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Profile reconstruction from neutron reflectivity data and *a priori* knowledge

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Abstract

The problem of incomplete and noisy information in profile reconstruction from neutron reflectometry data is considered. In particular methods of Bayesian statistics in combination with modelling or inverse scattering techniques are considered in order to properly include the required *a priori* knowledge to obtain quantitatively reliable estimates of the reconstructed profiles. Applying Bayes theorem the results of different experiments on the same sample can be consistently included in the profile reconstruction.

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1. Introduction

Neutron reflectometry is a widely applied technique in materials science [1]. It is well known that a unique reconstruction of the depth profile requires the knowledge of the complete reflection coefficient (modulus and phase). However, measurements with standard neutron reflectometer setups yield only the reflectivity (modulus), but not the phase of the reflection coefficient. Several proposals to solve this so-called *phase problem* of specular neutron reflectometry have been worked out [2–6], but for various reasons none of them has become an established technique so far. To our knowledge only the method of Ref. [6] has been implemented experimentally for a specific case [7]. Because of the missing phase information additional assumptions are required to uniquely reconstruct the depth profile from experimental reflection data. For example, we assumed knowledge of the nuclear depth profile and developed an iterative procedure to uniquely reconstruct magnetic profiles from polarized neutron reflectometry data [8]. However, in general the analyses of neutron reflectometry data rely on modelling, which make use of *a priori* knowledge in a usually uncontrolled way to by-pass the phase problem. Therefore, it is difficult to quantitatively estimate the reliability of the extracted profile.

In this contribution we study the method of profile reconstruction in detail. With regard to recent developments [8] we restrict ourselves to nuclear profiles. The unique reconstruction of profiles from experimental reflection data is hampered not only by the missing data on the phase of the reflection coefficient, but also by the limitations of the accessible momentum range as well as by the uncertainties of the measurements. In particular the latter represent a severe difficulty because of the *ill-posedness* of inverse scattering techniques (reconstruction techniques), which result in instabilities of the reconstructed profiles. *A priori* knowledge is required to overcome these difficulties and to obtain quantitatively reliable profiles. Bayesian statistics [9] offers a well-defined procedure to incorporate the *a priori* information into the reconstruction process. Within this concept of statistics we will consider reconstruction via modelling as well as via inverse scattering techniques.

In Section 2 we briefly recall the basic relationships of Bayesian statistics and apply them to the analysis of neutron reflectometry data. In Section 3 we follow the idea of Inguva and Baker-Jarvis [10] and propose a statistical procedure

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based on the Marchenko equation with noisy data. A summary and final conclusions are given in Section 4.

2. Modelling and statistics

In specular neutron reflection one is dealing with one-dimensional quantal scattering by a potential V which is proportional to the scattering-length density profile perpendicular to the surface. Hence, we assume a layered sample whose potential varies only with the coordinate x perpendicular to the surface. The scattering of a neutron beam with normal wave number q is then characterized by the complex reflection and transmission coefficients $R(q)$ and $T(q)$, respectively. For a known potential $V(x)$ the calculation of $R(q)$ and $T(q)$ is straightforward. The analysis of experimental data requires inferences in the opposite direction, i.e. from the data to the potential, and is usually denoted as an inverse problem, whose solution is generally more involved. Therefore most analyses of reflectometry data rely on iterative calculations of $r(q)$ with a parametrized ansatz of $\tilde{V} = V(r, \underline{a})$. In this so-called modelling procedure the parameters are adjusted by a χ^2-fit to the experimental $r(q)$-values.

In standard reflectometer setups only the reflectivity $r(q) = |R(q)|^2$ is measured with uncertainty $\sqrt{\Delta^2 r(q)}$ in a limited momentum range $0 \leqslant q \leqslant q_{\max}$. Hence, with regard to a unique reconstruction the information is incomplete and additional information is required (Section 1). Important *a priori* information is entered via the ansatz of the parametrized form of the potential. Without restriction to a specific experiment we can make the following assumptions: (1) the sample is mounted on a known substrate, (2) the maximal thickness a of the sample, (3) the material at the surface of the sample and (4) the minimal thickness of a layer are known. In the region between $0 \leqslant x \leqslant a$ the potential $V(x)$ is not determined and we can give only a probability distribution. In Fig. 1 we show the associated potential with its uncertainties.

