Scanning Microscopy Vol. 12, No. 2, 1998 (Pages 301-308)
Scanning Microscopy International, Chicago (AMF O'Hare), IL 60666 USA
0891-7035/98$5.00+.25

SIGNAL FORMATION, SIMULATION AND INVERSE PROBLEM IN SCANNING FLUORESCENT X-RAY MICROSCOPY USING FOCUSED BEAMS FOR ANALYSIS OF THE SURFACE RELIEF

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(Received for publication April 14, 1996 and in revised form January 19, 1997)

Abstract

The problem of quantitative determination of the surface microrelief from an X-ray fluorescent signal is investigated in this paper. The integro-differential equation, which connects the relief of the surface with the registered signal, is obtained. The inverse problem (the reconstruction of the surface form by using the X-ray fluorescent signal) is solved. The results of the relief reconstruction for both the pure signal and for the signal with the noise are obtained. The introduced concept of spatial resolution allows one to conceive of a minimal size of the region, where the method is sensitive to the relief changes in nanometer scale. It is shown that if the angle, at which the detector is positioned tends to 0°, direct measurements of the derivative of the function describing the relief become possible.

The method proposed does not require standard samples and belongs to the nondestructive control method.

Key Words: X-ray fluorescent scanning microscopy, surface microrelief, reconstruction of surface relief, spatial resolution.

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Introduction

X-ray fluorescent scanning microscopy [2] based on employing focused X-ray beams is a comparatively new direction in microdiagnostics. Its appearance is due to the creation of powerful radiation sources of nanometer scale, on one hand, and successful development of methods for fabrication X-ray optic elements, on the other hand [3, 8]. The range of diagnostic problems solved by X-ray fluorescent scanning microscopy expands constantly, owing to a continuous perfection of equipment and development of mathematical models and techniques for signal processing which allows covering a wider range of objects and fit different configurations of microscopes (the reciprocal location of the source, object and detector, in particular). For example, a model of fluorescent signal formation from a planar layered specimen has been developed [1]. The scanning procedure can be realized either by moving a sample (a stage) [5] or the beam [6]. X-ray microprobes with a submicron beam diameter have been used to obtain high spatial resolution maps of the element composition [3, 8]. Progress in the creation of X-ray optics elements results in the size of the focal spot in the micron and submicron (up to 55 nm [4]) [3, 5, 8] range. In the micron and submicron range, where the beam spot becomes comparable or less than the fundamental absorption lengths, the presence of a surface relief influences the X-ray fluorescent signal like the presence of the balk non-homogeneity does. Therefore, one of the classical microscopy problems is the observation and quantitative description (at the microlevel) of the surface of an object under investigation. Optical, electron and ion microscopes are employed to solve this problem. However, none of the existing techniques gives a comprehensive solution. Therefore, a search for new approaches to this problem seems urgent.

The problem of quantitative determination of the surface micro-relief from an X-ray fluorescent signal is investigated in this work. A mathematical model (signal equation) is developed and the direct problem (the signal simulation from the known signal) and inverse problem (the reconstruction of the surface form) are solved.

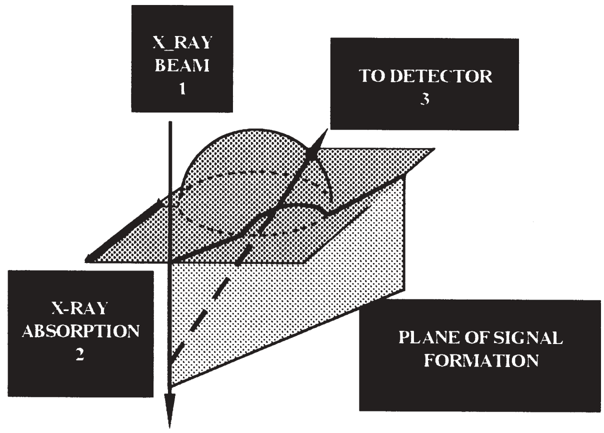


Figure 1. The scheme of the fluorescent signal.

