Deconvolution method for obtaining directly the original in-depth distribution of composition from measured sputter depth profile

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Abstract

A new method is proposed to determine the original in-depth distribution of composition directly by the deconvolution of measured depth profiling data using the MRI (Mixing-Roughness-Information) depth resolution function. To overcome the ill-conditioned problem that is often experienced in the deconvolution procedure, the regularization method and the TV-Tikhonov algorithm are employed. The method is applied for the case that the original depth-concentration distribution is either a smooth function or a square wave function and the corresponding arithmetic mean errors are determined accordingly. The noise effects from “measured” data on the deconvoluted profile are also quantitatively evaluated.

1. Introduction

Sputter depth profiling is a technique that combines ion bombardment and surface analysis. The primary aim of this technique is to determine the depth-concentration distribution of elemental composition in thin films. The study of quantification of sputter depth profile started in the 1970ies by introducing the concept of depth resolution, which marks the distortion degree of the measured depth profile as compared to the original layered structure. In 1994, Hofmann put forward an advanced model that is considered the three basic effects in any depth profiling technique: atomic Mixing due to ion sputtering, Roughness due to sample characteristics, and Information depth due to the analysis method [1]. Therefore it is called the MRI (Mixing-Roughness-Information) model, which is widely used to quantify measured AES, XPS and SIMS depth profiling data [2,3]. However, it is conventional that, using the MRI model, the original layered structure is determined indirectly from measured depth profiling data by the convolution method described in Section 2.1.

The aim of this paper is to demonstrate that the original layered structure can be obtained directly from measured depth profiling data using deconvolution method in combination with the MRI model. The two binary systems that the original layered structures are presented respectively by a smooth function and a square wave function are chosen for the deconvolution of “measured” depth profiling data with different noise levels. The current deconvolution method is also compared with the one presented in literature. Finally, as an example, the measured AES depth profiling data of Ni/Cr multilayer are deconvoluted and the original Ni/Cr multilayered structure is obtained accordingly.

2. Quantitative analysis of measured depth profile

2.1. Convolution method

The convolution method was first proposed for quantification of measured sputter depth profile by Ho and Lewis [4]. They proposed that the measured and normalized signal intensity $I(z)/I_0$ upon sputter depth profiling can be expressed by the convolution of the real in-depth distribution of composition $X(z)$ with a depth resolution function $g(z)$ as shown by Eq. (2.1)

$$\frac{I(z)}{I_0} = \int_{-\infty}^{+\infty} X(z')g(z-z')dz'$$

(2.1)

where $z$ is the sputtered depth, $z'$ is integral variable. The depth resolution function $g(z')$ contains all the distortion factors induced by sputtering process. The depth resolution function needs to meet the requirement of normalization, i.e.

$$\int_{-\infty}^{+\infty} g(z')dz' = 1$$

(2.2)
Several approaches for the depth resolution function were elaborated during the past 30 years [5–10], in which the MRI model is regarded as one of the most popular models for the quantification of sputter depth profiling data [1,9–11]. If the depth resolution function is known, there are two methods for the reconstruction of measured depth profiling data, i.e., the convolution and deconvolution methods. Generally speaking, the deconvolution of Eq. (2.1) is so complicated that may yield a large scatter enhanced by insufficient data precision (low signal to noise ratio) [2,11–14]. Therefore, it is customary to take the convolution method for obtaining the original layer structure indirectly by assuming a “real” depth-composition distribution $X(z)$ and comparing the calculated signal intensity $I(z)/I_0$ according to Eq. (2.1) with the measured one. This process is repeated until an optimum fit to the measured profile is obtained [15–17].

### 2.2. MRI model

The depth resolution function in the MRI model consists of three functions: $g_w$, $g_0$, and $g_t$, which are characterized by three parameters: atomic mixing length $w$, roughness parameter $\sigma$ and information depth $\lambda$, respectively, and are given by Ref. [1].

\[
g_w = \frac{1}{w} \exp[-(z - z_0)/w]
\]

\[
g_0 = \frac{1}{\sqrt{2\pi} \sigma} \exp \left[-\frac{(z - z_0)^2}{2\sigma^2}\right]
\]

\[
g_t = \frac{1}{\lambda} \exp[-(z_0 - z)/\lambda]
\]

These three MRI parameters have the respective physical meaning and can be predicted or measured independently. Usually, the atomic mixing length is of the order of the mean value of ion projected range, which can be obtained by the SRIM code [18] or measurement [3]. The (surface) roughness can be easily measured by atomic force microscopy (AFM). The information depth is based on the effective attenuation length of the detected signal [19].

### 3. Deconvolution method

#### 3.1. Inverse problem

The deconvolution method applied to Eq. (2.1) is referred to the so-called inverse problem, which is so sensitive to the noise of measured depth profiling data. The numerical solution of the inverse problems is usually solved by a linear algebraic system as:

\[
AX = b
\]  

(3.1)

Here, $A$ is an $n \times n$ matrix with respect to the depth resolution function $g(z)$, $X$ and $b$ are the $n$-vectors that represent the in-depth distribution of composition and the signal intensity $I(z)/I_0$ respectively.

