Comparison of Fourier and laser spectroscopy in the far-infrared–submillimeter range

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Successful spectroscopy in the submillimeter–far-infrared (far-IR) region is a difficult task and unconventional methods have commonly been employed in this frequency range. Fourier spectroscopy replaced the then-conventional grating techniques some 15 years ago and still dominates the field. During the transition from grating to Fourier spectroscopy the new method had to prove itself, and several comparisons and performance studies were published.

Recently a new device, the optically pumped far-IR laser, has shown promise as a high-power spectral source. Now, careful comparisons between the established Fourier technique and the new laser spectroscopy are called for. In this Letter we compare state-of-the-art Fourier spectrometers with a recently built laser system by describing measurements made on the same sample, a thin film of the metallic superconductor V3Si deposited on a sapphire substrate. The very low transmission of this sample provided a demanding test of spectral performance, and the comparison clearly shows the strengths and weaknesses of the two methods.

Two Fourier spectrometers, located at the Ohio State University, were employed in these measurements. A lamellar grating interferometer covered 6–30 cm⁻¹, while a Michelson interferometer was used over 50–200 cm⁻¹. These instruments have effective numerical apertures of f/1.6, employ 1.27-cm-diameter light-pipe optics, and use the same type of mercury arc lamp source (General Electric UA-5). In the 6–30-cm⁻¹ region these gratings have an apparent color temperature of 4000 K and emit into the aperture \(5 \times 10^{-6} \text{ W}\) of far-IR power. In the 50–200-cm⁻¹ region the color temperature is lower, approximately 1000 K, and the total power delivered is \(2 \times 10^{-4} \text{ W}\).

In the lamellar grating interferometer, beam division and interferometric modulation are achieved by two sets of interleave facets—one fixed and one movable. Because these sets have the same area, the efficiency of this beam splitter is nearly unity. The Michelson interferometer uses a Mylar beam splitter (6.3-μm thick in the present case) and has cat's-eye retroreflectors in the interferometer arms. The beam splitter has a maximum efficiency of 0.6 at 170 cm⁻¹ and an average efficiency over 50–200 cm⁻¹ of 0.3.

The far-IR radiation was detected by a germanium bolometer operating at 1.2 K. This type of detector typically has a noise equivalent power of \(5 \times 10^{-13} \text{ W (Hz)}^{-1/2}\) and a responsivity of 10 V/W. The detector is in a cryostat which also contains the sample under investigation. The overall efficiency of the Fourier systems, including losses in the windows and long high-efficiency filters, is estimated to be 0.008 for the Michelson and 0.02 for the lamellar grating.

The laser spectrometer located at Emory University uses a 20-W cw CO₂ laser to drive a waveguide-type of far-IR cavity. An internal Fabry-Perot interferometer provides line tuning in the cavity, and additional line filtering and wavelength measurement are provided by a second external Fabry-Perot. Some of the far-IR power is introduced into a feedback loop which uses a PZT piezoelectric element to stabilize the CO₂ laser, although some lines lase with sufficient stability that this feedback is not needed. Further stabilization is provided by a source compensation scheme, where the output of a detector following the sample is electronically divided by the output of a detector preceding the sample to give a ratioed quantity with power fluctuations removed. The two detectors are commercial room-temperature Golay cells with typical responsivities of \(2 \times 10^{8} \text{ V/W}\) and noise equivalent powers of \(10^{-10} \text{ W (Hz)}^{-1/2}\). Standard lock-in amplification of the detector outputs is used with the reference frequency of 11 Hz provided by a mechanical chopper interrupting the far-IR beam. Exact measurements of the laser output power are not available, since there are no well-calibrated far-IR power measuring devices, but estimates based on the above information suggest that the output power remains below \(10^{-10} \text{ W}\) in the 6–30-cm⁻¹ region, and can be as high as \(10^{-9} \text{ W}\) in the 50–200-cm⁻¹ region.
Theoretical analysis is given elsewhere.\(^5\) At 21-24 cm\(^{-1}\) and the low-frequency line shape, the full data for purposes of comparison. The data are in sub-

The comparative results for \(T_s/T_n\) between 6 cm\(^{-1}\) and 180 cm\(^{-1}\) are shown in Fig. 1, which also shows a theoretical fit to the data for purposes of comparison. The data are in sub-

The Fourier data were obtained with a resolution of 1.5 cm\(^{-1}\) and an integration time of 4 sec/point. The data shown are from the average of six interferograms in both superconducting and normal states with the lamellar grating and the average of three interferograms in both states with the Michelson. In this latter case, measurements were made up to 300 cm\(^{-1}\). The intensity maximum was at \(\approx 180\) cm\(^{-1}\) for the Michelson and at 24 cm\(^{-1}\) for the lamellar grating. These frequencies are determined by a number of factors, including the spectral emittance of the source, the beam-splitter efficiency, and the long pass filter employed.