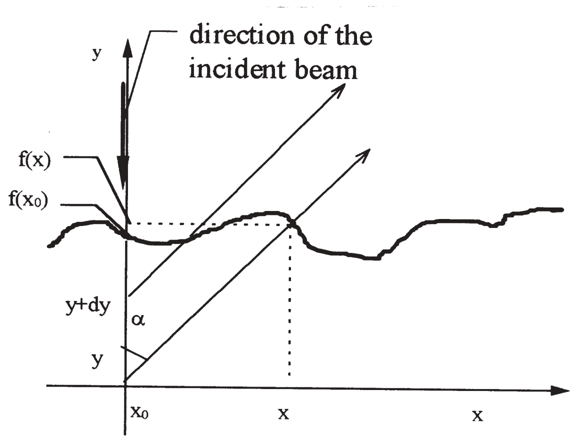


Figure 2. The plan of the signal.

Signal Equation

Figures 1 and 2 demonstrate the scheme of fluorescent signal formation in an X-ray microscope. The reciprocal locations of the source (1), investigated object (2), and detector (3) are shown in the Figure 1. The X ray absorption region is much smaller than the distance from the source to the object and from the object to the detector. Therefore, we can assume that the angle of the direction "object-source" with the direction "object-detector" α is equal for all points of the absorption region. Because the signal is formed in one plane, a three-dimensional (3D)-problem is reduced to a two-dimensional (2D)-problem (Fig. 1). We may introduce the Cartesian coordinate system for the plane 123, so that the direction of OY axis coincide with the direction 21. The origin of the coordinates is of no significance. Let f(x) be the function describing the surface relief in the plane 123. In what follows, we will assume that f(x) is differentiable and satisfies the condition:

$$\sup(f'(x)) \leq ctg\alpha \tag{1}$$

(this condition ensures that the registered part of fluorescent radiation intersects the object surface only once). Let x_0 denote the coordinate of the initial ray intersection with the surface (Fig. 2). The magnitude of the fluorescent radiation generated on the intercept ($y+dy,y$) is proportional to the absorbed X-ray radiation

$$v\exp(-v(f(x_0) - y))dy \tag{2}$$

where n_f is the yield of the fluorescence and v is the attenuation coefficient of the initial X-ray radiation for an object material.

Here, we consider the part of fluorescent radiation generated on the intercept ($y+dy,y$) which falls onto the detector and consequently, contributes to the signal (because the generated radiation is isotropic, the magnitude of the produced part is proportional to the quantity in relation (2) above).

Let x denote the coordinate of the intersection point of a fluorescent ray with the surface. Then, the distance, the fluorescent ray passes in the sample material, is equal to

$$(f(x) - y)/\cos\alpha \tag{3}$$

where x satisfies the equation

$$x - x_0 = (f(x) - y)tg\alpha \tag{4}$$

Thus, the contribution of fluorescent radiation generated on the intercept ($y+dy,y$) to the signal is equal to

$$n_f\, v \exp(-v(f(x_0) - y)) \exp\left(-\mu \frac{f(x) - y}{\cos\alpha}\right) dy \tag{5}$$

where μ is the attenuation coefficient of the fluorescent radiation. Integrating with respect to y from infinity to $f(x)$, we obtain

$$S(x_0) = n_f v \int_{-\infty}^{f(x_0)} \exp(-v(f(x_0) - y)) \times \exp\left(-\frac{\mu}{\cos\alpha}(f(x) - y)\right) dy \tag{6}$$

