

The Principles of Interferometric Spectroscopy

John Chamberlain

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Completed, collated and edited by
G. W. Chantry and N. W. B. Stone
*National Physical Laboratory,
Teddington, Middlesex.*

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Preface

When John Chamberlain died suddenly in October 1974, his coworkers lost not only a much-loved friend but also a most able and diligent colleague. During his brief career John had made major contributions to spectroscopy and seemed set to achieve an eminent position and to play a senior role in international science. His main field was asymmetric or dispersive Fourier transform spectrometry and he was, in fact, together with E. E. Bell in the USA, mainly responsible for the development of this branch of the art. It is indeed sad that these two, who had known each other for such a brief time, should die within a year or two of each other. John had known that his life was at risk from the high blood pressure to which he was subject, but he never allowed this to influence him in any way and was always a cheerful and considerate companion whose sensitivity and regard for the feelings of others was common knowledge.

Outside his scientific work, John's great passion was music and he built up a large collection of records, together with a superb reproduction system to indulge it. However, much of his spare time at home was taken up with a major project to which he was devoted—the writing of a comprehensive textbook on Fourier transform spectrometry. This he had been painstakingly assembling and at the time of his death there were twenty chapters in various stages of completion. His colleagues felt that the best memorial they could raise to him, as a scientist, would be to ensure that this book was published. However the text, as it stood, required considerable modification and pruning. Moreover there were very few diagrams and those mostly consisted of thumbnail sketches.

We decided therefore to edit the book down to manageable proportions, to supply the necessary diagrams, and to revise the text in accord with modern advances and with the newer ideas that had emerged since 1974. To do all this was a major task and we soon realized that we could not do a simple excision and grafting job. In the end we decided to redraft several sections of the book completely, but guided by John's original text and by the concepts which he had obviously chosen. We have done our best to remain faithful to his original conception of the book within these limitations and we hope that, were he alive now, he would approve of what we have done. We must however point out that Chapter 10 is entirely ours. It was a chapter that we felt was necessary, but the advances that have occurred in the field of computation made John's original sketches completely out of date. In the light of this, a completely new chapter, *ab initio*,

seemed in order, but we take sole responsibility for it. Throughout the period when we were redrafting and revising the text we received help from several of our colleagues, but we would especially like to acknowledge the assistance of J. R. Birch and J. W. Fleming whose careful reading and criticism of each section of the book as it appeared helped to eliminate many errors and obscurities.

We dedicate this book therefore to our late much-missed colleague in the hope that it will prove helpful to present and future generations of interferometric spectroscopists and in some small way replace the personal help which John would undoubtedly have given had he lived.

Summer, 1978

G. W. CHANTRY

N. W. B. STONE

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Introduction

1.1. SPECTROSCOPY

Spectroscopy is the term describing the study of spectra; *spectrometry* is the quantitative measurement of the form of a spectrum. Fellgett¹ has proposed the term *spectrology* as a possibly more precise description of the subject. Strictly speaking, spectroscopy implies the visual observation of a spectrum, but the term is so widely understood in the more general sense to describe the subject as a whole, that it will be used with that meaning here. Spectrometry will have the meaning given above; spectrology will not be used.

1.1.1. Absorption spectrometry

The quantitative investigation of spectra involves the determination of precise information about the energies (that is, frequencies or wavelengths) at which systems absorb or emit electromagnetic radiation and the extent to which the absorption or emission occurs (that is the intensity). Infrared and long-wave spectroscopists deal, for the most part, with passive or absorbing systems on account of the feeble spontaneous emission at low frequencies. The specimen under study is probed, therefore, with radiation, the exact state of which before and after interaction with the system yields the spectral information required. The optical properties of a specimen are completely specified when the complex refractive index $\hat{n}(\sigma)$ at wavenumber σ is known.[†] This quantity may be expressed in either of the alternative forms

$$\hat{n}(\sigma) = n(\sigma)[1 - i\kappa(\sigma)] = n(\sigma) - i\tilde{\kappa}(\sigma) \quad (1.1a)$$

or

$$\hat{n}(\sigma) = n(\sigma) - i(\alpha(\sigma)/4\pi\sigma), \quad (1.1b)$$

[†] Note: the spectral energy variable will be generally measured by the wavenumber σ having the dimensions of reciprocal length and the other variables, i.e. frequency ν (Hz), wavelength (in *vacuo*) λ (m), circular frequency ω (rad s⁻¹), are related to this by

$$\sigma = \frac{1}{\lambda} = \frac{\nu}{c} = \frac{\omega}{2\pi c},$$

where c is the speed of light (299.8×10^6 m s⁻¹). The SI unit for σ is m⁻¹, but the cm⁻¹ persists as the practical unit. In the literature the symbols $\tilde{\nu}$ and sometimes ν will be encountered with the same meaning as σ used here.

