



A subnanometric resolution method for studying local atomic structure of interface and surface of multilayered nanoheterostructure thin films

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ABSTRACT

An experimental method for studying the local atomic structure with a depth resolution in low-contrast multilayered thin films is proposed. We consider combination of the X-ray reflectivity and the extended X-ray absorption fine structure spectroscopy with angular resolution. To obtain the structural characteristics, the most advanced methods for solving inverse ill-posed problems are used, namely, the Tikhonov regularization method in the case of linear integral equations and the Levenberg-Marquardt algorithm for nonlinear problems. The proposed mathematical algorithms do not require *a priori* information about the studied system, such as the width and shape of the interface, the width and depth of the bands of the individual elements, or detailed information about the atomic structure. A software for the method has been developed. It allows us to obtain the most complete information about the local atomic structure of interface and surface. To evaluate the capabilities of the method, we used the system of cluster-layered nanostructures $\text{Al}_2\text{O}_3/\text{Cr}/\text{Fe-Cr}/\text{Cr}/\text{Fe-Cr}/\text{Cr}$ as a model. A depth resolution of 1–2 Å was reached.

1. Introduction

Multilayer structures are an important class of nanomaterials with unique magneto-transport properties. Surface and interface play a leading role in determining the properties of multilayers. In order to achieve the desired functionality, it is important to determine not only atomic but also electronic and magnetic structures of the interface in the multilayers. Because the width of the interface area varies from a fraction of a nanometer to several nm's, it is necessary to use experimental methods with depth resolution of 1 nm or better. A well-known method of X-ray reflectivity (XRR) is the main method of determining the dimensional characteristics of films and the depth profile of a certain type of atoms. It is only one side of the problem. On the other side, the short-range order is responsible for the magnetic properties of multilayers.

The X-ray techniques such as EXAFS (extended X-ray absorption fine structure) spectroscopy, X-ray emission spectroscopy, X-ray magnetic circular dichroism (XMCD), and Mossbauer spectroscopy are powerful methods for studying the local order around a certain type of atom, but these methods do not allow one to find the point of location of an excited atom (interface or layer). These methods do not have the depth resolution. There is no single experimental method that would satisfy

two criteria simultaneously: depth resolution and short-range order. This is a global problem of multilayer nanoheterostructure studies including superlattices Fe/Cr having a giant magnetoresistive (GMR) and the Kondo effects [1].

We have developed a model-independent method for investigation of the local atomic structure with depth resolution. The method gives us the most complete information on the atomic structure even for $[\text{Fe}/\text{Cr}]_n$ systems including cluster-layered ones with overlapping coordination spheres. The method consists in solving the inverse problems using the experimental data of the X-ray reflectivity and the angle-resolved EXAFS spectroscopy.

The paper is structured as follows. In Section 2, we briefly described a theory of an approach in the X-ray reflectivity [2]. Here, in order to test the proposed algorithm, the numerical simulation has been performed. We show the results of numerical simulations for the periodic, aperiodic multilayer systems and for cluster-layered Fe/Cr nanoheterostructures. Section 3 is devoted to describing a complete scheme of the method, which consists in solving inverse ill-posed problems without using any theoretical models. The method is performed in three steps: (1) determination of the concentration element i profile $p_i(z)$ with the depth z from X-ray reflectivity data; (2) determination of the X-ray absorption spectrum in fluorescence yield $\mu_i(z, E)$ for the element i as a

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function of depth z and photon energy E using the angle-resolved EXAFS data $I_i^f(E, \theta_i)$; and (3) determination of partial correlation functions $g_{ij}(z, r)$ as a function of depth z by solving the integral equations for functions $\chi_i(z, k)$ obtained from $\mu_i(z, E)$. All steps of the proposed method are demonstrated on a model example of cluster-layered Fe–Cr nanostructures $\text{Al}_2\text{O}_3/\text{Cr}/\text{Fe}-\text{Cr}/\text{Cr}/\text{Fe}-\text{Cr}/\text{Cr}$. In conclusion, perspectives for further development of the method are discussed.

2. Determination of selective atomic canonical functions in X-ray reflectivity of layered systems

2.1. Mathematical formalism

Let us define the depth profile of element i for a multilayered sample. The atomic density of element i at a certain depth z is written as

$$n_i(z) = \frac{N_i(z)}{V}. \quad (2.1)$$

Here, V is a volume (for example, unit cell volume), $N_i(z)$ is the number of atoms of type i at the depth z in this volume. We denote the atomic density n_{oi} for a pure layer of the element i . Then we can introduce the function

$$p_i(z) = n_i(z)/n_{oi}, \quad (2.2)$$

where $p_i(z)$ is the selective atomic canonical function [3].

We consider that the depth profile $p_i(z)$ of a multilayered sample has the following asymptotic behavior:

$$\begin{cases} p_i(z) = 0 & \text{at } z < 0 \\ 0 \leq p_i(z) \leq 1 & \text{at } 0 \leq z \leq L \\ p_i(z) = 0 & \text{at } z > L \end{cases}, \quad (2.3)$$

where $z = 0$ is a surface boundary and L is a sputtered film thickness.

Let us consider a multilayered sample consisting of alternating layers of elements, for example, A and B . The properties of this function are the following:

- $p_A(z) = 1$ in the layer A without mixing of atoms;
- $p_A(z) = 0$ in the layer B ;
- At the interface, $p_A(z)$ increases (decreases), *i.e.*, fulfill condition C:

$$0 \leq p_A(z) \leq 1.$$

The nature of increase (decrease) can be various; it depends on the symmetry, smoothness, and shape of the interface.

