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Gaseous effluent monitoring and identification using an imaging Fourier transform spectrometer

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ABSTRACT

We are developing an imaging Fourier transform spectrometer for chemical effluent monitoring. The system consists of a 2-D infrared imaging array in the focal plane of a Michelson interferometer. Individual images are coordinated with the positioning of a moving mirror in the Michelson interferometer. A three dimensional data cube with two spatial dimensions and one interferogram dimension is then Fourier transformed to produce a hyperspectral data cube with one spectral dimension and two spatial dimensions. The spectral range of the instrument is determined by the choice of optical components and the spectral range of the focal plane array. Measurements in the near UV, visible, near IR, and mid-IR ranges are possible with the existing instrument. Gaseous effluent monitoring and identification measurements will be primarily in the "fingerprint" region of the spectrum, ($\lambda = 8$ to $12 \mu\text{m}$). Initial measurements of effluent using this imaging interferometer in the mid-IR will be presented.

1. INTRODUCTION

The goal of this research is to develop a spectrally resolving, 2 dimensional imaging system for a wide variety of remote sensing applications including the identification and mapping of gaseous effluents. We are developing an imaging Fourier transform spectrometer (IFTS) for several reasons. The primary advantages of the IFTS are the capacity to acquire more than an order of magnitude more spectral channels than alternative systems with more than an order of magnitude greater étendue than for alternative systems. The primary disadvantage of Fourier transform spectroscopy is the sensitivity to random or periodic temporal fluctuations.

In general, imaging spectrometers acquire a three dimensional "data cube" consisting of 2 spatial and 1 spectral dimension. There are many different spectrometer designs capable of assembling such a data cube. Any non-imaging spectrometer may raster its field of view across a scene to produce a spatially and spectrally resolved data cube. This technique tends to be very inefficient. The availability of two dimensional focal plane arrays enables highly efficient acquisition of two dimensional imaging data. An imaging grating spectrometer (IGS) disperses light in 1 spatial dimension, and provides imaging in an orthogonal direction. One can scan the field of view of the instrument in one spatial dimension and concatenate a succession of 2 dimensional (1 spectral and 1 spatial) images and form a 3 dimensional data cube. This is the well known "pushbroom" geometry. An Echelle grating spectrometer (EGS) uses 2 orthogonal directions for coarse and fine spectral resolution, and is intrinsically not well suited for imaging spectroscopy, other than by rastering. An imaging Echelle grating spectrometer (IEGS) is possible based on temporally multiplexing the openings of a programmable slit array at the entrance aperture. If each pixel element is varied at a discrete multiple of a fundamental frequency, then FFT can be used to efficiently disentangle the image in the focal plane. Such a scheme has high optical throughput, but is very cumbersome. An imaging Fizeau interferometer (IFT) produces a spatially extended interferogram in 1 dimension, and has imaging in an orthogonal direction. A variable spectral filter spectrometer (VFS), such as a filter wheel or tunable filter acquires a full 2 dimensional image each frame, with successive frames viewed through successive filters.

Finally, an imaging Fourier transform spectrometer acquires a full 2 dimensional image per frame, with successive frames associated with different positions of a moving mirror in an interferometer. This is illustrated in figure 1, where a set of frames of a complete 2 dimensional scene are shown in a stack. The variation of light intensity for a single pixel in the scene as a function of the

position of the moving mirror is indicated by the interferogram curve in figure 1. The raw data cube for an IFTS consists of the interferogram in 1 dimension and the 2 dimensional scene in the 2 orthogonal directions. Fourier transformation of the interferograms for each pixel in the field of view then produce the spectral dimension of the data cube, as is illustrated by the bottom curve in figure 1. Most

Figure 1. IFTS raw data cube.

of the existing imaging spectrometers are IGS, e.g. HYDICE, ASAS, AVIRIS, CASI, with a few VFS systems, e.g. MUSIC, and some IFI systems, e.g. SMIFTS.

