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Abstract - A general theory, of second order in the film thickness and surface roughness, is developed of the optical properties of a thin continuous film. These properties are described by a small number of electromagnetic constitutive coefficients. Formulae for these coefficients are derived in terms of the height-height correlation functions of the upper and lower surfaces of the film, and its average thickness. The reflectance, transmittance and ellipsometric coefficient are expressed in terms of the constitutive coefficients, for arbitrary angles of incidence. The results of this work are also studied in the long wave length limit of surface roughness and compared with results, obtained earlier by Ohlidal and co-workers.

1. INTRODUCTION - Optical properties of surfaces are strongly influenced by roughness and the presence of thin films. We shall study these influences by considering a thin continuous isotropic film (liquid or solid, average thickness \(d<<\text{wave-length } \lambda\)), with dielectric constant \(\varepsilon\), covering the rough surface of a substrate (liquid or solid) with dielectric constant \(\varepsilon'\). The function \(z=f_+(\mathbf{r}_\parallel,t)\) and \(z=f_-^{\pm}(\mathbf{r}_\parallel,t)\) are assumed to be single valued and differentiable with local normals \(\hat{\mathbf{v}}_+^{\pm}(\mathbf{r}_\parallel,t)\) at the interfaces. The boundary conditions for the electro-magnetic fields \(\mathbf{E}\) and \(\mathbf{H}\) and the displacement fields \(\mathbf{d}\) and \(\mathbf{f}\) at the interfaces are given by

\[
\begin{align*}
\mathbf{E}_+^{\pm}(\mathbf{r}_\parallel,t) \cdot \hat{\mathbf{v}}_+^{\pm}(\mathbf{r}_\parallel,t) & = 0, \\
\mathbf{H}_+^{\pm}(\mathbf{r}_\parallel,t) \cdot \hat{\mathbf{v}}_+^{\pm}(\mathbf{r}_\parallel,t) & = 0,
\end{align*}
\]

where the subscripts + and - denote the fields in the substrate and ambient respectively, whereas fields without superscripts are within the film. From the Maxwell equations, together with the boundary conditions (1) and the statistical properties...
of the rough surfaces, it is possible to calculate the optical properties of the system. This will be done in the next sections.

2. DESCRIPTION WITH FLUCTUATING SURFACE SUSCEPTIBILITIES - The system described in section 1 can be replaced by the following system: a flat substrate with dielectric constant $\varepsilon^+$, filling the half space $z>0$, an ambient with dielectric constant $\varepsilon^-$, filling $z<0$, and singular polarization and magnetization densities $\delta^+(r, t)$ and $\delta^-(r, t)$, respectively. These densities, which can be calculated from the excess polarization and magnetization in the film /1/, produce the same fields in the regions where $z>f^+(r, t)$ and $z<f^-(r, t)$, as were present in the original system, described in section 1 (c.f. also ref /2/). The surface polarization and magnetization densities $\delta^S$ and $\delta^n$ can be expressed in terms of the extrapolated fields /1/ $\delta^S = \pm (\varepsilon^+_x, \varepsilon^+_y, d^S_z)$ and $\delta^n = (\varepsilon^-_x, \varepsilon^-_y, b^S_z)$ on both sides of $z=0$. Up to second order in $f^-$ one finds

$$\delta^S(r, t) = \sum_{i=1}^{3} \xi_0^{(i)} \nabla^i \delta^+(r, t) \cdot \delta^-(r, t) + c^{-1} \xi_0^{(4)} \nabla^4 \delta^+(r, t) \cdot \delta^-(r, t) / \partial t,$$

$$\delta^n(r, t) = c^{-1} \xi_0^{(4)} \nabla^4 \delta^+(r, t) \cdot \delta^-(r, t) / \partial t,$$

where $\nu = \pm$ or $-$. The fluctuating surface susceptibilities are given by

$$\xi_0^{(1)} (r, t) \equiv \varepsilon^-(\varepsilon^+-\varepsilon^-) \nabla^1 \delta^+(r, t) \cdot \delta^-(r, t) / (2 \varepsilon^+),$$

$$\xi_0^{(2)} (r, t) \equiv \varepsilon^+(\varepsilon^+-\varepsilon^-) \{ \nabla^2 \delta^+(r, t) - \varepsilon^-(\varepsilon^+-\varepsilon^-) \nabla^2 \delta^-(r, t) \},$$

$$\xi_0^{(3)} (r, t) \equiv \varepsilon^+(\varepsilon^+-\varepsilon^-) \{ \nabla^3 \delta^+(r, t) - \varepsilon^-(\varepsilon^+-\varepsilon^-) \nabla^3 \delta^-(r, t) \},$$

$$\xi_0^{(4)} (r, t) \equiv \varepsilon^-(\varepsilon^+-\varepsilon^-) \{ \nabla^4 \delta^+(r, t) - \varepsilon^-(\varepsilon^+-\varepsilon^-) \nabla^4 \delta^-(r, t) \}.$$

