FIZIKA Volume 21, Supplement 1, 1989.
Proceedings of the XI Yugoslav SYMPOSIUM OF THE PHYSICS OF CONDENSED MATTER.
M. Davidović and M. Lj. Napijalo Editors, Donji Milanovac, October 3–7, 1988.

Determination of Optical Constants of Solid State Specimens by Dispersive Fourier Transform Spectroscopy

R.Gajić
Institute of Physics
Beograd , P.O.Box 57

Dispersive Fourier transform spectroscopy (DFTS) is a broad band technique for determination of the optical constants of materials,usually in millimetre and submillimetre region[1, 2, 3, 4, 5] This technique uses two-beam interferometers as normal Fourier transform spectroscopy (FTS) does.In FTS a specimen is placed in front of the detector,or rarely,between the source and the beamsplitter.In DFTS,however,a specimen is placed in one of the active arms of the interferometer.Usually,it is the arm with the fixed mirror.
In contrast to FTS,where the reflectance R (transmittance T) of a specimen is determined,DFTS enables determination of the complex amplitude coefficients $\hat{r}$ ($\hat{t}$) of a specimen!
This means that reflectance R and phase ϕ_r (or T and ϕ_t) are determined simultaneously.On that way the problems connected with the fitting and the use of the Kramers-Kronig relations are avoided!
The DFTS measurements depend significantly on specimen quality, geometry and the degree of its transparency.Therefore,three different configurations are used as it is illustrated in Fig.1.

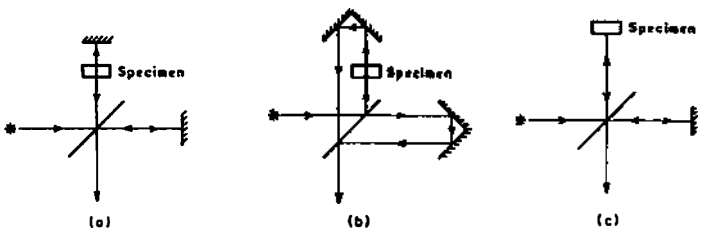

Fig.1. The three basic configurations used in DFTS

Using Fig.2. it is possible to give the explanation of the function of the DFTS spektrometer.

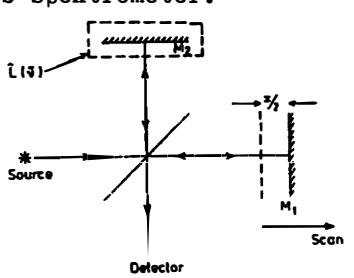

Fig.2. The generalized scheme of the DFTS measurement

Firstly, there is a need for the definition of the new quantity, so-called, the complex insertion loss of a specimen as :

$$\hat{L}(\sigma) = L(\sigma)\exp(i\phi_L(\sigma)) \tag{1}$$

Here, σ represents a wave number.
$\hat{L}(\sigma)$ is the complex factor by which the amplitude of a wave is changed when a reference material such as a vacuum or a mirror is replaced by the specimen.
The amplitude of the electric wave in free space is given by :

$$\hat{E}(z) = \int_{-\infty}^{+\infty} \hat{E}_0(\sigma)\exp(i2\pi\sigma(z-ct))d\sigma \tag{2}$$

Now, we assume that z is the position of the beamsplitter, and t=o for the sake of simplicity.
The amplitude of a wave from the arm with a specimen is :

$$\hat{E}_1(z) = \int_{-\infty}^{+\infty} L(\sigma)\hat{E}_0(\sigma)\exp(i(2\pi\sigma z)+\phi_L(\sigma)+\phi_0(\sigma))d\sigma \tag{3}$$

where ϕ_0 is a small residual phase difference due to an inherent asymetry of the interferometer.
The amplitude of a wave from the arm with the moving mirror is :

$$\hat{E}_2(z,x) = \int_{-\infty}^{+\infty} \exp(i\pi)\hat{E}_0(\sigma)\exp(i2\pi\sigma(z+x))d\sigma \tag{4}$$