The number of spectral channels practical with either IGS, or IFI is limited by the width, in number of pixels, of the detector system. In practice the number of spectral channels is at most a few hundred. Similarly, VFS systems are limited in practice to a few hundred discrete filter elements. For applications where the spectral features are fairly broad, such resolution is sufficient. Most of the existing imaging spectrometers are sensitive in the visible, or near visible, and indeed do not require high resolution for most applications. For spectral regions where the spectral features are very narrow, and spread over a wide spectral range, it is advantageous to have higher resolution than is practical with IGS, VFS and IFI systems. In particular, in the infrared, molecular spectra of vapors in the atmosphere are often a fraction of 1

cm^{-1} in width, and it is important to have high resolution over a broad range of wavenumbers in order to discriminate chemical compounds. The great advantage of an IFTS system is the very large number of spectral elements combined with imaging. It is natural to develop IFTS for the infrared region.

2. BASIC PRINCIPLES OF IFTS

Fourier transform spectrometers are based on the Michelson interferometer, with one fixed mirror and one moving mirror. The light transmitted through the interferometer is measured as a function of the displacement of the moving mirror from the zero phase difference position. The Fourier transform of this "interferogram" yields the spectrum. An imaging spectrometer is obtained by "looking through" the interferometer with a camera, and constructing the Fourier transform of the variations in light intensity at each pixel as a function of the position of the moving mirror. The optical layout of an IFTS is sketched in figure 2.

Typical rays emerging from two representative points are drawn. One point is located on the optical axis. The second point is displaced by a distance y from the optical axis. The object plane is located at a distance equal to the focal length f of the input collimating lens. The object plane may be a real emitting surface, or may be the focal plane of a telescope or microscope. Alternately, one could replace the input lens with an aperture of diameter D . This corresponds to the case $f = \infty$. The focusing lens, with focal length f' produces an image with magnification factor f/f' . The depth of field of the focusing system should accommodate twice the maximum travel distance of the moving mirror in the interferometer in order to maintain a sharp focus in the image plane. The beam splitter transmits a fraction T of the incident light and reflects a fraction R . These coefficients in general depend on the polarization of the incident light, the angle θ , and the wavelength.

For monochromatic light of wavenumber $k=1/\lambda$, the intensity of light at the image point y' relative

Figure 2. A sketch of the optics of an Imaging Fourier Transform Spectrometer

to the intensity emerging from the object point y is proportional to $R \cdot T \cdot \cos^2(\delta/2)$. The phase difference δ is determined by the round trip optical path difference x for the moving mirror with respect to its zero phase difference point, and the angle $\theta = y/f = y'/f$, between the collimated rays and the optic axis,

$$\delta = 2\pi kx \cos(\theta) \quad (1)$$

A point source in the object plane produces an extended spot in the image plane. The Raleigh diffraction limit on the degree of focus is $1.22 \lambda f/D'$. For reasonably fast optics, this limit is significantly smaller than the typical pixel size. Thus spatial resolution is limited by pixel size rather than by diffraction. For a non-monochromatic point source, the observed light intensity $I(x, y')$ in the focal plane is a function of both mirror position x and the distance from the optic axis y' . For a spectral distribution with intensity for wave numbers between k and $k+dk$ given by $S(k)dk$, and a detection efficiency E , the observed light intensity in the focal plane is given by

$$I(x, y') = \int S(k)/2 E \cdot R \cdot T \cdot (1 + \cos(2\pi k x \cos(\theta))) dk \quad (2)$$

The light intensity at any given radius y' is thus simply related to the Fourier transform of the spectrum. Introducing the modified frequency

$$k' = k \cos(\theta), \quad (3)$$

and the modified spectrum $S'(k') \int E \cdot R \cdot T \cdot S(k) = E \cdot R \cdot T \cdot S(k' \sec(\theta))$, the observed intensity is