3. SURFACE CONSTITUTIVE COEFFICIENTS - These are found by expressing the average surface polarization and magnetization densities $\bar{\delta}^S$ and $\bar{\delta}^n$ in terms of the average fields $\bar{\delta}^S = \langle \delta^S \rangle$ and $\bar{\delta}^n = \langle \delta^n \rangle$. One obtains, up to second order in the surface roughness and film thickness /3/

$$\bar{\delta}^S = \sum \xi_e \langle \delta^S \rangle_e + c^{-1} \xi_m \langle \delta^n \rangle_m / \partial t,$$

$$\bar{\delta}^n = c^{-1} \xi_m \langle \delta^n \rangle_m,$$

with

$$\xi_e \equiv \sum_{i=1}^{3} \xi_e \langle \delta^S \rangle_e + \xi_e \langle \delta^n \rangle_m,$$

$$\xi_m \equiv \xi_m \langle \delta^n \rangle_m.$$
versal. For \( \gamma_\perp \) one finds

\[
\gamma_\perp (k_\parallel, \omega) = 2\nu (\varepsilon - \varepsilon') < \nu' > + \frac{1}{\nu} \sum_{\nu'} \nu\nu' (\varepsilon - \varepsilon') (\varepsilon - \varepsilon')
\]

\[
\times \left\{ \cos^2 \phi \delta_1^+ \delta_1^- (\varepsilon_1^+ - \varepsilon_1^-)^{-1} + \sin^2 \phi (\omega'/c)^2 (q_1^+ + q_1^-)^{-1} \right\} \equiv 2\nu (\varepsilon - \varepsilon') < \nu' > + \gamma_\perp \gamma_\perp (k_\parallel, \omega), \tag{8}
\]

where \( S_{\nu\nu'} (k_\parallel, \omega) \equiv S_{\nu\nu'} (k_\parallel)^{2\pi6}(\omega) \) is the Fourier transform of eq. (6), \( q_1^\pm \equiv \left\{ \varepsilon (\omega'/c)^2 - k_\parallel^2 \right\}^{-1} \) and \( \phi \) is the angle between \( k_\parallel \) and \( k_\parallel' \). Similar expressions can be derived for \( \gamma_{\parallel\parallel}, \beta_{\perp}, \delta_{\perp}, \eta_{\perp} \) and \( \tau_{\perp} \) and are given in ref. /3/. The constitutive coefficients can in principle be evaluated, if the 4 correlation functions \( S_{\nu\nu'} \) are known, together with \( d, \varepsilon^+, \varepsilon^- \) and \( \varepsilon \).

4. REFLECTANCE, TRANSMITTANCE AND ELLIPSOmetric COEFFICIENT - Using the constitutive equations (4) and the boundary conditions for the fields in \( z=0 \) (see e.g. /4/), one can express the reflectance \( R \), transmittance \( T \) and ellipsometric coefficient \( r \) in terms of the surface constitutive coefficients, using eq. (7). One then obtains for \( s^+ \) and \( p^- \) polarized light:

\[
R_{s,p} = \left| r_{s,p} \right|^2, \quad T_{s,p} = \left| t_{s,p} \right|^2 (\cos \Theta_\perp)/(\cos \Theta_\perp) \quad r = r_{s,p} / r_{p,s}, \tag{9}
\]

with the amplitudes, valid up to second order in the surface roughness and film thickness:

\[
t_{s} = t_{s,t}^0 (d) \left\{ 1 + i (\omega / c) \gamma_{\perp} (c) \left( \cos \Theta_\perp + \cos \Theta_\perp \right) \right\}^{-1} - (\omega / c)^2 (n^+ \cos \Theta_\perp + n^- \cos \Theta_\perp)^{-1} (n^+ \cos \Theta_\perp + n^- \cos \Theta_\perp)^{-1} \quad r_{s} = r_{s,t}^0 (d) \left\{ 1 + i (\omega / c) \gamma_{\perp} (c) 2 n^+ (\cos \Theta_\perp)^{-1} \cos \Theta_\perp \right\} + (\omega / c)^2 (n^+ \cos \Theta_\perp)^{-1} (n^- \cos \Theta_\perp)^{-1} \cos \Theta_\perp \right\} \tag{10}
\]

and analogous expressions for \( t_p \) and \( r_p \) /3/. In the above equations \( \Theta \) and \( \Theta_\perp \) are the angles of incidence and transmission, \( t_{s,t}^0 (d) \) and \( r_{s,t}^0 (d) \) the transmission and reflection amplitudes for the plane parallel film with thickness \( d, n^\pm \equiv \sqrt{\varepsilon^\pm} \), whereas \( \gamma_{\perp} (c) \equiv \frac{1}{2} \gamma_{\perp} (c) \) and \( \tau_{\perp} (c) \equiv \frac{1}{2} \tau_{\perp} (c) \) describe the influence of surface roughness (cf. eq. (8)).

