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ABSTRACT

Sum rules are derived for the logarithm of the absolute value and the phase angle of the amplitude for normally reflected radiation.

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Many sum rules, involving different optical parameters, have been obtained up to date[1-4]. Unfortunately, the methods employed in the derivation of these sum rules were not directly applicable in the case of the near normal reflectivity which is the experimentally most "accessible optical quantity over a wide frequency range"[1]. The purpose of this note is to fill the gap presenting such sum rules for the reflectivity coefficients.

Some time ago Jahoda[5], following earlier suggestions[6], pointed out the advantages of the reflectivity method for obtaining the absorption coefficient. What is directly measured is the normally reflected intensity which is proportional to the reflectance coefficient $R=|r|^2$; r being given by the Fresnel equation for the amplitude of normally reflected radiation, namely

$$r = (N-1)/(N+1) = |r|e^{i\theta} \quad . \tag{1}$$

Here, $N=n+i\kappa$ is the complex index of refraction.

Were $R(\omega)$ known for all frequencies, the phase $\theta(\omega)$ could be obtained from the dispersion relation[5]

$$\theta(\omega) = \frac{\omega}{\pi} P \int_0^\infty \frac{\ell n \ R(\omega')d\omega'}{\omega^2 - \omega'^2} \quad , \tag{2}$$

where P stands for the principal value. It would then be easy to calculate the (real) refractive index n and the extinction coefficient κ by solving the sistem of equations

$$R = \left[(n-1)^2 + \kappa^2\right]/\left[(n+1)^2 + \kappa^2\right] \quad ,$$

$$\tan\theta = 2\kappa/(n^2-1+\kappa^2) \quad . \tag{3}$$

Since it is not possible in practice to measure R for all frequencies, its behavior beyond the available frequency range has to be somehow estimated[7,8]. It is for this reason that sum rules might be very useful in putting restrictions on possible extrapolations for R.

In order to obtain sum rules for some function it is, of course, not sufficient to know that it obeys a dispersion relation. It is also necessary that the function vanishes sufficiently fast at infinity. The knowledge of the asymptotic behavior of most optical functions is based on the well justified assumption that at very high frequencies any medium should respond like a free electron gas. This condition is easily translated into

$$\lim_{\omega\to\infty} \omega^2[n(\omega)-1] = -\frac{1}{2}\,\omega_p^2 \quad , \quad \lim_{\omega\to\infty}[\kappa(\omega)]\,\omega^2 = 0 \quad , \tag{4}$$

where ω_p is the plasma frequency. It is slightly less general to assume, for the extinction coefficient, the asymptotic behavior

$$\kappa(\omega) \to \bar{\gamma}(\omega_p^2/\omega^3) \quad , \tag{5}$$

where $\bar{\gamma}$ is a non-negative constant. In the Lorentz model[9] $\bar{\gamma}$ is the average of the damping coefficients.

Our derivation of the sum rules will be based on the following function:

$$F(\omega) = (4/\omega_p^2)\ [b^2\omega_p^2 - \omega^2 - ia\,\omega_p\omega]\ r(\omega) \quad , \tag{6}$$

where a and b are constants to be determined later. The asymptotic behavior of its logarithm is

$$\ell n\ F(\omega) \to \Big\{\big(\tfrac{1}{4}-b^2\big)(\omega_p/\omega)^2 + a\bar{\gamma}(\omega_p/\omega^2) +$$

$$+ (i/\omega)\left[a\,\omega_p - \bar{\gamma} + (\omega_p/\omega)^2\big(b^2\bar{\gamma} - \tfrac{1}{2}\bar{\gamma} + \tfrac{a}{4}\omega_p\big)\right]\Big\} \quad , \tag{7}$$

and, besides, $\ell n\ F(\omega)$ is analytic in the upper half of the ω complex plane. By the Phragmēn-Lindelöf theorem the asymptotic behavior (9) holds in any direction of that half plane. Furthermore, the crossing relations $F^*(\omega) = F(-\omega)$, $[\ell n\ F(\omega)]^* = \ell n\ F(-\omega)$ can be shown to hold. These conditions are sufficient to guarantee the existence of the relations[9]

$$Re\ \ell n\ F(\omega) = \frac{2}{\pi}\, P \int_0^\infty \frac{d\omega'\ \omega'\ Im\ \ell n\ F(\omega')}{\omega'^2 - \omega^2} \quad , \tag{8}$$

$$Im\ \ell n\ F(\omega) = -\frac{2\omega}{\pi}\, P \int_0^\infty \frac{d\omega'\ Re\ \ell n\ F(\omega')}{\omega'^2 - \omega^2} \quad . \tag{9}$$

When we multiply Eq. (8) by ω^2 and take the $\omega\to\infty$ limit, using Eq. (7), we get

$$\int_0^\infty d\omega\ \omega\ Im\ \ell n\ F(\omega) = -\frac{\pi}{2}\,\omega_p\big(\tfrac{1}{4}\omega_p + a\bar{\gamma} - b^2\omega_p\big) . \tag{10}$$