Since we are dealing with noisy and incomplete data, any reconstruction can only yield probability distributions for the profiles. In probability theory we have to deal with two fundamental relationships for the probability distribution p

$$p(\underline{v}\,|\,\underline{r}, M) + p(\bar{\underline{v}}|\,\underline{r}, M) = 1, \tag{1}$$

$$\begin{aligned} p(\underline{v}, \underline{r}\,|M) &= p(\underline{v}\,|\,\underline{r}, M)p(\underline{r}\,|M) \\ &= p(\underline{r}\,|\,\underline{v}, M)p(\underline{v}\,|M), \end{aligned} \tag{2}$$

where $\underline{v}, \underline{r}, M$ are propositions which are '*true*' and $\bar{\underline{v}}$ stands for the proposition $\underline{v}$ is '*false*'. Here, we have introduced the propositions relevant in neutron reflectometry, i.e. $\underline{v}$ refers to the set of the parameters in the model potential, $\underline{r}$ to the set of experimental data and M refers to the model expressed in terms of the conditions listed above. The product rule (2) reflects the symmetry of p with regard to the parameters $\underline{v}$ and observables $\underline{r}$ and leads directly to

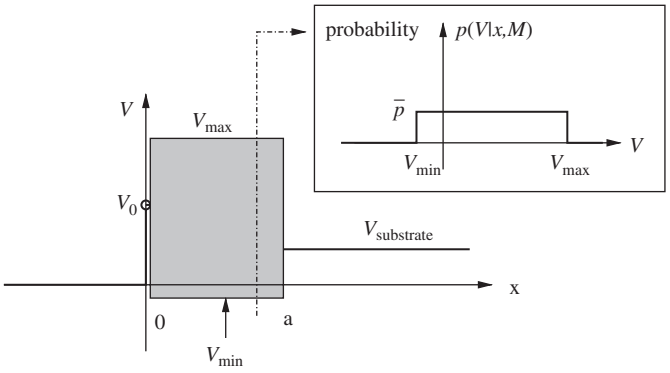

Fig. 1. *A priori* knowledge on the sample. The shaded area gives the possible variations of the potential in the regions $0 \leqslant x \leqslant a$. The boundaries are determined by the largest and smallest scattering length densities of all materials. In addition, the material on the surface is given, i.e. $V_0 = V(0)$. The inset shows the *a priori* probability distribution of the potential values at a given x, i.e. $p(V|x, M)$.

Bayes theorem [9]

$$p(\underline{v}\,|\,\underline{r}, M) = \frac{p(\underline{r}\,|\,\underline{v}, M)}{p(\underline{r}\,|M)} p(\underline{v}\,|M) \tag{3}$$

which allows to update the *prior* $p(\underline{r}\,|M)$ with the information of a new measurement expressed in the *likelihood function* $p(\underline{r}\,|\,\underline{v}, M)$. Thus the *a posteriori distribution* $p(\underline{v}\,|\,\underline{r}, M)$ is the expectation on the set of parameters taking consistently into account the experimental and the *a priori* knowledge.

The likelihood function gives the probability distribution of the data $\underline{r}$ for a model M with parameters $\underline{v}$ and is of the form

$$\begin{aligned} &p(\underline{r}\,|\,\underline{v}, M) = \hat{N} \exp[-\tfrac{1}{2}\chi^2] \\ &\quad \text{with } \chi^2 = (\underline{r} - \underline{r}^M(\underline{v}))^{\mathrm{T}} \mathscr{B}^{-1} (\underline{r} - \underline{r}^M(\underline{v})) \end{aligned} \tag{4}$$

where $\hat{N}$ is an appropriate normalization constant and $\mathscr{B}$ is the experimental covariance matrix with elements $B_{ij} = \langle \Delta r_i \Delta r_j \rangle$.

