Precise Dielectric Measurements of Low-Loss Materials at Millimeter and Submillimeter Wavelengths

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Abstract—**Transmission dispersive Fourier transform spectrometry (DFTS) has been used for the measurements of both real and imaginary parts of the complex relative permittivity of low-loss materials at millimeter and submillimeter wavelengths. The materials investigated include cis and trans decalin and polypropylene.**

I. Introduction

THE first indication that nonpolar materials might show dielectric absorption and dispersion in the high-frequency region came from the microwave work of Whiffen [1] on nondipolar liquids such as carbon tetrachloride and benzene. Wyss *et al.* [2] later detected the infrared "tail" of this absorption in the frequency region above 50 cm^{-1} but it was only with the work of Chantry *et al.* [3] and of Gabelnick and Strauss [4] that the absorption bands were completely characterized. They were then shown to be similar in profile to the bands observed for polar liquids [5] but about one order of magnitude weaker. Since that time much theoretical effort has been deployed in attempts to understand the origin of this absorption process. These theories are usually couched in terms of the complex relative permittivity $\hat{\varepsilon} = \varepsilon' - i\varepsilon''$ whereas the quantity usually measured at submillimeter frequencies is the complex refractive index $\hat{n} = n - i\alpha/4\pi\tilde{\nu}$ where n is the refractive index, α is the power absorption coefficient (usually expressed in Nepers/centimeter) and $\tilde{\nu}$ is the wavenumber (in cm^{-1}). The two systems are, however, readily related by Maxwell's celebrated equation

$$\hat{\varepsilon} = (\hat{n})^2. \tag{1}$$

Apart from its theoretical interest, the absorption, in the submillimeter region by nonpolar material is of great practical importance since transparent materials are required as transmissive media for high-frequency communication systems [6]. The simple theory of crystalline nonpolar polymers, such as polypropylene $[\text{CH}_2 \cdot \text{CH}(\text{CH}_3)]_n$, just as does the theory of simple nonpolar liquids, predicts no absorption in the millimeter-wave region. However, finite absorption is detected [7] for all practical polymers, and it is now suspected that even were one able to make bulk specimens with 100-percent crystallinity, they too would show absorption in this band.

In the light of this, we have over the last few years been developing techniques which permit us to make precise measurements of both n and α for low-loss liquids and solids. These involve the use of advanced Michelson-type interferometric spectrometers used in the asymmetric or dispersive mode [8]. The Fourier transformation of the interferograms produced by these instruments gives both n and α directly [8], [9]. The principal difficulty in measurements on low-loss materials is that the reflection losses and artefacts due to nonparallel radiation may easily exceed the absorptive loss in the specimen. However, the fully developed dispersive method [10] permits a full treatment of all interface effects and therefore results in very accurate determinations. This is true provided the absorption is not so small that an impossibly thick specimen is required to give measurable absorption. In practice, we can measure α values down to about 0.08 Np/cm, but this could be extended (with a consequent loss in precision) down to about 0.05 Np/cm by going over to straight transmission methods. The absolute precision we can achieve is about ±1 percent for α and ±0.01 percent for n, but this at the limit of our present technique.

II. Experimental

Transmission dispersive Fourier transform spectrometric techniques can be used to measure simultaneously both $n(\tilde{\nu})$ and $\alpha(\tilde{\nu})$ of liquids and solids [10]. The specimen rests in one of the active arms of the two-beam interferometer and gives a distorted and shifted interferogram. This is compared with an empty cell interferogram to give the complex insertion loss. If $F_S(x)$ and $F_0(x)$ are the interferograms recorded with and without the specimen, the complex insertion loss is given by

$$\hat{\mathscr{L}}(\tilde{\nu}) = \frac{\mathscr{F}\{F_S(x)\}}{\mathscr{F}\{F_0(x)\}} \tag{2}$$

where $\mathscr{F}\{\ \}$ indicates Fourier transformation. The interferogram $F_S(x)$ is a sum of signatures $F_R(x)$, $F_T(x)$, and $F_M(x)$. $F_R(x)$ is the reflection signature from the surface, $F_T(x)$ is the transmission signature, and $F_M(x)$ is the multiply reflected signature. All the signatures carry information and $n(\tilde{\nu})$ and $\alpha(\tilde{\nu})$ can be found from any of them, but the $F_T(x)$ signature is the most attractive. For low-loss materials the signatures are well separated and one can readily use the $F_T(x)$ signature without having to have recourse to editing techniques [9]. $F_T(x)$ can then be combined with $F_0(x)$ to yield both $\alpha(\tilde{\nu})$ and $n(\tilde{\nu})$.

