{i+1}+d_{i+1}), \\ \partial_z\psi_i(z_i) &= \partial_z\psi_{i+1}(z_{i+1}+d_{i+1}), \end{aligned} \tag{3.11}$$

where

$$d_i := z_{i-1} - z_i \tag{3.12}$$

is the thickness of layer i. With the solution (3.4), conditions (3.11) become

$$\begin{aligned} a_i + b_i, &= a_{i+1}\mathrm{e}^{iKf_{i+1}d_{i+1}} + b_{i+1}\mathrm{e}^{-iKf_{i+1}d_{i+1}}, \\ a_i f_i - b_i f_i, &= a_{i+1}\mathrm{e}^{iKf_{i+1}d_{i+1}} f_{i+1} - b_{i+1}\mathrm{e}^{-iKf_{i+1}d_{i+1}} f_{i+1}. \end{aligned} \tag{3.13}$$

We introduce the vector notation

$$c_i := \begin{pmatrix} a_i \\ b_i \end{pmatrix}, \tag{3.14}$$

to write (3.13) as

$$F_i c_i = F_{i+1} D_{i+1} c_{i+1} \tag{3.15}$$

with the matrices

$$F_i := \begin{pmatrix} 1 & 1 \\ f_i & -f_i \end{pmatrix}, \text{ and } D_i := \begin{pmatrix} \delta_i & 0 \\ 0 & \delta_i^* \end{pmatrix}, \tag{3.16}$$

and the phase factor

$$\delta_i := \mathrm{e}^{iKf_i d_i}. \tag{3.17}$$

We define the transfer matrix

$$M_i := F_{i-1}^{-1} F_i D_i, \tag{3.18}$$

to obtain the recursion

$$c_i = M_{i+1} c_{i+1}. \tag{3.19}$$

Straightforward computation yields

$$M_i = \frac{1}{2f_{i-1}} \begin{pmatrix} \delta_i(f_{i-1}+f_i) & \delta_i^*(f_{i-1}-f_i) \\ \delta_i(f_{i-1}-f_i) & \delta_i^*(f_{i-1}+f_i) \end{pmatrix} = \frac{1}{2} \begin{pmatrix} \delta_i(1+\rho_i) & \delta_i^*(1-\rho_i) \\ \delta_i(1-\rho_i) & \delta_i^*(1+\rho_i) \end{pmatrix} \tag{3.20}$$

with the ratio

$$\rho_i := \frac{f_i}{f_{i-1}}. \tag{3.21}$$

The recursion starts from the semi-infinite substrate layer N where there is no upwards propagating wave, hence $a_N = 0$. For layer i, the solution is

$$c_i = M_{i+1} \cdots M_N c_N. \tag{3.22}$$

3.2.4 Currents

Currents shall be computed in relative units as

$$\mathbf{J} := \psi^*(\mathbf{r}) \frac{\boldsymbol{\nabla}}{2i} \psi(\mathbf{r}) + \text{c.c.} \tag{3.23}$$

We are interested in the component J_z. With (3.4), we obtain in layer i

$$\begin{aligned} J_{zi} &= \tfrac{1}{2}\left(a_i^* \mathrm{e}^{-ik_{\perp i}(z-z_i)} + b_i^* \mathrm{e}^{ik_{\perp i}(z-z_i)}\right) k_{\perp i} \left(a_i \mathrm{e}^{ik_{\perp i}(z-z_i)} - b_i \mathrm{e}^{-ik_{\perp i}(z-z_i)}\right) + \text{c.c.} \\ &= \left(|a_i|^2 - |b_i|^2\right) K f_i, \end{aligned} \tag{3.24}$$

provided $k_{\perp i}$ is real. Obviously, the $|a_i|^2$ term is due to the upward beam and the $|b_i|^2$ term to the downward beam.

In the absence of absorption we expect that the net current is the same in all layers. We verify this as follows. Using the vector notation (3.14), we write (3.24) as a sesquilinear form

$$J_{zi} = c_i^\dagger \begin{pmatrix} K f_i & 0 \\ 0 & -K f_i \end{pmatrix} c_i. \tag{3.25}$$

Using the recursion (3.19) for c_i, we obtain a recursion for J_{zi}:

$$\begin{aligned} J_{zi} &= c_i^\dagger \begin{pmatrix} K f_i & 0 \\ 0 & -K f_i \end{pmatrix} c_i \\ &= c_{i+1}^\dagger M_{i+1}^\dagger \begin{pmatrix} K f_i & 0 \\ 0 & -K f_i \end{pmatrix} M_{i+1} c_{i+1} \\ &= c_{i+1}^\dagger \begin{pmatrix} K f_{i+1} & 0 \\ 0 & -K f_{i+1} \end{pmatrix} c_{i+1} \\ &= J_{z,i+1}, \end{aligned} \tag{3.26}$$

which means that the net current is constant.