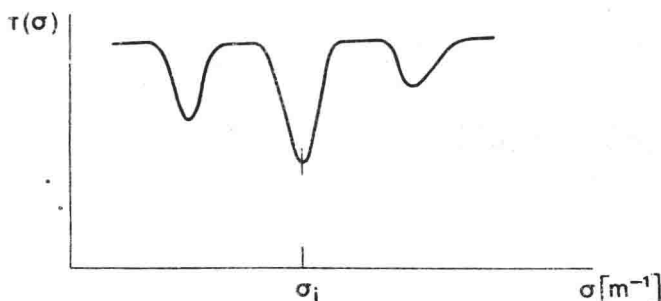


Figure 1.1. Example of a transmission spectrum $\tau(\sigma)$ showing the wavenumbers such as σ_i that are of interest to the spectroscopist

where $n(\sigma)$ is the real refractive index, $\kappa(\sigma)$ is the extinction coefficient, $k(\sigma) = n(\sigma)\kappa(\sigma)$ is the absorption index and $\alpha(\sigma)$ is the power absorption coefficient (neper m^{-1}).[†]

The real refractive index is related to the speed $v(\sigma)$ of the waves in the medium according to the relation

$$n(\sigma) = c/v(\sigma) \quad (1.2)$$

given by Maxwell; the absorption coefficient measures the intensity (or power) attenuation of the electromagnetic waves after travelling unit length within the (isotropic) medium.

The transmissivity of a bounded specimen of thickness d is given in terms of the incident and transmitted powers $I_0(\sigma)$ and $I(\sigma)$, respectively, by

$$\tau(\sigma) = I(\sigma)/I_0(\sigma) \quad (1.3)$$

and has values lying in the range $0 \leq \tau(\sigma) < 1$. The spectroscopist frequently measures the variation of $\tau(\sigma)$ with σ so that he can locate the positions of minima or points of inflection in $\tau(\sigma)$ —Figure 1.1. These two types of turning point are taken to represent the locations of absorption bands or lines. The information obtainable from measurements of the positions of absorption features is adequately discussed in textbooks dealing explicitly with spectra and their interpretation.³ However, to derive the maximum amount of information from a spectroscopic experiment it is desirable to determine an absolute measure of the absorption. In practice, the apparent loss of energy which the beam experiences in traversing the specimen arises from single and multiple reflection effects at the interfaces in addition to the true absorption mechanisms. One may write, therefore,⁴

$$\tau(\sigma) = \tau_A(\sigma)\tau_R(\sigma, d), \quad (1.4)$$

[†] The use of the neper (Np) to measure power absorption is discussed by Chamberlain and Chantry.²

where

$$\tau_A = \exp [-\alpha(\sigma)d] \quad (1.5)$$

is the transmissivity corresponding to the purely absorptive loss and $\tau_R(\sigma, d)$ represents the transmissivity ascribable to all causes other than pure absorption, principally the reflection effects (see ref. 4 for detailed treatment). Equation (1.5) is the mathematical formulation of the Bouguer-Lambert Law. It follows from (1.3), (1.4), and (1.5) that

$$\alpha(\sigma) = \frac{1}{d} \ln \left(\frac{I_0(\sigma)}{I(\sigma)} \right) - \frac{1}{d} \ln \left(\frac{1}{\tau_R(\sigma, d)} \right) \quad [\text{Np m}^{-1}]. \quad (1.6)$$

The second term represents a correction which must be applied to the combination

$$\frac{1}{d} \ln \left(\frac{I_0(\sigma)}{I(\sigma)} \right) = \alpha^\circ(\sigma) \quad [\text{Np m}^{-1}] \quad (1.7)$$

of the measured quantities in order to arrive at the true absorption coefficient. Unfortunately, it is very common to find that absorption coefficients quoted in the literature are of the erroneous $\alpha^\circ(\sigma)$ kind, and there is seldom sufficient additional information from which the correction can be inferred.

