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LIGHT SCATTERING OF DIELECTRIC MIRRORS

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ABSTRACT

The Rayleigh scattering of dielectric mirrors and dielectric components was measured at 441.6 nm wavelength. The light scattering of the TiO_2-SiO_2 and ZnS-MgF_2 film systems can be explained by scattering at the interfaces according to the Elson model. By comparing curves of the light scattering as a function of scattering angle with the results of a numerical computing method the statistical parameters of the interfaces were obtained and were found to be in good agreement with electron microscope results.

АННОТАЦИЯ

Измерено рассеяние света на диэлектрических зеркалах и диэлектрических компонентах при длине волны 441,6 нм. Угловое распределение рассеянного света на тонких пленках состава TiO_2-SiO_2 и ZnS-MgF_2 удалось объяснить с помощью модели Элзона, которая описывает рассеяние на граничных поверхностях. Статистические параметры граничных поверхностей, полученные из сравнения экспериментальных и теоретических результатов угловой зависимости интенсивности, находятся в согласии с данными, полученными раньше из электронно-микроскопических исследований.

KIVONAT

Dielektrikum tükrök és dielektrikum komponensek fényszórását mértük 441,6 nm-en. A TiO_2-SiO_2 és ZnS-MgF_2 összetételü vékonyréteg rendszerek fényszórásának szögeloszlását a határfelületek fényszórását leiró Elson modellel sikerült magyarázni. Az elméleti modell és a mért szórási szög függés összehasonlításának eredményeképpen kapott határfelületi statisztikai paraméterek a korábbi elektronmikroszkópos eredményekkel összhangban vannak.

INTRODUCTION

In the calculation of the optical properties of multilayer thin film structures, ideally planar and homogeneous layers are supposed. The thermodinamical properties determining layer deposition and growing cause development of bulk and surface inhomogenities /e.g. rough boundaries caused by the column structure of the film/. A usual way to detemine the average column size is investigation of the thin film by electron microscope [1] [2]. The wavefront of a light beam travelling in an inhomogeneous media is distorted. The travelling becomes possible in other directions than determined by the laws of the geometrical optics, namely the light is scattered. The specular reflectivity of the laser mirrors is limited by the light scattering [3] [4] [5] [6].

Some experimental aspects of light scattering of multilayer thin films was described in the last years. The angular distribution of the light scattering of ZnS-MgF_2 laser mirrors was measured by Günther, Gruber and Pulker [4] [5]. The experimental curves were explained according to the Beckman theory /for the surface/ and the Debye theory /for the volume inhomogenities/. The Airy method was generalised for non planar boundaries by Eastman [8]. Gourley and Lissberger developed a matrix formulation of the theory of optical properties of multilayer thin films which is suitable for characterising the effects of the interfacial roughness [9] [10]. The differential scattering cross section of thin film systems containing identical or statistically independent interfaces is given by Elson [1] [12] [13].

This method gives a possibility to connect in an explicit way the measurable angular distribution of the scattered light to the statistical parameters of the boundaries.

THE BASIC THEORETICAL MODEL

A good theoretical approximation of the scattering properties of the thin film systems can be based on the knowledge of the thin film structure.

Fig. 1 shows our basic conception about the thin film structure and the boundaries.

Δn_v characterises the internal /volume/ fluctuation of the refractive index caused by dust particles and structural defects. Δn_s means the refractive index fluctuation appearing on the interface consisting of two materials having different refractive indices. It can be supposed, that $\Delta n_s \gg \Delta n_v$ so it is a reasonable supposition, that the scattering properties of the multilayer thin films are defined by the interfaces in most cases. The morphology of the interfaces are characterised by the σ rms height and the T mean correlation length of the grains. Naturally, not only one σ and T pair belong to one surface /eg. the substrate surface is not ideally planar, this causes a roughness with $\sigma_{sb} \ll \lambda$ and $T_{sb} \geq \lambda$ parameters/. This type of roughness causes an observable scattering in small angles /less than 10^o/. The roughness because of the columned film structure has $\sigma \ll \lambda$ and $T < \lambda$ statistical parameters.

The angular distribution of the scattered light of a coherent source is determined by the interference of the beams, coming from the different centres and interfaces /specle noise/. The matematical description of the boundary fields is very complicated in the case of non planar boundaries. Elson solved this problem by using a non orthogonal coordinate transformation, as follows:

$$\begin{aligned} u_1 &= x \\ u_2 &= y \\ u_3 &= z-\xi(x,y) \end{aligned} \qquad (1)$$

where u_1, u_2 and u_3 are the non orthogonal coordinates, the function $\xi(x,y)$ describes the non planar surface in the orthogonal system. In the new coordinate system the incident wave fronts are deformed according to $\xi(x,y)$, but the boundaries are ideally planar. So the problem is reduced to the propagation of a slightly

