

**Determination of the Line Shape of a Fourier Transform Infra-Red Spectrometer
Using an Infra-Red Helium-Neon Laser and a Low Pressure Gas Cell.**

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CONTENTS

1. INTRODUCTION 1

2. CHOICE OF EMISSION SOURCE..... 1

3. CONSTRUCTION OF THE LASER EMISSION SOURCE 2

4. MEASUREMENTS OF THE LASER EMISSION SOURCE..... 3

5. SELECTION OF THE GAS FOR LOW PRESSURE CELL..... 3

6. MEASUREMENTS USING THE LOW PRESSURE GAS CELL 3

7. SUMMARY AND FURTHER WORK.....

ACKNOWLEDGEMENTS 5

FIGURES 6

Determination of the Line Shape of a Fourier Transform Infra-Red Interferometer Using an Infra-Red Helium-Neon Laser and a Low Pressure Gas Cell.

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1. INTRODUCTION

Fourier-transform infra-red spectrometers are used regularly for measurements of stratospheric trace gases. However, in order to facilitate the retrieval of accurate vertical column abundances and, more importantly, concentration profiles of stratospheric trace species from Fourier-transform infra-red (FTIR) spectra, it is necessary to know the true response function or instrumental line shape (ILS) of the spectrometer used for the measurements. It is possible to calculate a theoretical ILS but this is usually not an accurate estimate of the true ILS for any given instrument, as small mis-alignments of the spectrometer may cause significant deviations from the theoretical predictions. Measurements of ILS can be made by using a spectral line (either in absorption or emission) which has a full-width-at-half maximum (FWHM) that is significantly narrower than the spectral resolution of the instrument being characterised.

FTIR atmospheric transmission spectra have been recorded on a Bruker IFS 120M spectrometer using the sun as a source. The output beam from a helium-neon laser, operating at $3.39\ \mu\text{m}$ ($2948\ \text{cm}^{-1}$), was used as a narrow bandwidth emission source which was introduced into the spectrometer coaxially with the solar beam to provide a measurement of the ILS of this instrument. In addition, a gas cell containing hydrogen bromide (HBr) at low pressure ($< 1\ \text{mbar}$) was inserted into the combined solar and laser beam and used both as a complementary ILS measurement technique and for the validation of the ILS measurements obtained using the laser.

2. CHOICE OF EMISSION SOURCE

The criteria for selection of the laser emission source were that:

- (i) the line width should be equal to or less than one tenth of the maximum resolution ($0.0027\ \text{cm}^{-1}$)¹ of the Bruker IFS 120M;

¹Derived from $0.7/(\text{maximum optical path difference})$ [Chamberlain, 'The principles of Interferometric Spectroscopy' p 142].

- (ii) the beam diameter should be larger than the input collecting mirror of the FTIR;
- (iii) the radial intensity profile of the laser beam in the plane of the field stop should be comparable with that of the solar beam;
- (iv) the frequency of the source should not drift significantly during the period of a full-resolution scan of the FTIR (103 seconds);
- (v) the emission line should preferably be at a wavelength which is in one of the routinely spectral windows measured using the FTIR, preferably in a broad region of saturated absorption.

A helium-neon gas laser was chosen for its narrow line width (limited by acoustic vibrations to $\sim 10^4 \text{ cm}^{-1}$) and the 2948 cm^{-1} line of this laser is convenient in the infrared in that it lies when making ground-level measurements in a large saturated atmospheric methane feature in the spectral window which is used for measurements of stratospheric nitrous oxide and hydrogen chloride - both of which are well suited to profile retrieval as they have narrow isolated absorption lines.

3. CONSTRUCTION OF THE LASER EMISSION SOURCE

The laser system consists of a red, helium-neon alignment laser ($0.633 \mu\text{m}$) and a further infrared helium-neon laser operating at $3.39 \mu\text{m}$ (commercial plasma tube integrated within an NPL 'in-house' 300 mm long cavity). This is coupled to a set of optics which expands the beam, flattens its radial intensity profile and monitors its power. The infra-red laser is enclosed in an aluminium box to prevent drafts which would cause temperature fluctuations that would, in turn, cause the laser wavelength to drift. A 5 mm diameter hole in each end of the box permits passage of the red and the infra-red laser beams. The whole system is also contained in another enclosure to improve the thermal stability of the laser system. This enclosure has an 80 mm diameter hole for the output beams.