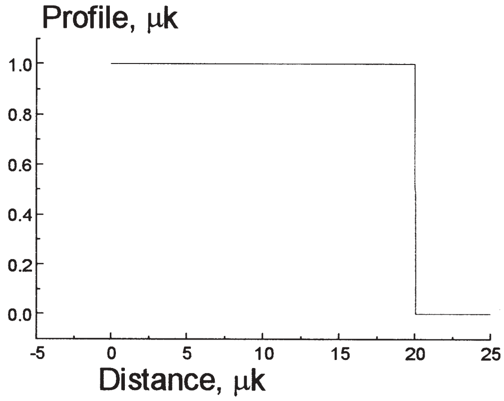


Figure 3. The smooth surface with the single step.

where $S(x_0)$ is the signal magnitude. Changing the terms in eq. (6) according to

$$y = f(x) - \{(x - x_0)/(tg\alpha)\} \tag{7}$$

we obtain the following equation for the signal at normal incidence of the X-ray beam (the surface is set perpendicularly to the beam)

$$S(x_0) = n_f \nu \int_{x_0}^{\infty} (ctg\alpha - f'(x)) \times \exp\left(-\nu(f(x_0) - f(x)) - \frac{(\nu\cos\alpha + \mu)}{\sin\alpha}(x - x_0)\right) dx \tag{8}$$

For the case of oblique incidence of the beam, the value of the signal can be calculated by the formula

$$S(x_0) = \left(\int_{X_0}^{\infty} \exp(A(B(x - x_0) - C(f(x) - f(x_0)))) \times \left(\frac{1 - f'(x)tg\alpha}{tg\alpha + tg\beta}\right) dx\right) n_f \nu \tag{9}$$

where

$$A = -\frac{1}{tg\alpha + tg\beta}; \quad B = \frac{\nu}{\cos\alpha} + \frac{\mu}{\cos\beta};$$

$$C = \frac{\nu tg\alpha}{\cos\beta} - \frac{\mu tg\beta}{\cos\alpha};$$

and β is the angle of the direction "source-object" with the normal. Since the derivation of eq. (9) is similar to the derivation of eq. (8), eq. (9) is written without derivation.

Equation (8) solves a direct problem, and gives the value of a signal as the function of the surface relief ($f(x)$, geometry (the angle α), and material constants (absorption lengths μ^{-1} and ν^{-1}). It can be used for the signal simulation in diagnosis.

The Inverse Problem

Another problem of diagnosis is reconstruction, using the measured signal. The problem is known as an inverse problem. From our point of view, the most interesting aim of the inverse problem is the reconstruction of the surface relief. This can be done only numerically, but in our case, the reconstruction problem can be solved analytically.

To solve the inverse problem, we differentiate signal eq. (8) with respect to x_0.

$$S'(x_0) = \left(\nu f'(x_0) + \frac{\nu cos\alpha + \mu}{\sin\alpha}\right) S(x_0) - n_f \nu (ctg\alpha - f'(x_0)) \tag{10}$$

Modulation techniques allow one to measure the signal derivative by, for example, oscillating a sample along the X-axis, so the direct solution of the reconstruction problem can be given in terms of the measured functions (signal and its derivative)

$$f'(x_0) = \frac{S'(x_0) - (\frac{\nu cos\alpha + \mu}{\sin\alpha})S(x_0) + n_f \nu ctg\alpha}{\nu(n_f - S(x_0))} \tag{10a}$$

After the integration of eq. (10), we obtain another presentation of the inverse problem solution, eq. (11), which can be used if the signal derivative is not available. This expression allows one to reconstruct the value of the profile function in each scan point, using the measured signal S.

$$f(x) = \frac{1}{\nu}\left(\frac{(\nu cos\alpha + \mu)X}{\sin\alpha} - \int_0^x \frac{n_f \mu dt}{\sin\alpha(S(t) - n_f)} + \ln\left|\frac{S(x) - n_f}{S(0) - n_f}\right|\right) \tag{11}$$