$$I(x, y') = \sec(\theta) \int S'(k')/2 (1 + \cos(2\pi k' x)) dk'. \quad (4)$$

By Fourier transform of the $I(x,y')$ as a function of x , the spectrum $S'(k')$ can be recovered, and thus $S(k)$ itself. Ordinarily, a range of values of y' are integrated, which has the effect of broadening and shifting a monochromatic line in the spectrum which is recovered from the Fourier transform of $I(x)$. This is the origin of the Jacquinot limit¹ on the resolution of a Fourier transform spectrometer. For a Jacquinot stop of radius r , to first order in r/f , the total width of the Jacquinot blurring is

$$dk/k = r^2/2f^2. \quad (5)$$

For points off the optic axis, this blurring varies linearly with distance from the optic axis. In terms of the angular position q and spread dq of a given pixel, this limit to the spectral resolution is given by

$$dk/k = \tan(\theta) \delta\theta. \quad (6)$$

Except for extremely high resolution instruments, this limitation is small compared to the limit on the resolution from the total travel distance of the moving mirror. For our prototype instrument in particular, $\theta < 0.15$, $d\theta = 0.0025$, so that this resolution limit is 0.4 cm^{-1} at 1000 cm^{-1} for objects at the extreme edge of the field of view, while at the center of the field of view, this resolution limit is only 0.003 cm^{-1} .

3. IFTS ADVANTAGES

In non-imaging Fourier transform spectrometers, high resolution can only be obtained with the use of a relatively small aperture for the Jacquinot stop. A small Jacquinot stop then limits the étendue of the conventional Fourier transform spectrometer. To first order, the étendue for a conventional Fourier transform spectrometer whose limiting aperture area (usually determined by the beam splitter) is A , is given by

$$2\pi A dk/k. \quad (7)$$

In contrast, a conventional grating spectrometer, with entrance slit of height h , having the same resolution dk/k , and the same focal length f , has an étendue which is less than that of a conventional FT instrument by the factor^{2,3}

$$h/2\pi f \quad (8)$$

Since the slit height is usually substantially less than the focal length in a grating spectrometer, a non imaging Fourier transform spectrometer enjoys 2 or 3 orders of magnitude larger étendue than a grating instrument. This advantage is known as the Jacquinot advantage of conventional Fourier transform spectrometers. For imaging spectrometers, the étendue per frame of the focal plane detector is less for IGS than for IFTS by approximately the same factor as for non-imaging spectrometers. The number of frames per data cube for the IGS is simply the number of angular resolution elements per total field of view. The number of frames per data cube for the IFTS is instead the number of spectral resolution elements. For equal volume data cubes, the IFTS throughput relative to IGS throughput is still given by approximately the ratio in equation (8). However, in an IFTS, the pixel size represents an effective Jacquinot stop, and the spectral resolution can be greatly increased with no sacrifice of étendue. Thus in an IFTS, a much larger volume data cube can be acquired for a given scene, with the extended dimension being the spectral dimension.

Another well known advantage of Fourier transform spectrometers, the Connes advantage, is that the wavenumber scale and instrumental line shape are precisely determined, and are independent of wavenumber. Because the off axis distortion of the spectra is given by the simple (and slight) linear

shrinking of the wavenumber scale expressed by equation. (3), the Connes advantage of Fourier transform spectrometers is maintained in the imaging implementation. It is practical to readily compare, or subtract, even very complex standard reference spectra taken with different Fourier transform spectrometers. In contrast, the instrumental line shape of grating spectrometers in general, and a fortiori IGS, is dependent on wavelength and position in the focal plane. It is therefore much more difficult to compare IGS data with complex standard reference spectra without extensive and precise calibration.

4. IFTS DISADVANTAGES

The sensitivity of Fourier transform instruments depends on the signal to noise ratio of the interferogram, the number of resolution elements N , and the form of the spectrum. For a one sided interferogram consisting of N points, the corresponding spectrum will contain N spectral channels equally spaced extending from 0 up to the Nyquist sampling limit, $k_{\max} = 1/(2\Delta x)$, where Δx is the sampling interval. Let M be the ratio of the total observed intensity to the average intensity per channel. M is roughly the ratio of the width of the spectrum to the spectral resolution, or the number of significant channels. If there is white noise in the interferogram, there will be white noise in the spectrum. Let f be the ratio of the rms noise in the interferogram to the maximum intensity in the interferogram. The rms spectral noise relative to the typical intensity in the spectrum is approximately given by $f \cdot M/\sqrt{N}$. For high resolution and high sensitivity it is important to have a Nyquist limit significantly greater than the extent of the detected spectrum.

