

MICHELSON INTERFEROMETERS FOR ABSORPTION SPECTROSCOPY AND REFRACTOMETRY

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INTRODUCTION

In this contribution I seek to give an account of the main items of progress made over the last two years by members of the group in Basic Physics Division of the National Physical Laboratory, Teddington, who are working in the field of interferometric spectroscopy (an account of earlier work is given by Sutherland¹). In that time, the use of commercially manufactured interferometers for spectroscopy in the far infrared has greatly increased, so that there are now between 35 and 40 instruments being used for a wide variety of research topics in laboratories all over the world, and a number of the contributions at the 1965 conference are based on observations made with them. At NPL we have two commercial instruments in use, but this paper is concerned with researches for which special equipment has been constructed. Some of these researches are still in progress and so no attempt will be made at giving a complete account here. One feature common to all the topics discussed is that general purpose digital computers (the NPL ACE and English Electric KDF9) have been used for data reduction. I believe that the flexibility of general purpose computers, especially when considering the new developments in refractive index and intensity measurements, are vital to the full exploitation of Fourier methods in spectroscopy.

HIGH RESOLUTION SPECTROSCOPY

Perhaps the most obvious development to be reported is the extension of interferometric techniques to obtain spectra with resolution in the range of hundredths of a reciprocal centimetre. To do this we have constructed an interferometer capable of giving a maximum path difference between beams of one metre, which corresponds to a maximum resolution, by the definition we customarily use, of one hundredth of a reciprocal centimetre. So far it has not been used to this full resolution but only to five hundredths. *Figure 1* shows a small part of the pure rotation spectrum of water vapour and it will be seen that the predicted lines are well reproduced and that their widths are as expected at this resolution. The signal-to-noise ratio is not particularly high in this spectrum but this will be improved by simply increasing the integration time when a serious attempt is made at recording the water spectrum to include the weaker features. In all the measurements reported here the detector used was a Golay cell.

The "Teratron" 337 μ source

The "Teratron" is a pulsed gas maser source giving monochromatic radiation of wavelength 337 μ at mean power levels of milliwatts and peak

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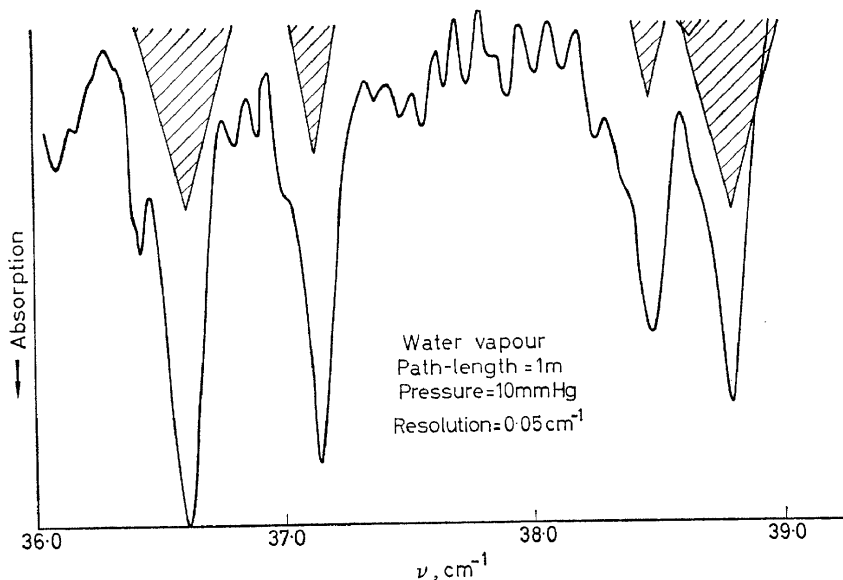