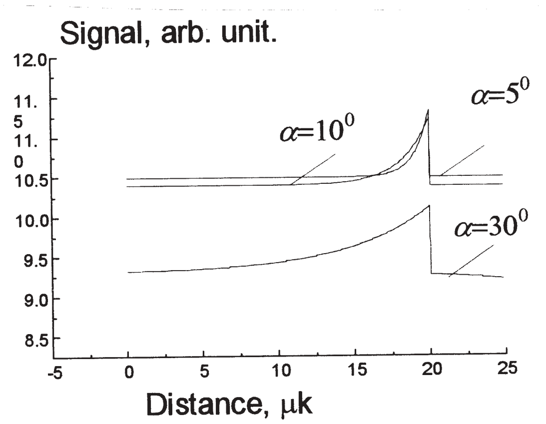


Figure 4. Signal diagrams at the various values of the angle (5°, 10°, 30°) for the profile with the single step.

Spatial Resolution

The signal equation allows the investigation of the image properties obtained in the microscope such as visibility (contrast) and resolution, depending on the conditions of the experiment. The most important of these properties is the value of the angle. Suppose that we record a signal from a smooth surface with the single step (Fig. 3). For the inlet point x_0 positioned far from the step, the equation of the signal can be rewritten to give

$$S(x_0) = n_f \nu \int_{x_0}^{\infty} \exp\left(-\frac{\nu \cos\alpha + \mu}{\sin\alpha}(x - x_0) \right) ctg\alpha dx \tag{12}$$

The same expression is valid when x_0 is on the right form the step. Equation (12) implies that signal S "feels" the step when the distance between x_0 and the step is of the order of magnitude of the exponent parameter

$$r = \frac{\sin\alpha}{\nu \cos\alpha + \mu} \tag{13}$$

Therefore, r is the natural characteristic of the spatial resolution. The spatial resolution is influenced by: (1) the angle between the normal direction and the detector; (20 the attenuation coefficient of incident X-ray radiation; and (3) the absorption coefficient of the fluorescent quanta. We can obtain the spatial resolution by decreasing the angle between the normal direction (Fig. 4) and the detector.

A Limiting Case

Consider the case where the angle between the direction of the incident beam and the direction towards the detector tends to 0°. Then, x tends to x_0, and the following expression holds

$$f(x_0) - f(x) \approx f'(x_0)\ (x_0 - x) \tag{14}$$

Factoring

$$S(x_0) = n_f \nu \,(ctg\alpha - f'(x_0)) \times \int_{x_0}^{\infty} \exp\left(-\nu f'(x_0) + \frac{\nu \cos\alpha + \mu}{\sin\alpha}(x_0 - x) \right) dx \tag{15}$$

with respect to $sin\alpha$ and retaining the first order, we obtain

$$S(x_0) = \frac{n_f \nu (1 - \sin\alpha f'(x_0))}{\nu + \mu - \nu \sin\alpha f'(x_0)} \tag{16}$$

These expressions show that if $\alpha = 0°$, the fluorescent signal is non-informative (does not dependent on $f(x)$), and, the reconstruction of relief is impossible.

However, direct measurements of $f'(x)$ become possible at small (although differing from 0) values of the angle.

A Special Method of Measurement

Differentiating eq. (16) with respect to $sin\alpha$, we obtain

$$\frac{dS(x_0)}{d \sin\alpha} = -\frac{n_f \nu \mu f'(x_0)}{(\nu + \mu)^2} \tag{17}$$

at small α.

From this expression, the derivative $f'(x)$ is found directly. The differentiation with respect to $sin\alpha$ can be implemented as the oscillation of the detector in the horizontal direction. Equation (17) can be considered as one more representation of the inverse problem solution for the limiting case of a small angle with the difference that for eq. (10), the derivative in eq. (10) is a derivative on the beam position x_0 (and the sample should oscillate to measure the derivative) where the detector should be moved to measure the derivative for eq. (17).

