Terahertz spectroscopy with a holographic Fourier transform spectrometer plus array detector using coherent synchrotron radiation

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With the development of coherent broadband terahertz (THz) synchrotron sources based on the radiation produced by short bunches of electrons in storage rings [1], interest has developed in the appropriate spectroscopic instrumentation for bunch length measurements [2,3], which correspond to the subpicosecond time scale. In addition to single shot spectroscopy measurements there is also interest in using this millimeter and submillimeter coherent wave radiation signature from the bunch for its structural analysis. Spectroscopic measurement of the bunch length by means of its coherent radiation signature becomes technically easier as the bunch gets shorter in length since the resulting radiation extends over a larger frequency interval. The determination of the bunch form factor using this technique has been described in some detail [4–8].

A candidate instrument for the types of study just outlined is a holographic Fourier transform spectrometer (HFTS), which has no moving parts and produces an interferogram in the spatial, not in the time domain. Thanks to its high throughput, the HFTS can be used in remote and time-dependent sensing areas, where it can outperform even multichannel spectrographs [9]. The extremely broad spectral coverage of the HFTS would be particularly beneficial in the THz region in which a diffraction grating cannot be used without elaborate optics to filter high diffraction orders. But the large interference angle required for a THz HFTS makes associated field aberrations too large for a realistic size of the array detector. This difficulty has been resolved by dividing the Fourier optics into two halves with a tilt interferometer design [10]. Here we present experimental tests of this new THz HFTS instrument using the coherent synchrotron radiation produced in the energy recovery linac of the Free Electron Laser Facility at Jefferson Laboratory.

A three-dimensional schematic of the THz HFTS optical design is shown in Fig. 1. The fore optics consisting of a 45° off-axis parabolic mirror accepts the collimated THz radiation. The intermediate focus is formed slightly behind a wire grid beam splitter, which also serves as the field angle-limiting aperture. The motor driven shutter in each arm provides the means to measure the intensity in each interferometer arm. The two tilted mirrors direct the beams toward the 30° off-axis parabolic mirrors, which serve as the mirror analog of the Fourier transform lens. Finally, the collimated beams converge on the
array detector located with its plane perpendicular to the axial beam bisector. An additional wire grid polarizer is located next to the detector (not shown) to mix the orthogonal polarizations that propagate along the interferometer arms.

Assuming that the amplitude transmission and reflection coefficients are \( \tau \) and \( 1 - \tau \) at the beam splitter for a specific polarization analyzed by the detector, the intensity produced in the detector plane \((x,y)\) by beams along the axial directions is given by the following relation [10]:

\[
I(x,y) = \int_0^\infty P^0(\sigma,x,y)\left\{\tau^2 + (1-\tau)^2 + 2\tau(1-\tau) \times \cos(2\pi\sigma y)\sin(\alpha/2)\right\}B(\sigma)d\sigma,
\]

where \( P^0(\sigma,x,y) \) is the intensity at frequency \( \sigma \equiv 1/\lambda \) (\( \lambda \) is the wavelength). The \( 2\sin(\alpha/2) \) factor in the cosine argument determines the scale of the interference pattern. This scaling factor determines the spectral resolution and the high frequency cutoff of the instrument for a given size and pixel dimension of the array detector. The only commercial array detector available today for the THz region is the Pyrocam III, which was used in these experiments. Its sensitivity is sufficiently low that this instrument could not be tested with incoherent sources of THz radiation such as a globar or mercury arc. All the experimental tests of the new spectrometer were performed using coherent THz synchrotron radiation.

Synchrotron radiation produced by short electron bunches in one of the bending magnets of the Jefferson Laboratory Free Electron Laser Facility [11] was delivered through a vacuum optical transmission line [12] into a class 4 laser shielded laboratory. Figure 2 shows the actual arrangement. After the diamond exit window (1), the THz radiation propagated in air and is collimated with a 90° parabolic reflector (2). The original horizontal polarization of the synchrotron radiation after reflections in the transfer line appeared at the optical table at a small angle from the vertical direction. (It is referred to as horizontal in the following.) For measurements of transmission spectra an intermediate focus was created between a pair of additional 90° parabolic mirrors (3) and (4). The tilt interferometer (7) is located in the collimated beam, and the array detector (8) is installed on a translation stage so that the center burst of the static interference pattern produced by the interferometer can be positioned at any pixel of the array.