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A. Liquids

For the transmission DFTS measurements on liquids, an interferometer has been constructed [9] in which the fixed mirror arm is held vertically downwards. The radiation is collimated and phase modulation has been used. The liquid under investigation rests on the stainless-steel mirror and forms a gravity-held plane parallel layer. The liquid compartment and the stainless-steel mirror are separated from the main body of the evacuated interferometer by means of a 50-μm-thick Mylar window. The interferogram with the liquid has signatures $R, T, M_1, M_2, M_3, \cdots$. The distance between the centers of the R and T signatures provides the apparent thickness d_S'. The true thickness d_S is given by $d_S = d_S' + \delta_R$ where δ_R is the path difference error caused by the phase shift $\theta_{0L}(\tilde{v})$ on reflection at the liquid surface. The term δ_R can be estimated from

$$\delta_R = \theta_{0L}(\tilde{v}) = \tan^{-1} \frac{\alpha(\tilde{v})}{2\pi\tilde{v}[n^2(\tilde{v}) + \{\alpha(\tilde{v})/4\pi\tilde{v}\}^2 - 1]} \tag{3}$$

using the approximate $n(\tilde{v})$ and $\alpha(\tilde{v})$ values obtained with d_S' and an iterative procedure, or else it can be canceled out by the use of the DFTS two-thicknesses method [9], [10]. This requires one thin (d_{s_1}) and one thick (d_{s_2}) layer of the liquid. The $F_{T_1}(x)$ and $F_{T_2}(x)$ signatures of the interferograms $F_{S_1}(x)$ and $F_{S_2}(x)$ can be used to give the complex (transmission) insertion loss $\hat{\mathscr{L}}_T(\tilde{v})$. The method is very attractive because it does not require any surface reflection loss corrections or multiple reflection loss corrections and cancels out the phase shift error δ_R to provide the absolute level of complex refractive index spectra.

The complex insertion loss is then

$$\begin{aligned}\hat{\mathscr{L}}_T(\tilde{v}) &= \frac{\mathscr{F}\{F_{T_2}(x)\}}{\mathscr{F}\{F_{T_1}(x)\}} = \frac{\hat{s}_{T_2}(\tilde{v})}{\hat{s}_{T_1}(\tilde{v})} = \frac{s_{T_2}(\tilde{v})}{s_{T_1}(\tilde{v})} \\ &\quad \cdot \exp\left[-i\{\text{ph } \hat{s}_{T_2}(\tilde{v}) - \text{ph } \hat{s}_{T_1}(\tilde{v})\}\right] \\ &= \exp\left[-4\pi i\tilde{v}\{\hat{n}(\tilde{v}) - 1\}(d_2 - d_1)\right]\end{aligned} \tag{4}$$

where $\hat{s}_{T_1}(\tilde{v})$ and $\hat{s}_{T_2}(\tilde{v})$ are complex spectra with moduli $s_{T_1}(\tilde{v})$ and $s_{T_2}(\tilde{v})$ and phases ph $\hat{s}_{T_1}(\tilde{v})$ and ph $\hat{s}_{T_2}(\tilde{v})$. $\alpha(\tilde{v})$ and $n(\tilde{v})$ are then calculated from

$$\alpha(\tilde{v}) = \frac{1}{d_{S_2}' - d_{S_1}'} \ln \frac{1}{|\hat{\mathscr{L}}_T(\tilde{v})|} = \frac{1}{d_{S_2}' - d_{S_1}'} \ln \frac{s_{T_1}(\tilde{v})}{s_{T_2}(\tilde{v})} \tag{5}$$

$$\begin{aligned}n(\tilde{v}) &= 1 + \frac{\text{ph } \hat{\mathscr{L}}_T(\tilde{v})}{4\pi\tilde{v}(d_{S_2}' - d_{S_1}')} = 1 + \frac{1}{4\pi\tilde{v}(d_{S_2}' - d_{S_1}')} \\ &\quad \cdot [\text{ph } \hat{s}_{T_2}(\tilde{v}) - \text{ph } \hat{s}_{T_1}(\tilde{v})].\end{aligned} \tag{6}$$

Spot frequency absorption measurements on liquids have also been carried out using the HCN (29.7 cm^{-1}) and H_2O vapor (84.27 cm^{-1}) lasers and an absorptiometer (a beam splitter arrangement) coupled to a set of cells with path lengths ranging from 5 to 25 mm. The construction of a long 25-mm travel variable path length cell is underway which with a water-vapor-free phase-modulated Mach–Zehnder interferometer will enable high precision refraction measurement to be made on the liquids as well.