The determination of the prior is the most important step for the application of Bayesian statistics. Here, we assume that the parameters $\underline{v} = (v_1, \ldots, v_M)$ correspond to values of the potential at specific mesh points $(x_1, \ldots, x_M)$. Thus it is reasonable to assume the uncorrelated probability

$$p(\underline{v}\,|M) = \prod_{i=1}^{M} p(v_i) \tag{5}$$

with $\bar{p} = (V_{\max} - V_{\min})^{-1}$ and

$$p(v_i) = \begin{cases} \bar{p} & 0 < x \leqslant a \\ 0 & x \leqslant 0 \text{ or } a < x. \end{cases} \tag{6}$$

If we know the materials within the sample, e.g. from the production process, we can reduce $p(v_i)$ to equally weighted discrete values.

Using this prior and the likelihood function (4) we obtain the probability distribution of the parameters $\underline{v}$ and

thus of the potential $V(x)$. The calculation of uncertainties and correlations of the potential is straightforward. If we want to take a further measurement into account, we can use the *a posteriori* distribution (3) as a prior and multiply with the new likelihood function to obtain after normalization the new *a posteriori* distribution. Thus also X-ray reflectivity data can consistently be included into the reconstruction procedure.

3. Inverse scattering and information entropy

The use of inverse scattering techniques for profile reconstruction has been extensively discussed in the literature, e.g. Refs. [11,12] and references therein. These methods are based on the solution of the Marchenko equation for $x>y$

$$K(x,y)+B(x+y)+\int_{-x}^{+x} \mathrm{d}z K(x,z)B(z+y)=0, \tag{7}$$

where the reflection coefficient $R(q)$ enters via the Fourier transformation

$$B(x)=\frac{1}{2\pi}\int_{-\infty}^{+\infty} \mathrm{d}q \mathrm{e}^{-\mathrm{i}qx}R(q) \tag{8}$$

for $x>0$, while $B(x)=0$ for $x<0$. The potential is obtained from the kernel $K(x,y)$ via

$$V(x)=2\frac{\mathrm{d}}{\mathrm{d}x}K(x,x-0) \quad \text{for } x>0. \tag{9}$$

In several studies on the phase problem in neutron reflectometry [2,3,13,14] reconstructions with noisy schematic data of the reflection coefficient $R(q)$ have been performed. The quality of the corresponding reconstructions as well as the depth resolution depended strongly on the noise of the data, while the limitation in $q\leqslant q_{\max}$ was only reflected in smeared out transitions and Gibbs oscillations at the interfaces of the layers.

The situation is different for the reconstruction of profiles from reflectivity data of standard experiments. Due to the missing phase information the ill-posedness of the inverse problem represents a severe difficulty for the reconstruction of the profile. In order to carry out the inversion procedure with noisy and incomplete information, Inguva and Baker-Jarvis [10] suggested a statistical procedure and assume that the actual data provide only a probability $p(\underline{v}\,|\,\underline{R})$ for the resulting potential.

In order to determine this probability we follow Ref. [10] and discretize the Marchenko equation (7). In particular we introduce a mesh in the coordinate x ($x_i,\ i=1,\ldots,N$). Thus the discretized Marchenko equation (7) can be cast into the form

$$\sum_{m<i} K_{i,m}[w_m^{(i)}B_{m,j}+\delta_{j,m}]=-B_{i,j}, \tag{10}$$

where $w_m^{(i)}$ are the weights for the chosen quadrature and $m<i$ refers to values of the kernel $K(x_i,y_m)$ with $y_m<x_i$. The linear equation (10) can easily be solved for the unknowns $K_{i,m}$ at each x_i. In the following we treat $\underline{K}$ as a matrix of random variables $K_{i,j}$ with the probability $p(\underline{K})$. Following Shannon [15] we introduce the information entropy

$$S=-\int \prod_{j<i=1}^{N} (\mathrm{d}K_{i,j})p(\underline{K})\ln p(\underline{K}). \tag{11}$$

