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## Use of Fourier transform in grazing X-rays reflectometry

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**Résumé.** — La technique de réflectométrie en rayons X rasants permet l'analyse d'empilements de couches minces. L'ajustement de la courbe de réflectivité par une méthode d'essai et erreur permet de déterminer les paramètres de ces couches. L'analyse de Fourier de la courbe de réflectivité expérimentale peut donner directement une idée du profil d'indice de l'empilement. A partir d'exemples réels, on montre la validité de la méthode.

**Abstract.** — Grazing X-ray reflectometry allows the analysis of thin layer stacks. The fitting of the reflectivity curve by a trial and error method can be used in order to determine the parameters of the films. Fourier analysis of the experimental reflectivity can directly give a rough determination of the profil index. Application to real examples shows the validity of the method.

### 1. Introduction.

For a long time grazing X-ray specular reflectivity has been used in order to characterize thin layer stacks [1]. In this case, the specular reflectivity of grazing X-rays depends on the thicknesses, the complex refractive indices of the layers, and on the roughness of the interfaces. Numerous authors have characterized the multilayers by fitting the experimental reflectivity curves. The fit can be obtained by a trial and error method (TEM) which consists in calculating the theoretical reflectivity and comparing the result with the experimental reflection.

In the case where the samples are flat enough, it was shown that it is possible to let a computer do the tedious work of trial and comparison [2]. For a stack which is made up of at least one layer, the reflectivity curve shows oscillations which reveals the different thicknesses of the stack. A Fourier transform of the reflectivity data can give directly the frequencies of these oscillations, and allow us to obtain quickly the thicknesses of each layer. Even if the Fourier transform give false results, these results are close enough to the real thicknesses to be used as good starting values for the trial and error computing, and to make this method more effective.

The goal of this study is to show that it is indeed possible to obtain almost directly the real thicknesses of the stack layers by using the Fourier transform.

## 2. Use of Fourier transform in grazing X-rays reflectometry : theoretical approach.

One of the main problems is that the reflectivity curve is not periodical and before using the Fourier transform it is necessary to make a preliminary transformation of the reflectivity data.

Sakurai and Iida [3] have subtracted an average curve from the logarithm intensity curve. This method is empirical, because it is necessary to define the criteria for the calculation of this average curve, and often in the results spurious orders remain.

Sivia *et al.* [4] use Fourier transform techniques to study neutron reflectivity curves recorded with a grazing incidence neutron reflectometer, with a scanned wavelength (Time-of-Flight technique).

Here, the FT is used with grazing X-ray reflectometry. Some limitations appear, due to the specific values of the complex indices for X-rays.

It is known that discrete Fourier transform has to be used with particular precautions in order to give satisfactory results.

OUTSTANDING PROPERTY OF THE REFLECTIVITY DATA. — We use the fact pointed out by some authors [4, 5] that for scattering vectors larger than the critical scattering momentum value  $Q_0$ , the reflectivity data relates to the derivative of the density profile by :

$$R(Q) \propto \frac{(4\pi)^2}{Q^4} \left| \int_{-\infty}^{+\infty} \frac{dn}{dz} \cdot e^{i Q \cdot z} dz \right|^2 \quad (1)$$

where  $Q$  the scattering momentum generally used is simply twice the normal component of the wave vector in the vacuum. As for us, we prefer to take into account the refraction and define  $Q$  by

$$Q = \frac{4\pi \cdot \sqrt{n^2 - \cos^2(\theta)}}{\lambda} \quad (2)$$

$\theta$  is the grazing angle,  $z$  is the depth and  $\lambda$  is the incident wavelength, and  $n$  is the root mean square index of the stack deduced from the critical angle of the total reflection as shown in reference [6].

The formula shows that  $R$  is related to the Fourier transform of the derivative of  $n$  with respect to the depth.

Then the auto correlation function (ACF) of the derivative of the density profile can be written :

$$\begin{aligned} \rho(z) &= \int_{-\infty}^{+\infty} n'(t) \cdot n'(t-z) \cdot dt \\ \rho(z) &\propto \frac{1}{(4\pi)^2} \int_{-\infty}^{+\infty} Q^4 R(Q) \cdot e^{-iQz} dQ \end{aligned} \quad (3)$$

where  $t$  is the depth variable.