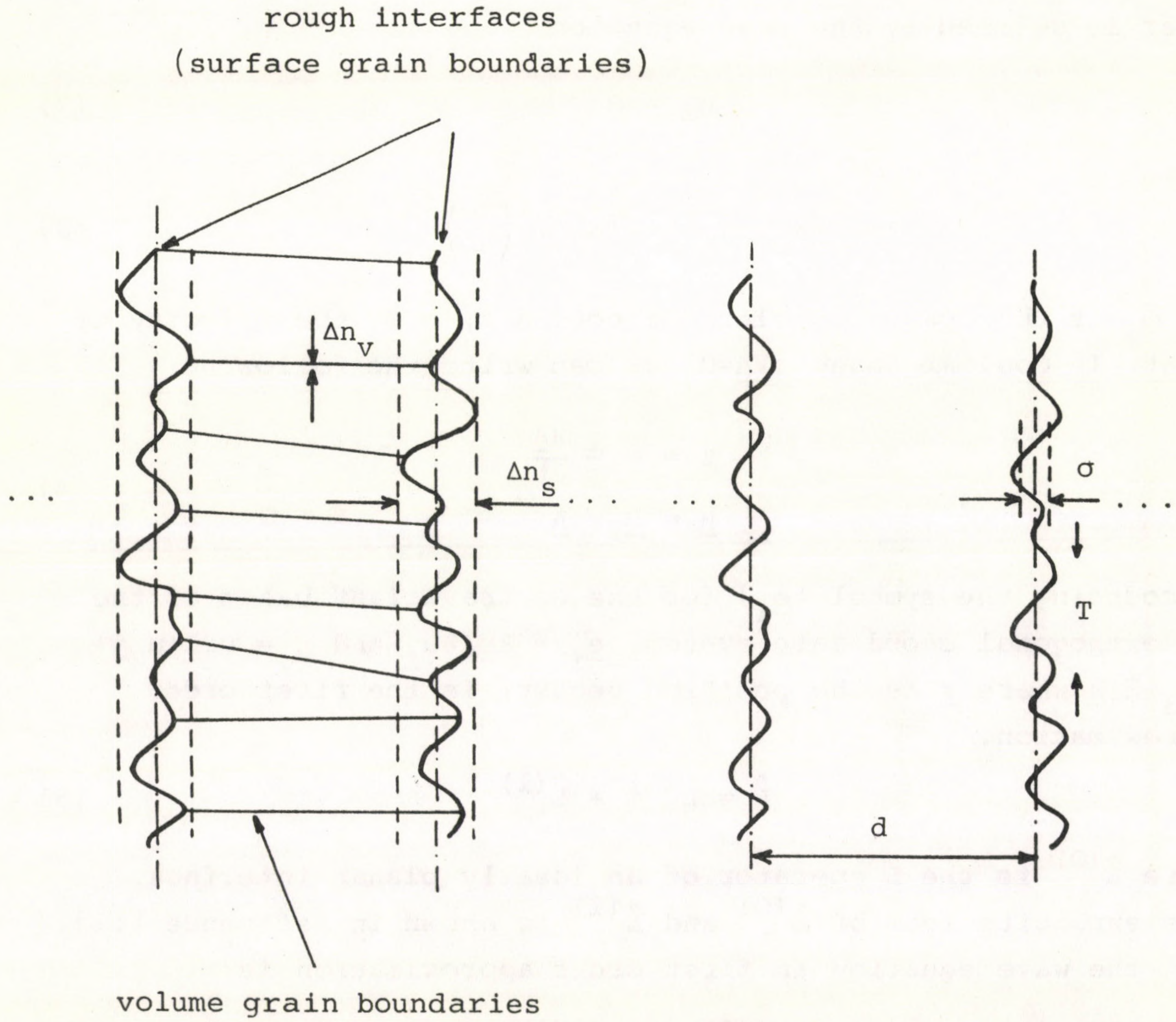


Fig. 1
The meaning of the basic quantities causing the light scattering effect according to our point of wiev

non planar wave front in a medium containing plane paralell boundaries. The first order approximation of the perturbation theory must be sufficient, since we supposed, however that $\sigma \ll \lambda$ /linear approximation/. The $\underline{A}=\underline{A}(\underline{r},t)$ vectorpotential in the i-th layer is defined by the wave equation,

$$\hat{L}\underline{A} = 0 \tag{2}$$

where

$$\hat{L} = \underline{\nabla}x\underline{\nabla}x-\varepsilon_i[z-\xi(x,y)]\left(\frac{\omega_o}{c}\right)^2 \tag{3}$$

and ω_i is the complex dielectric constant, c is the velocity of light. In Coulomb gauge ($\underline{\nabla A}=0$) we can write the following:

$$\begin{aligned} \underline{E} &= -\frac{1}{c}\frac{\partial A}{\partial t} \\ \underline{H} &= \underline{\nabla} \times \underline{A} \end{aligned} \tag{4}$$

Introducing the symbol $\{\underline{e}_i\}$ for the contravariant basis of the non orthogonal coordinate system, $\underline{e}_i = \partial\underline{r}/\partial u_i$ and $\underline{r} = u_1\underline{i}+u_2\underline{j}+$ $+(u_3+\xi)\underline{k}$ where $\underline{r}$ is the position vector, in the first order approximation.

$$\hat{L} = \hat{L}^{(0)} + \hat{L}^{(1)} \tag{5}$$

where $\hat{L}^{(0)}$ is the $\hat{L}$ operator of an ideally planar interface. /The explicite form of $\hat{L}^{(0)}$ and $\hat{L}^{(1)}$ is shown in reference [11]./ Then the wave equation in first order approximation is

$$\hat{L}^{(0)}\ \underline{A} = -\hat{L}^{(1)}\ \underline{A} \tag{6}$$

We can look for the solution with iterative approximation. In the zero order /if $\xi=0$/ it follows:

$$\hat{L}^{(0)}\ \underline{A}^{(0)} = 0 \tag{7}$$

Comtining with (6)

$$\hat{L}^{(0)}\ \underline{A}^{(1)} = -\hat{L}^{(1)}\ A^{(0)} \tag{8}$$

Then the solution

$$\underline{A} = \underline{A}^{(0)} + \underline{A}^{(1)} \tag{9}$$

We can find $\underline{A}^{(1)}$ using the Green function method. Then the problem is the following:

$$\hat{L}^{(0)}G(\underline{u},t,\underline{u}',t') = \hat{I}\delta^3(\underline{u}-\underline{u}')\delta(t-t') \qquad (10)$$

where $\hat{I}$ is the unit operator. Then the Green matrixis:

$$G(\underline{u},t,\underline{u}',t') = \frac{1}{(2\pi)^3}\int d^2k d\omega g(u_3,u_3')e^{i[\underline{k}(\underline{\rho}-\underline{\rho}')-\omega(t-t')]} \qquad (11)$$

where

$$\underline{\rho}\equiv(u_1,u_2),\ \underline{k}\equiv(k_1,k_2)$$

Evaluating G we can obtain $\underline{A}_1$

$$\underline{A}^{(1)}(\underline{u},t) = -\int d^3u'dt'G(\underline{u},t,\underline{u}',t')\hat{L}^{(1)}\underline{A}^{(0)}(\underline{u}',t') \qquad (12)$$