The resultant electric field amplitude toward detector is :

$$\hat{E}_R(x) = \hat{E}_1+\hat{E}_2 =$$

$$= \exp(i2\pi z)\int_{-\infty}^{+\infty} \hat{E}_0(\sigma)(L(\sigma)\exp)i\phi(\sigma))+\exp(i(2\pi\sigma x+\pi)))d\sigma \tag{5}$$

where

$$\phi(\sigma) = \phi_L(\sigma)+\phi_0(\sigma) \tag{6}$$

The intensity I in the detector is proportinal to $\hat{E}_R^*\hat{E}_R$:

$$I = \int_{-\infty}^{+\infty} \hat{E}_0^*(\sigma)\hat{E}_0(\sigma)(1+L^2(\sigma))d\sigma +$$

$$+ 2\int_{-\infty}^{+\infty} \hat{E}_0^*(\sigma)\hat{E}_0(\sigma)\, L(\sigma)\cos(2\pi\sigma x+\pi-\phi(\sigma))d\sigma \tag{7}$$

We are only interesting in the second term which is usually called the interferogram(F(x)). It is not dificult to see that :

$$F(x) = \int_{-\infty}^{+\infty} L(\sigma)B_{bg}(\sigma)\exp(i\phi(\sigma)-i\pi)\exp(i2\pi\sigma x)d\sigma \tag{8}$$

where $B_{bg}(\sigma)$ is a background spectrum (a specimen is absent) :

$$B_{bg}(\sigma) = 2\hat{E}_0^*(\sigma)\hat{E}_0(\sigma) \tag{9}$$

After taking the inverse Fourier transform we get :

$$\int_{-\infty}^{+\infty} F(x)\exp(-i2\pi\sigma x)dx = p(\sigma)-iq(\sigma) \tag{10}$$

$$p(\sigma) = L(\sigma)B_{bg}(\sigma)\cos(\phi(\sigma)-\pi) \tag{11}$$

$$q(\sigma) = L(\sigma)B_{bg}(\sigma)\sin(\phi(\sigma)-\pi) \tag{12}$$

Now, it is easy to find $L(\sigma)$ and $\phi_L(\sigma)$ as :

$$L(\sigma) = (p^2(\sigma)+q^2(\sigma))^{1/2}/(p_0^2(\sigma)+q_0^2(\sigma))^{1/2} \tag{13}$$

$$\phi_L(\sigma) = \arctan(q(\sigma)/p(\sigma))-\arctan(q_0(\sigma)/p_0(\sigma)) \tag{14}$$

Calculation of Optical Constants from $\hat{L}(\sigma)$

1. Reflection from an Opaque Solid State Specimen

In this case holds $\hat{L}(\sigma) = r(\sigma)\exp(i\phi_r(\sigma)) = \hat{r}(\sigma)$ so, we imediately get the complex refractive index as :

$$\hat{n}(\sigma) = (1+\hat{r}(\sigma))/(1-\hat{r}(\sigma)) \tag{15}$$

2. Transmission through the Specimen of the Thickness d

In the case of the single-pass measurement (Fig.1.b) we have :

$$\hat{L}(\sigma) = \hat{t}_R(\sigma)\exp(-0.5\alpha(\sigma)d)\exp(i2\pi\sigma(n(\sigma)-1)d) \tag{16}$$

For the double-pass measurement we have :

$$\hat{L}(\sigma) = t_R^2(\sigma)\exp(-\alpha(\sigma)d)\exp(i2\pi\sigma\Delta(\sigma)+i2\phi_R(\sigma)) \tag{17}$$

$\hat{t}_R(\sigma)$ stands for reflection losses and phase shifts caused by internal multiple reflections within a specimen. Also, we define

$$\Delta(\sigma) = 2(n(\sigma)-1)d \quad \text{and} \quad \alpha = 4\pi\sigma k \tag{18}$$

The inverse transformation gives :

$$n(\sigma) = 1-(\phi_L(\sigma)+2\phi_R(\sigma)+\phi_o(\sigma))/(4\pi\sigma d) \tag{19}$$

$$t_R^2(\sigma)\exp(-\alpha(\sigma)d) = (p^2+q^2)^{1/2}/(p_o^2+q_o^2)^{1/2} \tag{20}$$

