High-resolution far-infrared spectroscopy at NSLS beamline U12IR

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Abstract

A Bruker model IFS 125HR Fourier transform interferometer has been installed and its performance tested using high-brightness, far-infrared synchrotron radiation. Results of absorption measurements for the rotational modes of water vapor demonstrate a nearly 10-fold improvement in signal-to-noise when compared with the instrument’s internal high-pressure Hg arc lamp source. © 2008 Published by Elsevier B.V.

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1. Introduction and background

We report on the performance of a high spectral resolution Bruker [1] IFS 125HR Fourier spectro-meter installed at NSLS infrared beamline U12IR [2]. This instrument features a 10 m maximum path difference and corresponding unapodized resolution of better than 0.001 cm⁻¹. A view of the beamline is shown in Fig. 1.

Such very high resolution is required for the study of narrow rotation/vibrational modes of gas molecules and other systems having long-lived resonances (e.g., cyclotron resonance in ultra-high mobility 2D electron gas systems and magnetic resonance in crystals). Achieving this resolution hinges upon producing good interferometric modulation over the entire optical path difference.

The most important factor for meeting this requirement is that the Jacquinot stop (source aperture) ensures sufficiently flat wavefronts and well-defined phases [3–5]. To meet this requirement, the aperture diameter d and the source-optic focal length, F must satisfy d < 2F (\(\delta\nu/\nu\))¹/² where \(\delta\nu\) is the spectral resolution at frequency \(\nu\). Although there is some freedom in the choice of focal length, practical values are in the 100–500 mm range; the IFS 125HR uses \(F = 418\) mm. Fig. 2 shows the required Jacquinot stop diameter as a function of frequency when operating at the highest resolution, \(\delta\nu = 0.001\) cm⁻¹.

The spectrometer optics are 75 mm in diameter, yielding a numerical aperture, NA = 0.093. Thus the diffraction-limited spot size at the Jacquinot stop is \(s = 1.22/(\nu \times NA)\), with \(\nu\) in cm⁻¹. Fig. 2 shows this diffraction-limited spot size as well as the ratio (Jacquinot/ Diffraction). Because the flux for most infrared synchrotron radiation sources is 100–1000 times greater than for a conventional thermal spectrometer source and because the flux scales as aperture area, the synchrotron source has an advantage over a thermal source when this ratio is less than 10. As can be seen, this is the case for all frequencies below 1000 cm⁻¹. Indeed, the ratio actually falls below 1 near 10 cm⁻¹ meaning that the Jacquinot requirement exceeds the diffraction limit.

In practice, one loses some of the intrinsic synchrotron source brightness at very low frequencies, e.g., below about 10 cm⁻¹ due to limits on the beamline’s extraction apertures. Fig. 3 illustrates this point, by showing the calculated brightness for the NSLS U12IR beamline. Thus, the spectral range where synchrotron radiation can be expected to offer the maximum advantage is over 20–1000 cm⁻¹. Even below 20 cm⁻¹, where the extraction limits the performance, the synchrotron emits more energy than the strongest thermal sources [2].
2. Measurements and results

As a test, we have measured the pure rotational spectrum of water vapor \([5-7]\) over 20–100 cm\(^{-1}\) using both the synchrotron source and the spectrometer’s internal high-pressure Hg arc lamp source. We used a 125 \(\mu\)m thick Mylar beamsplitter and a low temperature \((T = 1.5 \text{ K})\) bolometer detector. The detector included a cold low-pass filter to limit its bandwidth to frequencies below 100 cm\(^{-1}\). The water vapor pressure was set to 4 Torr using a fine metering valve and absolute pressure gauge. At this pressure, a 1 m path length was found to produce good absorbance spectra (most features not saturated). Spectra were collected with the spectrometer set to produce 0.001 cm\(^{-1}\) resolution. A Jacquinot stop aperture of 4 mm diameter was used to achieve this resolution at 100 cm\(^{-1}\). Two scans of the interferometer were used for each spectrum, with identical collection for the Hg arc lamp and the synchrotron source. Some example transmission spectra are shown in Figs. 4–6. Fig. 4 shows a relatively wide spectral range spanning four absorption features of varying intensity.

As can be immediately seen, the synchrotron spectrum has significantly lower noise, a consequence of the 1000x higher source brightness and corresponding larger signal at the detector. Note that only the lowest frequency mode in this sequence (at 85.63 cm\(^{-1}\)) is observed with the Hg arc lamp source; even then, the S/N is not sufficient to obtain an accurate line width. Similar results are obtained for other absorption modes, with several illustrated in Fig. 5.

We note that, to resolve these features requires a path difference approaching 10 m. This retardation is not only larger than the electron bunch length (\(\approx 50 \text{ cm during stretched bunch operation}\)) but also exceeds the 5.7 m bunch-to-bunch spacing of the storage ring. This result confirms that overlapping of the storage ring’s light pulses at the sample or detector is not necessary to achieve interferometric modulation, a result first reported by Schweizer \([8]\).

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Fig. 1. Bruker IFS 125HR installed at beamline U12IR, shown configured with a 16 T superconducting solenoid for magneto-spectroscopy.

Fig. 2. Calculated Jacquinot aperture and diffraction-limited source size for the IFS-125HR at 0.001 cm\(^{-1}\) resolution. Also shown is the ratio of these two dimensions. The throughput becomes diffraction limited when the ratio is equal to unity.

Fig. 3. Calculated brightness for the synchrotron radiation (SR) source at beamline U12IR and a 3500 K blackbody. Note the logarithmic scales. The SR source falls below the ideal brightness at frequencies <30 cm\(^{-1}\) due to the limiting extraction aperture and the beamline UHV window.

Fig. 4. Transmission spectra for water vapor. The curves have been offset by +/- 0.2 for clarity.
Nor do any problems arise when pulses from two distinct bunches overlap (in contrast to what would occur with a coherent synchrotron radiation mode having excellent bunch-to-bunch phase stability). Indeed, it is the sample itself that defines and controls the interference necessary to resolve a narrow feature.

A sample with a very narrow spectral feature acts as a filter that rings a sufficiently long time to cause coherent overlap of the light from the two arms of the interferometer. This ringing occurs independent of the pulsed nature of the source (assuming a coherence length much less than the bunch length).

3. Conclusions

In summary, we demonstrate very high resolution spectroscopy using synchrotron radiation from NSLS beamline U12IR and show a significant performance advantage over the instrument’s conventional source. A signal-to-noise advantage of better than 10 implies measurements could be completed in 1% of the time for the same spectral quality. This instrumentation is also used regularly for time-resolved pump-probe spectroscopy in combination with a synchronized Ti:sapphire laser source [9].

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References