280(2): One Photon Reflection Theory with the Memory Function Theory of Molecular Dynamics

Consider the reflection of one photon from a material of refractive index n_1. The equations of conservation of energy and momentum are:

$$\hbar\omega = \hbar\omega_1 + \hbar\omega_2 \quad -(1)$$

and

$$\hbar\underline{\kappa} = \hbar\underline{\kappa}_1 + \hbar\underline{\kappa}_2 \quad -(2)$$

where ω is the incident angular frequency and ω_1 and ω_2 are the refracted and reflected angular frequencies. The incident wavenumber is $\underline{\kappa}$ and the refracted and reflected wavenumbers are $\underline{\kappa}_1$ and $\underline{\kappa}_2$. Therefore:

$$\omega_1 = \omega - \omega_2 \quad -(3)$$

and

$$\underline{\kappa}_1 \cdot \underline{\kappa}_1 = \underline{\kappa} \cdot \underline{\kappa} + \underline{\kappa}_2 \cdot \underline{\kappa}_2 - 2\,\underline{\kappa} \cdot \underline{\kappa}_2 \cos 2\theta \quad (4)$$

where θ is the angle of incidence, equal to the angle of reflection by Snell's Law.

Assume that the incident wave travels in air at phase velocity c. The phase velocity of the refracted wave is v_1, and the phase velocity of the reflected wave is c. We have:

$$\kappa_1 = \frac{\omega_1}{v_1}, \quad \kappa = \frac{\omega}{c}, \quad \kappa_2 = \frac{\omega_2}{c} \quad -(5)$$

and: $$\frac{1}{v_1^2} = \frac{n_1^2}{c^2} \quad -(6)$$

Therefore:

$$n_1^2 \omega_1^2 = \omega^2 + \omega_2^2 - 2\omega\omega_2 \cos 2\theta \quad -(7)$$

From eq. (3):

$$\boxed{n_1^2 (\omega - \omega_2)^2 = \omega^2 + \omega_2^2 - 2\omega\omega_2 \cos 2\theta} \quad -(8)$$

So ω_2 can be found in terms of the variables ω, θ and n_1.

The real part of n_1 is given by:

$$n_1'^2 = \frac{1}{2}\left(\epsilon_{1r}' + \left(\epsilon_{1r}'^2 + \epsilon_{1r}''^2\right)^{1/2}\right) \quad -(9)$$

where ϵ_{1r}' is the relative permittivity of the material from which reflection takes place, and ϵ_{1r}'' is its dielectric loss.

<u>The Memory Function Theory.</u>

In general this expresses the Laplace transform of a spectral function as a continued fraction:

3) $$\tilde{C}(p) = \cfrac{C(0)}{p + \cfrac{K_0(0)}{p + \cfrac{K_1(0)}{\ddots \, \cfrac{K_n(0)}{p + K_{n+1}(p)}}}} \quad -(10)$$

For the Debye theory:

$$\tilde{C}(p) = (\epsilon_0 - \epsilon_\infty) \frac{K_0(0)}{p + K_0(0)} \quad -(11)$$

where

$$K_0(0) = 1/\tau \quad -(12)$$

where τ is the Debye relaxation time. Here:

$$p = -i\omega. \quad -(12)$$

The dielectric loss is given by:

$$\epsilon'' = \frac{\omega}{K_0(0)} Re\left(\tilde{C}(-i\omega)\right) \quad -(13)$$

and the permittivity by:

$$\epsilon' = \epsilon_0 + \frac{K_0(0)}{\omega} Im\left(\tilde{C}(-i\omega)\right) \quad -(14)$$

Here: $$\tilde{C}(-i\omega) = (\epsilon_0 - \epsilon_\infty) \frac{K_0(0)}{-i\omega + K_0(0)} \quad -(15)$$

4)

$$= \left(\epsilon_0 - \epsilon_\infty\right) \kappa_0(0) \frac{\left(i\omega + \kappa_0(0)\right)}{\omega^2 + \kappa_0^2(0)}$$

So:

$$\mathrm{Real}\left(\tilde{c}(-i\omega)\right) = \left(\epsilon_0 - \epsilon_\infty\right) \frac{\kappa_0^2}{\omega^2 + \kappa_0^2}$$

$$= \frac{\left(\epsilon_0 - \epsilon_\infty\right)}{1 + \omega^2 \tau^2} \quad -(16)$$

and

$$\epsilon'' = \left(\epsilon_0 - \epsilon_\infty\right) \frac{\omega \tau}{1 + \omega^2 \tau^2}$$

which is the Debye theory, QED.