For a broad band spectrum, with a series of absorption lines, the noise equivalent absorbance varies as $\sqrt{N}$, whereas the depth of the absorption line varies linearly with the resolving power, and is therefore proportional to N , up to the point that the resolution is approximately equal to the line width. Thus it is advantageous to operate with spectral resolution matched to the width of the spectral features of interest. For relatively broad spectral features for which the resolution of an IGS or VFS is adequate, the sensitivity of an IFTS is likely to be inferior. For narrow spectral features, however, the sensitivity of an IFTS can be substantially larger than that of an IGS or VFS.

The greatest disadvantage of the IFTS is the difficulty of observing fluctuating sources. Since a Fourier transform spectrometer inherently uses time domain multiplexing of the spectral data, with spectral intensity vs. modulation frequency translated into spectral intensity vs. wavenumber, any significant temporal fluctuations in the observed light intensity within the frequency range corresponding to the spectral features of interest represents noise in the derived spectrum. Furthermore, periodic fluctuations produce line shifts of the observed spectral features, with the implication that it is very difficult, for example, to measure ac modulated calibration sources. Under certain circumstances however, this aspect of an IFTS may be used to one's advantage. For example, it is possible to use the broadening of ordinarily narrow spectral features as a measure of the degree of temporal fluctuation of the source term, and this can be used to distinguish transient or erratic spectral features.

5. APPLICATIONS OF IMAGING FOURIER TRANSFORM SPECTROMETRY

There are many applications of an IFTS. The remote observation, identification and visualization of visible and non-visible gaseous effluents such as environmental pollutants, chemical warfare agents, and effluent indicative of clandestine activity is an important application. There are many other areas where spectrally resolved imaging of gaseous and non-gaseous substances is important including: astronomy, forensics, biological and medical imaging, military applications, environmental monitoring. The applications range from chemical identification to spectrally resolved, infrared thermography to imaging cellular structures. All applications have in common the need to spectrally resolve features in the emission or absorption spectra to characterize and understand a complicated image.

6. PROTOTYPE DATA

We have begun to address several of the important technical issues of effluent detection with the IFTS by utilizing commercially available spectrally and spatially resolving instruments to observe standard gas plumes. With these instruments we have studied the detectabilities of various inert gases, the distinguishability of these gases and their near-source dispersal. Figure 3 shows a spatially resolved image of a freon-22 (chlorodifluoromethane) gas plume with clear night sky in the background. This image was taken with a commercially available thermal infrared scanner with a HgCdTe detector. The scanner is sensitive throughout the 8-12 μm region of the spectrum, the water window. In order to easily see the plume these images display the excess emission with respect to a reference image taken just before the gas was released. The gas plume is invisible to the naked eye, even in bright daylight, beyond a few inches from the end of the nozzle. The images are also invisible to a similar IR scanner with sensitivity from 2-5 μm . The gas plume is visible in the 8-12 μm region of the spectra because of excess emission, above the sky emission, in the 8-12 μm region of the spectrum.

Figure 4 shows a series of spectra taken with a high resolution Fourier transform spectrometer

Figure 3. Freon gas plume imaged with an 8-12 μm infrared scanner.

looking at a fixed area in the image above. The resolution of these spectra is 4 cm^{-1} . The three traces in figure 4 show the observed freon-22 excess emission in the water window region between 8 and 13 μm (750 to 1250 cm^{-1}). These emission spectra are actually the result of subtracting a reference spectra (cold night sky only) taken just before the release of the freon gas. As the gas plume enters the field of view of the spectrometer, and thus the thickness of the gas plume within the field of view increases, the intensity of excess emission increases.