The calculation of $\tau_R(\sigma, d)$ in the general case is very difficult and it is desirable, if possible, to eliminate it from the calculation by a suitable modification of the experimental technique. For a suitably thick specimen, not only will $\tau_R(\sigma, d)$ be much reduced relative to $\tau_A(\sigma)$, but it becomes possible virtually to eliminate the multiple-beam interference effects and hence make $\tau_R(\sigma, d)$ essentially independent of d . If then we have that $\tau_R(\sigma, d) \rightarrow \tau_R(\sigma)$, it follows that, by making measurements of the powers transmitted by two specimens of identical composition but differing thicknesses d_1 and d_2 , it is possible to determine $\alpha(\sigma)$ directly since

$$\alpha(\sigma) = \frac{1}{d_2 - d_1} \ln \left(\frac{I_1(\sigma)}{I_2(\sigma)} \right) \quad [\text{Np m}^{-1}]. \quad (1.8)$$

From this equation it will be seen that reliable determination of $\alpha(\sigma)$ depends on accurate measurement of specimen thickness. This becomes difficult when $\alpha(\sigma)$ is large, for then d will have to be very small, and consequently hard to measure accurately, if $\tau_A(\sigma)$ and $\tau(\sigma)$ are not to be too small for meaningful measurement.

The scope of modern spectroscopy has recently been considerably extended in that, in addition to being able to determine the characteristic wavenumbers σ_i , it is now possible to determine the integrated absorption strengths

$$A_i = \int_i \alpha(\sigma) d\sigma \quad [\text{Np m}^{-2}]. \quad (1.9)$$

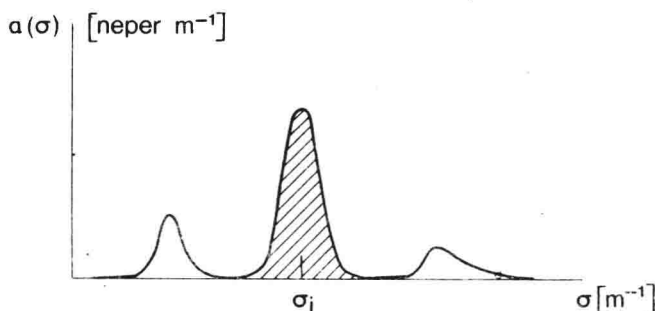


Figure 1.2. Absorption spectrum corresponding to the transmission spectrum of Figure 1.1. The shaded area yields the integrated absorption strength $A_i = \int \alpha(\sigma) d\sigma$ of the feature at σ_i

A_i is the area of the i th absorption feature and is a measure of the strength of the oscillator of frequency $\nu_i = c\sigma_i$.

1.1.2. Emission spectrometry

Emission spectra may be continuous or have a discrete structure. While the observer may be concerned with both the shape and the magnitude of the spectrum in either case, he is frequently interested in the absolute power level of the continuous spectra and in the shapes and relative intensities of the features in the line or band spectra. Since the measurements concern the source rather than any passive absorbing system, it is important to know the transmission characteristic of the complete spectrometric system over the range of interest. This knowledge is not easily acquired.

1.1.3. Resolution and resolving power

The amount of detail seen in an experimental spectrum is limited and can be described in terms of the resolution R or the resolving power \mathcal{R} . The resolution R is measured in frequency or wavenumber units and represents the smallest spectral interval that can be meaningfully discerned. Hence, the *smaller* the numerical value of R the more detailed the spectrum and the *better* the resolution. Every spectroscopic instrument has a scanning function (or apparatus function) which we can regard as the observed spectral record obtained for a strictly monochromatic input to the spectrometer. The function has a finite width which is related to the resolution limit. The choice of definition of the criterion for the resolution limit is to some extent arbitrary, but more detailed treatment, in Chapters 6 and 8, discusses the various definitions that may be employed.

