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THE TECHNIQUES OF HIGH RESOLUTION, DOUBLE-BEAM, TWO-BEAM INTERFEROMETRY WITH A FAR INFRARED LAMELLAR GRATING INTERFEROMETER

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Résumé. — Nous décrivons un interféromètre du type lamellaire destiné à la haute résolution dans l'infra-rouge lointain. Les avantages obtenus en faisant la différence entre deux faisceaux pour réduire l'étendue dynamique nécessaire sont illustrés par des interférogrammes pris dans l'infra-rouge lointain et les spectres à haute résolution correspondants de CO, H₂O, DCl et NO. Dans le spectre de rotation pure de DCl le dédoublement isotopique de la transition $J = 2 \rightarrow 3$ $(\Delta \nu \sim 0,09 \text{ cm}^{-1})$ dû à 35 Cl et 37 Cl est nettement résolu. Certains problèmes et inconvénients rencontrés dans l'emploi de la technique de différentiation à deux faisceaux sont discutés.

Abstract. — The instrumentation of a high resolution, far infrared lamellar grating interferometer is discussed. The advantages of double-beam differencing to minimize the dynamic range problem is illustrated with far infrared interferograms and corresponding high resolution spectra of Carbon Monoxide (CO), water vapor (H₂O), Deuterium Chloride (DCl) and Nitric Oxide (NO). In the pure rotational spectrum of DCl the ³⁵Cl-³⁷Cl isotopic splitting of the $J = 2 \rightarrow 3$ transition ($\Delta \nu \sim 0.09 \text{ cm}^{-1}$) is clearly resolved. Some of the problems and disadvantages encountered using the double-beam differencing technique are discussed.

Instrumentation. — A high resolution far infrared lamellar grating interferometer which operates in either a single-beam or a double-beam differencing mode of operation is described. At present the instrument covers a range of 10 to 125 cm⁻¹ (1 000 to 80μ). The performance of the instrument is discussed and illustrated with results from the pure rotational spectra of H₂O, CO, HCl and its isotopic species, and NO.

The optical diagram of the lamellar grating interferometer is shown in figure 1. The source of radiation is a high pressure, mercury arc lamp. The source output is divided into two beams, the reference and sample beams. The two beams are caught and focused by two ellipsoidal mirrors M_1 and M_2 . The upper beam, the sample beam, is folded by two flats M_4 and M_5 and then passed through the sample chamber. Mirror M_5 is a polished aluminum flat. The beam comes to a focus in the middle of the sample chamber allowing the use of minimum size samples. The lower beam, the reference beam, passes under the sample chamber and comes to a focus on the flat M_3 . A focus at this position is useful for reflection studies. Flat M_3 is located on a six position slide assembly. This allows changing between the mirror and any of five 2.5 cm diameter samples for reflection studies. This can be done from outside the tank without breaking the vacuum in the chamber. The flat M_3 folds the reference beam up perpendicular to the sample beam. The two beams are then recombined by the polished aluminum semicircular chopper mirror M_6 which rotates at 19 cps. So that intensities of the two beams can be matched precisely, motorized iris diaphragms have been placed in equivalent positions in both beams approximately 18 cm before the first focus. Mirror M_7 refocuses the combined beams. At positions M_9 and M_{13} are placed coarse (< 20 lines/mm) gratings which provide the high frequency cutoff filtering of the source radiation.

The interferometer optics consist of two 45.7 cm diameter spherical mirrors and the lamellar grating. The two spherical mirrors are arrayed in a Czerny-Turner configuration which allows the grating to be used in parallel light in the zeroth order.

After the optical path difference has been introduced by the grating, the beam is reflected to the second Czerny-Turner mirror and is refocused. The beam is then directed by the diagonal flat M_{13} (scat-





FIG. 2. - Side View of Lamellar Grating Interferometer.

ter grating) through an exit aperture mounted on the end of a light pipe. The detector, a gallium doped germanium bolometer which operates at 4.2 °K is mounted on the other end of the light pipe.

The interferogram can be recorded either singlebeam or double-beam. In double-beam operation the outputs of the reference and sample beams are differenced automatically by the broadband AC preamplifier following the detector. This difference signal is recorded. In single-beam operation one of the two beams is blanked off and the signal obtained is the difference between this opaque shutter and the beam.

The gas samples to be studied must be contained in a conventional gas cell, and the windows of the sample cell are wedged crystalline quartz. To compensate for the absorption in the sample beam, two nearly identical pieces of crystalline quartz are placed in the reference beam. These are located between M₃ and M₆. The crystalline quartz absorbs the intermediate infrared radiation of the source. The quartz windows in the reference beam are slightly thinner - by a few hundredths of a millimeter - than those on the sample cells. The reference beam windows are mounted on a rotating mount. This allows the effective thickness of these pieces to be variable so that a precise equivalence of quartz in both beams can be obtained. This is necessary to ensure complete balance of the spectral transmission of both beams. Slight deviations in balance lead to anomalies in intensity for the region of zero optical path difference.