The function $p_A(z)$ is called the depth profile of the element A in the sample.

Recently, a method for determination of the depth profile $p_i(z)$ of element i by XRR data for low-contrast multilayered heterosystems was proposed [2]. Consideration is carried out in terms of the canonical atomic distribution functions [3]. It is a key idea of the approach. The function $p_i(z)$ means the probability to find the element i at a depth z from the surface of the sample (see in details [2]). It does not require *a priori* structure information such as the positions and widths of the interfaces, as well as, their lineshapes. The method is confirmed experimentally [4].

An integral equation is derived in the framework of the first Born approximation. It describes the relationship of the reflection coefficient $R(s)$ for a multilayered sample. The equation is written as

$$R(s) = \frac{4\pi r_e}{s^2} \sum_{i=1}^n n_{oi} f_i(s, E) \int_0^\infty \left(\frac{dp_i(z)}{dz} \right) \exp(isz) dz, \quad (2.4)$$

where s is the component z of the scattering vector, n is the number of types of atoms in the sample, n_{oi} is the density of i element at the depth of pure layer, r_e is the classical electron radius, the X-ray form factor $f_i(s, E)$ is a complex function, $f_i(s, E) = f_i^0(s) + f_i'(E) + if_i''(E)$, with the atomic form factor $f_i^0(s)$, the dispersion $f_i'(E)$, and absorption

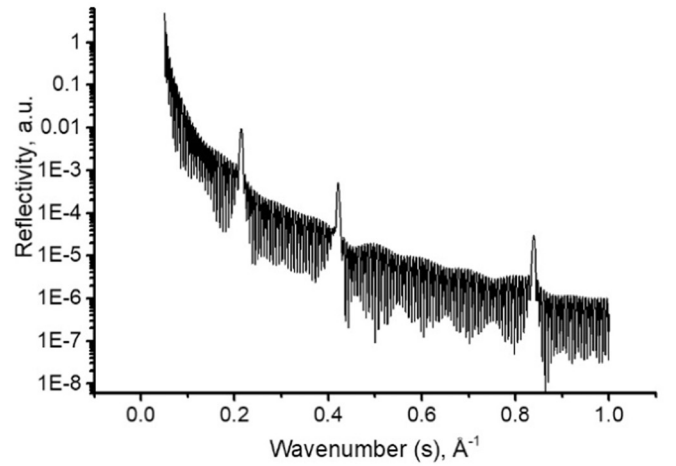
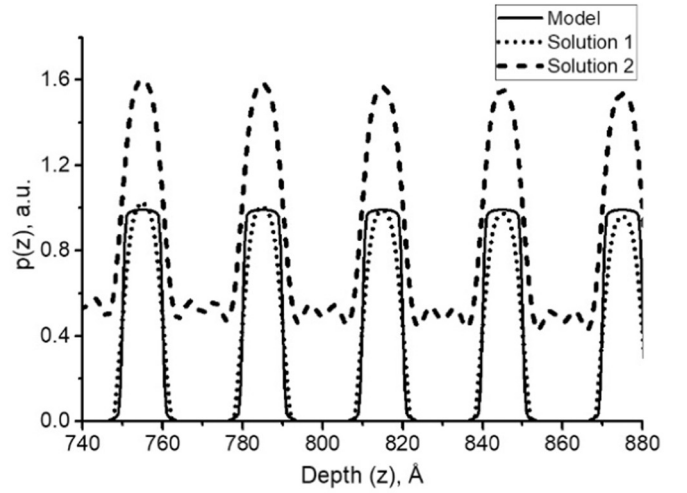


Fig. 1. $\text{Al}_2\text{O}_3/\text{Cr}(70 \text{ \AA})/[\text{Fe}(20 \text{ \AA})/\text{Cr}(10 \text{ \AA})]_{30}/\text{Cr}(20 \text{ \AA})$ film with periodic Fe layers: model curve $p_C(z)$, the solution 1 at fulfilling the requirement C: $0 \leq p_C(z) \leq 1$ (the residual norm RN is 0.11%), and the solution 2 without the requirement C (RN is 0.22%), top; input reflectivity data, bottom.

corrections $f_i''(E)$ depending on the radiation energy E , which is regarded as a parameter. The summation is performed over the types of atoms in the sample.

For the concentration profiles of elements A and B of the neighboring layers, between which the transition occurs at the interface, the condition is

$$p_A(z) + p_B(z) = 1 \text{ or } p'_A(z) + p'_B(z) = 0. \quad (2.5)$$

In the case of two layers, we rewrite (2.4) in the form.

$$R(s) = \frac{4\pi r_e}{s^2} (n_{0A} f_A(s, E) - n_{0B} f_B(s, E)) \int_0^\infty \frac{dp_A(z)}{dz} \exp(isz) dz. \quad (2.6)$$

Solving the inverse problem (2.6), we obtain the derivative of depth profile $p'_i(z)$. After subsequent integration, the solution $p_i(z)$ may differ from the true one up to a constant. In that case, the condition C does not fulfill. The corresponding graphics show in Figs. 1–3 as solution 2.

To eliminate this drawback, we transform Eq. (2.6). Taking the integral by parts and taking into account the asymptotic properties of the function $p_i(z)$ (2.3), we have

$$R(s) = -i \frac{4\pi n_0 r_e}{s} (n_{0A} f_A(s, E) - n_{0B} f_B(s, E)) \int_0^L \exp(isz) p'_A(z) dz, \quad (2.7)$$

where $f_i(s, E) = f_i^0(s) + f_i'(E) + if_i''(E) \equiv f_i^*(s, E) + if_i''(E)$.

Let us write the real and imaginary parts of the reflection coefficient

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