Generally $\hat{t}_R$ is a complicated function of $n(\sigma)$ and (σ) .

$$t_R(\sigma) = f(n(\sigma),\alpha(\sigma)) \tag{21}$$

$$\phi_R(\sigma) = g(n(\sigma),\alpha(\sigma)) \tag{22}$$

However, in special cases when $\alpha(\sigma)$ is small or large relations (21) and (22) have relatively simplified forms .
$n(\sigma)$ and $\alpha(\sigma)$ could be found by iterative procedure.
Firstly, $n(\sigma)$ is chosen then from (20) and (21) it is possible to determined $\alpha(\sigma)$. Now, $\phi_R(\sigma)$ is found from (22) and finally the new value of $n(\sigma)$ is obtained from (19). The process will be repeated untill the values for $n(\sigma)$ converge.
Between many different applications of DFTS we shall emphasize few. With DFTS, it is possible to make accurate low transmittance measurements. Suppose one measures $t(\sigma)=0.1$ with an error of $\Delta t=0.01$, then the transmittance $T=t^2=0.01$ with an error $2t\Delta t=\pm 0.002$![3]
Refractive index of some materials (e.g. quartz) was determined with a high accuracy-5 or 6 significant digits.[6]
As another example we will take DFTS spectra of alkali halide crystals : KCl, NaCl, KBr, KI which have strong anharmonic effects. Johnson and Bell were first who reported DFTS measurements on single crystals KBr and KCl[7,8,9]. They were also the first who compared their experimental values of optical constants with theoretical models of the complex dieletric response of an anharmonic crystal obtained with the help of the Green's functions[10,11]. The reflection measurements revealed the details in phase spectra which were not noticed before using the Kramers-Kronig relations !

Besides, alkali halides DFTS was used for the analysis of multiphonon structure in CdTe[12].
Due to great sensitivity to weak spectral features, DFTS was used for an analysis of imperfect crystals as KBr containing o.3% Cl[13].
At the end of this article, which has an introductory character, it is necessary to emphasize an important fact that many details of DFTS[11,15,16] and FTS[17,18] are droped here.

REFERENCES

1. J.R.Birch, T.J.Parker, Infr. and Millim. Waves, Vol.2, Chap.3 (ed.by K.J.Button), 1979.
2. J.Chamberlain, Principles of Interferometric Spectroscopy, Wiley, 1979.
3. R.J.Bell, Introductory Fourier Transform Spectroscopy, Academic Press, 1972.
4. E.E.Bell, Infr.Phys., Vol.6., 57-74, 1966.
5. E.E.Russel, E.E.Bell, Infr.Phys., Vol.6., 75-84, 1966.
6. G.J.Simonis, Handbook of Optical Constants of Solids, (ed.by E.D.Palik), Chap.8., 1985.
7. A.Memon, T.J.Parker, Int.Jour.Infr.Millim.Waves, Vol.2., No.4, 1981.
8. T.J.Parker, D.A.Ledsham? W.G.Chambers, Infr.Phys., Vol.16, 293-297, 1976.
9. D.A.Ledsham, W.G.Chambers, T.J.Parker, Infr.Phys., Vol.16, 515-522, 1976.
10. K.W.Johnson, E.E.Bell, Phys.Rev., Vol.187., 1044-1052, 1969.
11. C.L.Mok, T.J.Parker, W.G.Chambers, Solid State Comm., Vol.34., 567-570, 1980.
12. T.J.Parker, J.R.Birch, C.L.Mok, Solid State Comm., Vol.36., 581-584, 1980.
13. K.A.Maslin, G.A.Gledhill, T.J.Parker, Infr.Phys., Vol.25., 439-441, 1985
14. J.R.Birch, C.E.Bulleid, Infr.Phys., Vol.17, 279-282, 1977.
15. J.R.Birch, T.J.Parker, Infr.Phys., Vol.19, 1979.
16. J.R.Birch, Infr.Phys., Vol.28, 345-352, 1988.
17. R.Gajić, Infr.Phys., Vol.27, 297, 1987.
18. R.Gajić, M.J.Merkle, Infr.Phys., Vol.28., 333-335, 1988.