With $F(\omega)$ being as in Eq. (6) and with the choice of a frequency Ω above which the behavior (7) approximately holds, Eq. (10) can be rewritten as

$$\int_0^\Omega d\omega\ \omega\left[\theta(\omega) - \tan^{-1}\frac{a\,\omega\,\omega_p}{b^2\omega_p^2 - \omega^2}\right] = -\frac{\pi}{2}\,\omega_p\big(\tfrac{1}{4}\,\omega_p + a\bar{\gamma} - b^2\omega_p\big)$$

$$+ \int_\Omega^\infty d\omega\ \left[a\,\omega_p - \bar{\gamma} + (\omega_p/\omega)^2\big(b^2\bar{\gamma} - \tfrac{1}{2}\bar{\gamma} + \tfrac{a}{4}\omega_p\big)\right] . \tag{11}$$

This equation only makes sence for $a = \bar{\gamma}/\omega_p$ in which case we have

$$\int_0^\Omega d\omega\ \omega\left[\theta(\omega) - \tan^{-1}\frac{\omega\,\bar{\gamma}}{b^2\omega_p^2 - \omega^2}\right] = -\frac{\pi}{2}\left[\omega_p^2\big(\tfrac{1}{4} - b^2\big) + \bar{\gamma}^2\right] + \frac{\bar{\gamma}\omega_p^2}{4\Omega}\,(1 - 4b^2) . \tag{12}$$

With $b^2=\bar{\gamma}^2/\omega_p^2$ in the last equation we get, in the $\Omega\to\infty$ limit,

$$\int_0^\infty d\omega\, \omega \left[\theta(\omega) - \tan^{-1}\frac{\omega\bar{\gamma}}{\bar{\gamma}^2-\omega^2}\right] = -\frac{\pi}{8}\omega_p^2 \quad , \tag{13}$$

which shows that asymptotically $\theta\to\pi$.

The crossing relation $N^*(\omega)=N(-\omega)$ leads to a similar relation for r , i.e., $r^*(\omega)=r(-\omega)$ or

$$|r(-\omega)| = |r(\omega)| \quad , \quad \theta(-\omega) = -\theta(\omega) \quad . \tag{14}$$

Thus, the square bracket in Eq. (13) is, for large ω , of order $O(\omega^{-3})$. An integration by parts of that equation yields a sum rule for the slope $\theta' = d\theta/d\omega$,

$$\int_0^\infty \omega^2\, d\omega \left[\theta'(\omega) - \frac{\bar{\gamma}\,(\omega^2+\bar{\gamma}^2)}{(\omega^2-\bar{\gamma}^2) + \omega^2\bar{\gamma}^2}\right] = \frac{1}{4}\pi\omega_p^2 \quad . \tag{15}$$

It is, of course, easy to obtain sum rules for higher derivatives by further integrating by parts.

Let us now proceed to derive sum rule for $\ln R$. If we multiply Eq. (9) by ω , take the $\omega\to\infty$ limit and use Eq. (7) we get

$$\int_0^\infty d\omega\ \mathrm{Re}\ \ln F(\omega) = \frac{\pi}{2}\ (a\omega_p-\bar{\gamma}) \ . \tag{16}$$

Let Ω be again a frequency above which $\ln F(\omega)$ behaves as in Eq. (7). Then, using Eqs. (6), (7) and (16) we have ,

for 2b=a,

$$\int_0^\Omega d\omega \ln\left(a^2+\frac{4\omega^2}{\omega_p^2}\right)^2 R(\omega) =$$

$$= \pi(a\omega_p-\bar{\gamma}) - \frac{\omega_p}{\Omega}\left[\frac{1}{2}\omega_p + 2a\bar{\gamma} - \frac{1}{2}a^2\omega_p\right], \tag{17}$$

for any number a. The sum rule that holds in the $\Omega\to\infty$ limit is now

$$\int_0^\infty d\omega \ln\left(a^2 + \frac{4\omega^2}{\omega_p^2}\right)^2 R(\omega) = \pi(a\omega_p-\bar{\gamma}) \ . \tag{18}$$

The sum rules we have derived, depending as they are on $\bar{\gamma}$, will be useful whenever that parameter could be estimated. On the other hand, a knoledge of the reflectance will allow us to compute $\bar{\gamma}$. From Eq.(18) we get, in the $a\to 0$ limit,

$$\bar{\gamma} = -\frac{1}{\pi}\int_0^\infty d\omega\ \ln\left[(2\omega/\omega_p)^4 R(\omega)\right] \ . \tag{19}$$

In deriving the sum rules we have ignored a possible Blaschke product[9-11]. As Stern has argued[11], Blaschke product contribution to the reflectivity coefficients can be ruled out on physical grounds. Stern also discusses at some length the dispersion relations for the reflectivity coefficients. On the other hand, the sum rules obtained in the present paper seem to be new.

According to the sum rule (18) (or (19)) at very high frequencies, R behaves as $(\omega_p/2\omega)^4$. But for that we did not need the sum rules; Eqs. (1), (3) and (4) almost immediately lead to that asymptotic behavior. The sum rules might be very usefull, however, in the region of intermediate energies (between the measurable frequency range and the asymptotic region). There, they might provide a decisive test for different interpolations. In Ref. (7), for instance, Philipp and Taft interpolated the reflectance of Ge between 11 and 30 eV with the function $R = R_o(E_o/E)^A$ with $A \approx 1.8$ while Rimmer and Dexter[8] used, in the same region, $R = R_o \exp a(E_o - E)$. It would be interesting to check the sum rule (18) with their data and interpolating functions.

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