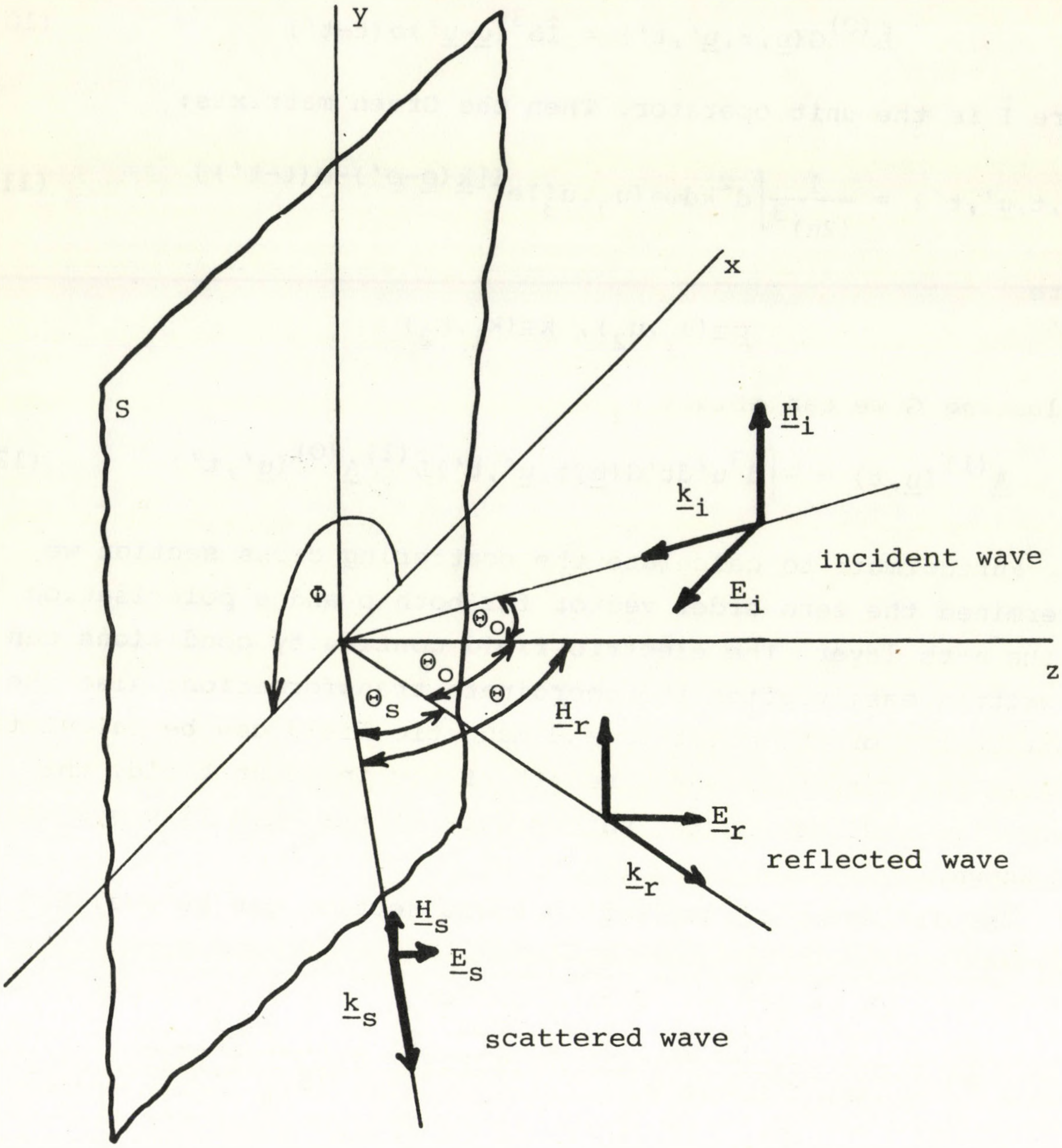
Furthermore to calculate the scattering cross section we determined the zero order vector for both p and s polarisation in the n-th layer. The electric field continuity conditions can be written easily after the coordinate transformation, also the distribution of the electric and magnetic field can be calculated, because the direction and strength of the incident field, the refractive indices and the thicknesses of the thin film systems are known.

The differential scattering cross section can be evaluted as follows:

$$\frac{dP}{d\Omega} = \frac{\left(\frac{\omega_o}{c}\right)^4|\xi(\underline{k}-\underline{k}_o)|^2}{(4\pi)^2I^2S\cos\Theta_o}\left[\frac{|a_\varphi|^2\cos^2\Theta}{|\xi_{11}^{(1,L+1)}|} + \frac{|a_\Theta|^2}{|\delta_{11}^{(1,L+1)}|}\right] \qquad (13)$$

where $d\Omega = \sin\Theta d\Theta d\varphi$ is the solid angle of the detector around the direction determined by the angles Θ and φ, $\xi(\underline{k}-\underline{k}_o)$ is the Fourier transformed form of $\xi(u_1,u_2)$ and S is the illuminated surface. The symbols are explained on *Fig. 2*. If $\xi(x,y)$ is a random function, then the mean value of $|\xi(\underline{k}-\underline{k}_o)|^2$ for the S surface can be easily written:

$$F = \frac{<|\xi(\underline{k}-\underline{k}_o)|^2>}{S} = \frac{\sigma^2}{(2\pi)^2}G(\underline{k}-\underline{k}_o) \qquad (14)$$


y
x
z
S
$\underline{H}_i$
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incident wave
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Φ
Θ_o
Θ_o
Θ_s
Θ
$\underline{H}_r$
$\underline{E}_r$
reflected wave
$\underline{k}_r$
$\underline{H}_s$
$\underline{E}_s$
$\underline{k}_s$
scattered wave

Fig. 2

where

$$G(\underline{k}-\underline{k}_o) = \iint\limits_{(S)} d^2\tau c(\underline{\tau}) e^{i(\underline{k}-\underline{k}_o)\underline{\tau}} \tag{15}$$

and in (15) $c(\underline{\tau})$ is the autocorrelation function of the surface. In the case of an isotropic gaussian autocorrelation

$$c(\tau) = e^{-\tau^2/T^2} \tag{16}$$

and

$$G(\underline{k}-\underline{k}_o) = \pi T^2 \exp\left[\frac{\left\{-\left(\frac{\omega_o}{c}\right)^2 (\sin\Theta - \sin\Theta_o)^2 T^2\right\}}{4}\right] \tag{17}$$

where Θ_o is the angle of the incidence, and

$$F = \frac{\sigma^2 T^2}{4\pi} e^{-\frac{S^2T^2}{4}} \tag{18}$$

$$S = \frac{\omega_o}{c} (\sin\Theta - \sin\Theta_o) \tag{19}$$

supposing, that $S^{1/2} >> \lambda$.

THE EXPERIMENTAL ARRANGEMENT

The experimental arrangement is shown on *Fig. 3*. A positive column He-Cd$^+$ laser operating at 441.6 nm was used as the light source. The CW energy was 30 mW with about 1% stability. The detector was a PIN-10 photodiode with 2.3×10^{-4} sr. solid angle. The angular distribution of the measured scattered intensity of the "empty" system and a well polished BK7 glass substrate are shown on *Fig.4* and *5*. The intensity distribution shown on *Fig. 4* was taken into correction in the further measurements as a noise level.