The Fourier data in Fig. 1 have error bars attached to representative points. These points include those near the intensity maxima, those where the intensity is half of and a quarter of these maxima, and the points where the data change from the lamellar grating to the Michelson (near 40 cm\(^{-1}\)). The noise levels are calculated as the standard deviations of the individual spectra which were averaged to give the data of Fig. 1. A check on this calculation exists because the sampling interval is about 20% shorter than the maximum allowable value, so the intensity is zero at the high-frequency end of the computed spectrum. The standard deviation of the data in the region gives the noise level, assuming a white-noise spectrum. These two estimates give similar values for the noise level.

For the laser measurements seventeen powerful lines of the many available\(^3,10\) were selected, giving an average frequency spacing of \(\approx 10\) cm\(^{-1}\) between 11.2 cm\(^{-1}\) and 175.4 cm\(^{-1}\). Typical lock-in time constants were 0.3 sec. The errors for the laser results, taken as the standard deviations of several measurements, are typically \(\pm 2\%\) of the ratio \(T_s/T_n\).

The two sets of data can be compared between 6 cm\(^{-1}\) and 30 cm\(^{-1}\) and between 50 cm\(^{-1}\) and 180 cm\(^{-1}\) but not in the peak region 30-50 cm\(^{-1}\) where the SNR in the Fourier data was quite low. The agreement between laser and Fourier data between 50 cm\(^{-1}\) and 180 cm\(^{-1}\) is excellent, with most of the laser points lying at the means of the oscillations appearing in the Fourier data. The low-frequency agreement is also good except at 21-24 cm\(^{-1}\). Here a pronounced shoulder appears in the laser results but not in the Fourier data. The Fourier results do show a slight convexity at the same frequency.

The comparison makes it obvious that there is no serious disparity between results from the older Fourier and the new laser methods. The disagreement at 21-24 cm\(^{-1}\) may be due to any of several reasons. It may be related to the small difference in superconducting temperatures, which would be of greatest importance at low frequencies. The most intriguing possibility, however, is that the relatively high laser power may cause some nonlinear effect not as yet understood.

Figure 1 clearly shows where each technique has its strengths. The Fourier method in general can give much higher resolution than is available from the quasi-tunable laser. It would probably be practical to double the number of laser lines shown in the figure to give an average spacing of about 5 cm\(^{-1}\), but a resolution much better than this is unlikely with present techniques. The resolution of the interferometers has been demonstrated to be 0.1 cm\(^{-1}\) for the lamellar grating system\(^2\) and 0.05 cm\(^{-1}\) for the Michelson system.\(^3\) The lowest laser line was at 11.2 cm\(^{-1}\), whereas the Fourier data extend to 4 cm\(^{-1}\). At comparable resolution, the random errors in the laser data are equal to those in the Fourier data with the advantage that the former were obtained without cooled detectors requiring costly liquid helium. Further proof of the excellent noise performance of the laser system is given by other measurements\(^5\) in a \(V_3Si\) film of much greater thickness, where the typical transmission is
0.01%. Here the laser spectrometer gave $T_r/T_a$ with an accuracy only slightly worse than that shown in Fig. 1. The Fourier systems, on the other hand, simply cannot be used with a specimen having this small a transmittance.

One feature of the laser system can prove to be either a handicap or an advantage. The narrowness of the laser lines means that interference fringes will appear strongly in cases where the broadband character of the Fourier blackbody source suppresses such effects. A clear advantage of laser spectroscopy is its independence from any computer transformation and analysis of the data and from the related questions of apodization and filtering. On the other hand, the operation of the laser system is at present more complex than that of a Fourier spectrometer. Another measure of the usefulness of the two spectroscopic methods is the actual laboratory time involved in gathering the data shown in the figure. The Fourier results, including setup, data acquisition, and computer data analysis, were obtained in ~20 man hours, while the laser data were obtained in ~60 man hours. While operating, both systems required the attention of one or two researchers.

Our comparisons show that the ideal far-IR–submillimeter spectral source remains elusive. However, the combination of Fourier and laser methods does give the flexibility of choosing the most effective approach for a given spectral measurement problem.

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References