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TITLE: BAYESIAN SPECTRAL ANALYSIS OF REFLECTIVITY DATA

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BAYESIAN SPECTRAL ANALYSIS OF REFLECTIVITY DATA

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ABSTRACT. The analysis of neutron and x-ray reflectivity data to obtain scattering-length density profiles is akin to the notorious phaseless Fourier problem, well-known in many fields such as crystallography. Current methods of analysis culminate in the refinement of a few parameters of a functional model, and are often preceded by a long and laborious process of trial-and-error. A discussion of the use of maximum entropy for obtaining "free-form" solutions of the density profile, as an alternative to the trial-and-error phase when a functional model is not available, and the suggestion for a novel experimental procedure designed to alleviate the ambiguity problems inherent in traditional reflectivity measurements, are given in Sivia *et al.* (1990). In this paper we consider the Bayesian spectral analysis approach, which is appropriate for optimising the parameters of a simple (but adequate) type of model when the number of parameters is not known.

1. Introduction

A wide variety of interfaces of scientific and technological interest exhibit depth profile structure on length scales of tens to thousands of Ångström. Examples include: model biological lipid membranes, analogous to the walls of living cells; semi-conductor multilayers of importance to the electronics industry; organic thin-film chemical sensors; conjugated polymers for electro-optic devices; and thin-film superconductors. This size range is an order of magnitude less than is accessible by conventional optical techniques. One method of probing interfacial structures on these length scales is to measure the interference effects of the depth profile on the reflection of short wavelength radiation. This extension of visible range optical techniques to shorter wavelengths has been vigorously pursued over the last few years in x-ray reflectometry, and also by the increasingly popular complementary technique of neutron reflectometry.

In Section 2 we consider reflectivity from a data analysis point-of-view and in Section 3 we present the Bayesian spectral analysis approach to the problem. This is a generalised form of model-fitting where the solutions are restricted to a broad class of models rather than to a particular model. The choice of the type of models encodes considerably more prior knowledge about the nature of the depth profile than does a free-form approach (such as MaxEnt), while still allowing considerably more flexibility than conventional model refinement. We conclude with Section 4.

Figure 2. A simple, but (hopefully) adequate, representation of the scattering-length density profile based on the prior knowledge that layered structures have histogram-like depth profiles.

profile by constructing a hypothesis space defined by the "heights and widths" of a few layers as illustrated in Fig. 2. We could make a more elaborate model for the density profile by assigning a "roughness" parameter to each interface, and a "tilt angle" to each layer, but we ignore such complications for simplicity in this paper (the generalisation being straight-forward). This is also in keeping with the spirit of Bayesian spectral analysis where we are trying to estimate the parameters of a *simple*, but *adequate*, type of model when the number of parameters is not known. By using our additional prior knowledge to construct a much smaller hypothesis space (than that used in the free-form approach) we hope to obtain more accurate estimates of the depth profile, the reliability of which is also easier to quantify.

For this formulation of the problem, our best estimate of the density profile is obtained by maximising the posterior p.d.f.:

$$\text{prob}[T_1, T_2, \dots, T_N, \beta_1, \beta_2, \dots, \beta_N | R(Q), \beta_s] \quad (1)$$

where $\{T_i, \beta_i\}$ are the set of heights and widths of the N layers describing the density profile, β_s is the scattering-length density of the substrate (assumed known) and $R(Q)$ are the reflectivity data. If the number of layers was known, this would be a straight-forward problem of parameter estimation (in principle). However, we do not know N ! So, we need to compute $\text{prob}[N | R(Q), \beta_s]$.

The mathematics presented below, to compute the posterior probability for the number of layers, is essentially the same as that found in Chapter 5 of Jeffreys (1939). It involves the use of Bayes' theorem and marginalisation, and requires a number of assumptions and approximations to be made in order to obtain an analytic solution. Although the assumptions and approximations are always open to question, they are not unusual in that they are always implicit when using the commonly-used least-squares analysis.

We begin by using Bayes' theorem:

Figure 3. (a) Simulated reflectivity data. (b) & (c) The posterior probability for the number of layers plotted on a logarithmic and linear scale respectively. (d) Best estimate of the density profile.

$$\text{prob}[N|R(Q)] = \frac{\text{Constant} \cdot N!}{[(\beta_{\text{max}} - \beta_{\text{min}}) \cdot T_{\text{max}}]^N} \times \frac{e^{-\chi^2_{\text{min}}/2}}{\sqrt{\text{Det}(\nabla\nabla\chi^2)}} \quad (9)$$

where $\text{Det}(\nabla\nabla\chi^2)$ is the determinant of the Hessian matrix.

AN EXAMPLE

Fig. 3(a) shows simulated reflectivity data generated from a density profile consisting of a few layers; the simulated data suffer from typical instrumental resolution broadening and noise. Figs. 3 (b) and (c) show the posterior probability for the number of layers, computed according to the formula derived above. T_{max} was taken to be the actual width of the sample (350 $\text{\AA}$) on the basis that, in general, it is very difficult to find the best-fit solutions if a good estimate of the overall width is not available. β_{max} was set to $10^{-5} \text{\AA}^{-2}$ in accordance with prior knowledge about the neutron scattering-lengths of atoms (the simulated density profile constructed to comply), β_{min} was set zero indicating prior

the complex topology of the likelihood function for this non-linear problem means that it is very hard to find the best-fit solution, even when one knows the number of layers; a full parameter-space search entails cosmological time-scales even for models with only 4 or 5 layers! In practice we have found that our chances of success are greatly increased (above zero) when we have a good estimate of the total width of the sample.

4. Conclusions

Bayesian spectral analysis is a very powerful technique. It is a generalised form of model-fitting, which is appropriate for optimising the parameters of a simple (but adequate) type of model when the number of parameters is not known. In this paper we have the example of estimating the density profile for a layered sample, when the number of layers is unknown, but there are numerous other applications. Gull (1988), for example, addresses the problem of fitting a polynomial to a pertinent set of finite and noisy data, (again) when the order of the polynomial is not known.

This method of analysis encodes more prior knowledge about the object of interest than does a free-form approach, while still allowing a considerable amount of flexibility over traditional model refinement. Although we are often hesitant to commit ourselves to (even a class of) simple functional models, the advantages of working in such a reduced *hypothesis space* are that we are able to obtain results which are more reliable and easier to quantify. It is in the nature of scientific inference that the best we can do is show that a theory (or choice of hypothesis space) is adequate (rather than "the truth"); if it is not adequate, probability theory will tell us so!

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