Estimation of the Sensitivity

We express the estimate of the intensity of the X ray fluorescent radiation hitting the detector as

$$I = Pn_f \frac{\nu}{\nu + \mu} \frac{\Omega}{4\pi} \tag{18}$$

where P is the flux of the radiation incident on the sample, Ω is the solid angle magnitude determined by the input window size of the detector and its position. The formula proposed implies that the attenuation of the intensity due to the absorption in the sample is of the order of unity. This value is negligible in comparison with the main reduction 3-4 orders, which results from a small value of solid angle Ω. Therefore, for the measurement error to be of the order of 3 percent, not less than 10^3 photons should be detected; this gives 10^6-10^7 as the total number of photons in the beam per pixel. High intensity synchrotron radiation is now available with a flux of 10^8-10^9 photons/s in the spot [7] and this makes it possible to realize the described analysis, promising low noise level in the range 0.1%.

Simulation of the Signal and Reconstruction of the Relief

The results of X-ray fluorescent signal simulation and the results of surface relief reconstruction are presented in Figures 3 to 12. We will concentrate on Figure 5 because the results illustrated in the Figures 3 and 4 were discussed previously. Three signals, obtained at three values of the angle α (5°, 15°, 30°, respectively), are shown in Figure 5. It can be readily seen that the spatial resolution increases with a decrease of the angle α, the diffusion of individual elements disappears, whereas the visibility (the inhomogeneity-induced signal change) decreases.

Therefore, a decrease in the angle is efficient when a detector with a low noise level is used. The signal contrast increases with an increase of the angle α (Fig. 5), and at small values of the angle, the curve of the signal tends to the derivative of the function describing the surface relief. Tending the signal contrast to zero at small angles and tending the signal shape to the profile derivative is in accordance with the predictions of the model for the limiting case of small angles {eq. (16)} where an alternating part of the signal is proportional to

$$n_f \frac{\nu\mu}{(\nu + \mu)^2} \sin \alpha f'(x_0) \tag{19}$$

An example of profile reconstruction is shown in Figure

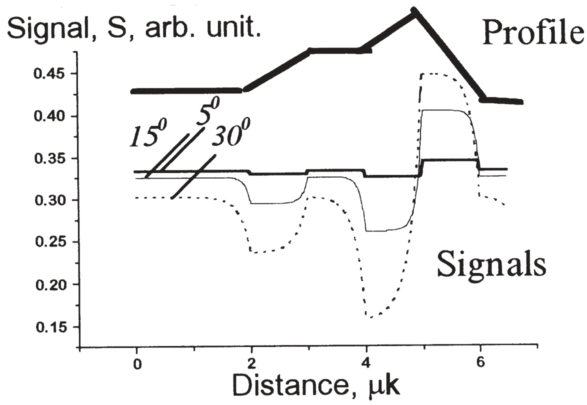


Figure 5. Signal diagrams at various values of the angle (5°, 15°, 30°).

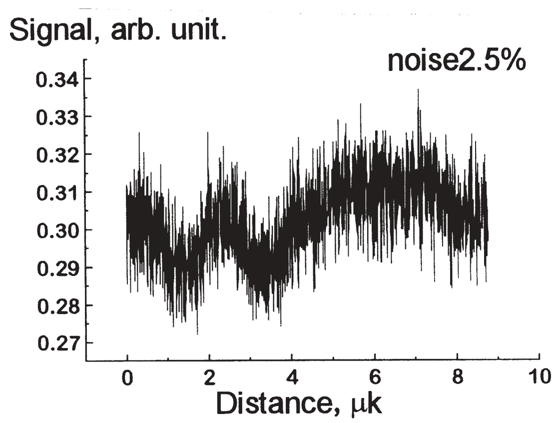


Figure 6. Signal with added Gaussian noise.