Similarly:

$$\mathrm{Im}\left(\tilde{c}(-i\omega)\right) = \frac{\kappa_0(0)\left(\epsilon_0 - \epsilon_\infty\right)\omega}{\omega^2 + \kappa_0^2(0)} \quad -(17)$$

so from eq. (14):

$$\epsilon' = \epsilon_0 + \left(\epsilon_0 - \epsilon_\infty\right) \frac{\kappa_0^2(0)}{\omega^2 + \kappa_0^2(0)} \quad -(18)$$

$$= \epsilon_0 + \frac{\left(\epsilon_0 - \epsilon_\infty\right)}{1 + \omega^2 \tau^2}$$

which is the Debye theory for dispersion, QED.

In general therefore:

5)

$$\epsilon'' = \frac{\omega}{K_0(0)} \mathrm{Re}\left(\tilde{C}(-i\omega)\right) \quad -(19)$$

and

$$\epsilon' = \epsilon_0 + \frac{K_0(0)}{\omega} \mathrm{Im}\left(\tilde{C}(-i\omega)\right) \quad -(20)$$

where the spectral function is given by the continued fraction (10).

In the two variable theory the continued fraction is truncated by:

$$K_1(t) = K_1(0)e^{-\gamma t} \quad -(21)$$

So

$$K_1(p) = \frac{K_1(0)}{p+\gamma} \quad -(22)$$

and:

$$\tilde{C}(p) = (\epsilon_0 - \epsilon_\infty) \frac{K_0(0)}{p + \frac{K_0(0)}{p + \frac{K_1(0)}{p+\gamma}}} \quad -(23)$$

$$= \frac{K_0(0)\left(p^2 + \gamma p + K_1(0)\right)}{p^3 + \gamma p^2 + \left(K_0(0) + K_1(0)\right)p + \gamma K_0(0)}$$

using eq. (12), $\tilde{C}(p)$ is transformed into a spectral function $\tilde{C}(-i\omega)$:

6)

$$\tilde{C}(-i\omega) = \frac{K_0(0)}{D}(A - i\omega\gamma)(B - i\omega C_1) \quad -(24)$$

where:

$$A = K_1(0) - \omega^2$$
$$B = \gamma(K_0(0) - \omega^2) \quad -(25)$$
$$C_1 = \omega^2 - (K_0(0) + K_1(0))$$

and:

$$D = \gamma^2(K_0(0) - \omega^2)^2 + \omega^2(\omega^2 - (K_0(0) + K_1(0))^2) \quad -(26)$$

It follows that:

$$\epsilon''(\omega) = \frac{(\epsilon_0 - \epsilon_\infty)}{D}\,\omega\gamma K_0(0)K_1(0) \quad -(27)$$

and

$$\epsilon'(\omega) = \epsilon_0 + \frac{K_0(0)}{\omega}\,\mathrm{Im}(\tilde{C}(-i\omega))$$
$$= \epsilon_0 - \frac{K_0^2(0)}{D}(\gamma B + A C_1) \quad -(28)$$

So the reflected frequency is calculated from eqns. (8), (9), (27) and (28).

Eq. (27) gave the first successful

7) explanation of the far-infra red absorption of liquids in 0020 on www.aias.us.

The far infra-red power absorption is:

$$\alpha(\omega) = \frac{\omega\epsilon''(\omega)}{n'(\omega)c} \quad -(29)$$

and the Debye theory produces a plateau in $\alpha(\omega)$ from eqs. (16) and (29):

$$\alpha(\omega)(\text{Debye}) = \frac{(\epsilon_0 - \epsilon_\infty)\omega^2\tau}{n'(\omega)c(1+\omega^2\tau^2)} \quad -(30)$$

$$\xrightarrow[\omega\to\infty]{} \frac{(\epsilon_0 - \epsilon_\infty)\tau}{n'(0)c}$$

which is a plateau, because at very high frequencies:

$$n_1'^2 \sim \epsilon_0 \quad -(31)$$

However, eq. (27) gives an almost perfect description of the far infra-red, as shown in 0020.

Computer algebra can be used to extend this continued fraction theory to any number of terms, at the expense of using parameters such as $K_0(0), K_1(0), \ldots, K_n(0)$