B. Solids

The liquid interferometer was suitably modified for the measurements of solids. The stainless-steel mirror and the liquid compartment were replaced by an adjustable fixed mirror and a specimen support. This was flushed with dry air throughout the measurement. Solid specimens with plane parallel surfaces are necessary in order to give three plane parallel interfaces: dry air–specimen, specimen–dry air, and dry air–mirror. For a low-loss material with large d_S, the reflection signatures at dry air–specimen $F_{R_1}(x)$, specimen–dry air $F_{R_2}(x)$, and transmission signature at dry air–mirror $F_T(x)$ interfaces are well separated.

The $F_T(x)$ signature is the most attractive.[1] Combining this with an empty cell background interferogram $F_0(x)$, the transmission insertion loss becomes

$$\begin{aligned}\hat{\mathscr{L}}(\tilde{v}) &= \frac{\mathscr{F}\{F_T(x)\}}{\mathscr{F}\{F_0(x)\}} = \frac{\hat{s}_T(\tilde{v})}{\hat{s}_0(\tilde{v})} \\ &= \{\hat{S}_{0S}'(\tilde{v})\}^2 \exp\left[-4\pi i\tilde{v}\{\hat{n}(\tilde{v}) - 1\}\, d_S\right].\end{aligned} \tag{7}$$

$\hat{s}_T(\tilde{v})$ and $\hat{s}_0(\tilde{v})$ are complex spectra with respect to $F_T(x)$ and $F_0(x)$, and $\{\hat{S}_{0S}'(\tilde{v})\}^2$ is the total contribution from the reflection losses at the surfaces and is given by

$$\begin{aligned}\{\hat{S}_{0S}'(\tilde{v})\}^2 &= [1 - \{\hat{r}_{0S}(\tilde{v})\}^2]^2 \\ &= [4\hat{n}(\tilde{v})/\{\hat{n}(\tilde{v}) + 1\}^2]^2.\end{aligned} \tag{8}$$

$n(\tilde{v})$ and $\alpha(\tilde{v})$ are then evaluated from

$$\begin{aligned}n(\tilde{v}) &= 1 + \frac{\text{ph } \hat{\mathscr{L}}_T(\tilde{v})}{4\pi\tilde{v}\, d_S} - \frac{\text{ph } \{\hat{S}_{0S}'(\tilde{v})\}^2}{4\pi\tilde{v}\, d_S} \\ &= 1 + \frac{\text{ph } \hat{s}_T(\tilde{v}) - \text{ph } \hat{s}_0(\tilde{v})}{4\pi\tilde{v}\, d_S} - \frac{\text{ph } \{\hat{S}_{0S}'(\tilde{v})\}^2}{4\pi\tilde{v}\, d_S}\end{aligned} \tag{9}$$

$$\begin{aligned}\alpha(\tilde{v}) &= \frac{1}{d_S} \ln \frac{1}{|\hat{\mathscr{L}}_T(\tilde{v})|} + \frac{1}{d_S} \ln \{S_{0S}'(\tilde{v})\}^2 \\ &= \frac{1}{d_S} \ln \frac{s_0(\tilde{v})}{s_T(\tilde{v})} + \frac{1}{d_S} \ln \{S_{0S}'(\tilde{v})\}^2.\end{aligned} \tag{10}$$

$\{S_{0S}'(\tilde{v})\}^2$ and ph $\{\hat{S}_{0S}'(\tilde{v})\}^2$ can be determined using approximate $n(\tilde{v})$ and $\alpha(\tilde{v})$ values and an iterative procedure [10].

The use of DFTS techniques brings a special benefit for the measurement of low-loss systems in that all the interface effects are treated exactly in the theory. In the familiar transmission nondispersive methods, defocusing of the beam and the effects of short paths through the specimen can give erroneous results even if a variable path length cell is being used.

III. Discussion

The three most transparent liquids so far known in the submillimeter region are cyclohexane, and cis and trans decalin. Cyclohexane is the least absorbing at microwave

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[1] A different method of determining $\hat{n}(\tilde{v})$ of solids has been described in [11], [12] in which $F_{R_1}(x)$ and $F_{R_2}(x)$ are used to evaluate $n(\tilde{v})$ and $\alpha(\tilde{v})$.