Due to the ill-posed nature of the inverse problem, the numerical solution of Eq. (3.1) is extremely sensitive to perturbations caused by observation and rounding errors [20–25]. Therefore, the regularization is needed for obtaining a reasonable approximated piecewise-continuous solutions, while the total variation (TV) regularization method is often applied [26–29].

#### 3.2. TV regularization method

In the TV regularization model, the solution for Eq. (3.1) is transformed to minimize the so-called TV function:

\[
T_{\alpha}(X) = \frac{1}{2} b^T (AX - b) + \alpha TV(X)
\]  

(3.2)

where $\alpha > 0$ is called the regularization parameter, $TV(X)$ is the total variation (TV) function defined as

\[
TV(X) = \sum_{i=1}^{n} |x_i - x_{i-1}|
\]  

(3.3)

Noted that Eq. (3.3) can be regarded as an approximation of

\[
\int_{I_0}^{\infty} \rho dt, \text{ where is the interval of integration.}
\]

Since the term $|x_i - x_{i-1}|$ in $TV(X)$ is non-smooth, it is approximated by $\sqrt{|x_i - x_{i-1}|^2 + \beta^2}$, where $\beta$ is a small positive parameter. Let

\[
D = \begin{pmatrix} 1 & 1 & \cdots & 1 \\ \vdots & \vdots & \ddots & \vdots \\ 1 & 1 & \cdots & 1 \end{pmatrix}_{(n-1) \times n}
\]  

(3.4)

and $D_i$ be the $i$-th row of $D$. Then, $TV(X)$ can be represented as

\[
J_\rho(X) = \frac{1}{2} \sum_{i=1}^{n} \psi((D_i X)^2)
\]  

(3.5)

where $\psi(z) = 2\sqrt{z^2 + \beta^2}$. It follows that the minimization of Eq. (3.2) turns to that of the following function:

\[
T(X) = \frac{1}{2} b^T (AX - b) + \alpha J_\rho(X)
\]  

(3.6)

To get the minimum of Eq. (3.6) by the Newton method, the gradient and the Hessian of $T(X)$ are needed, which can be expressed by ([26])

\[
\text{Grad}T(X) = L(X) X
\]  

(3.7)

with

\[
L(X) = \Delta z D^T \text{diag}(\psi'(X)) D
\]  

(3.8)

and

\[
\text{Hass}T(X) = L(X) X + L'(X) X
\]  

(3.9)

By Eqs. (3.7) and (3.8), the gradient of $T(X)$ is given by

\[
\text{Grad}T(X) = A^T (AX - b) + \alpha L(X) X
\]  

(3.10)

By Eqs. (3.9)–(3.11), the Hessian of $T(X)$ is given by

\[
\text{Hass}T(X) = A^T A + \alpha L(X) X + \alpha L'(X) X
\]  

(3.12)

Here is the TV-Tikhonov algorithm:

1. Assign $j = 0$;
2. Choose an initial iterative solution $X^{(0)}$ (say, the zero vector);
3. Calculate the gradient: $g_l = A^T (AX - b) + \alpha L(X) X$;
4. Calculate the Hessian: $H = A^T A + \alpha L(X) X + \alpha L'(X) X$;
5. Calculate the down direction: $S_j = -H^{-1} g_l$;
6. Search the optimal step length: $\xi_j = \text{argmin}_{0 < \xi_j <1} T(X^{(j+1)}) + \alpha S_j$;
7. Calculate the $(j+1)$-th iterative solution: $X^{(j+1)} = X^{(j)} + \xi_j S_j$;
8. Update the iteration number: $j = j + 1$;
9. If the solution meets the requirements, the iteration stops.

Otherwise the process goes to step (3) for further calculation.

### 4. Results and discussion

The deconvoluted simulation is performed based on the TV-Tikhonov algorithm in combination with the MRI model. The original layer structure for the simulation of the MRI depth profile is taken as a square wave function or a smooth function. A sandwich layer structure of $A(40\text{nm})/B(30\text{nm})/A(30\text{nm})$ is taken as the square wave function as shown as dashed line in Fig. 1a. The smooth function is generated by the convolution of a Gaussian function with the square wave function as shown as dashed line in Fig. 1b. The detailed steps are as following:

1. Taking the original structure as the square wave function and the
smooth function as dashed lines in Fig. 1;

(2) Calculating the depth profile (dash-dotted line in Fig. 1) by the MRI model with the parameters of \( w = \sigma = \lambda = 5 \) nm (dash-dotted line in Fig. 1. The three MRI parameters are intentionally taken as large as the reality ones in order to verify the validity of the current method);

(3) Deconvoluting the calculated MRI depth profile by the TV-Tikhonov algorithm with \( \alpha = 0.01 \) and \( \beta = 0.001 \) (solid line in Fig. 1). Noted that the parameters \( \alpha \) and \( \beta \) of TV-Tikhonov algorithm are obtained by the L-curve method [30].

