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CONTRAST IMPROVEMENT IN PHOTOACOUSTIC AND DIFFUSE LIGHT FOURIER TRANSFORM SPECTROSCOPY

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Abstract - A noticeable contrast improvement is obtained by filtering the interferogram in photoacoustic or diffuse transmittance spectroscopy. The sensitivity of both methods is also compared.

When spectroscopy recordings lead to the observation of weak narrow lines superimposed with a large broadband background, the presence of this background is often a severe limitation to the sensitivity. One can observe such effect, e.g., in a PA experiment in which residual window, wall (or sample) induced absorptions are present or in a simple one beam transmission experiment with a weakly absorbing sample (Fig. 1a). One can avoid the contribution of the background to the signal, when the spectrum under study is constituted of narrow lines by using wavelength modulation spectroscopy; but such method implies an optimum modulation amplitude in order to get a good signal to noise ratio without line shape distortion /1/. One known the numerous advantages of Fourier Transform Spectroscopy both for conventional (absorption or emission) or for photoacoustic and photothermal measurements /2/ /3/. We are going to show how this technique coupled with a suitable filtering of the interferogram allows to get easily a noticeable background rejection. Indeed one can cancel the large background contribution by eliminating the central part of the interferogram which is mainly due to this background as shown in Fig. 1b. More quantitatively in the case of a phase compensated interferometer (either by construction or by a numerical procedure) recorded from \( \delta_1 \) to \( \delta_2 \) the modulus of the Fourier Transform of the interferogram \( I(\delta) \) is given by:

\[
|B(\sigma)|^2 = \left[ B_{\cos}(\sigma) \ast (\delta_2 - \delta_1) \left\{ \frac{\sin \pi \sigma (\delta_2 - \delta_1)}{\pi \sigma (\delta_2 - \delta_1)} \cos \pi \sigma (\delta_2 + \delta_1) \right\}^2 + B_{\cos}(\sigma) \ast (\delta_2 - \delta_1) \left\{ \frac{\sin \pi \sigma (\delta_2 - \delta_1)}{\pi \sigma (\delta_2 - \delta_1)} \sin \pi \sigma (\delta_2 + \delta_1) \right\}^2 \right]
\]

where \( B_{\cos}(\sigma) = \int_{-\infty}^{+\infty} I(\delta) \cos 2\pi \sigma \delta \, d\delta \)

In the case of a very narrow line located at \( \sigma_0 \) (Dirac function) one can notice that:
which, as expected, shows that the resolution is only dependent on the amplitude of the displacement of the moving mirror: $(\delta_2 - \delta_1)/2$.

Nevertheless as shown in Fig. 2, in the case of a Gaussian line whose width is $\Delta \sigma$ one can notice that the shape and the amplitude are strongly dependent of $(\delta_1 + \delta_2) \times \Delta \sigma$.

As far as this product is smaller than 1, $|B(\sigma)|$ is given by formula (2) multiplied by the total intensity of the line. When $\Delta \sigma$ is of the order of $1/(\delta_2 + \delta_1)$ or larger (and not of $1/(\delta_2 - \delta_1)$ which is the limiting factor for the resolution) the line shape is affected and the amplitude is reduced.

The samples that we have studied with our home-made FT visible and near IR spectrometer /3/ consist of uncompacted solid solutions of $\text{Er}_2\text{O}_3$ in $\text{SiO}_2$ $(10^{-1}$ to $10^{-5}$ in volume). The Fourier transform diffuse transmittance spectra of a 1 mm thick sample (dilution $10^{-3}$) are represented in Fig. 3, the detector being a germanium photodiode. One can hardly guess the presence of the lines (arrows) on the transmitted light intensity (Fig. 3a) whereas a paramount improvement of the contrast is observed by using the filtering (Fig. 3b).

The PA spectrum of the same sample is represented on Fig. 3. As expected for this weakly absorbing sample, the background is much lower compared to the single beam transmittance experiment because the light deposited into the sample itself represents the main contribution to the signal. Nevertheless at these low concentrations, an important background, due to residual impurities and dust, (Fig. 3c) is present which is eliminated by filtering (Fig. 3d).

At last, Fig. 4 shows in more detail a part of the spectrum obtained in the same experimental condition (source, time constant ...) by using the two methods. One can observe that a better signal to noise ratio is obtained for the diffuse transmittance experiments. The noise equivalent signal corresponds to a variation of $3 \times 10^{-5}$ of the transmittance, which is close to $10^{-5}$ as expected from shot noise calculation.

By this very simple procedure we have shown both theoretically and experimentally, that a strong rejection of broad band background can be achieved. Let us recall that the method is particularly suitable when one deals with lines which are narrower than the resolution limit of the Fourier transform Spectrometer.

REFERENCES

This method can be easily extended to the usual case of symmetrical recordings of interferograms (e.g. from $-\delta_2$ to $+\delta_2$). By nulling a narrow part of the interferogram (e.g. from $-\delta_1$ to $\delta_1$) one can obtain the filtering which is described above. Indeed because FT is a linear transformation one can write (Fig. 5):

$$I(\delta) = I(-\delta_2, +\delta_2) - I(-\delta_1, +\delta_1)$$

So:

$$B(\sigma) = B_2(\sigma) - B_1(\sigma)$$

where

$$B_2(\sigma) = \int_{-\infty}^{+\infty} I(-\delta_2, +\delta_2) \cos 2\pi \sigma \delta \, d\delta$$

and

$$B_1(\sigma) = \int_{-\infty}^{+\infty} I(-\delta_1, +\delta_1) \cos 2\pi \sigma \delta \, d\delta$$

$$B(\sigma) = B_{\cos}(\sigma) \ast \left[ 2\delta_2 \frac{\sin 2\pi \sigma \delta_2}{2\pi \sigma \delta_2} - 2\delta_1 \frac{\sin 2\pi \sigma \delta_1}{2\pi \sigma \delta_1} \right]$$

$$B(\sigma) = 2B_{\cos}(\sigma) \ast \left[ (\delta_2-\delta_1) \frac{\sin \pi \sigma (\delta_2-\delta_1)}{\pi \sigma (\delta_2-\delta_1)} \cos \pi \sigma (\delta_2+\delta_1) \right]$$

which is the first part of equation (1), the second being zero because the calculation is achieved on symmetrical interferograms.
Fig. 1 - Contribution to the interferogram of a broad band background superimposed with a weak narrow line.

Fig. 4 - Extended view of the Er\textsubscript{2}O\textsubscript{3} in SiO\textsubscript{2} (6 \times 10^{-5}) line at 12300 cm\textsuperscript{-1}. a) Diffuse transmittance. b) Photoacoustic. The small stray line (arrow) in (a) is induced by the transmission of the interferometer.
Fig. 2 - Influence of a non-symmetric interval $|\delta_1 \delta_2|$ in the calculation of the Fourier Transform of an interferogram.

Response to a monochromatic excitation.

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Fig. 3 - Fourier transform spectra obtained with unfiltered (left) and filtered (right) spectra.

a) and b) Diffuse transmittance,
c) and d) Photoacoustic.

Powdered sample: Er₂O₃ in SiO₂ (10⁻³).