For a single interface between two semi-infinite media, wave amplitudes are related by

$$\begin{pmatrix} a_0 \\ b_0 \end{pmatrix} = M_1 \begin{pmatrix} 0 \\ b_1 \end{pmatrix} = \frac{\delta_1^* b_1}{2f_0} \begin{pmatrix} f_0 - f_1 \\ f_0 + f_1 \end{pmatrix}. \tag{3.27}$$

The reflectivity of the interface is

$$R = \left| \frac{a_0}{b_0} \right|^2 = \left| \frac{f_0 - f_1}{f_0 + f_1} \right|^2, \tag{3.28}$$

which agrees with Fresnel's result for s-polarized light.

3.2.5 Damped waves in absorbing media or under total reflection conditions

If layer i absorbs radiation, its index of refraction n_i has a positive imaginary component. By (3.5), $k_\parallel^2 + k_{\perp i}^2$ then also has a positive imaginary component. From the continuity of ψ and $\boldsymbol{\nabla}\psi$ across layer interfaces it still follows that $\mathbf{k}_\parallel$ is constant. We assume that the top layer 0 is not absorbing. Hence $\mathbf{k}_\parallel$ is real. To fulfill (3.5), it is necessary that $k_{\perp i}$ has a positive imaginary component. In consequence, the intensities of the forward and backward travelling beams (3.4) decrease exponentially in $\pm z$.

Snell's law of refraction (3.8) cannot be fulfilled if

$$n_i < n_0 \cos \alpha_0. \tag{3.29}$$

In this case, total reflection occurs at the top interface of layer i, accompanied by an evanescent wave within layer i. Strictly speaking, total reflection is only possible if layer i is not absorbing. Otherwise, some intensity would be dissipated by the evanescent wave, and the reflection would not be total. Also, the unequality (3.29) is undefined if n_i has an imaginary component.

With definition (3.9), total reflection occurs if f_i is a pure imaginary number. For an absorbing medium, f_i has a positive imaginary part and a non-zero real part. Therefore it is appropriate to treat total reflection as a special case of refraction by a medium with complex f_i.

For complex f_i, the theory developed above remains applicable, with the following exceptions: The geometric interpretation of the wavevectors $\mathbf{k}_\pm$ in Eqs. (3.6–3.8) is untenable, and the computation of currents in Eqs. (3.24–3.26) is invalid because it relies on $\operatorname{Im} k_{\perp i} = 0$.

3.2.6 Numerical considerations

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3.3 Reflection with polarization-dependent interactions

3.3.1 Wave equation and propagation within one layer

To allow for polarization-dependent interactions, we replace the squared index of refraction n^2 by $1+\underline{\underline{\chi}}$, where $\underline{\underline{\chi}}$ is a 2×2 susceptibility matrix. The wave equation (3.1) for layer i becomes

$$(\Delta + K^2 + K^2 \underline{\underline{\chi}}_i)\underline{\psi}(\mathbf{r}) = 0, \tag{3.30}$$

where $\underline{\psi}(\mathbf{r})$ is a two-component spinor wave function, with components $\psi_\uparrow(\mathbf{r})$ and $\psi_\downarrow(\mathbf{r})$. At interfaces between layers, both spinor components of $\underline{\psi}(\mathbf{r})$ and $\boldsymbol{\nabla}\underline{\psi}(\mathbf{r})$ must evolve continuously.

The reasons for the factorization (3.2) still apply, and so we can write

$$\underline{\psi}(\mathbf{r}) = \underline{\psi}(z)\mathrm{e}^{i\mathbf{k}_\parallel \mathbf{r}_\parallel}. \tag{3.31}$$

As before, $\mathbf{k}_\parallel$ is constant across layers. The wave equation (3.30) reduces to

$$\left(\partial_z^2 + K^2 + K^2\underline{\underline{\chi}}_i - k_\parallel^2\right)\underline{\psi}(z) = 0. \tag{3.32}$$

We abbreviate

$$\underline{\underline{H}}_i := K^2(1+\underline{\underline{\chi}}_i) - k_\parallel^2 \tag{3.33}$$

so that the wave equation becomes simply

$$\left(\partial_z^2 + \underline{\underline{H}}_i\right)\underline{\psi}(z) = 0. \tag{3.34}$$

The solution is

$$\underline{\psi}_i(z) = \sum_{j=1}^{2} \underline{x}_{ij}\left(\alpha_{ij}\mathrm{e}^{ip_{ij}(z-z_i)} + \beta_{ij}\mathrm{e}^{-ip_{ij}(z-z_i)}\right), \tag{3.35}$$

where the $\underline{x}_{ij}$ are eigenvectors of $\underline{\underline{H}}_i$ with eigenvalues p_{ij}^2:

$$\left(-p_{ij}^2 + \underline{\underline{H}}_i\right)\underline{x}_{ij} = 0 \quad \text{for} \quad j = 1,2. \tag{3.36}$$

In a reproducible algorithm, the eigenvectors $\underline{x}_{ij}$ must be chosen according to some arbitrary normalization rule, for instance

$$|\underline{x}_{ij}| = 1, \quad x_{ij\uparrow} \text{ real and nonnegative.} \tag{3.37}$$

Similarly, a rule is needed how to handle the case of one degenerate eigenvalue, which includes in particular the case of scalar interactions.