Here and in the following $i>j$ refers to all unknown $K_{i,j}$-values up to $i=N$ as outlined in the explanation of Eq. (10). At fixed reflection coefficient $R(q)$ the most probable distribution $p(\underline{K})$ is given by the maximum of the information entropy under the constraints

$$1=\int \prod_{j<i=1}^{N} (\mathrm{d}K_{i,j})p(\underline{K}) \tag{12}$$

$$-B_{i,j}=\langle K_{i,j}\rangle+\sum_{m<i} w_m^{(i)}\langle K_{i,m}\rangle B_{m,j} \tag{13}$$

with

$$\langle K_{i,j}\rangle=\int \prod_{j<i=1}^{N} (\mathrm{d}K_{i,j})p(\underline{K})K_{i,j}. \tag{14}$$

Using the technique of Lagrange multipliers $\lambda_{i,j}$, $j<i$ one obtains the probability

$$p(\underline{K})=\frac{1}{Z}\exp\left(-\sum_{j<i} K_{i,j}\Gamma_{i,j}\right) \tag{15}$$

with

$$Z=\int \prod_{j<i=1}^{N} (\mathrm{d}K_{i,j})\exp\left(-\sum_{m<k} K_{m,k}\Gamma_{m,k}\right) \tag{16}$$

$$\Gamma_{i,j}=\sum_m [w_j^{(i)}B_{j,m}+\delta_{j,m}]\lambda_{i,m}. \tag{17}$$

Following Ref. [10] we assume uncorrelated values $K_{i,j}\in[-B,\infty[$, where $-B$ is a reasonably chosen lower bound. Thus we are able to perform the integrations in Eqs. (15) and (16) and obtain

$$p(\underline{K})=\prod_{j<i}\Gamma_{i,j}\exp\left(-\sum_{m<k} X_{k,m}\right) \tag{18}$$

with

$$X_{k,m}=\Gamma_{k,m}(K_{k,m}+B) \tag{19}$$

and

$$\langle K_{k,m}\rangle=\frac{1}{\Gamma_{k,m}}-B. \tag{20}$$

The Lagrange multipliers can be determined by the solution of

$$\sum_m (w_m^{(i)}B_{m,j}+\delta_{m,j})[\Gamma_{m,j}^{-1}-B]=-B_{i,j}. \tag{21}$$

Thus the probability distribution (18) is completely known and one can determine $\langle K_{i,j}\rangle$, the potential $V(x)$ and the corresponding probability $p(\underline{V})$.

Following Ref. [10] we have implicitly assumed to know the phase of $R(q)=\sqrt{r(q)}\exp(\mathrm{i}\varphi(q))$ with some uncertainty. At first glance this condition is not satisfied because standard neutron reflectometer experiments yield only $r(q)$. However, due to the existence of dispersion relations [16,17] the reflection coefficient can be given in the form

$$R(q)=\sqrt{r(q)}\exp(\mathrm{i}\varphi_{\mathrm{H}}(q))\prod_{i=1}^{\tilde{M}}\frac{q-a_i}{q+a_i}\frac{q+a_i^*}{q-a_i^*} \tag{22}$$

with the Hilbert phase

$$\varphi_{\mathrm{H}}(q)=-\pi-\frac{q}{2\pi}\int_0^\infty \mathrm{d}q'\frac{\ln r(q')-\ln r(q)}{q^2-q'^2}. \tag{23}$$

Thus the determination of the phase is reduced to the determination of the complex valued zero points a_i of the reflection coefficient, which occurs always in pairs because of $R(-q)=R^*(q)$ for real potentials. In addition, there is a relationship between the potentials with equivalent reflectivity by the recursive procedure for $n=1,2,\ldots,\tilde{M}$

$$V_n(x)=V_{n-1}(x)-2\frac{\mathrm{d}^2}{\mathrm{d}x^2}\ln\left\{\mathrm{Im}\left[f_{n-1}^+(a_n,x)\frac{\mathrm{d}}{\mathrm{d}x}f_{n-1}^+(-a_n^*,x)\right]\right\} \tag{24}$$

with $V_0(x)=V_{\mathrm{H}}(x)$ and $V(x)=V_{\tilde{M}}(x)$. Here $V_{\mathrm{H}}(x)$ is the potential associated with the Hilbert phase $\varphi_{\mathrm{H}}(q)$ and $f_{i-1}^+(a_i,x)$ are the Jost solutions of the Schrödinger equation with $V_{i-1}(x)$ at the momentum $k=a_i$. However, we do not obtain any information about the number $\tilde{M}$ and the zero points of $R(q)$ from standard experiments.