Sometimes it is more convenient to describe the quality of the resolution in terms of the resolving power \mathcal{R} , which is a measure of the ability to

separate close spectral lines. \mathcal{R} is a pure (dimensionless) number given by

$$\mathcal{R}(\sigma) = \sigma/R. \quad (1.10)$$

It is obvious that \mathcal{R} is numerically greater the better the detail seen in the spectrum.

1.2. FOURIER TRANSFORM SPECTROMETRY

It is, of course, possible to measure $I(\sigma)$, and hence $\tau(\sigma)$, directly using a number of well-established techniques. There are, however, alternatives, of which an important one is the method of Fourier transform spectroscopy (FTS). This may be defined as the technique whereby a spectrum is determined by the explicit application of a Fourier transformation to the output of an optical† apparatus—generally a two-beam interferometer.

The procedure is, roughly, to divide a beam of radiation from the source into two parts; make provision for the introduction of a known, variable phase delay of one part relative to the other; and then detect the resultant power when the two beams are recombined. This power is recorded as a function of the phase delay and shows fluctuations that are basically periodic. When all the detailed fluctuations of this power record are subject to Fourier transformation we obtain a *fully characterized spectrum*; when only the trends shown by the fluctuations are subject to Fourier transformation we obtain a *partially characterized spectrum*. This latter technique is suitable for application only to spectra having a narrow bandwidth.

1.2.1. Fully characterized spectrometry

In fully characterized spectrometry, the required spectrum, the power distribution of the detected radiation as a function of frequency (or wavenumber), is obtained by Fourier transformation of the variable part of the record of power versus phase delay. As we shall see below, we call the total record the *interference function* and the variable part of it the *interferogram*. The spectrum is, therefore, the Fourier transform of the interferogram. That there is an explicit dependence of the interferogram on the spectrum may be simply seen from the following discussion.

Consider the schematic interferometer shown in Figure 1.3. The partial beam that has traversed the path XP_1Y to the detector has travelled a distance that is greater than that travelled by the other beam along XP_2Y by an amount x ($0 \leq x < \infty$) which we call the path difference. By assuming the interferometer to be otherwise symmetrical and evacuated, the optical and geometrical path differences are identical. Because of the path difference x ,

† We shall use the term 'optical' to mean an array of lenses, mirrors, etc. when pertaining to apparatus, and to mean some part of $\hat{n}(\sigma)$ when pertaining to constants; the terms 'light' and 'visible' will be applied only to radiation detectable by the eye. Optical path length is defined, as usual, by the product refractive index \times path length.

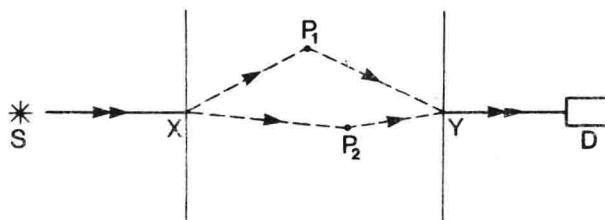


Figure 1.3. Schematic arrangement for a two-beam interferometer. The radiation from the source S is divided, within the interferometer, into two partial beams passing, respectively, via P_1 and P_2 . These beams recombine before falling on the detector, which they reach with a relative phase delay determined by the path difference $XP_1Y - XP_2Y$ within the interferometer

the two beams arrive at the detector with a phase delay $2\pi\sigma x$ for any component σ and show interference which is governed by this delay. As we shall see below, the power at the detector is, in fact,

$$I(x) = B(\sigma) d\sigma + B(\sigma) \cos 2\pi\sigma x d\sigma \quad [W] \quad (1.11)$$

for each spectral component of power $B(\sigma) d\sigma$ in the interval σ to $\sigma + d\sigma$. We assume the contribution from each path SP_1D and SP_2D to be the same, $\frac{1}{2}B(\sigma) d\sigma$.

When the source is strictly monochromatic and of wavenumber σ_0 , the power from each partial beam is $\bar{\epsilon}_0$ (the Russian symbol $\bar{\epsilon}$ is known as 'buki')

$$I(x) = \bar{\epsilon}_0(1 + \cos 2\pi\sigma_0 x) \quad (1.12a)$$

$$= 2\bar{\epsilon}_0 \cos^2 \pi\sigma_0 x \quad [W] \quad (1.12b)$$

and is the interference function. It has the familiar form of cosine fringes which extend to infinite values of x without change of either $I_{\max} = 2\bar{\epsilon}_0$ or $I_{\min} = 0$ (Figure 1.4). If, however, the source is made more realistic and given a finite, but small, width $\Delta\sigma$ the fringes are still basically cosinusoidal of period σ_0^{-1} ; the interference between components from either side of the feature is now constructive at $x = 0$, but becomes increasingly destructive as x increases. This has the effect of modulating the fringes whose I_{\max} falls to zero at about $x = \Delta\sigma^{-1}$.