In the case of homogeneous layers the auto correlation function is simply.

$$\rho(z) = \sum_k \sum_{\ell} (n_{k+1} - n_k) \cdot (n_{\ell+1} - n_{\ell}) \cdot \delta_{|x_k - x_{\ell}|}(z) \quad (4)$$

$k$  and  $\ell$  are the layer numbers,  $n_k$  are the layer indices.  $x_k$  is the depth between the  $k$ -th surface and the origin.  $\delta_h$  is the Dirac function centered around  $z = h$ . These analytical results are illustrated by figure 1.

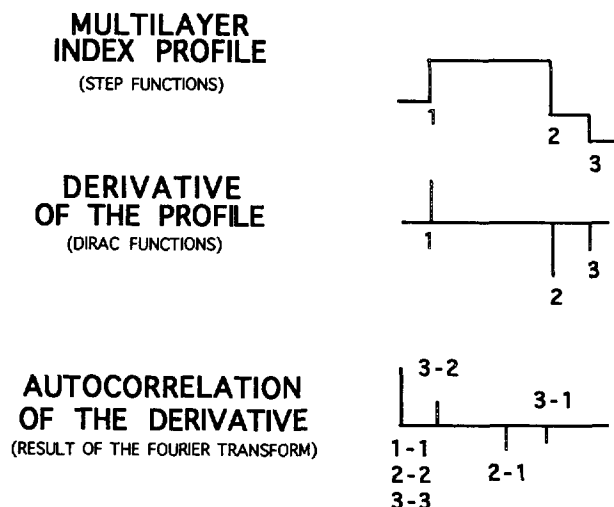


Fig. 1. — Index profile, derivative of the index profile and auto correlation of the derivative of the index profile in the case of a perfect stack.

If  $i$  is the number of interfaces, this function  $\rho$  has  $i \cdot (i - 1)/2$  peaks associated with the couples of interfaces. The position of these peaks are the distances between the interfaces. There are  $i$  other peaks which are superimposed at the origin, these peaks are associated with the self coupled interfaces.

Note that the auto correlation function  $\rho$  is different from the index derivative profile defined by :

$$n'(z) = \sum_k (n_{k+1} - n_k) \cdot \delta_{z_k}(z) \quad (5)$$

this last function has  $i$  peak only. The abscises of the derivative peaks coincide with abscises of the interface. The auto correlation function has additional peaks the positions of which are the differences between the positions of the density discontinuities.

For real thin layer stacks, the roughness leads to a widening of the peaks in the Fourier spectrum, Gaussian functions replacing the dirac functions in (5). A theoretical study of this effect has been developed in a previous paper [7].

### 3. Experimental results.

In this paper we will use the FT in three more and more complicated cases : a simple layer, a bilayer and a periodical stack.

The experimental reflectivity curves are obtained by means of a goniometer the principle of which is shown in figure 2 [1, 8]. The source unit is made of an X-ray source with a copper anode. The  $K\alpha$  radiation ( $\lambda = 0.15405$  nm) is obtained by the means of a 200 LiF crystal. The sample is positioned at a fixed stand, the source unit and the detector are fixed on arms which are moving around the same axis. The reflectivity curve is obtained by varying the grazing incidence angle while tracking the reflected beam.

**3.1 MONOLAYER.** — Figure 3a shows the experimental reflectivity of a gold monolayer deposited onto a float glass substrate. The grazing angle is given in arc seconds while the intensity is given in logarithmic scale. The Fourier transform of these experimental data is