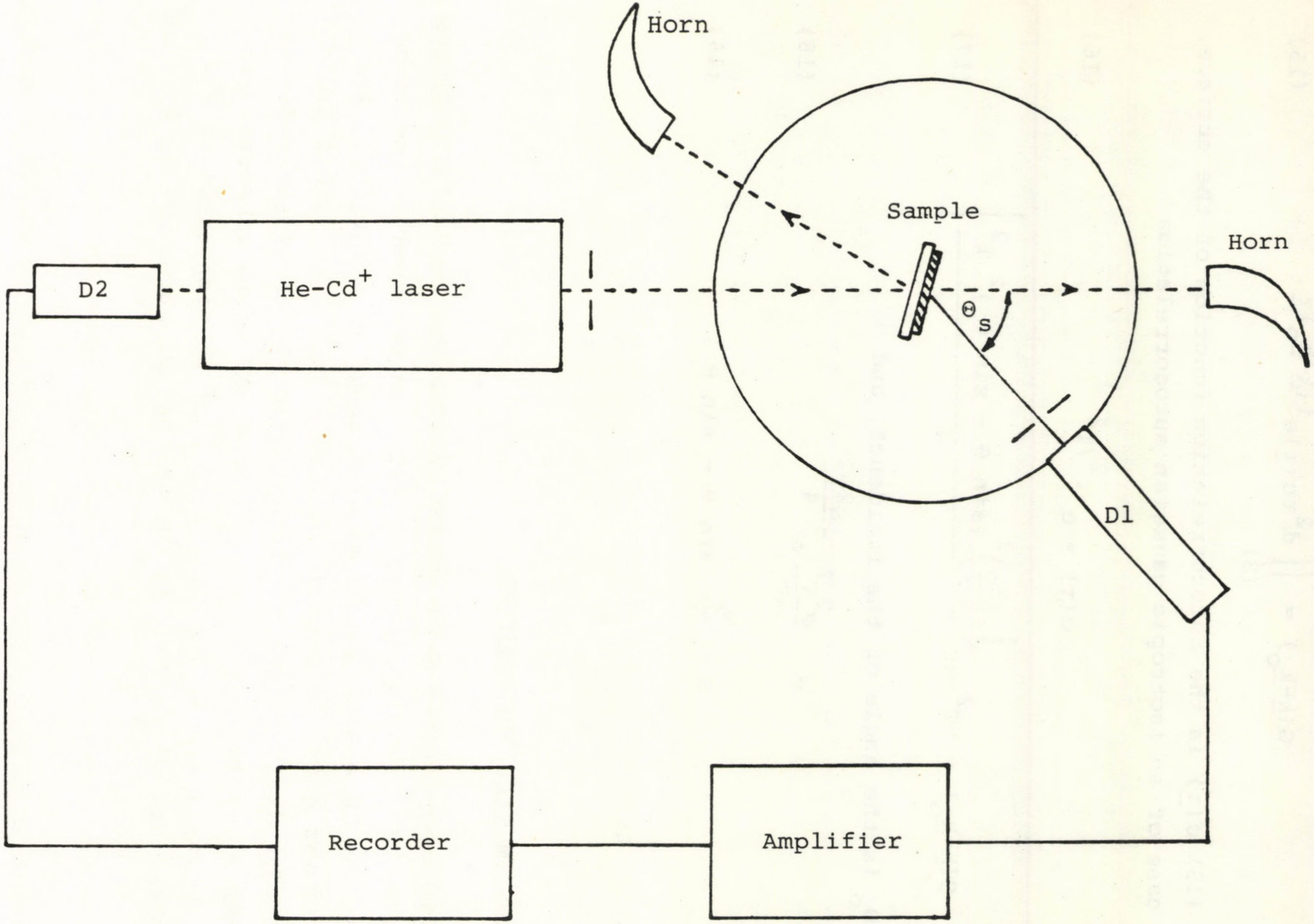

Fig. 3
Experimental arrangement for scattering measurements

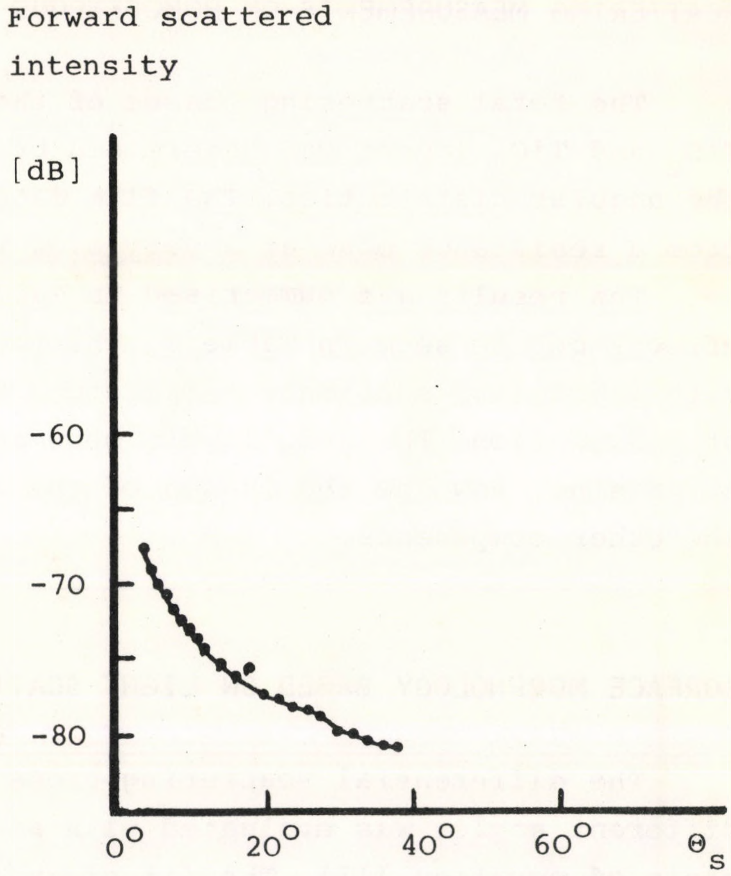

Fig. 4
The angular distribution of the scattered light intensity in the "empty" system

