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The phase problem for X-ray specular reflectivity from thin films: A new approach

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ABSTRACT

A new approach to solving the phase problem for X-ray specular reflectivity is proposed. An iterative algorithm is used to solve the nonlinear Fredholm integral equation describing the intensity of the X-rays reflection from the sample surface and to determine the concentration depth-profile of element. Effectiveness of the proposed method was tested by numerical experiments for the thin film. We compare the results obtained for the Cr film experimental data using the Parratt model, phaseless inverse scattering method (logarithmic dispersion relations), and the proposed method.

1. Introduction

Determination of the electron density profile (EDP) perpendicular to an interface by specular X-ray reflectivity (XRR) has become a routine tool in surface and thin film science. Reflectivity data have been traditionally analyzed by the trial-and-error and non-linear least squares fitting methods using Parratt's recursive reflectivity calculation method [1] to obtain the final EDP. The main advantage of this method is that it involves relatively simple computation, but often the result is highly subjective. It is known that in this case there is no stability and no uniqueness [2–4]. Dependency of these fitting methods on an *a priori* postulated model follows from the fact usually only the amplitudes of the reflected waves are measured. This is so called the phase problem.

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Majkrzak et al. have proposed an exact theoretical approach to the phase retrieval problem in neutron and X-ray reflectivity. This approach is known as the reference layer method [5]. It is based on the following statement: (1) there are three samples composed of the same unknown layered structure and of different, but known, reference layers (substrates), (2) complex amplitude reflectivity of the unknown structure can be found exactly by measuring the reflectivity from all three samples. The approach, although being correct from a mathematical point of view, has a number of practical disadvantages. First, it is only valid for non-absorbing materials. Second, the optical properties of the different substrates or reference layers should be known *a priori*. At last, it is necessary to guarantee the absolute identity of the unknown structure within the three samples in spite of the presence of different substrates or reference layers.

Methods that do not use additional measurements have been proposed by Klibanov and Sacks [6] and by Clinton [7]. They used a logarithmic dispersion relation. The phase determined by this formula is unique if the reflection coefficient has no zeros in the upper half-plane. These methods are called the phaseless inverse scattering (PIS) methods, since they calculate the EDP from the experimental data without explicit phase information from elsewhere. Phase information is assumed to be implicitly present in the amplitude information and is extracted by mathematical means. Of course, the advantage of PIS ones is that the system to be studied is not disturbed by presence of a reference layer and that the data collection can be obtained, in general, in any laboratory equipped with a commercial diffractometer. Another advantage of PIS is that the mathematical data treatment is less laborious than that of two- or three measurements. Clinton wrote [7] that for multilayer systems the condition of absence of zeros in the upper half-plane can be provided in most cases and determined phase is unique. When using the high-quality Al_2O_3 substrates and the molecular beam epitaxy (MBE) for sputtering the samples, this condition is satisfied. Possibilities of solving ill-posed inverse problems by PIS methods from experimental data for the X-rays specular reflectivity were first demonstrated by Van der Lee [8]. The drawback is that some physical pre-information is required to judge the uniqueness of the result. However, it should be noted, that the methods proposed to determine the phase uniquely quite often give two alternative solutions as well, from which one has to be rejected on physical grounds as well [9]. A second disadvantage is that not every system is suitable for the EDP determination by PIS.

Kozhevnikov [10–12] proposed a new method of exact phase retrieval that is free from the shortcomings mentioned above. The method is applicable to a layered film, for which the reflectivity has been measured in-situ during growth, so that, at a point t in time, both the reflectivity R and the derivative dR/dt are known. In this case, both the real and imaginary parts of the amplitude reflectivity can be found exactly.

But there is a principally different approach. It is known that the equation describing the intensity of the X-ray specular reflection from the sample surface is a nonlinear Fredholm integral equation of the first kind. In this paper, we propose to solve this equation using the Levenberg–Marquardt method. Recently, this method was modified and was applied successfully to solving an inverse problem of gravimetry and magnetometry [13,14]. It is an effective way to avoid the phase problem. In our case, it is no need to find the scattered wave phase and not necessary to perform additional measurements. The proposed method can play an important role in laboratory experiments.

This paper is structured as follows. In Section 2, the nonlinear Fredholm integral equation for the XRR inverse problem is briefly described. Section 3 is devoted to description of mathematical algorithm. In Section 4, in order to test the proposed algorithm, the numerical simulation has been performed. In Section 5, we give the results of experimental studies obtained using the synchrotron radiation of the Russian Research Center "Kurchatov Institute" (Moscow). We compare the results of the processing in three cases: the Parratt model, PIS methods, and the proposed method.

2. A nonlinear integral equation for X-ray reflectivity

Recently, a new method for determination of the depth-profile of element $p_j(z)$ by XRR data for low contrast multilayer heterosystems was proposed. Consideration is carried out in terms of the canonical atomic distribution functions. It is a key idea of the new approach. Function $p_j(z)$ means the