Summarizing these facts, Eq. (18) leads to a conditioned probability $p(\underline{V}\,|\,\underline{r},\underline{\varphi}_{\mathrm{H}},I)$, where I stands for a specific combination of zero points $\underline{a}$. In order to obtain a probability distribution similar to Eq. (3) one has to evaluate first

$$p(\underline{v}\,|\,\underline{r})=\tilde{N}\sum_I\int \mathrm{d}\,\underline{r}\int \mathrm{d}\underline{\varphi}_{\mathrm{H}}p(\underline{v}\,|\,\underline{r},\underline{\varphi}_{\mathrm{H}},\underline{a},I) \tag{25}$$

$$\times p(\underline{\varphi}_{\mathrm{H}}\,|\,\underline{r})p_{\mathrm{exp}}(\underline{r})p(\underline{a}\,|I) \tag{26}$$

where $p_{\mathrm{exp}}(\underline{r})$ is the probability distribution of the measured reflectivities. The probability $p(\underline{\varphi}_{\mathrm{H}}\,|\,\underline{r})$ refers to the fact that the measurement of $r(q)$ is limited to $q\leqslant q_{\max}$ and leads therefore to some uncertainty in the evaluation of the $\varphi_{\mathrm{H}}(q)$ via Eq. (23). Finally, the probability $p(\underline{a}\,|I)$ accounts for the distribution of the zero points of $R(q)$ for a given model I, characterized by the number of a_i-values. In practical applications one will assume equally distributed values of the real and imaginary part in the interval $0\leqslant q\leqslant 2q_{\max}$. The constant $\tilde{N}$ provides the normalization (1) of the probability.

So far we have not used any of the four constraints on the model potential given in Section 2. In order to include these constraints we transform $p(\underline{v}\,|\,\underline{r})$ into $p(\underline{v}\,|\,\underline{r},M)$ by applying $p(\underline{v}\,|M)$ as a filter, i.e.

$$p(\underline{v}\,|\,\underline{r},M)=\begin{cases}Np(\underline{v}\,|\,\underline{r}) & \text{if } p(\underline{v}\,|M)\neq 0\\ 0 & \text{if } p(\underline{v}\,|M)=0.\end{cases} \tag{27}$$

Here N is again a proper normalization of the final probability in order to satisfy Eq. (1). It is expected that the model conditions of Section 2 will select admissible zero points of $R(q)$, thus significantly reducing the ambiguities.

In principle the probability of Eq. (27) obtained via the inverse scattering techniques contains essentially the same data and *a priori* information as the probability distribution (3) obtained by modelling. Therefore, despite the differences in procedure the final result should reveal the same characteristics of the depth profile. A verification of this supposition would be very helpful with regard to extensions of these procedures to other model constraints.

4. Conclusions

We have considered the reconstruction of depth profiles from neutron reflection data within a statistical context in order to consistently include *a priori* information. Assuming constraints on the model potential the application of Bayesian statistics to modelling is straightforward. Following Inguva and Baker-Jarvis [10] we developed a statistical procedure for profile reconstruction, which is based on the Marchenko equation. The method is much more involved, but should finally lead to a comparable result without explicitly fitting the potential. At present work is in progress to demonstrate the feasibility of the method and to compare its result with modelling. Both solutions are promising with regard to uniqueness and quantitative estimate of the extracted profile. In addition, they allow a consistent update with other experimental data, e.g. from X-ray reflectivity measurements.

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