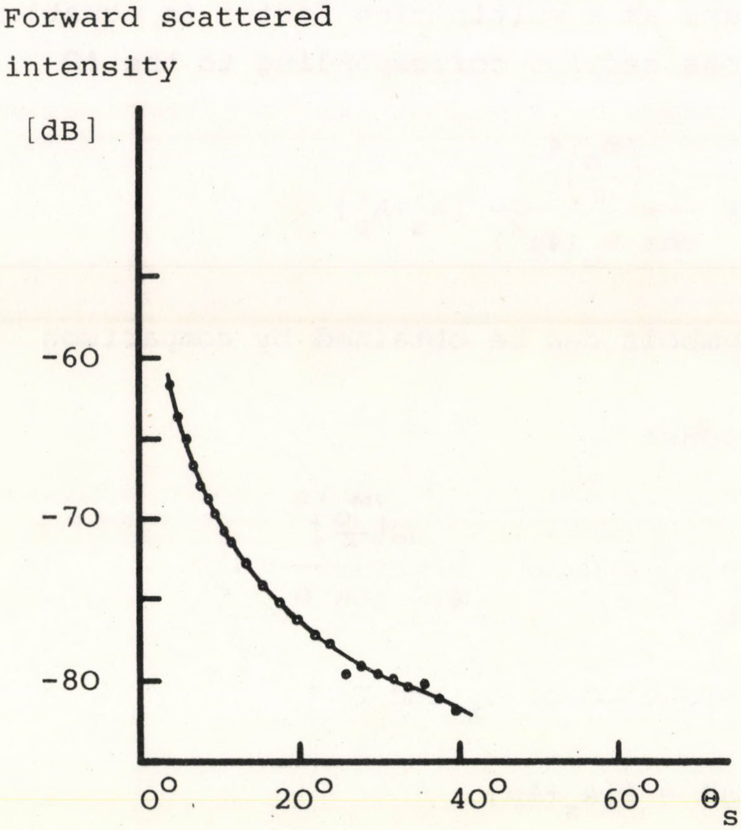

Fig. 5
Light scattering of the BK7 substrate

SCATTERING MEASUREMENTS ON HOMOGENEOUS LAYERS

The total scattering losses of the homogeneous ZnS, MgF_2, SiO_2 and TiO_2 layers was determined by numerical integration of the angular distribution. The film materials were evaporated from a resistance boat at a very slow rate.

The results are summarised in *Table 1.* Some remarkable effects can be seen in Table 1. The total losses usually increase with increasing substrate temperature and by decreasing the rate of evaporation. The CeO_2 layers show an opposite effect. It is surprising, how low the losses of the SiO_2 layers are compared to the other components.

SURFACE MORPHOLOGY BASED ON LIGHT SCATTERING

The differential scattering cross section belonging to the different angles was evaluated with an ESR-40 computer on the basis of equation (13). The fit of experimental data with the theoretical one was simple because the approximation is linear, eg. the F from factor appears as a multiplying factor in equation (13). The ΔP scattering cross section corresponding to the ΔΩ solid angle is

$$\Delta P = \Delta\Omega \cdot F \frac{\left(\frac{\omega_o}{c}\right)^4}{\cos \Theta_o (4\pi^2)} [A_s' + A_p']$$

where the meaning of the symbols can be obtained by comparison with equation (13).

Introducing the notations:

$$A_s = \frac{\Delta\Omega\left(\frac{\omega_o}{c}\right)^4}{4\pi^2 \cos \Theta_o} A_s' \text{ and } Ap = \frac{\Delta\Omega\left(\frac{\omega_o}{c}\right)^4}{4\pi^2 \cos \Theta_o} A_p'$$

we can obtain ΔP by multiplication of A_i and F

$$\Delta P = F[A_s + Ap]$$

Table 1

Light scattering losses of homogeneous films

Sample number	Material	Substrate temperature /^{o}C/	Geometrical thickness /nm/	Deposition rate /nm.sec^{-1}/	Pressure /torr/	Total losses /%/
1	ZnS	20	543	.57	5×10^{-6}	.114
2		20		.48	10^{-3}	.36
3		300		.48	10^{-5}	1.36
4	MgF_2	300	906	1.89	5×10^{-6}	.11
5	SiO_2	250	856	.31	10^{-3*}	.05
6				.34	10^{-4*}	.06
7				.68	10^{-5*}	.08
8	TiO_2	100	279	.23	$8 \times 10^{-4*}$	
9		250	558	.21		.10
10		400	558	.17		.54
11	CeO_2	50	284	1.2	8×10^{-5}	.40
12		50		.32	5×10^{-5}	1.8
13		200		.95	10^{-5}	.43
14		200		.24	10^{-5}	.10
15		400		.63	3×10^{-5}	.12
16		400		.18	3×10^{-5}	.15

*The pressure means the oxigen partial pressure

Table 2

Light scattering losses of multilayer mirrors

Sample number	Materials	Structure	Substrate temperature	Total deposition time /sec/	Total losses /%/
17	H = ZnS	$/HL/^2$	20	360	.13
18	L = MgF_2	$/HL/^2$	300	300	.62
19		$/HL/^5$	20	900	1.65
20		$/HL/^5$	300	1500	6.72
21		$/HL/^8$	220	1136	4.71
22		$/HL/^8$	300	1568	10.66
23-28*	H = TiO_2 L = SiO_2	$/HL/^5H$	300		.15

*The oxigen partial pressure during deposition was $7x10^{-4}$ torr

The input data for the calculation of A_i were the following:

number of the quaterlambda layers /L/
refractive index of the substrate /$n_s = 1.52$/
refractive index of the layer components /$n_H = 2.3$/
/$n_L = 1.4$/
refractive index of the incidence medium /$n_o = 1.0$/
the tuning wavelength of the quaterlambda layers
/$\lambda_o = 500$ or 441 nm/
angle of incidence /$\Theta_o = 0^o$/
the measuring angle was changed between 5^o and 85^o with 5^o steps.

The angular distribution of the scattering of three ZnS/MgF_2 mirrors fitted with the appropriate theoretical curves is shown on *Fig. 6.*

The most correct fit could be obtained with the shown σ and T surface parameters. The influence of the higher substrate temperature is shown on *Fig. 7.* The angular distribution could be fitted with the theoretical curves containing higher σ and T in all the three cases of the number of layers. *Fig. 8* shows the angular distribution of the light scattering of $(HL)^6H$ TiO_2/SiO_2 laser mirrors tuned to 441.6 nm fitted with the theoretical curves. Comparing this Figure with Figs. 6-7 the high correlation length of the surface irregularities is surprising. We could obtain much smaller correlation length in the case of 500 nm thick homogeneous TiO_2 films, as it is shown on *Fig. 9* /the light scattering properties of the films depend on the subsequent annealing process/.

THE EFFECT OF MULTILAYER INTERFERENCE

The angular distribution of scattered light must be determined not only by the specle noise from the interface irregularities, but also by the cross-correlation caused by the multilayer interference in a tuned thin film system. In the case of multilayer mirrors, this cross correlation causes exterma of the scattered intensity at higher angles if the tuning wavelength and