Figure 2 is a side view of the lamellar grating interferometer supported on a dolly which allows the instrument to be rolled out of the vacuum tank for servicing and/or major modification. There are a number of access ports on the vacuum tank which allow access for minor servicing.

The lamellar grating consists of two sets of interleaving plane mirrors and is shown in figure 3. There are 24 facets in each set, each facet being 0.635 cm wide and 30.5 cm long, giving a total area of 30.5×30.5 cm for the grating. The individual facets of each grating are flat and coplanar to two fringes of the 5 461 Å line of Hg, except for the two outside facets of each grating which deviate by about four fringes. The lower, moving set of facets of the grating is mounted on a platform which is suspended from the platform holding the stationary set of facets. The lower platform is attached to the upper platform by three flexure hinges similar to those used by John Strong [1]. With proper adjustment and restraints the pitch and yaw errors are less than ± 1 arc sec



FIG. 3. - Front View of Lamellar Grating Interferometer.

over the 83 mm of geometric travel. The position of one grating relative to the other is measured with a linear metric inductosyn scale with an accuracy of better than $\pm 1.5 \,\mu$.

Figure 4 shows an over-all view of the vacuum tank (interferometer inside) and the electronics console. The entire vacuum tank is vibrationally isolated by air suspension mounts under each of the six legs.

The interferogram is not recorded continuously but is sampled at equally spaced increments. The system is designed to sample at multiples of 10 microns in optical path difference. The actual sampling interval is dictated by the highest frequency present.

The grating is allowed to sample each point for a time long enough to largely average out the noise, approximately two to eight seconds. During this time the output of the detector, suitably amplified and synchronously detected, is fed into a voltage-tofrequency converter. The voltage output of the detector is converted linearly into a frequency which is then counted for the gate or sampling time. At the end of this time, the number of counts (0-9999) accumulated in the counter together with the number of the point is punched on paper tape. The grating then advances to the next point and the process is



FIG. 4. - Overall View.

repeated. This process is continued until the entire interferogram has been punched out on the paper tape. The paper tape output is then processed by the data reduction center and the Fourier cosine transformation computed yielding the frequency spectrum.

Dynamic range is a serious problem in interferometry. This situation can be discussed in terms of the pure rotational spectra of diatomic and linear molecules. The pure rotational spectra of such molecules are quite simple, consisting of very nearly equally spaced absorption lines (every 2 B_0 cm⁻¹). As is well known [2, 3] the interferograms of such spectra are also simple in appearance. Characteristic features called signatures are seen every $(2 B_0)^{-1}$ cm in optical path difference. In general the amplitudes of these signatures are quite small in comparison to the central maxima; however it is just these features one wants to bring out, since they represent most of the information about the absorber. Increasing the amount of absorption of the sample by raising the pressure of the gas will lead to other difficulties such as line broadening. Usually narrow lines are desired, especially if wavelength measurements are to be made. Also if the lines are broader the signatures decay in amplitude more rapidly [4] and little or no gain will be achieved.

Double-beaming will largely circumvent this problem. In double-beaming one uses two beams, one of which passes through the sample cell (sample beam) and the other through a matched reference cell (reference beam), and then the sample beam intensity (or signal) is subtracted from the reference beam intensity (or signal). When this is done information about the spectral distribution of energy from the source is lost, but such information may be recovered by a prior and/or subsequent recording of single-beam interferograms of the source alone. In practice this is not difficult or inconvenient as the single-beam source interferogram need only to run out to 3-5 mm optical path difference and can be obtained in a time interval short compared with the higher resolution double-beam interferograms where the optical path difference typically runs out to 100 to 160 mm.

Performance and results. — The first ten signatures of carbon monoxide as shown in figure 5 were obtained under the double-beam mode of operation. If the interferogram had been obtained single-beam, keeping the central maximum fixed, the first signature would be down in intensity by a factor of ~ 20 .

The amplitudes of these signatures, when corrected for the distortion (due to the fact the pure rotational spectrum of CO is not made up of exactly equally spaced lines, but rather converges due to centrifugal distortion in the molecule), decay exponentially with increasing optical path difference. The rate of decay (argument of the exponential) depends on the effective half width of the pure rotational lines.