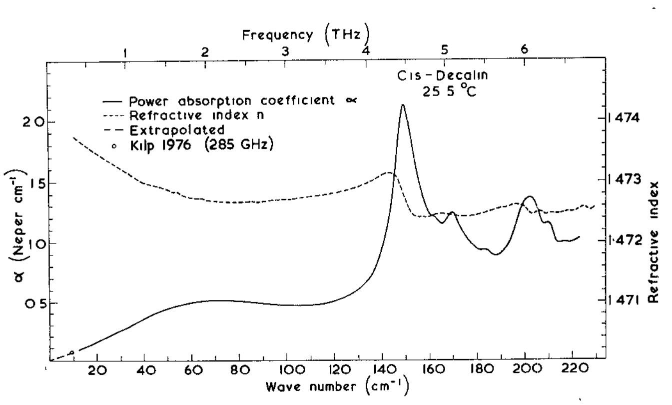

Fig. 1. Power absorption coefficient and refraction spectra of cis decalin at 25.5°C.

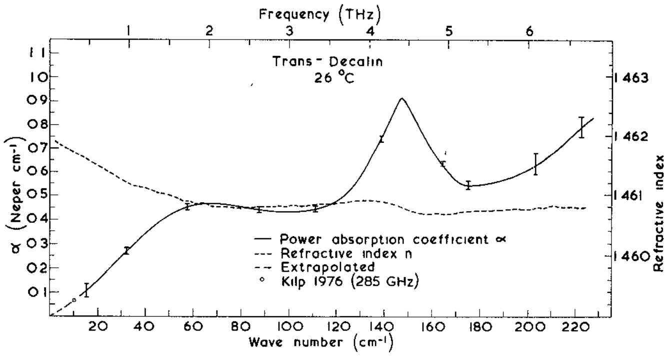

Fig. 2. Power absorption coefficient and refractive index spectra of trans decalin at 26°C.

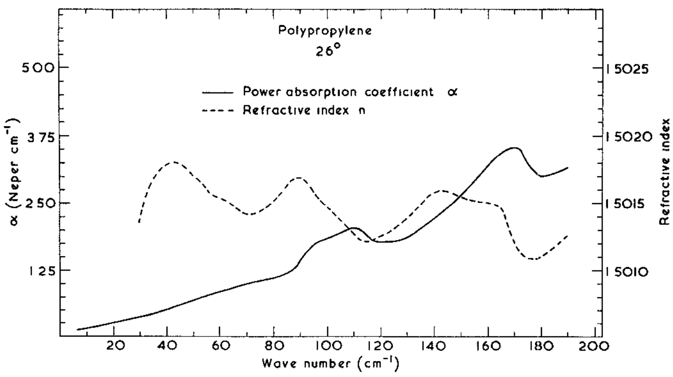

Fig. 3. Power absorption coefficient and refractive index spectra of polypropylene at 26°C.

frequencies, but at 50–100 cm^{-1} (1.5–3 THz) the situation is reversed and both decalins are most transparent. Thus the peak of the submillimeter absorption band for cyclohexane reaches 0.68 Np/cm (at a frequency of 2.5 THz, i.e., 83.33 cm^{-1}) whereas the peaks for both decalins lie below 0.5 Np/cm. The reason for this is not at all clear. The power absorption and refraction spectra of cis decalin at 25.5 °C and trans decalin at 26 °C obtained using the DFTS two-thickness method and the interferometer in the polarization mode [8] are shown in Figs. 1 and 2.

The extrapolation of our curves to zero frequency passes very close to the values determined at 9.5 cm^{-1} (285 GHz) by Kilp [13] using microwave free space techniques. The four most promising low-loss polymers are polyethylene, polypropylene, TPX (poly-4 methyl pentene-1), and PTFE (polytetrafluorethylene) [7]. A great deal of work has been done on polypropylene because of its attractive mechanical properties and because of its ease of fabrication. The power absorption coefficient and refractive index spectra of polypropylene at 26 °C are shown in Fig. 3.

It will be seen that the absorption levels so far achieved with this and with the other polymers are still too high for effective use of the materials in communication systems, but they are low enough for them to be used as windows, and in the fabrication of optical components such as lenses. However, the processes, which cause the microwave and millimeter-wave loss, peak in the submillimeter-wave region, and measurements in this region will be very helpful to the engineers trying to make acceptably low-loss polymers.

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