When the source has a broad bandwidth, such as is required for most spectroscopic measurements, all components are in phase at $x = 0$, but the detected intensity fluctuates rapidly to zero as x is increased and we have the equivalent of white light fringes observed in broad-band visible interferometry. The detected power is given by adding all the components represented singly by equation (1.11):

$$I(x) = \int_0^\infty B(\sigma) d\sigma + \int_0^\infty B(\sigma) \cos 2\pi\sigma x d\sigma \quad [W] \quad (1.13)$$

Because

$$I(0) = 2 \int_0^{\infty} B(\sigma) d\sigma \quad [W] \quad (1.14)$$

when $x=0$, we may write

$$I(x) = \frac{1}{2}I(0) + \int_0^{\infty} B(\sigma) \cos 2\pi\sigma x d\sigma \quad [W] \quad (1.15)$$

for the interference function, noting that it consists of a part $\frac{1}{2}I(0) = I$ invariant with x , and a variable part $I(x) - I = F(x)$. We call this variable part the *interferogram* and rewrite equation (1.15) in terms of it as

$$F(x) = \int_0^{\infty} B(\sigma) \cos 2\pi\sigma x d\sigma \quad [W], \quad (1.16)$$

which shows that the interferogram depends on the spectrum according to a

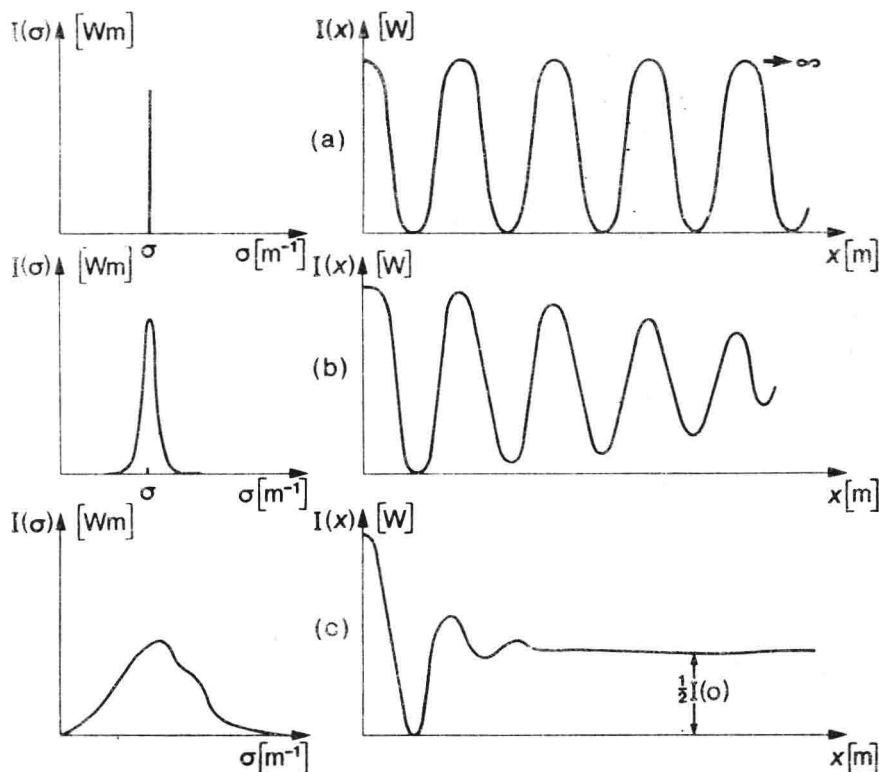


Figure 1.4. Relation between the detected spectrum $I(\sigma)$ and the two-beam interference signal $I(x)$ produced by it: (a) monochromatic spectrum; (b) quasi-monochromatic spectrum; (c) broad-band spectrum.