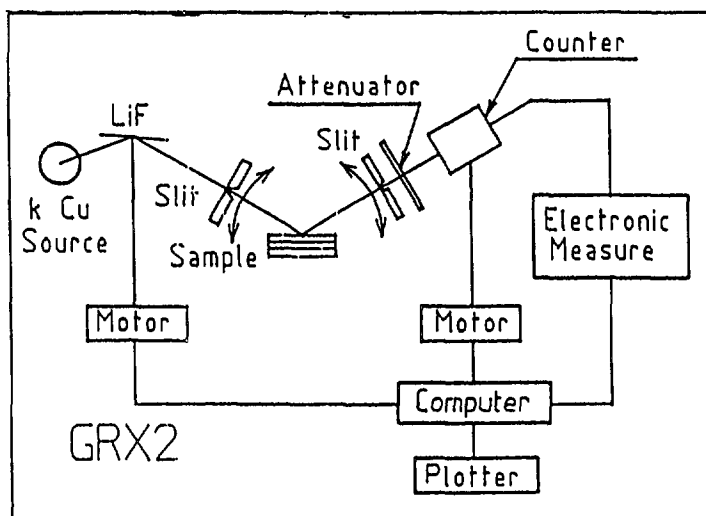
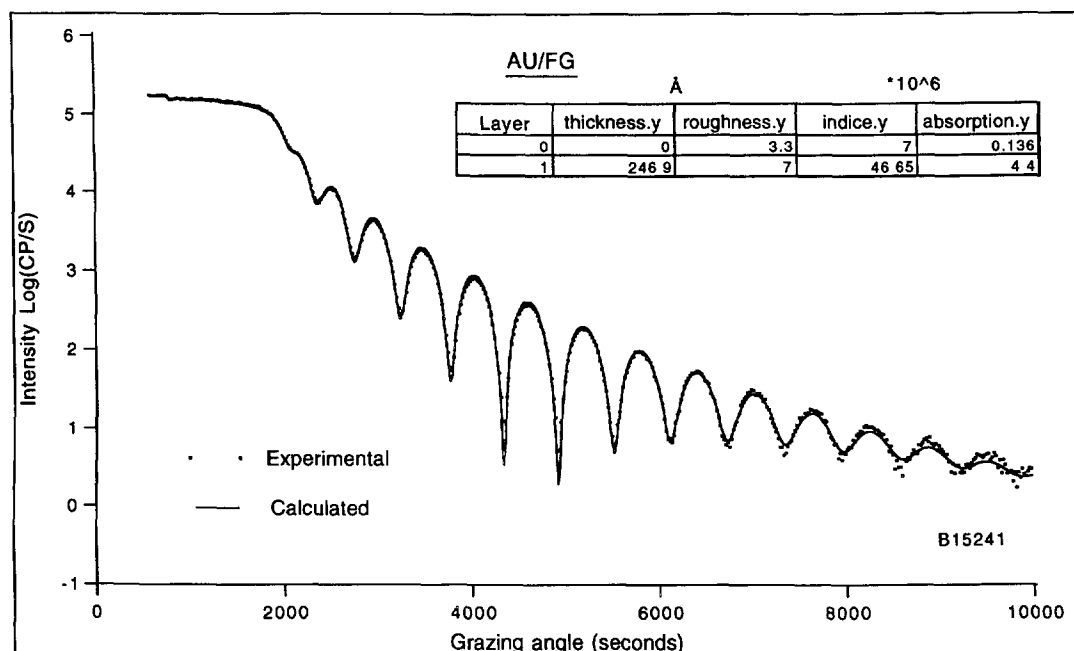


Fig. 2. — Sketch of X-ray goniometer.



a)

Fig. 3. — a) Experimental reflectivity curve for a Au layer (dotted line), fitted with a calculated curve (continuous line), with parameters given in the table. The layer n 0 corresponds to the glass substrate. b) Fourier transform of experimental data of figure 3a. On the x-axis are reported the values of the depth  $z$ , in Å units. The y-axis is related to the auto correlation function (ACF( $z$ )) defined by the equation (3), so the units are here in Å<sup>-5</sup> and is proportional to the index variation.