The integrated intensity over the range shown in figure 4 was measured as a function of time and is shown in figure 5. For this data the gas jet was released in a pattern of two short bursts followed by a longer burst. This time history is clearly evident in figure 5. Also shown in figure 5 is the intensity of excess emission seen with the infrared scanner integrated over a 6" square area near the center of the field of view of the FTIR spectrometer. The correspondence between the time dependence of the integrated intensity of excess emission as measured by the two instruments is excellent. The difference between the two curves can be attributed to the imperfect overlap of the fields of view of the two instruments.

A comparison of the measured freon-22 emission spectra from the thickest part of the gas plume with a quantitative reference spectrum⁴ is shown in figure 6. The calibrated reference spectrum has been offset in the figure for ease of comparison. In the region of the spectra with the highest specific

Figure 4. Excess emission spectra for freon-22 gas plume with sky background.

Figure 5. Time history of excess freon-22 emission as measured by an IR imager and a FTIR spectrometer.

absorbance (near 1117 cm^{-1}), the emission spectrum is near saturation. That is, the gas plume is nearly optically thick in this region of the spectrum. The fraction of the spectrum that is nearly in saturation is so small, however, that the integrated emission intensity over the water window, from 755 cm^{-1} to 1255 cm^{-1} , is nearly a linear measure of the thickness of the gas plume.

The relative signal to noise ratio for the spatially resolved and spectrally resolved data is of critical importance. We can calculate the noise level for both the thermal imager and the FTIR spectrometer using the fluctuations in the intensity level shown in figure 5 just before the gas plume enters the

instrumental field of view. The signal to noise level for the FTIR spectrometer was 620. The signal to noise level for the infrared imager was 360. The observability of an infrared emitting gas plume with a high-efficiency non-imaging FTIR spectrometer is approximately the same as the observability with a high efficiency, imaging, but not spectrally resolving, infrared camera. The critical difference is that the infrared camera can profile the spatial extent of the plume but cannot identify the gas, while the spectrometer can identify the gas but cannot measure the spatial extent of the plume.

Figure 6. Measured and reference spectrum for freon-22 gas plume.

The observability of gaseous effluent with a warm background has also been investigated using the IR imager and FTIR spectrometer. In this case, the infrared signature in the thermal region of the spectrum depends on a radiance contrast between the gas plume in the foreground and the emission from the background. Typically, the

background emission has a spectrum well characterized by a gray body, i.e. a black body with emissivity less than unity. In the simplest case where the background is a black body, there will be an infrared signature for the gas plume only if the gas temperature is hotter or colder than the background. If the gas is hotter, then emission lines characteristic of the gas are observed. If the gas is colder than the background, an absorption spectra will be observed. In practice, the natural environment is never isothermal, and it is almost always the case that radiation temperatures of a few degrees are easily observable within a field of view.

As a simple test of the variation in detectability of a simple gas plume with a solid backlighter, we acquired simultaneous IR images and FTIR spectra of a freon gas plume with a building as a background source. Within the field of view of the IR imager there was an open metal door. The inside of the building, viewed through the open door, served as a background with temperature warmer than the gas plume. The open door surface served as a solid background whose temperature was colder than the gas plume. Figure 7 shows one infrared image of the excess emission with respect to a reference image taken just before the gas was released.

The left side of the image is backlit by the open door and the gas is clearly observable as a source of absorption. The right side of the image is backlit by the surface of the metal door and the gas plume is observable as a source of excess emission.

Figure 8 shows the measured emission and absorption spectra for the freon-12 gas plume with the building in the background. The baseline of the reference spectrum soybean offset for display purposes. The quantity plotted in figure 8 is the ratio of the measured emission at a particular time to a background spectrum taken shortly before the gas plume entered the field of view. The lower curve shows the spectrum with the surface of the open door in the background. In this case, the gas plume is warmer than the background and a freon-12 emission spectrum is measured. The upper curve shows the measured spectrum with the open doorway in the background. In this case, the interior of the building is warmer than the gas plume and the presence of the gas is characterized by it's absorption spectrum. Note that the spectrum is significantly different from the previously measured freon-22 spectra and it is quite easy to distinguish between the two gasses.