Interferograms of CO were obtained at four pressures (521, 440, 375, and 254 Torr) and the ampli-



OPTICAL PATH DIFFERENCE, cm FIG. 5. — First Ten Signatures From the Pure Rotational Double-Beam Interferogram of Carbon Monoxide.

tudes of the signatures analyzed for the effective half width Δv . The results are shown in figure 6. The line width parameter (as deduced from the interferogram) is plotted as a function of pressure. The resulting curve is a straight line and extrapolates through the origin, indicating negligible instrumental broading. It has been shown [5] that the slope of this curve represents an average upper limit for Δv for carbon monoxide.



FIG. 6. — Line Width Parameter Δv (cm⁻¹) vs p(Torr) for Carbon Monoxide.

Figure 7 shows the pure rotational spectrum of carbon monoxide as realized from the Fourier transformation of the complete interferogram. The cosine modulated baseline (which should be flat) is a channel spectrum generated by regions of plane parallelness of the quartz envelope of the lamp. More will be said about this later.



FIG. 7. -- Pure Rotational Spectrum of Carbon Monoxide

A double-beam interferogram of water vapor is shown in figure 8 (P = 15.5 Torr, L = 15 cm). A few observations on the H₂O interferogram are in order.



First, there is a small, long term drift which is instrumental in nature. This causes no observable difficulty in the spectrum except in the very low wavenumber region, since a long term drift of this type corresponds largely to low frequency Fourier components. Second, the interferogram is very complicated, and the over-all variations do become less as x is made larger. At the end of the interferogram the estimated signal (variation about 1/2 I(0))-to-noise ratio is ~ 1. The spectrum between 15 cm⁻¹ and 115 cm⁻¹ is shown in figure 9. All the strong rotational lines predicted for H₂O have been observed and all individual lines resolved except six doublets and one triplet, some of which are partially resolved.

Two of the unresolved doublets are shown in figure 10 in larger detail. These doublets should have been resolved with the optical path difference attained for the interferogram. In order to determine if these lines were pressure broadened, another interferogram (with approximately the same optical path difference) was obtained at lower pressure (P = 10 Torr) and the same wavenumber regions are shown in figure 11. The doublets are clearly resolved, confirming that the lines were pressure broadened.

To further evaluate the performance of the instrument an interferogram of DCl was obtained. Portions of this interferogram are shown in figure 12. From x = 0 out to x = 2.6 cm the interferogram is characteristic of those obtained for diatomic and linear molecules [2, 3], that is, signatures spaced $\sim (2 B_0)^{-1}$ cm.



FIG. 9. — The Pure Rotational Spectrum of Water Vapor From 15 to 115 cm⁻¹.

FIG. 10. — The 38.5 and 59.9 cm⁻¹ Regions of the Water Vapor Spectrum (L = 15 cm, p = 15 Torr).



FIG. 11. — The 38.5 and 59.9 cm⁻¹ Regions of the Water Vapor Spectrum (L = 15 cm, p = 10 Torr).



FIG. 12. — The Interferogram of DCl (L = 15 cm, p = 83 Torr).

The region between 2.6 cm and 3.3 cm is somewhat confused but after that the signatures reappear, somewhat broader and decreased considerably in intensity. This is due to the two isotopic species of DCl present ($D^{35}Cl$ and $D^{37}Cl$). There are two nearly equal rotational constants, B_0 , and thus two separate sets of signatures which, for small x (low resolution), are in step but for larger x get out of step and « destructively interfere » and for still larger x « interfere constructively » again. The fall off of intensity for those signatures between x = 0 and x = 2.6 cm is indeed much faster than one would expect because of the breadth of the lines (4, 6). The disappearance and subsequent reappearance of the signatures is indicative of at least some of the isotope splittings of the pure rotational spectrum of DCl being resolved. The allowed transitions of the pure rotational spectral DCl can be calculated from the data given by Cowan and Gordy [6] who studied the $J = 0 \rightarrow 1$ transition for D³⁵Cl and D³⁷Cl in the millimeter region of the microwave spectrum and constants obtained from the near infrared data of Pickworth and Thompson [7]. The pure rotational spectra of DCl should consist of a series of closely spaced doublets spaced approximately 10.7 cm⁻¹ throughout the far infrared region. The separation

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of the doublets becomes progressively larger as one goes to the higher J transitions $(J = 0 \rightarrow 1, \Delta v \approx 0.031 \text{ cm}^{-1}; J = 1 \rightarrow 2, \Delta v \approx 0.063 \text{ cm}^{-1}; J = 2 \rightarrow 3, \Delta v \approx 0.094 \text{ cm}^{-1}; J = 3 \rightarrow 4,$

 $\Delta v \approx 0.126 \text{ cm}^{-1}$; etc.). Three regions of the transformed interferogram (the spectrum) are shown in figure 13, the 21.5 ($J = 1 \rightarrow 2$), the 32.3 ($J = 2 \rightarrow 3$),