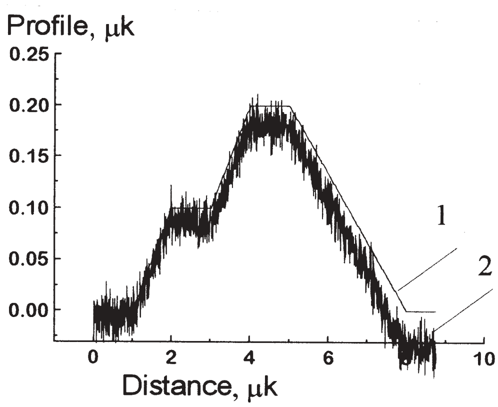
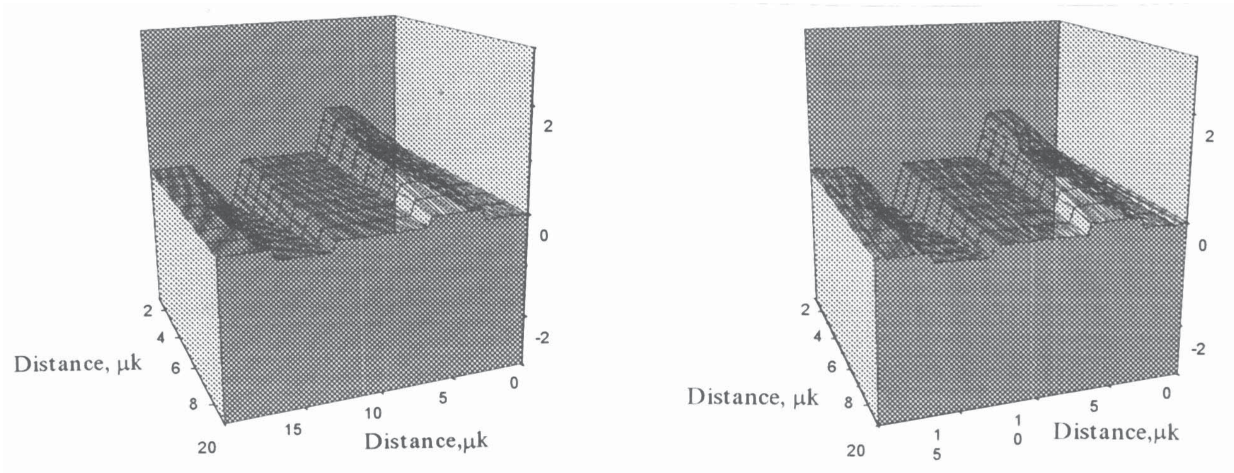
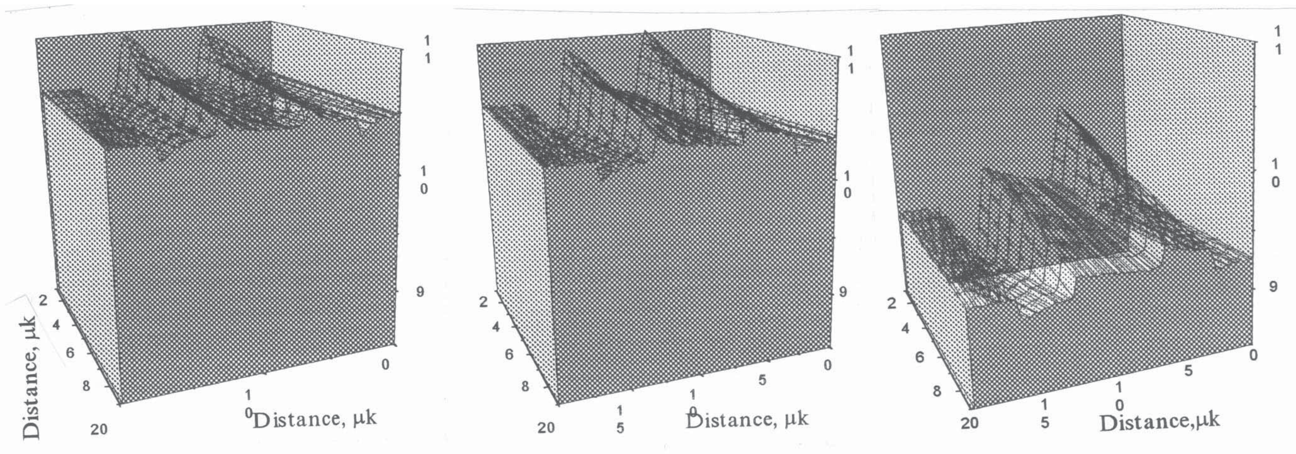


Figure 7. The initial (**1**) and reconstructed (**2**) profiles.