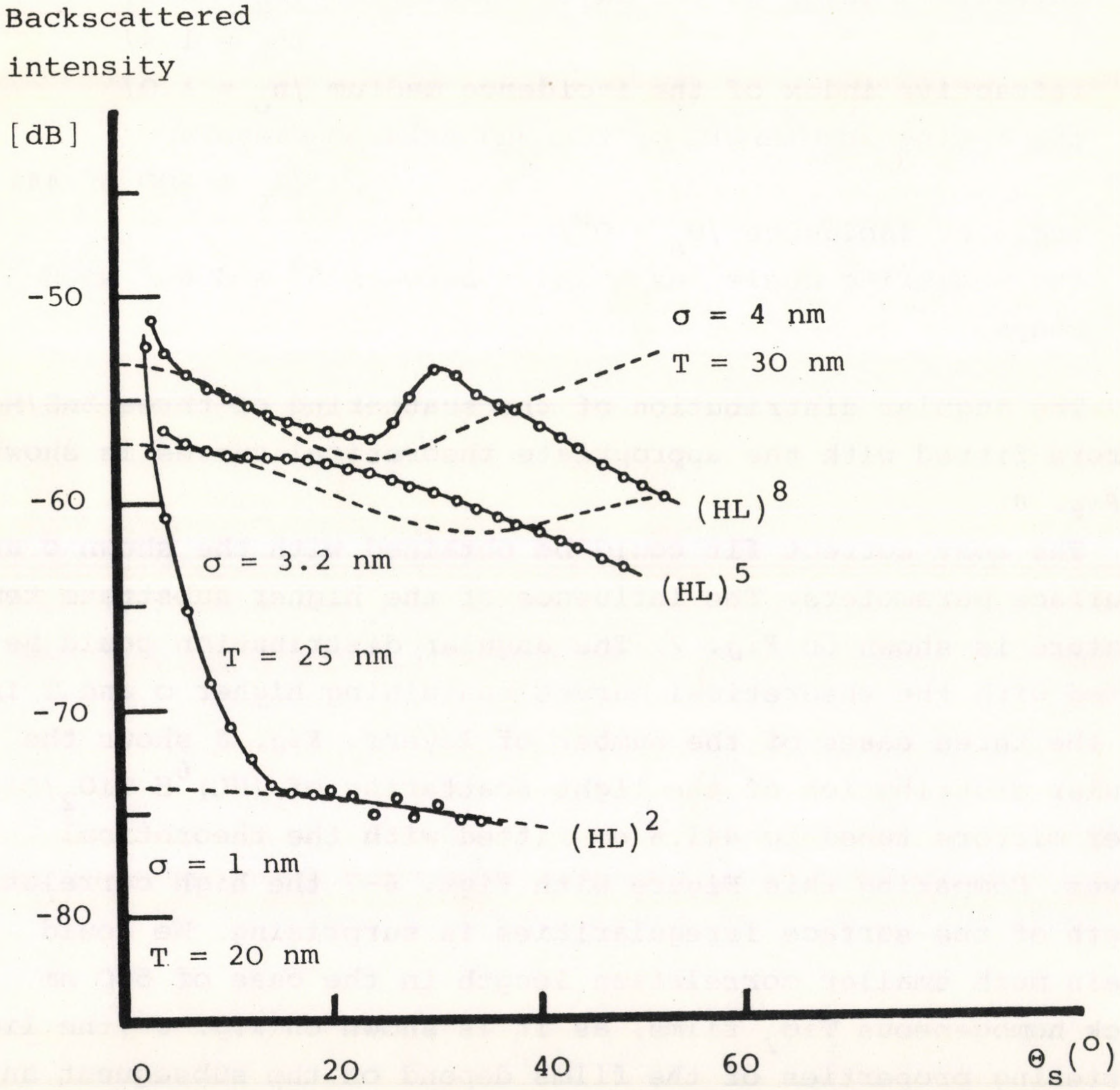

Fig. 6

The measured light scattering of three ZnS/MgF$_2$ interference mirrors /——/ fitted with the theoretical curves /----/

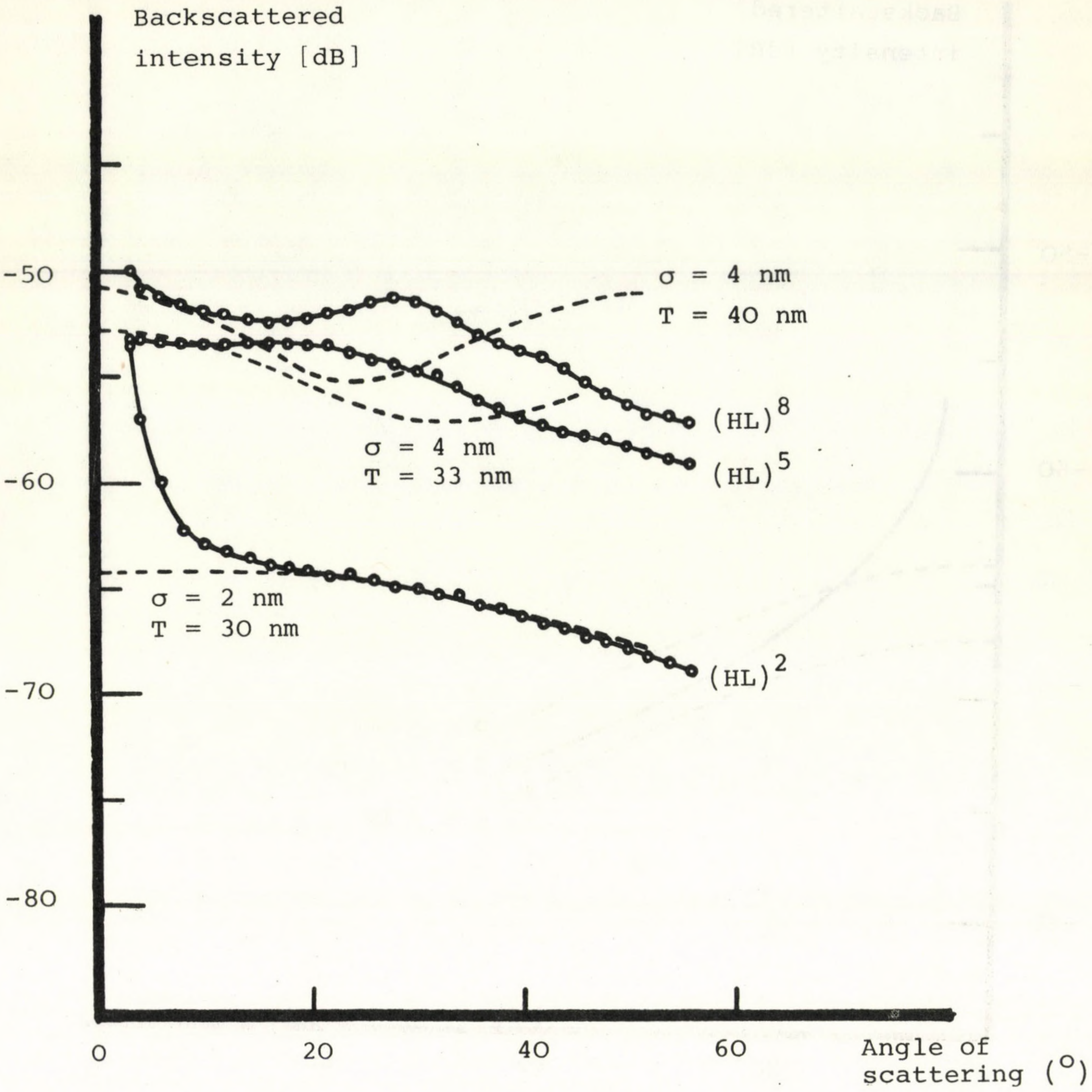

Fig. 7
The result of the fitting procedure in the cases of higher substrate temperature