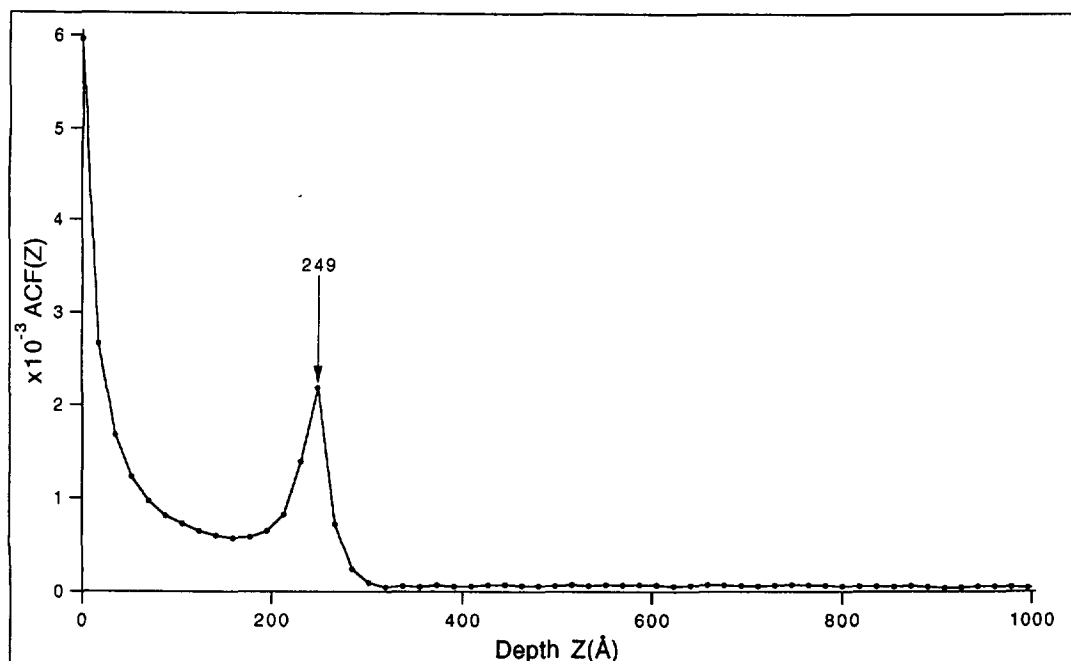


Fig. 3b.

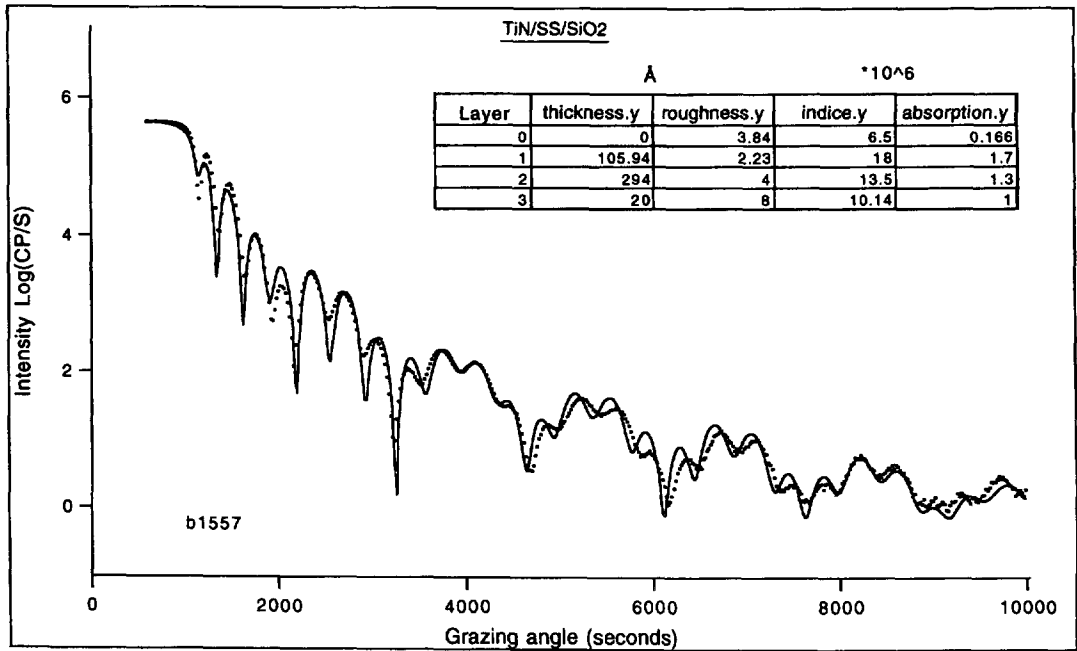
shown in the figure 3b. The  $y$  scale is given in arbitrary units and the  $x$  scale in  $\text{\AA}$ . There is one evident peak at  $249 \text{ \AA}$ . This result agrees very well with the value  $247 \text{ \AA}$  obtained by the trial and error method figure 3a-table. Moreover the TEM which is a heavier method gives the roughness and the index.