Figures 8 (at left) and 9 (at right). Initial (**Fig. 8**) and reconstructed (**Fig. 9**) profiles.



Figures 10 (at left), 11 (center) and 12 (at right). Signals at $\alpha = 5°$ (**Fig. 10**), 10° (**Fig. 11**) and 30° (**Fig. 12**).

7. The X-ray fluorescent signal was calculated by eq. (8) for the test profile (Fig. 7, curve 1). Gaussian noise was added to the value of the signal obtained (Fig. 6). In order to reconstruct the surface profile (Fig. 7, curve 2), eq. (11) was used. It is easily seen that the solution of the inverse problem is stable with respect to the noise.

Three-dimensional-images of the surface relief, the reconstructed profile and the noise signals for different angles are presented in Figures 8 to 12. The fundamental constants (absorption lengths μ^{-1} and ν^{-1}) have been calculated for silicon and the wavelength of the incident X ray beam corresponds to $CuK_{\alpha 1}$ radiation (1.54 Å). The obtained 3D-images correspond to the scan in two directions (along X and Z axes). For each point on the Z axis, the 2D-problems (direct and inverse) have been solved. The signal has been calculated at three values of the angle α: 5° (Fig. 10), 10° (Fig. 11), and 30° (Fig. 12). Although the absorption lengths are about 10 μm, the profile features 0.1-1 mk high can be reconstructed successfully. Thus, the fluorescent signal can be used for a quantitative relief characterization in the nanometer scale.

Summarizing, the results of the simulation allow one to evaluate the value of a signal and to choose an optimal position for the detector, depending on the relief magnitude and sample material.

Acknowledgements

Research supported in part by Russian Foundation of

Fundamental Research (Grant No. 96-02-19798).

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Discussion with Reviewers

M.L. Rivers: A major weakness is the lack of discussion of the effect of finite angular acceptance of a real detector. The authors claim {just after eq. (18)} that the fractional acceptance is about 3° to 5° in each direction. Many of their simulations are at detector angles of 5°. However, they assumed EXACTLY 5°, whereas in the real case it will be 5° plus or minus 2-3°. What is the effect of this finite solid angle on the resolution function for determining the surface relief?

Authors: We restricted the consideration by asymptotic case of infinitely small detector to concentrate on physics and derivation of the signal equation. The signal equation for finite angular acceptance can be easily obtained from the basic formulae by integration over a solid angle of the detector. Particularly, it is seen that in the first approximation, the finite acceptance $\Delta\alpha$ will contribute a linear term $\propto\Delta\alpha/\alpha$. As to simulation at 5°, the quantitative conclusion (spatial resolution increases with a decrease of the angle α, whereas the signal contrast increases with an increase of α) arrived at with this simulation is valid for the detector of finite size.

M.L. Rivers: There is no discussion of the assumption behind the proposed technique. One obvious unstated assumption is that there is a uniform concentration of the element whose fluorescence is being measured. If that concentration is heterogeneous, then it will appear as a false topography signal.

