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ELLIPSOMETRIC FORMULAS FOR AN INDEX PROFILE OF SMALL AMPLITUDE BUT ARBITRARY SHAPE

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Résumé - Le plus souvent les réflectances de couches non homogènes sont calculées par résolution numérique des équations de Maxwell. Pour certains problèmes physiques cette procédure ne permet pas bien de résoudre le problème inverse, c'est-à-dire de déterminer le profil d'indice $n(z)$ à partir des mesures ellipsométriques (ψ et Δ). Ici nous calculons les réflectances explicitement pour n'importe quelle forme de $n(z)$ par une approximation de Born, valable au premier ordre en $n(z) - n_0$ (où n_0 est l'indice du milieu d'incidence). Par contre, l'effet de la paroi réfléchissante (en $z = 0$) est incorporé dans le problème non perturbé. ψ et Δ sont ainsi exprimés en fonction de la transformée de Fourier complexe $\Gamma(2q) = \Gamma' + i\Gamma''$ du profil (où q est la composante normale du vecteur d'onde incident). Pour des couches épaisses ($e \gg \lambda/4\pi$) ceci doit permettre une reconstruction complète du profil. Pour des couches minces ($e \ll \lambda/4\pi$) on détermine seulement les premiers moments du profil d'indice. Pour illustrer ces techniques, nous discutons deux exemples qui font intervenir un profil d'indice lentement décroissant : (i) effets de paroi sur un mélange binaire critique; (ii) adsorption de polymères flexibles à partir d'un bon solvant.

Abstract - The reflectance of non homogeneous layers is usually calculated by numerical solution of the Maxwell equations. This requires a specific model for the layer structure. We are interested here in the inverse problem : to find the refraction index profile $n(z)$ from the ellipsometric data (ψ and Δ). We have calculated the reflectances explicitly in a 1st Born approximation (i.e. to first order in $n(z) - n_0$ where n_0 is the index of the pure liquid). The effect of the reflecting wall at $z = 0$ is incorporated exactly. Finally we express ψ and Δ in terms of the complex Fourier transform $\Gamma(2q) = \Gamma' + i\Gamma''$ of the profile (where q is the normal component of the incident wave vector). For thick diffuse layers ($e \gg \lambda/4\pi$) this should allow for a complete reconstruction of the profile. For thin layers ($e \ll \lambda/4\pi$) what is really measured is the moments Γ_0 and Γ_1 (of order 0 and 1) of the index profile. To illustrate these methods, we discuss two specific examples, which are associated with a slowly decreasing index profile : (i) wall effects in critical binary mixtures; (ii) polymer adsorption from a good solvent.

Reflectance measurements can give us a rather detailed information on the structure of diffuse interfaces, even when the thickness e of the interface is somewhat smaller than the optical wavelength λ : we can probe conveniently values of $e \sim \lambda/4\pi \sim 500 \text{ \AA}$. For oblique incidence, one can define two complex reflectance coefficients, R_s and R_p corresponding to the two polarisation states "s" and "p". The quantity of experimental interest is the ratio : $R_p/R_s \equiv \tan \psi e^{i\Delta}$. For simple situations, such as a wall covered by a slab of thickness e and constant refractive index n_1 , we have relatively simple formulas for R_s and R_p [1,2]. There are a number of cases, however, where this model is not satisfactory. We shall quote two examples of current interest a) a critical binary liquid mixture, near a wall, shows a profile of a concentration

ϕ (versus distance to the wall, z) of the form /3,4/ : $\phi(z) - \phi(\infty) = (\text{const.})(a/z)^{\beta/\nu}$ where β and ν are critical exponents ($\beta/\nu \sim 0.5$ in three dimensions) and a is a molecular size;

b) an adsorbed layer of flexible polymers (in a good solvent) is expected to show a concentration profile /5,6/ : $\phi(z) = \text{const.}(a/z)^{4/3}$. In both cases (a) and (b), $\phi(z)$ is a slowly decreasing function of z , and it is not at all permissible to replace the profile by a slab of finite thickness e and constant index n_1 : if we did this, the apparent thickness e would become a function of the experimental conditions (wavelength, angle of incidence).