3.3.2 Wave propagation across layers

Generalizing (3.14), we introduce the coefficient vector

$$c_i := (\alpha_{i1}, \alpha_{i2}, \beta_{i1}, \beta_{i2})^{\mathrm{T}}. \tag{3.38}$$

To match solutions for neighboring layers, continuity is requested for both spinorial components of $\underline{\psi}$ and $\nabla\underline{\psi}$. As before (3.15), we have at the bottom of layer i

$$F_i c_i = F_{i+1} D_{i+1} c_{i+1}, \tag{3.39}$$

where the matrices are now

$$F_i := \begin{pmatrix} x_{i1\uparrow} & x_{i2\uparrow} & x_{i1\uparrow} & x_{i2\uparrow} \\ x_{i1\downarrow} & x_{i2\downarrow} & x_{i1\downarrow} & x_{i2\downarrow} \\ x_{i1\uparrow} p_{i1} & x_{i2\uparrow} p_{i2} & -x_{i1\uparrow} p_{i1} & -x_{i2\uparrow} p_{i2} \\ x_{i1\downarrow} p_{i1} & x_{i2\downarrow} p_{i2} & -x_{i1\downarrow} p_{i1} & -x_{i2\downarrow} p_{i2} \end{pmatrix} \tag{3.40}$$

and

$$D_i := \mathrm{diag}(\delta_{i1}, \delta_{i2}, \delta_{i1}^*, \delta_{i2}^*) \tag{3.41}$$

with the phase factor

$$\delta_{ij} := \mathrm{e}^{i p_{ij} d_i}. \tag{3.42}$$

Note that matrix F_i has the block form

$$F_i = \begin{pmatrix} \underline{\underline{x}}_i & \underline{\underline{x}}_i \\ \underline{\underline{x}}_i \underline{\underline{P}}_i & -\underline{\underline{x}}_i \underline{\underline{P}}_i \end{pmatrix} = \underline{\underline{x}}_i \cdot \begin{pmatrix} \underline{\underline{1}} & \underline{\underline{1}} \\ \underline{\underline{P}}_i & -\underline{\underline{P}}_i \end{pmatrix}, \tag{3.43}$$

with

$$\underline{\underline{x}}_i := \left(\underline{x}_{i1}, \underline{x}_{i2}\right), \quad \underline{\underline{P}}_i := \mathrm{diag}\left(p_{i1}, p_{i2}\right). \tag{3.44}$$

This facilitates the computation of the inverse

$$F_i^{-1} = \frac{1}{2} \begin{pmatrix} \underline{\underline{1}} & \underline{\underline{P}}_i^{-1} \\ \underline{\underline{1}} & -\underline{\underline{P}}_i^{-1} \end{pmatrix} \cdot \underline{\underline{x}}_i^{-1}, \tag{3.45}$$

which is needed for the transfer matrix M_i, defined as in (3.18). With the new meaning of c_i and M_i, the recursion (3.19) and the explicit solution (3.22) hold as derived above. To resolve (3.22) for the reflected amplitudes α_{0j} as function of the incident amplitudes β_{0j}, we choose the notations

$$\underline{\alpha}_i := \begin{pmatrix} \alpha_{i1} \\ \alpha_{i2} \end{pmatrix}, \quad \underline{\beta}_i := \begin{pmatrix} \beta_{i1} \\ \beta_{i2} \end{pmatrix}, \quad M := M_1 \cdots M_N =: \begin{pmatrix} \underline{\underline{m}}_{11} & \underline{\underline{m}}_{12} \\ \underline{\underline{m}}_{21} & \underline{\underline{m}}_{22} \end{pmatrix}, \tag{3.46}$$

where the $\underline{\underline{m}}_{jk}$ are 2×2 matrices. Eq. (3.22) then takes the form

$$\begin{pmatrix} \underline{\alpha}_0 \\ \underline{\beta}_0 \end{pmatrix} = \begin{pmatrix} \underline{\underline{m}}_{11} & \underline{\underline{m}}_{12} \\ \underline{\underline{m}}_{21} & \underline{\underline{m}}_{22} \end{pmatrix} \begin{pmatrix} \underline{0} \\ \underline{\beta}_N \end{pmatrix}, \tag{3.47}$$

which immediately yields

$$\underline{\alpha}_0 = \underline{\underline{m}}_{12} \underline{\underline{m}}_{22}^{-1} \underline{\beta}_0. \tag{3.48}$$

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