**3.2 BILAYER.** — The second example is a little more complicated. It is a bilayer of TiN and stainless steel deposited on a  $\text{SiO}_2$  substrate. The Fourier transform of the reflection data (Fig. 4a) is shown in figure 4b). Three peaks can be seen, a peak at  $106 \text{ \AA}$ , a peak at  $318 \text{ \AA}$  and one at  $424 \text{ \AA}$ . These peaks correspond to the different thicknesses obtained by coupling the different interfaces. From the FT it is difficult to deduce directly the order of the different layers. The FT results agree well enough with the ones of the trial and error method figure 4a-table. Nevertheless the TEM seems to reveal 3 layers. But if one looks at the figure 4c one can see that the superficial layer of about  $20 \text{ \AA}$  is kind of transition layer that the FT cannot display.

**3.3 PERIODIC STACKING.** — The third example is a periodic stacking with 10 bilayers. For confidential reasons the nature of the materials are not communicated, but it does not matter for our study. The reflectivity is given by the figure 5a. The explanation of Fourier spectrum is more difficult, the Fourier spectrum is shown in figure 5b. All the peaks correspond, with a good enough agreement, to the combinations of interfaces at the depth  $0, 36.1, 107.2, 143.3, 214.4, 250.5, \dots, 1\ 035.9, 1\ 072 \text{ \AA}$  given by the TEM figure 5a-table. In fact the initial values chosen to start the fit were the ones given by the FT.

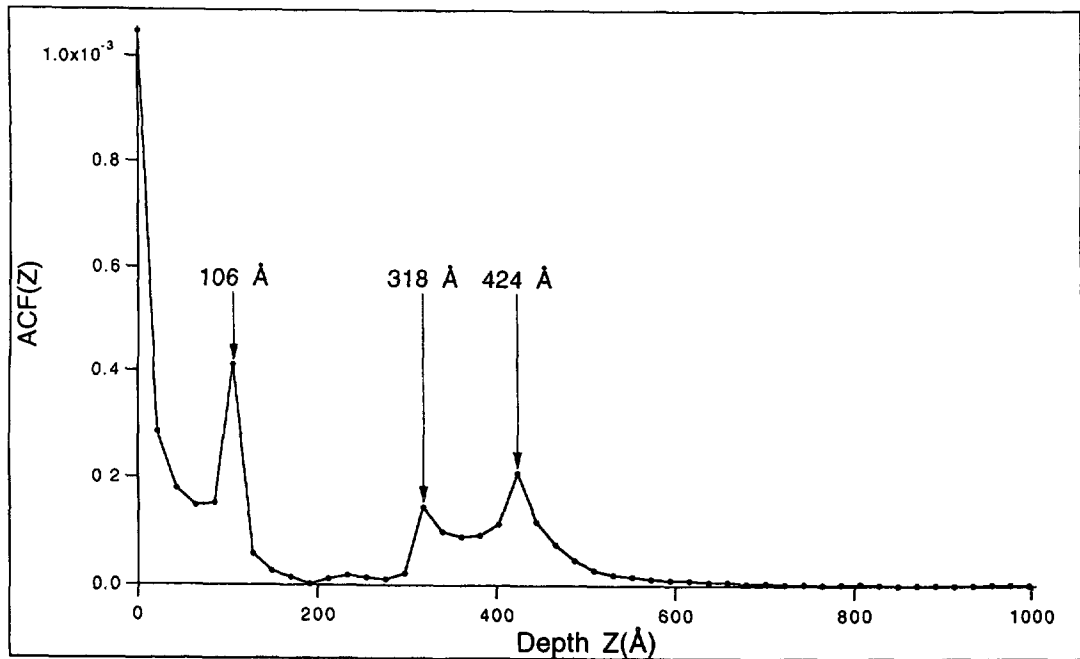
#### 4. Conclusions.