To compensate for the nonuniform illumination of the detector area, the following calibration procedure was used. Three separate measurements were performed for various positions of the shutters blocking the beam in each arm of the interferometer; see Fig. 1. First, the background intensity distribution \( I_0(x,y) \) in the array was measured with both shutters closed. Then two intensities, \( I_1(x,y) \) and \( I_2(x,y) \), were measured for shutters blocking only one of the interferometer arms. These distributions, \( I_1(x,y) - I_0(x,y) \) and \( I_2(x,y) - I_0(x,y) \), where the scattered parasitic light is subtracted from each arm of the interferometer, were added and normalized to give the correction factor \( k(x,y) \) between 0 and 1. The corrected intensity distribution \( I_c(x,y) \) was calculated from the signal \( I(x,y) \) with both shutters open according to the following relation:

\[
I_c(x,y) = \frac{I(x,y) - I_0(x,y)}{k(x,y)}. \tag{2}
\]

The same correction factors were used for the reference and for the sample spectra during subsequent transmission measurements. This procedure ignores any frequency dependence of the beam splitter efficiency and assumes that the intensity distributions produced by the two interferometer arms differ only by a constant factor. Generally, the latter point is not true for a polarizing beam splitter with different intensity distributions for horizontal and vertical polarizations of the synchrotron radiation. To avoid this problem, an additional polarizer transmitting only
horizontal polarization was installed before the interferometer.

Measurements were performed in a quasi-continuous mode using the internal chopper of the Pyrocam detector to interrupt the incoming THz beams at 48 Hz. A laptop computer running LabVIEW software controlled the operation of the shutters and of the array detector. Spectra were calculated and displayed in real time. Two positions were used for the center burst of the spatial interferograms. When the maximum possible resolution was desired, it was located at the 10th pixel; when a better phase correction was required, it was located at the 20th pixel from the edge. The Pyrocam III detector has $124 \times 124$ square pixels with a 100 $\mu$m pitch so the resulting spectral resolution is 1.2–1.3 cm$^{-1}$, with a maximum nonaliased frequency of 114 cm$^{-1}$.

Examples of corrected static interference patterns and corresponding calculated spectra are shown in Fig. 3. Notable are the periodic oscillations in the spectra. These measurements were performed with the following parameters of the energy recovery linac: current 1 mA, bunch charge 100 pC, repetition rate 9.36 MHz, and beam energy 114.65 MeV. The Pyrocam III accumulation time was 1 s. Detuning the bunch energy by changing the gradient field in the RF cavities varied the electron bunch length. The sharpest center burst is observed for the $-70$ to $-140$ keV detuning, hence the narrowest electron bunch is observed at this beam energy.

To determine the spectral response of the array detector it was used together with a scanning Michelson interferometer to analyze the same synchrotron radiation. Both the scanning Michelson and the HFTS spectral results are shown in Fig. 4 by dot–dash and solid curves, respectively. Apart from a difference in resolution, the two spectra are quite similar, confirming that the nonuniform spectral response of the THz HFTS measurement is due to the detector array. For an additional comparison the same spectral region was measured with a Golay cell in the scanning Michelson interferometer (dashed curve in Fig. 4). Crossed wire grid polarizers were used to attenuate the radiation to the level acceptable for the Golay cell. The most probable explanation of the spectral nonuniform characteristics of the array detector is the production of a channel spectrum in its pixels. The 15 cm$^{-1}$ measured period corresponds to a 50 $\mu$m thickness for the lithium tantalate used in the detector elements. Note that similar nonuniform spectral response due to a channel spectrum has also been reported in some single element pyroelectric detectors [8].

To examine the high frequency characteristics of the THz HFTS a transmission spectrum of a band-pass THz filter (QMC Instruments) was measured as well. The results for the HFTS are compared with those from a lamellar interferometer with a Golay detector in Fig. 5. The angular distribution of the coherent synchrotron radiation is wavelength dependent due to the electron beam vacuum chamber screening effects [13]. For the effective focal length of 10 cm used here the angular distribution is sufficiently narrow at the filter transmission maximum.

Fig. 3. Coherent synchrotron radiation interference patterns for different electron bunch lengths and corresponding coherent synchrotron spectra. The larger the frequency interval of the spectrum, the shorter the bunch. The narrowest bunches occur when the gradient field in the RF cavities is between $-70$ and $-140$ keV. The Pyrocam array has a highly nonuniform spectral response that is due to the channel spectrum produced in the elements.

Fig. 4. Comparison of coherent synchrotron spectra measured with a scanning Michelson interferometer and for the THz HFTS. Solid curve, THz HFTS; dot–dash curve: Michelson interferometer with the same detector; dashed curve, Michelson interferometer with a Golay cell.
(28 cm\(^{-1}\)) so that the collimated beam did not cover the entire area of the array detector at this frequency. This resulted in the effective degradation of the spectral resolution of the HFTS but the general shape of the bandpass filter spectrum is similar for both kinds of spectrometer.