R. Gauvin: Real materials are non-homogeneous. So, what do you propose to do with your method when $C(x) = f(x,y,z)$?

Authors: Of course, there are specimens containing both spatial inhomogeneities and surface relief. One of the main motivations of the paper is to estimate a contribution of the relief in the fluorescent signal with hope to find ways to eliminate or minimize the contribution. The first step on this way is deriving the signal equation for spatially uniform specimen with surface relief. On the other hand, the formation of the signal from uniform specimen with the relief is a significant scientific problem by itself, solution of which could be a theoretical base for diagnostic and metrological applications of the focused X-ray beams. Signal equations for different situations with spatial non-homogeneities (e.g., a thin film of variable thickness on a substrate, or planar impurity distribution covered by a film) will be published elsewhere.

M.L. Rivers: There is no discussion of the potential advantages, if any, of this technique (which requires a synchrotron source, sophisticated microfocusing optics, and slow mechanical scanning) over the existing techniques (such as, secondary electron imaging) for determining surface relief.

Authors: A synchrotron source, sophisticated micro-focusing optics and slow scanning are obvious draw-backs of the techniques in application to relief investigation which could be improved or overcame in the future. But, there is an unbeatable advantage. Physics of the signal formation is uniquely simple and transparent as it is based on exponential attenuation along straight trajectories. The advantage becomes obvious after a comparison of derivation and investigation of the signal equation for specimen with relief that we have developed for scanning electron microscopy [9]. Here, the basic physical phenomenon is a random walk (of electron) with elastic scattering depending on currently decreasing electron energy. So, investigators need to use either phenomenological model for generation zone or Monte-Carlo simulation with well known inherent drawbacks of the both approaches.

R. Gauvin: Since you need a synchrotron radiation to get a photon flux greater than 10^6 photons/second, I think that few people will be able to use your method. Can you comment on this?

Authors: It is difficult to comment on how many people will use the approach to investigate surface profile by itself but we hope that solutions of direct and inverse problems presented in the paper will be useful at least to estimate an influence of the surface relief and roughness on fluorescent signal in X-ray micro-analysis. Consideration of such idealized situation is necessary to analyze more complicated cases as the film on the substrate or planar impurity distribution covered by the film (as mentioned above in answer to second comment of Dr. Rivers).

R. Gauvin: Real surfaces generally have $df(x)/dx$ = infinity, and this condition is not allowed in your method. What can you do to improve that in order to study fracture surfaces of technological materials for example? Have you considered the use of the concepts of fractal geometry in this context? Also, will you improve your model to consider the case when a photon crosses the surfaces several times (very irregular surfaces)?
Authors: We considered an important class of surfaces with one intersection only as the first step, we plan to develop a signal equation for surfaces with more intersections (this includes the case $df(x)/dx$ = infinity). As to fractal geometry and characterization of roughness by fractal dimension, we used such a concept while investigating backscattered electron signals [9] and we will use these ideas of statistical approach in fluorescent X-ray microscopy as well.

R. Gauvin: What is the time needed to get an analysis?
Authors: We estimate the time per pixel as 0.1-1.0 seconds at flux 10^8 photons/second in beam and at statistical scattering in the signal about 1%.

R. Gauvin: It is not surprising that your model works because you deconvoluted a simulated spectrum which is computed with the same theory as your inverse reconstruction technique. You should validate your method with experimental spectrum of known shape to prove it in the correct way.
Authors: Really, we applied reconstruction procedure to signals generated according to the signal model developed. By this, we did not validate model of signal formation (of course, it should be compared with experimental data), but, by this, we check stability of reconstruction method to small noise in the signal. An important feature was illustrated (and this was really surprising): the problem of surface restoration considered in the paper is stable to experimental noise, and thus, it essentially differs from inverse problems arising, for example, in absorption X-ray tomography.

An estimate can be deduced from the formula for spatial resolution {eq. (13)}, it can be read as following to resolve profile details with length l one needs angular precision $\Delta\alpha$ about

$$\Delta\alpha = l(\cos\alpha + \mu)^2 / (\nu + \cos\alpha) \qquad (20)$$

The estimate is a good qualitative supplement to the answer to the first comment of Dr. Rivers above.

Additional Reference

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