The optical beam from this laser system is incident on a zinc-selenide beam splitter which redirects about 10% of the laser output through a mechanical chopper and onto a zinc selenide lens. This focuses the radiation onto an indium-antimonide detector. The output from this is amplified and used to monitor the laser power. The remainder (90%) of the radiation passes through the beam splitter and is expanded by an off-axis parabolic mirror onto a diffusive reflector (flame-sprayed aluminium). This diffuser is surrounded by a hemispherical reflector (with holes drilled in it to let in the incident and let out the scattered beams). The diffuser and reflector are used to increase the throughput of the system and improve the flatness of the beam's intensity profile. The scattered radiation from the diffuser is then collected and collimated by a large off-axis paraboloid, and directed out of the outer enclosure.

4. MEASUREMENTS OF THE LASER EMISSION SOURCE.

After nine hours of operation in a temperature-controlled room at Lauder² (long-term drift: ~ 0.06 K/hour over ten hours, with short-term oscillations of ± 0.6 K over a 40 minute period) the cavity temperature was stabilised and had a maximum temperature drift of not greater than 0.018 K in any 100 second interval (duration of one scan at maximum resolution) and a long term drift of 0.06 K/hour. This enabled repeatable measurements of the ILS to be obtained (figure 3). The effects of various laser frequency drifts on the measured line shape of the FTIR have been modelled. For low drift rates (less than one part in 3×10^7 or 26 MHz during the course of a scan), it has been shown that whilst the FWHM of the measured laser line will not change significantly, it will become asymmetric. Asymmetry of the measured ILS is defined here as the absolute value of the difference between the depths of the first minima either side of the central peak expressed as a percentage of that peak height. Figure 1 shows two spectra taken thirty minutes apart. It can be seen that although there is approximately 30% asymmetry in the lines, the drift in the laser frequency was less than 13 MHz during the course of a single scan. From the modelling work it can be shown that this drift rate should produce an asymmetry of approximately 10% which is significantly less than that shown in figure 1. This implies that, these scans may not be precise measurements of the ILS of NPL's Bruker 120M at these times, and that there is some other instrumental effect producing an additional asymmetry.

5. SELECTION OF THE GAS FOR LOW PRESSURE CELL.

As noted above, an alternative technique for determining the ILS was also investigated - the use of a low pressure gas cell.

The criteria used to select the gas for the low pressure cell were that it should have a simple spectrum consisting of narrow, isolated lines and that these should be in one of the routinely measured spectral windows, preferably close to the laser frequency of 2948 cm^{-1} . It should also be a species which is not found in the atmosphere in significant quantities and should have some spectral lines clear of atmospheric absorptions. These resulted in the choice of hydrogen bromide which has a Doppler line width of 0.0033 cm^{-1} at 293 K. A survey of the P and R branches of the HBr spectrum, were recorded with the sun as a source. This was done to establish the best lines to use for this ILS work, and the doublet at 2412.7 cm^{-1} and 2413.0 cm^{-1} was selected from this measurement.

6. MEASUREMENTS USING THE LOW PRESSURE GAS CELL

The selected lines are not ideal since the Doppler width is comparable to, rather than much smaller than, the resolution of the instrument but they should, nevertheless,

²Intercomparison with NIWA's Bruker IFS 120M; Lauder, New Zealand, 19th February 1997.