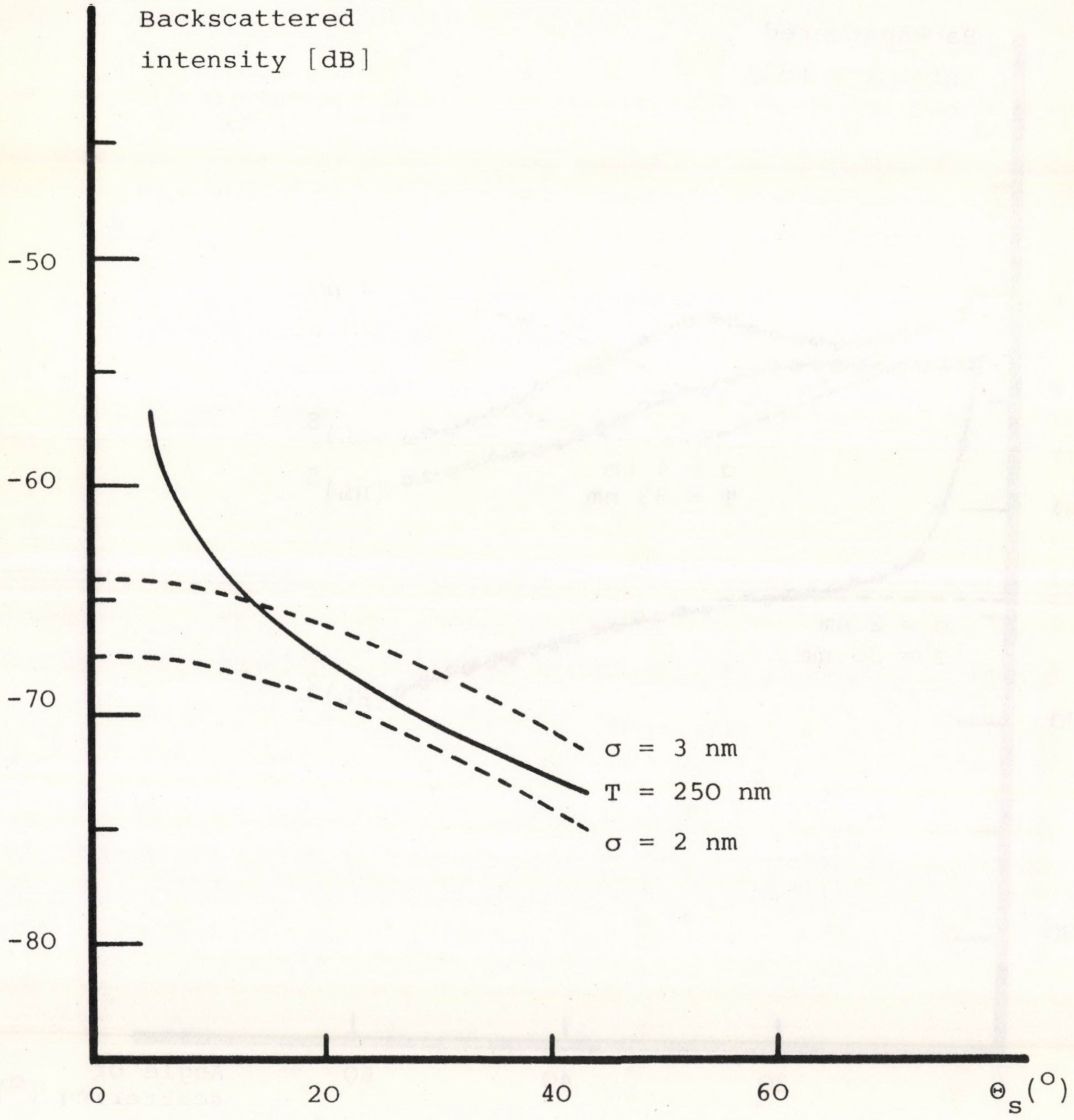

Fig. 8
Light scattering of TiO_2/SiO_2 laser mirrors

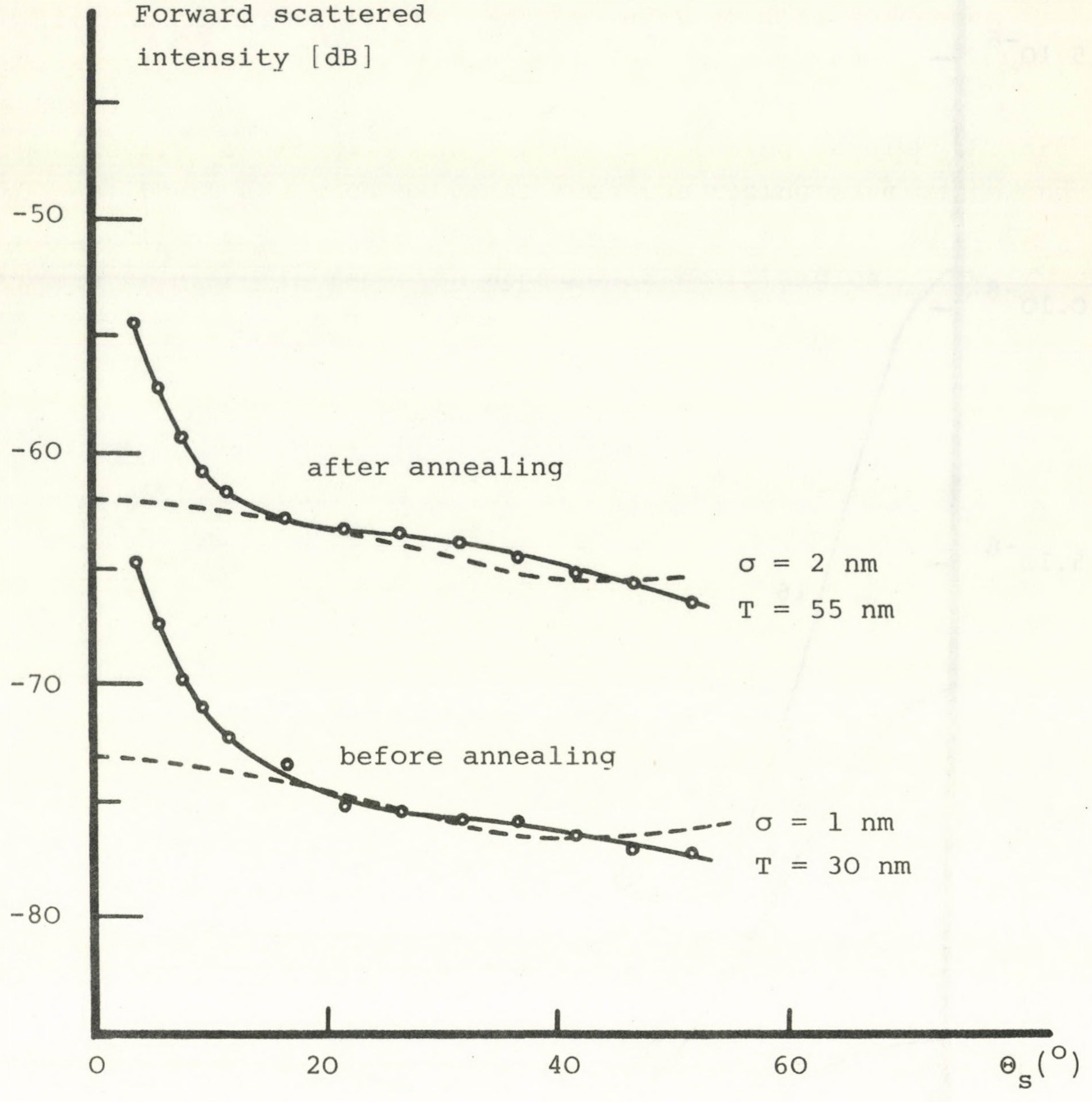

Fig. 9

Light scattering of a homogeneous TiO_2 *film before and after annealing*

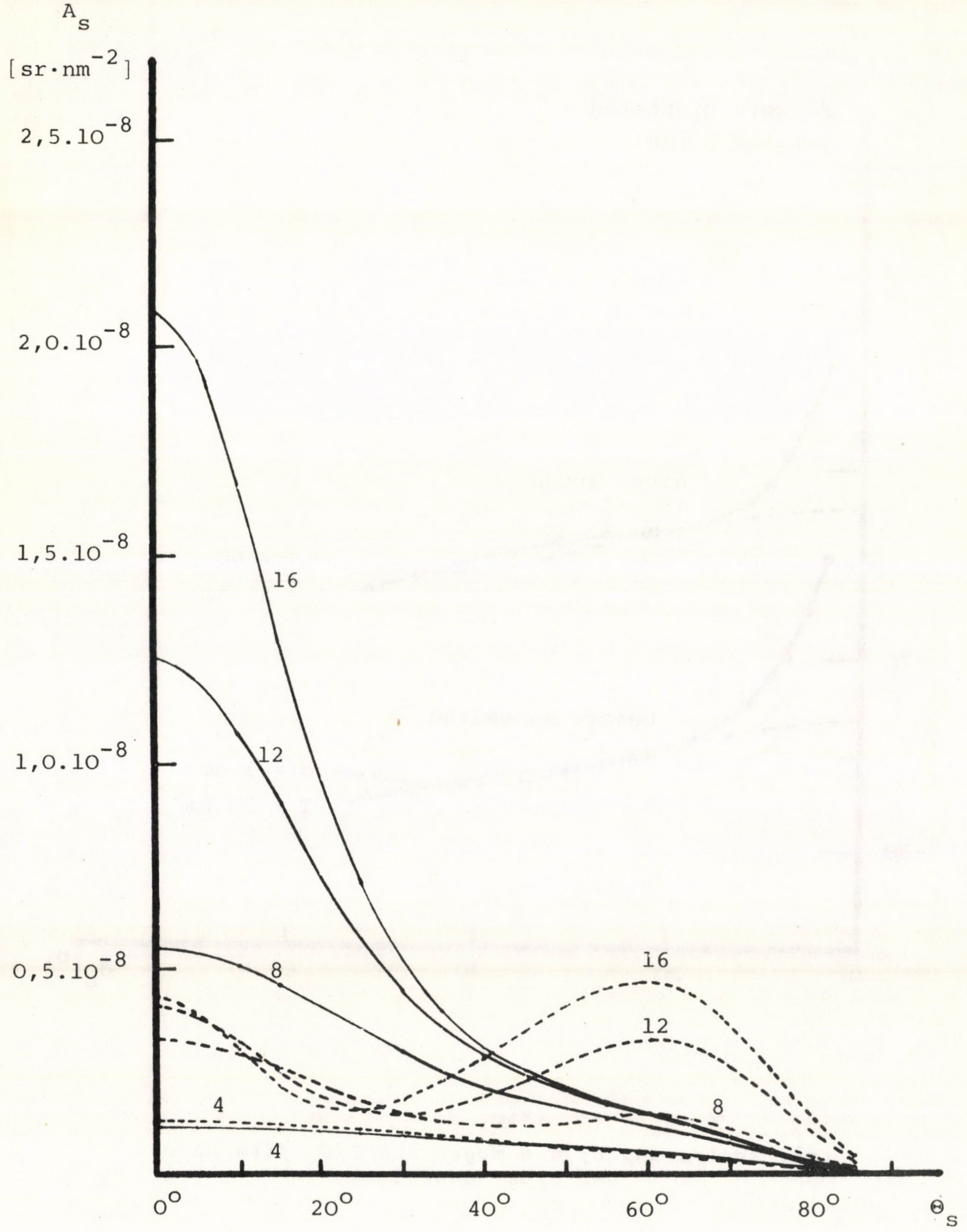

Fig. 10

The cross correlation effect in light scattering appearing in the case of multiple beam interference the light wavelength differ from each other. The evaluated As versus angle of scattering is shown on *Fig. 10* in the cases of different number of layers. In the cases of good coherence between the wave fronts /identical film structures/ extremum appears.

Really, on Figs. 6 and 7 where the tuning wavelengths are 500 nm, there are such extrema. When the tuning wavelength and the light wavelength are nearly equal /Fig. 8/ this modification of the angular distribution appears at small angles.

SUMMARY, CONCLUSIONS, PROBLEMS

The Elson model was employed for describing light scattering of multilayer thin films. After developing a computer program and fitting the measured angular distributions of the scattered light to the computed curves, we could obtain the statistical parameters of the surface roughnesses. These parameters were similar to those obtained from results of electronmicroscopic examinations. Increasing the number and the thickness of the layers the roughness of the interfaces increased.

The properties of film scattering show, that the cross correlation caused by multiple beam interference increases the intensity in directions other than the ones defined by geometrical optics.

In our calculations a constant surface statistics was supposed for each boundary. Although this assumption is not perfectly correct since according to our results the statistical parameters of the boundaries change interface by interface - the model seems to be suitable for characterising the distribution of the scattered light, because the fluctuation of the surfaces is small enough and does not destroy the coherence properties of the scattered light.

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