The Fourier transform of the reflectivity data multiplied by the  $Q^4$  factor has a theoretical justification as shown by equations (1) to (4), so that the FT gives results having physical



IOTA GRAZING X-RAYS REFLECTOMETRY

a)



b)

Fig. 4. — a) Experimental reflectivity curve for a Ti/stainless steel/SiO<sub>2</sub> sample (dotted line), fitted with a calculated curve (continuous line), with parameters given in the table. The layer n 0 corresponds to the glass substrate. b) Fourier transform of experimental data of figure 4a. c) Index profile calculated from the results displayed in the table of the figure 4a.

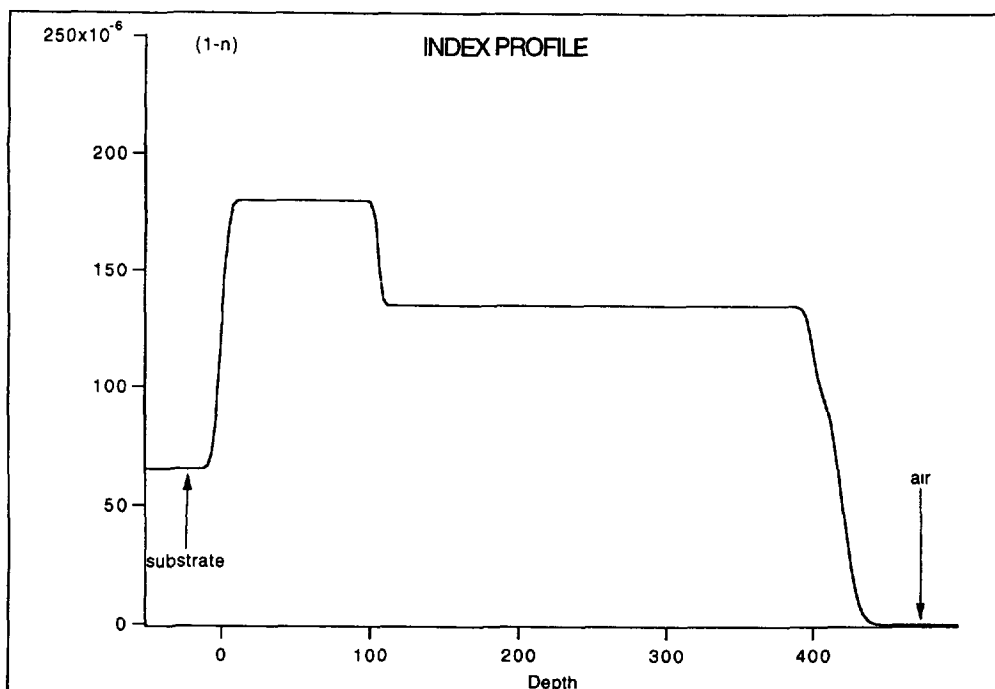
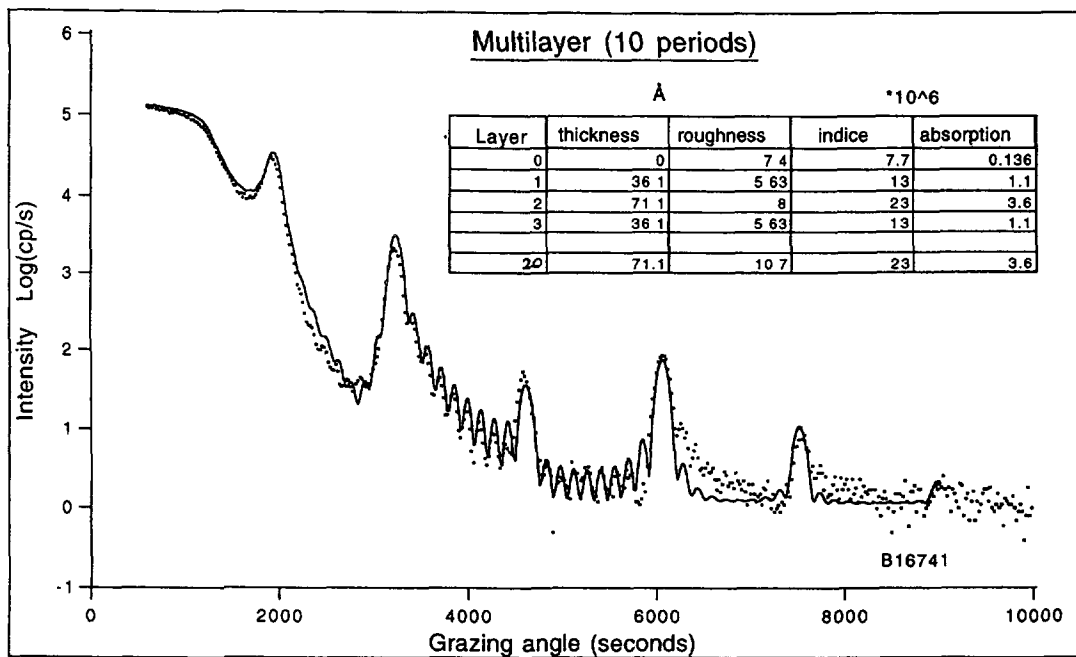