In the case of the gas plumes backlit by a solid object near ambient temperature, the signal to noise ratio is much reduced from the case where the plume is backlit by clear sky. In this particular case the signal to noise ratio for the IR imager is about 100 and the signal to noise ratio for the FTIR spectrometer is between 25 and 30. The expected signal to noise is clearly a function of the temperature difference between the gas plume and the background.

Figure 7. Image of excess IR emission of a Freon-12 gas plume with a building in the background. The area to the left of the dividing line has a warm room in the background, the area to the right of the divider has a cold metal door in the background.

Figure 8. Emission and absorption spectra for freon-12 gas plume with a building in the background. Also shown is the freon-12 reference emission spectra.

7. IFTS DATA

It is clear from the above data that there is value in both the spatial imaging of a gas plume to determine its extent and the spectroscopic measurement of the plume to determine the composition of the plume. We have constructed and begun tests with a prototype imaging Fourier transform spectrometer (IFTS) which combines 2-dimensional imaging and high resolution spectroscopy.

In order to illustrate both the spatial and spectral resolution simultaneously, a set of gas cells 6 inches long by 2 inches in diameter, containing various vapors, were positioned 1 foot from the entrance aperture of the IFTS instrument. A hot plate set at 350°C was positioned 2 feet behind the gas cells to provide IR backlighting. The gas cells had silicon windows which were nearly transparent to the IR. The instrument total field of view was approximately 9°, and contained about 3,000 spatial pixels. Figure 9 shows the image seen at the zero phase difference mirror position, where the interferometer transmits all wavelengths equally. This image is equivalent to that seen directly by an infrared camera. The brightest points in this image correspond to the corners of the hot plate, which are unattenuated by the gas cells. The 3 dimmer circular features surrounding the center in this image correspond to the circular faces of the partially transmitting cylindrical gas cells. About 1500 interferogram points were measured, thus the volume of the data cube was nearly 5×10^6 points. By Fourier transform of the interferogram dimension of the raw data cube, a processed data cube consisting of 2 spatial and 1 spectral dimension was obtained.

Representative spectra from 3 different single pixels, located within parts of the image corresponding to Acetone, Propanol, and Methyl Ethyl Ketone gas cells are shown in figure 4. The spectral band pass of the system for this data extended over the range from 800 to 2500 cm^{-1} . The Nyquist upper limit on wavenumber was 7899 cm^{-1} . The spectral resolution was 6 cm^{-1} , independent of

wavenumber. The spectral noise observed was "white", at an rms level for various pixels in the different samples ranging from 1% to 2% relative to the intensity at the peaks of the spectra. Averaging of the interferograms for N neighboring pixels within a region corresponding to a given spectrum reduces the spectral noise in proportion to $\sqrt{N}$. In the spectra, prominent absorption of the hot plate emission by ambient water vapor and CO_2 present in the 3 feet of air between the hot plate and the IFTS has been indicated in the figure. Note that the spectra for the three different chemicals are very distinct, especially in the "IR fingerprint region" between 800 cm^{-1} and 1600 cm^{-1} .

Figure 9. IR image of gas cells

Using the interferograms for the three pixels whose spectra are shown in figure 10, a cross correlation with all of the other pixels in the image can be computed. Specifically, for each pixel, the individual values in its interferogram may be regarded as the components of a vector in an N dimensional space. The cosine of the angle between this vector and corresponding vectors for all of the other pixels in the image can be calculated from the scalar product of the two vectors. A plot of this overlap factor for each pixel in the field of view then is greatest where the spectra are most similar, and becomes small for dissimilar spectra. In figure 11, the correlation images for spectral features characteristic of the 3 vapors, Acetone, Propanol, and Methyl Ethyl Ketone are shown.

It can clearly be seen in figure 11 that the cross correlation images strongly enhance the contrast for a particular chemical species.

Figure 10. Spectra for three different pixels in the field of view of the IFTS.

MEK

Propanol

Acetone

Figure 11. Cross correlation images corresponding to three different spectra; MEK, Propanol and Acetone.

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