5. LONG WAVE LENGTH LIMIT OF SURFACE ROUGHNESS; COMPARISON WITH EARLIER WORK -

We shall assume that in this limit the correlation functions \( S_{\nu\nu'} (k_\parallel) \) may be approximated by \( \delta \)-functions in eq. (8) and corresponding equations for \( \gamma_{\perp}, \beta_{\perp}, \delta_{\perp}, \ \eta_{\perp}, \ \tau_{\perp} \), so that integrations can easily be performed. For normally incident light we then obtain /5/ the same result for \( R \) as Ohldal, Navrát, and Lukeš /6/.

In order to calculate the ellipsometric coefficient \( r \), we have to make the better approximation

\[
S_{\nu\nu'} (k_\parallel) \approx 4\pi \delta (\Delta ^2) > 0 \delta_{\perp} (k_\parallel) \times 4\pi^2 \left\{ < \Delta ^2 > 2 \right\} \{ \delta (k_\parallel) \delta (k_\parallel) + \delta (k_\parallel) \delta (k_\parallel) \} \tag{11}
\]

in eq. (8) and corresponding equations for \( \gamma_{\perp}, \beta_{\perp}, \delta_{\perp} \), etc. Otherwise we would find no influence of surface roughness. We assume here identical films, i.e. all 4 correlation functions \( S_{\nu\nu'} \) are equal with same \( < \Delta ^2 > 2 \) and same correlation length \( \ell \) (\( \ell > > \lambda \) in considered limit). The symbol \( \delta'' \) denotes second derivative of the \( \delta \)-function. With eq. (11) it is again possible to evaluate \( \gamma_{\perp}, \beta_{\perp} \), etc. /5/. One then obtains, using eqs. (9) and (10), the following second order expression for \( r \):

\[
r = r_{s,t}^0 (d) + r_{p,t}^0 (d) + (< \Delta ^2 > 2 ) F (\Theta_\perp, \varepsilon^-, \varepsilon^+) \tag{12}
\]
where \( r^0(d)/r^0_{B}(d) \) is the ellipsometric coefficient of the plane parallel film with thickness \( d \), whereas the last term describes the effect of surface roughness. The function \( F(\Theta, \varepsilon^-, \varepsilon^+) \) is rather complicated and given explicitly in ref. [5]. For the Brewster angle \( \Theta_B^0 = \arccos \frac{1}{n} \left( \varepsilon^+ \right)^{-\frac{1}{2}} \), one gets

\[
F(\Theta_B^0, \varepsilon^-, \varepsilon^+) = -\frac{1}{2} \left( \varepsilon^+ \right)^{-3} \left( \varepsilon^- \right)^{-1} \cdot \left\{ \left( \varepsilon^+ \right)^2 - \left( \varepsilon^- \right)^2 \right\} \cdot \left\{ \left( \varepsilon^+ \right)^2 + 4\varepsilon^+ \varepsilon^- + 2\left( \varepsilon^- \right)^2 \right\}.
\]

(13)

This result is different from that obtained by Ohlidal and Lukes, using the Helmholtz-Kirchhoff integral [7], but also from the result obtained by the same authors, using the Stratton-Chu-Silver integral [8]. In the first case these authors obtain a contribution to \( r \) from surface roughness, which does not vanish for \( \Theta=0 \), as it should do for isotropic surfaces. So we only have to compare our result with that of reference [8]. It can be shown [5] that the difference in results is due to an inconsistency in the assumptions made by Ohlidal and Lukes [7,8] in order to calculate the local electro-magnetic field on a roughness surface. We show [5], by a direct calculation of this field, up to second order in the surface roughness, that it differs from the local field, postulated by Ohlidal and Lukes [7,8]. Repeating their calculations with our fields, we find that both methods (H.K. and S.C.S.) lead to our result, eqs. (12) and (13). We furthermore prove in ref. [5] that the local field on a rough surface, postulated by Ohlidal and Lukes [7,8], can be obtained by neglecting all terms with second derivatives of \( F^x \) with respect to \( x \) and \( y \) in our calculation. This is in agreement with the fact that these authors neglect local curvature of the rough surfaces. We prove, however, in ref. [5], that it is inconsistent to make this approximation in a second order calculation of the fields. It seems to be a pure coincidence that for the numerical example of the Si-Si\(_2\) system, given by Ohlidal and Lukes [7], our formulae (12) and (13) give, within a few per cents, the same results as theirs.

REFERENCES