provide a useful consistency check on the laser measurement when fitted with a simulated line hydrogen bromide line derived from the measured laser line and convoluted with a theoretical, Doppler-broadened HBr line. A problem was then encountered, however, which was that all the measured HBr lines were broader than calculated. It is known, however that broadening of these HBr lines arises from nuclear hyperfine splitting of the energy levels in the $J \rightarrow J+1$ and $J \rightarrow J-1$ transitions between the ground ($v=0$) and first excited ($v=1$) vibrational states. It is also expected that this broadening effect would be greatest for low values of J . Figure 2(a) shows the measured effective widths of the 38 HBr lines visible in a spectrum recorded with an infrared glow-bar source. The x-axis is plotted at 0.0033 cm^{-1} intervals to indicate the theoretical, Doppler-broadened width of the lines in the absence of hyperfine splitting. Figure 2(b) shows the relative absorption of each of the lines, showing that the effect is not correlated to the strength of the absorption features and is, therefore, the result of nuclear hyperfine splitting. This supports the choice of the doublet at 2412.7 cm^{-1} and 2413.0 cm^{-1} as these lines are broadened the least by this hyperfine effect.

7. SUMMARY AND FURTHER WORK

A mid infra-red laser system has been designed and built with the aim of routinely measuring FTIR spectrometer line shapes and thereby facilitating the retrieval of vertical distribution profiles for stratospheric trace gases. Measurements of HBr in a low pressure cell have been made simultaneously with laser and solar spectra in order to validate these ILS measurements. Problems have been encountered with the laser frequency drifting and work to improve the temperature stability of the system is ongoing. The effects of laser frequency drift on measured FTIR spectra has been modelled. While the use of HBr spectra to validate laser measurements of ILS has produced some encouraging initial results, further work must be devoted to modelling and quantifying the nuclear hyperfine splitting which has the effect of broadening the measured HBr lines. This is under investigation with the aim of validating the technique.