Fig. 4c.



a)

Fig. 5. — a) Experimental reflectivity curve for a periodic multilayer (dotted line), fitted with a calculated curve (continuous line), with parameters given in the table. The layer n 0 corresponds to the glass substrate. b) Fourier transform of experimental data of figure 5a.



## FOURIER TRANSFORM

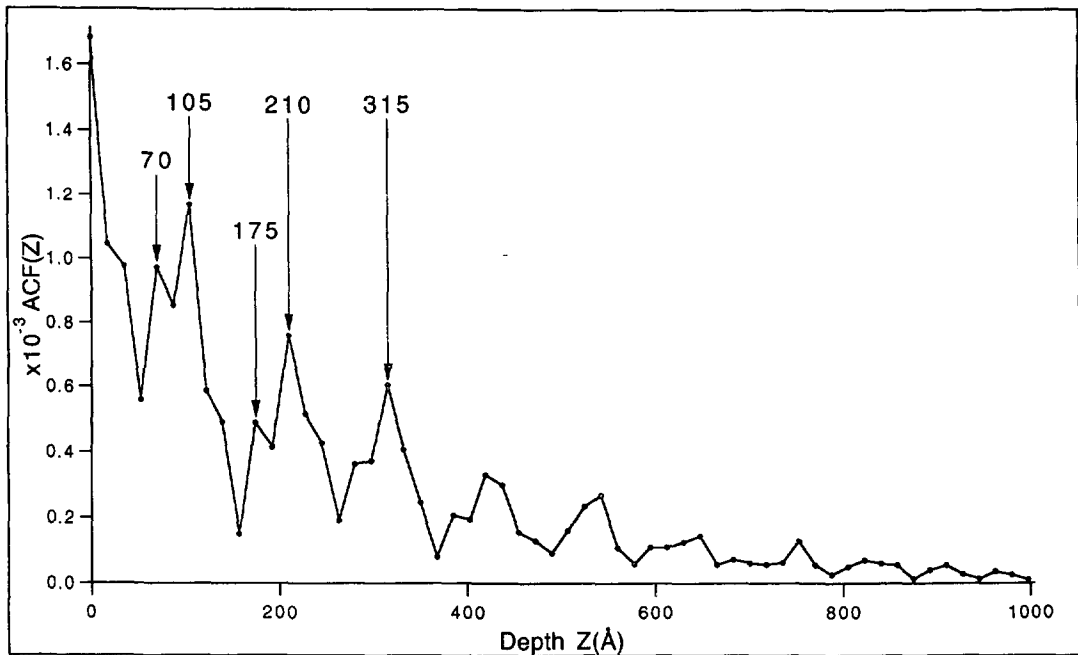


Fig. 5b.

meaning. The FT allows us an easy determination of the distances which separate the different interfaces. These results can be useful in making a choice of a starting model to fit the curve and to determine the parameters of the stack. Note that, because it gives an auto correlation function, the FT does not allow the direct determination of exact order of the stacking. But in the actual conditions this order is usually known *a priori*.

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