Figure 1. Part of the pure rotation absorption spectrum of water vapour at a resolution of 0.05 cm^{-1} with theoretical line positions indicated

levels of watts^{2,3}. It has proved to be a very valuable ancillary to our interferometric techniques particularly in the following applications. The first is in determining the "spectral window" of the interferometer at any chosen resolution. The spectral window corresponds to the slit function in a prism or grating spectrometer, but in a Michelson interferometer has its accuracy almost completely determined by the accuracy of the mirror drive. The curve obtained by Fourier transformation of an interferogram made using the monochromatic radiation to illuminate the interferometer may therefore be thought of either as the spectral window or as the spectrum of screw intervals. Ideally, these intervals would be constant, the spectrum would be peaked at one frequency, and would have a half width inversely proportional to the length of the screw used. Figure 2 shows some actual results obtained with the high resolution instrument mentioned above and the "Teratron" source. The upper curve shows peaks additional to the single maximum expected and at frequencies indicative of periodic error in the screw drive. The lower curve shows the same record after the drive had been modified and it can be seen that the error has been reduced so as to give "ghosts" of less than one half per cent intensity. The second application of the "Teratron" is in obtaining single frequency values of absorption coefficient and refractive index. These single point measurements can be made with high precision because of the large power available and they provide particularly valuable complementary data to the wide-range refractive index measurements to be described in the next section. Results of refractive index measurements on air⁴, some polar liquids⁵ and crystalline quartz⁶ have already been reported.

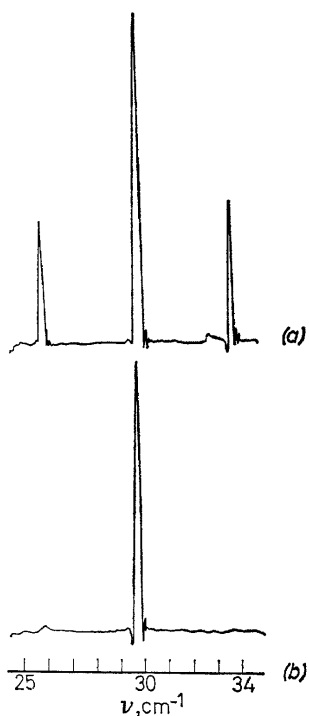


Figure 2. The "spectral window" of a high resolution interferometer determined from observations of a monochromatic line at 29.7 cm^{-1} from a "Teratron" source. (a) Bad drive; (b) improved drive

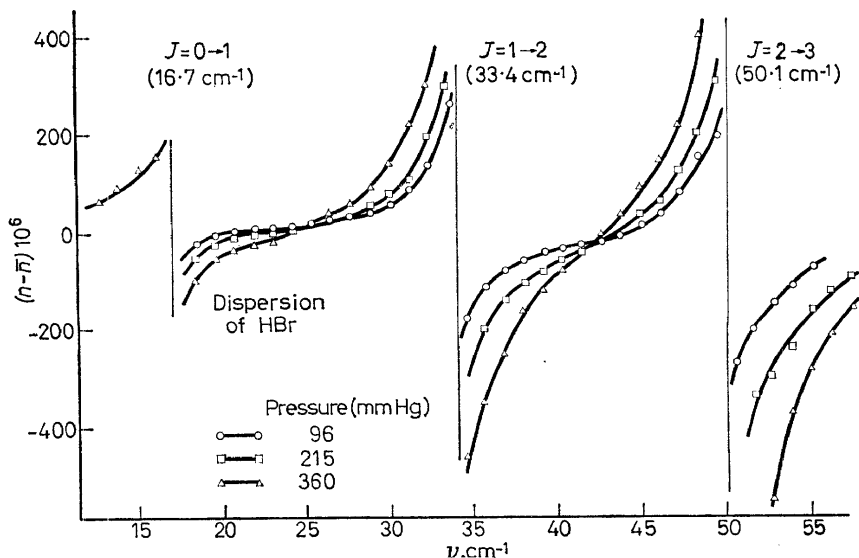


Figure 3. The dispersion associated with pure rotational transitions in hydrogen bromide found by Michelson interferometer observations. Temperature $\sim 300^\circ\text{K}$

WIDE RANGE REFRACTIVE INDEX MEASUREMENTS BY THE MICHELSON INTERFEROMETER

The use of a Michelson interferometer with Fourier analysis for obtaining refractive index spectra was reported some time ago⁷, but measurements have now been made which include dispersive absorption bands in solids and of pure rotation lines in gaseous hydrogen halides. *Figure 3* shows the dispersion of hydrogen bromide in the region of the first few rotation lines. Apart from the direct usefulness of the index data as such, we believe that this technique will be of increasing value in obtaining absolute line strengths, since it avoids the necessity for very high resolution and curve of growth to determine line width.