For a more detailed spectral test of the HFTS plus array detector the channel spectrum of a silicon slab was also measured and compared with that determined from a lamellar interferometer plus Golay measurement. The results are shown in Fig. 6, where a clear difference between the two spectra is evident. To understand this difference first the parameters for the standard lamellar spectrum are determined. The silicon wafer of \(d = 401 \mu m\) thickness has only one side polished; the other surface is rough with a characteristic grain size of \(\sim 50 \mu m\), determined with the aid of an optical microscope, so only a two-pass interference spectrum is expected. The equation that describes the silicon wafer spectrum fit with one two-pass interference beam is

\[
T(\sigma) = \frac{16\pi^2}{(n+1)^4} \left[ 1 + \frac{(n-1)^4}{(n+1)^4} + 2 \frac{(n-1)^2}{(n+1)^2} \cos(4\pi nd + \varphi) \right],
\]

where the constant phase angle \(\varphi\) has been included to take into account the change produced by the free carrier contribution at frequencies below this range. The exact properties of the rough wafer surface are not included in this model expression. Fitting Eq. (3) to the lamellar spectrum gives \(n = 3.43 \pm 0.03\) and \(\varphi = -1.2 \pm 0.4\) for the silicon refractive index and the phase at THz frequencies. This spectrum is represented by the dotted trace in Fig. 6. Clearly, the modulation occurs at the correct frequency intervals but the experimentally measured amplitude is greatly reduced because of the surface roughness.

Next, consider the irregular frequency shifts that appear in the THz HFTS spectrum. These frequency shifts are consistent with a beating between two different silicon channel spectra. One spectrum is described by Eq. (3) and the second has a different scale resulting from the beams incident on the array at a slightly different angle. The result is Eq. (4):

\[
T(\sigma) = A + C \{ D \cos(4\pi nd + \varphi) + (1 - D) \times \cos(4\pi nd \sin(\alpha/2) / \sin(\alpha/2 + \delta) + \varphi) \},
\]

where \(\delta\) is the change in the angle transmitted through the pixels, \(D\) describes the relative strength of the beating spectra, \(\alpha = 24.4^\circ\) is the tilt interferometer designed angle. Parameters \(A\) and \(C\) account for the nonideal reflections from the rough silicon surface. The result of the fit using Eq. (4) is shown in Fig. 7. The following parameters were found to be optimal: \(A = 0.47 \pm 0.01, \; C = 0.28 \pm 0.04, \; D = 0.58 \pm 0.02, \; \text{and} \; \delta = 1.67^\circ \pm 0.04^\circ\). The poor agreement near 15 cm\(^{-1}\) is due to the large error in the transmission spectrum as a result of the small intensity at this frequency, as shown in Fig. 4. It should be noted that neither the angular distribution of the radiation at the silicon sample nor the angular dependence of the scaling factor in the THz HFTS can generate a second scaling factor.

How can one account for an extra angle that appears to be necessary? It is clear from Fig. 4 that multiple reflections occur within the pixel array...
demonstrating that the THz radiation passes through the lithium tantalate pixels. Because of the birefringence of the pixel material and polarization mixing inside the detector structure, a beam from an interferometer arm can split into two beams with a few degrees difference. After reflection from the back metal plate of the array, these beams will fall on the pixels from the backside again and will produce a rescaled interferogram component. This change in angle will result in scaling factors $2 \sin(a/2)$ and $2 \sin(a/2 + \delta)$ for the incident and reflected beam paths, respectively. The fitted angle $\delta = 1.67^\circ$ is compatible with the expected lithium tantalate birefringence effect. This simple model is not a substitute for the rigorous treatment taking into account the near field effects in the real three-dimensional array detector structure. Such a treatment is not possible because of proprietary restrictions on the array construction; however, the model illustrates what happens when the array is not properly designed for holographic spectral measurements at THz frequencies.

To optimize this spectroscopic system at least two changes in the array detector fabrication are recommended. The pixels should be vacuum impedance matched in the THz region to increase the sensitivity and an efficient absorber should be placed behind the pixels to avoid complications that are due to backreflections of the beams. Since the pixel spacing in the array sets the shortest wavelength limit of the HFTS, the pixel size should be reduced so that a broader spectral range can be covered.

Summarizing, the experimental test of the holographic FTS for the THz region demonstrates the following.

1. A new spectrometer design for the THz, the HFTS built around the tilt interferometer, using an array detector and coherent synchrotron radiation has been demonstrated.
2. A qualitative characterization of the electron bunch length with the new instrument has been shown.
3. These experimental tests of the new spectrometer demonstrate its viability as a spectral device and reveal that its performance is strongly dependent on the details of the array detector construction.

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