Comparing the deconvolution results (solid lines in Fig. 1) with the original bilayer structure (dashed lines in Fig. 1), the arithmetic mean errors are 1.49% and 0.36%, respectively, for the square wave function (Fig. 1a) and the smooth function. These comparisons with the deviation less than 2% indicate that the regularization method based on the TV-Tikhonov algorithm is suitable for solving the inverse problem.

It is well known that the deconvolution procedure is often ill-posed and it is challenging to obtain a reasonable solution due to the noise and/or insufficient number of measured data points. In the following, the noise imposed on the simulated MRI depth profile (as measured depth profiling data points) is generated in two ways: (a) Half-Random (HR) noise: taking randomly half of the data points of the simulated depth profile and multiplying their values with a constant noise level (as a controlling parameter) and \( \sin(x) \). The insufficiency of “measured” data points (i.e. the simulated MRI depth profile) is considered by taking only one data point every fixed intervals, e.g. the notation of gap(n) means taking only one data point every \( n \) intervals of the \( n + 1 \) data points and casting out the other data points.

The simulation results for the half-random noise with different noise levels are shown as solid lines with different symbols for the square wave function in Fig. 2a and the smooth function in Fig. 2b, as well as the convoluted MRI depth profile (solid line without symbol) with the noise level of 20%. Fig. 2 shows that, when the noise level is 5%, the deconvoluted layer structure is almost the same as the one without noise. If the noise level is 10%, the arithmetic mean errors are 4.88% and 3.87%, respectively, for the square wave and smooth functions, and become 7.09% and 7.19% if the noise level increases to 20%. Clearly, the deviation is smaller for the smooth function than that for the square wave function.

The simulation results for the sine-type noise with different noise levels are shown as solid lines with different symbols for the square wave function in Fig. 3a and the smooth function in Fig. 3b, as well as the convoluted MRI depth profile (solid line without symbol) with the noise level of 20%. Fig. 3a shows that, when the noise level is less than 10%, the deconvoluted layer structure for the square wave function...
The simulation results for the insufficient number of “measured” data points with different gap values are shown as solid lines with different symbols for the square wave function in Fig. 4a and for the smooth function in Fig. 4b, respectively, as well as the convoluted MRI depth profile (solid line without symbol) with the gap(10) (i.e. taking only one point every 10-point intervals). Fig. 4 shows that the deconvoluted layer structure is almost the same as the original one for the gap(2) and gap(5). When taking the gap(10), the arithmetic mean errors are 6.75% and 2.95%, respectively, for the square wave and the smooth functions. It is concluded that the TV-Tikhonov algorithm to deal with the inverse problem by using the deconvolution method has good stability for the different-type noises and the insufficient number of “measured” data points.

In the literature, an iterative constrained algorithm had been proposed for the deconvolution of measured SIMS depth profiling data [31,32]. The current study with the TV-Tikhonov method is compared with the iterative deconvolution method. The deconvolution results of the MRI depth profile (w = σ = λ = 5 nm, i.e. the dash-dotted line in Fig. 1) without noise are shown as dotted line for the iterative method.
deconvolution method and as dash-dotted line for the TV-Tikhonov method in Fig. 6. As compared to the original structure, the arithmetic mean errors are 9.48% and 1.49%, respectively, for the iterative deconvolution method and the TV-Tikhonov method. When taking the Half-Random noise of HR(5%), the deconvolution results of the MRI depth profile (w = σ = λ = 5 nm, i.e. the dash-dotted line in Fig. 1) are shown as dotted line for the iterative deconvolution method and as dash-dotted line for the TV-Tikhonov method in Fig. 6. As compared to the original structure, the arithmetic mean errors are 16.75% and 3.17%, respectively, for the iterative deconvolution method and the TV-Tikhonov method. When taking the Half-Random noise level more than HR(10%), the iterative deconvolution method cannot give any reasonable result. Clearly, the TV-Tikhonov method is much more powerful than the iterative method for the deconvolution of measured depth profiling data with different-type noises and the insufficient number of “measured” data points.

5. Conclusion

The original in-depth distribution of composition can be obtained directly by the deconvolution of measured depth profiling data using the TV-Tikhonov algorithm in combination with the MRI depth resolution function. By deconvoluting the simulated MRI depth profile with different types of noise and an insufficient number of depth profiling data points, it shows that the TV-Tikhonov method is more stable and powerful than the iterative method used in the literature. A rectangular layer structure can be reproduced by the deconvolution of “measured” depth profiling data.

Acknowledgement

The authors gratefully thank for the discussion with Dr. X.M. Fang, School of Mathematics andStatistic, Zhaoting University, Zhaoting, China and Prof. S. Hofmann, Max Planck Institute for Metals Research, Stuttgart, Germany. The work is supported by Science and Technology Planning Project of Guangdong Province, China (NO. 2017A010103021).

References


Fig. 6. The comparison of the TV-Tikhonov method with the iterative method for the deconvolution with the HR(5%) noise level.

Vacuum 166 (2019) 196–200