ABSORPTION MEASUREMENTS ON GASES WHERE INTRINSIC ABSORPTION IS VERY WEAK

In an attempt to record the magnetic dipole pure rotation spectrum of molecular oxygen which is intrinsically about 10^5 times weaker than spectra involving electric dipole interactions we have used a multiple pass absorption cell following the design of White⁸. The great advantage of an interferometer in this work is that its high efficiency as compared with a grating instrument offsets to some extent the large intensity loss with the inevitably small aperture ratio of the multipass cell. Specifically the cell used had an aperture ratio of $f/16$ with 6 in. diameter mirrors; it gave absorption lengths up to about 100 m and could be pressurized to 5 atm. *Figure 4* shows some pre-

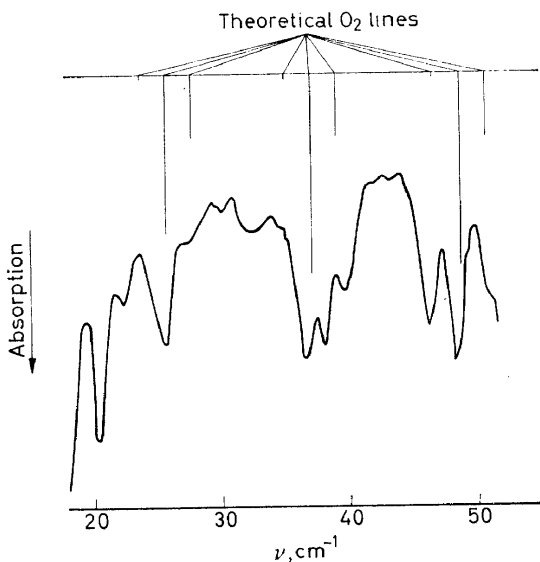


Figure 4. An absorption spectrum of about 350 metre atmospheres of dry oxygen at 5 atm pressure. Theoretical line positions are indicated

liminary results obtained with this apparatus. The observed spectrum shows fair agreement with prediction and though several unsatisfactory features remain to be understood the results show that this type of weak absorption experiment is possible by interferometry.

USE OF HIGH PRESSURES WITH FAR-INFRARED SPECTROSCOPY

To extend our understanding of some recently observed liquid phase bands in polar liquids, we have combined high pressure absorption cells with interferometers. One apparatus uses a conventional piston intensifier in a

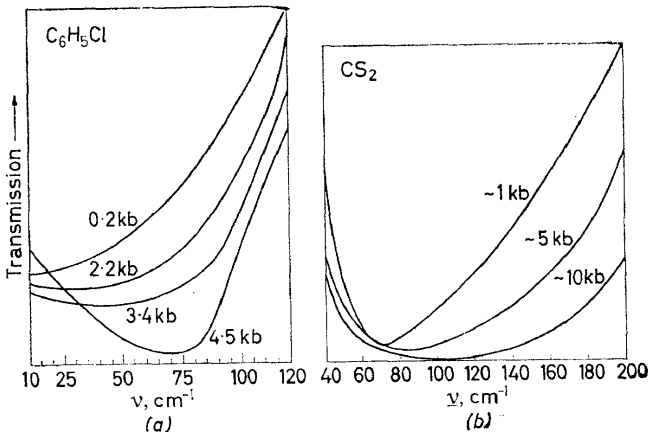


Figure 5. Preliminary observations of the effect of pressure on the far-infrared absorption bands observed in (a) liquid chlorobenzene and (b) liquid carbon disulphide at room temperature

cell with crystal quartz windows and in it liquids can be subjected to hydrostatic pressures up to 5 kb and transmission measurements made in a vacuum interferometer. A second apparatus is capable of subjecting specimens to pressures up to 30 or 40 kb. This is of the anvil and cone type with the specimen contained in a pyrophyllite cube also fitted with crystal quartz windows. One pair of opposing anvils has holes to allow a beam through and again this cell is built into an interferometer system. The aperture is limited by strength design requirements but with a specimen of about 2 mm diameter and a radiation cone of $f/10$, we can record satisfactory spectra in the range 10–50 cm^{-1} which includes the interesting lattice band region. Figure 5 shows some early results for liquid chlorobenzene and liquid carbon disulphide showing that marked changes in the form of these “pseudo-lattice” liquid absorption bands are observed when we subject them to pressure.

I am grateful to my colleagues in Basic Physics Division for permission to quote their results, and to the Acting Director of the National Physical Laboratory for permission to publish this work.

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