ACKNOWLEDGEMENTS

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FIGURES

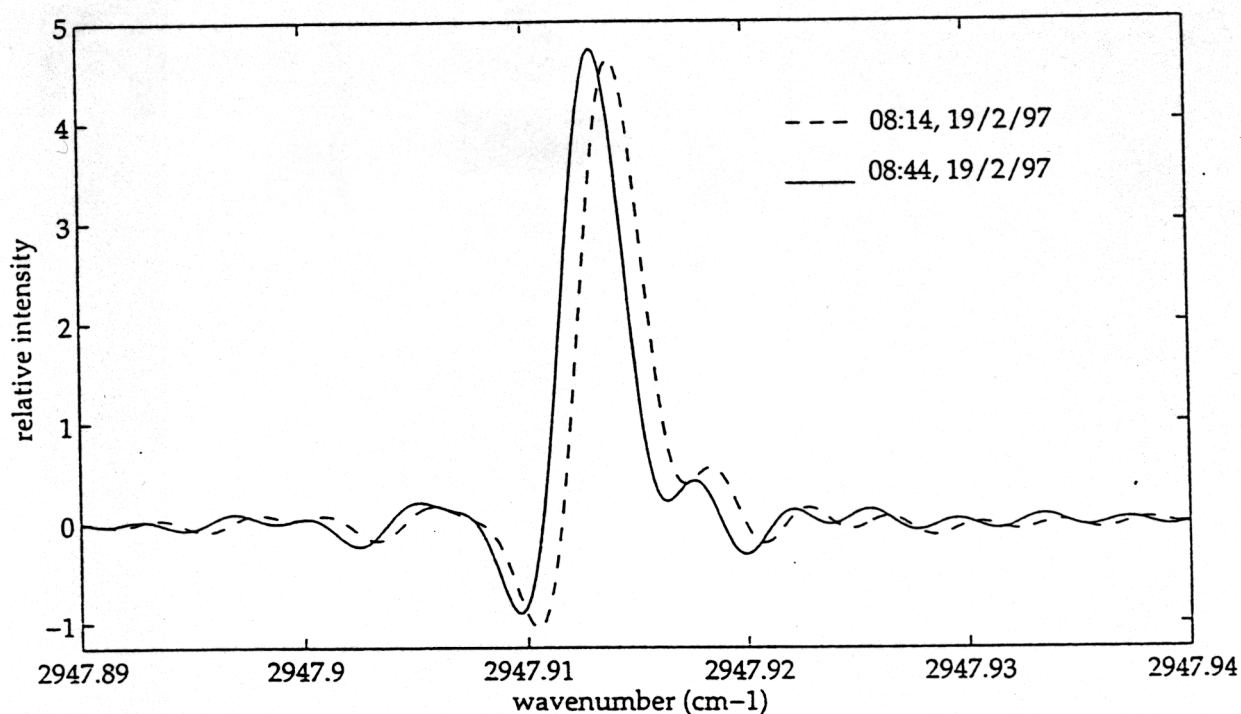


Figure 1. Laser lines from solar transmission spectra recorded 30 minutes apart.

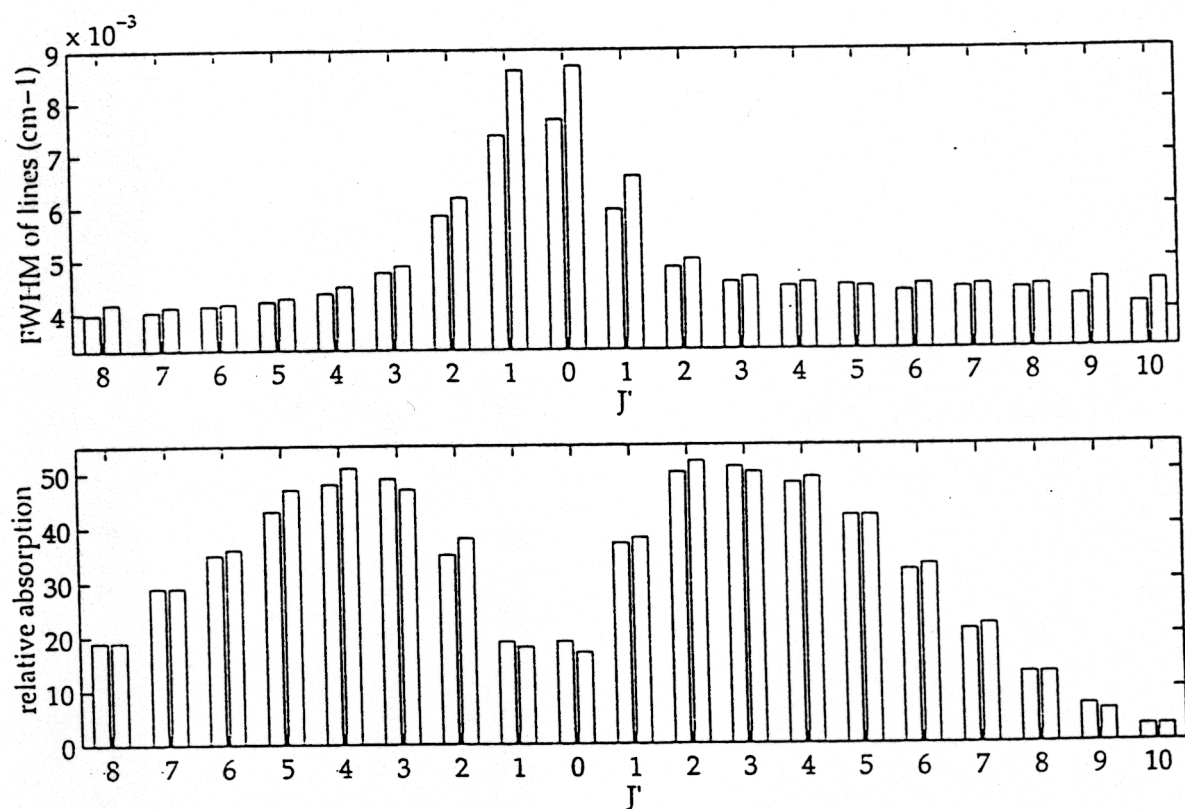


Figure 2. (a) Plot of measured H^{79}Br and H^{81}Br line widths in P and R branches. The x-axis is plotted at 0.00328 cm^{-1} , the Doppler width of HBr in the absence of hyperfine splitting. (b) Relative absorption strength of each of the above lines.