Another important feature of cases (a) and (b) is that there is a cutt off $z_{\max}$ to the profile. In case (a), $z_{\max}$ is the correlation length ξ and is finite when we are not exactly at the critical point. In case (b), $z_{\max}$ is the coil size /6/. One of the questions to be clarified is the influence of this cut off on Δ and ψ .

This has led us to search for a general formulation where ψ and Δ would be written explicitly as functionals of the profile. It is not too hard to realise this program when the local perturbation $\phi(z) - \phi(\infty)$ is small. We derive the reflectance amplitudes to first order in this local perturbation, by a procedure reminiscent of the Born approximation in quantum mechanics /7/. The problem is not entirely classical, however, because we incorporate in the 0 order wave functions the effect of the reflecting wall (at $z = 0$).

After some calculation we can write the ellipsometric ratio to first order in δn as :

$$R_p/R_s = \tan \psi e^{i\Delta} = r_p/r_s (1 + iA\Gamma(-2q) + iB\Gamma(2q)) \quad \text{where :}$$

- r_s and r_p are the reflectances for the wall in direct contact with a half space of index n_0 (without diffuse layer);

- A and B are two known functions of q and of the unperturbed reflectances :

$$A(q) = a(q)(r_p^{-1} - r_s^{-1}) - b(q) r_p^{-1} \quad \text{with} \quad a(q) = n_0(w/c)^2 q^{-1}$$

$$B(q) = a(q)(r_p - r_s) - b(q) r_p \quad b(q) = 2 n_0^{-1} q$$

- and $\Gamma(k)$ the Fourier transform of the index profile

$$\Gamma(k) = \int_0^\infty dz \delta n(z) e^{ikz} = \Gamma^*(-k).$$

Our discussion of what is really measured in an ellipsometric experiment is restricted to diffuse layers where the refractive index is not too different from the bulk value. But within these limits we can discuss arbitrary shapes for the index profile. Two limiting cases are of particular interest :

(i) thick layers where one can reconstruct the profile from the data by standard Fourier transform procedures. (Note that the Kramers Kronig relations give us some extra information);

(ii) thin layers (thickness $z_{\max}$ below $\lambda/4\pi$ where λ is the optical wavelength) where one may expand R_p/R_s in powers of $(4\pi z_{\max}/\lambda)$. The data give only two informations : the first moments Γ_0 and Γ_1 of the profile. We recover here a set of relations between reflectances and moments which was first constructed by Lekner /8/.

For this second case, it is sometimes instructive to define an apparent thickness e_a for the layer : $e_a = 2\Gamma_1/\Gamma_0$. For a homogeneous slab of thickness e , we return to $e_a = e$ exactly). When we analyse the moment structure for the examples of diffuse layers :

a) for a critical binary fluid, where the profile decreases slowly like $z^{-1/2}$, the (0)

moment is very dependent on the cut off ξ (the correlation length) $r_0 \sim \xi^{1/2}$ and the apparent thickness is of order ξ : in this case e_a is a rather good image of the overall size;

b) for flexible polymer adsorption, the profile is slightly steeper ($z^{-4/3}$) and this leads to very different conclusions. The $\langle 0 \rangle$ moment is insensitive to the cut off R_F (the coil size), and the effective thickness e_a is not proportional to R_F , but to a weaker power $e_a \sim R_F^{2/3}$. Thus this second example shows very vividly the pitfalls of all models based on homogeneous slabs.

References

- /1/ - AZZAM R., BASHARA N.B., "Ellipsometry and polarised light", North Holland, Amsterdam (1977).
- /2/ - BORN M., WOLF E., "Principles of optics", Pergamon Press (1975).
- /3/ - FISHER M., de GENNES P.G., C.R. Acad. Sci. (Paris) B287 (1978) 207.
AU-YANG H., FISHER M., Phys. Rev. B21 (1980) 3956.
AU-YANG H., FISHER M., Physica 101A (1980) 255.
- /4/ - FRANK C., SCHNATTERLEY S., Phys. Rev. Lett. 48 (1982) 762.
- /5/ - de GENNES P.G., "Scaling concepts in polymer physics", Cornell U. Press, Ithaca (NY) (1979).
- /6/ - de GENNES P.G., Macromolecules 14 (1981) 1637.
- /7/ - MORSE P.M., FESHBACH H., "Methods of theoretical physics", Mc Graw Hill, ch. 9.3 (1953).
- /8/ - LEKNER J., Physica 112A (1982) 544.