FIG. 13. — The 21.5, 32.2, and 43.0 cm⁻¹ Regions of the Pure Rotational Spectrum of DCl.

and the 43.0 $(J = 3 \rightarrow 4)$ cm⁻¹ regions. The calculated isotope splitting of the $J = 2 \rightarrow 3$ transition is 0.0946 cm⁻¹. These lines are clearly resolved and, in

fact, it is obvious that the resolution displayed is better than the separation of the lines. The transition $J = 1 \rightarrow 2$ has a calculated isotope splitting of 0.0631 cm⁻¹ and, although the splitting is not observed, there is no doubt that this is an unresolved line made up from at least two closely spaced lines. It is clear from the more than adequately split $J = 2 \rightarrow 3$ transitions and the unresolved but clearly asymmetric $J = 1 \rightarrow 2$ transitions that the theoretical resolution of 0.074 cm⁻¹ is approached.

The DCl sample was contaminated with HCl and the HCl pure rotational spectrum was also observed. The observed frequencies in vacuum wavenumbers and their deviation from the calculated values using the rotational constants obtained in the microwave and near infrared investigation (6, 7, 8) are shown in Table I. In each case the $J = 0 \rightarrow 1$ transition was not observed and the $J = 1 \rightarrow 2$ transitions are not resolved for the Cl³⁵-Cl³⁷ pair. All the other doublets are resolved. The average deviation is 4×10^{-3} cm⁻¹ and the maximum deviation is 11×10^{-3} cm⁻¹.

As mentioned previously the channel spectrum due to the lamp envelope is troublesome, especially in the low wavenumber region. Channel spectra effects are localized narrowly in the interferogram; in fact, one sees a « channel signature » as is shown in figure 14 at the top. The section of the interferogram is that for NO (nitric oxide). The signature was edited out by running the empty cell out to ~ 4 mm and subtracting this interferogram from the NO interferogram.

Trans.	Species	Obs, Far Infrared	Δ (obs- calc) (^a) × 10 ³	Trans.	Species	Obs, Far Infrared	\triangle (obs- calc) (^a) × 10 ³
$1 \rightarrow 2$	DCl ³⁷ DCl ³⁵	21.554		6 → 7	DCl ³⁷ DCl ³⁵	75.086 75.307	2
$2 \rightarrow 3$	DCl ³⁷ DCl ³⁵	32.241	- 3	$3 \rightarrow 4$	HCl ³⁷	83.272	11
$1 \rightarrow 2$	HCl^{37}	41.742		$7 \rightarrow 8$	DCl^{37}	85.748	3
$3 \rightarrow 4$	DCl ³⁷ DCl ³⁵	42.977	0	$8 \rightarrow 9$	DCl ³⁷ DCl ³⁵	96.387 96.667	8
4 <i>→</i> 5	DCl ³⁷ DCl ³⁵	53.697	- + 0 3	$4 \rightarrow 5$	HCl^{37}	103.989	8
$2 \rightarrow 3$	HCl ³⁷	62.492	2	$9 \rightarrow 10$	DCl ³⁷	106.982	- 3
$5 \rightarrow 6$	DCl ³⁷ DCl ³⁵	62.586 64.401 64.593	2 1 4	10 → 11	$\frac{DCl^{37}}{DCl^{35}}$	107.299 117.557 117.913	$-\frac{1}{2}$ 11

TABLE I. — Calculated and Observed Frequencies in Vacuum Wave Numbers for the Pure Rotational Spectra of HCl³⁵, HCl³⁷, DCl³⁵ and DCl³⁷

(a) Calculated values are determined from rotational constants obtained from microwave and near infrared absorption data [Ref. 6, 7, 8].



FIG. 14. — The First 8 mm (Optical Path Difference) of the Interferogram of Nitric Oxide, with the Channel Spectrum Signature (upper) and After It is Edited Out.



FIG. 15. — The Pure Rotational Spectrum of Nitric Oxide from 20 to 65 cm^{-1} Showing the Result of Eliminating the Channel Signature.

Both interferograms (with and without editing) were transformed to obtain the spectra. The results are shown in figure 15. Eliminating the channel signature has straightened out the baseline considerably.

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DISCUSSION

E. Bell. — I wish to congratulate Dr. Dowling upon the development of the technique for obtaining the difference of the interferogram between a background and a sample. The results speak for themselves.

H. GEBBIE. — Why did you pick NO? Let me suggest NO_2 as a more interesting molecule. It is an asymetric rotor with magnetic splitting. Interferogram signatures are not likely to be useful in this case.

J. M. DOWLING. — NO was chosen since we had a sample on hand and secondly we knew the interferogram would be quite interesting; in fact one can see all qualitative aspects of the pure rotational spectrum of NO from the interferogram, including the fact that the rotational quantum number is half integral.