Far-Infrared and Terahertz Research from 1950 to the present

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Abstract: A sequence of research activities in the spectral range between light waves and microwaves carried out at Osaka University and KARC / NICT is described, starting from the times of far-infrared grating spectroscopy through the times of far-infrared Fourier-transform spectroscopy to the present stimulated by the terahertzpulse technology.Activities on terahertz-wave optoelectronics at KARC / NICT and related groups in Japan are an extension of the studies initiated at Osaka University.

Keywords: FIR, FTS, THz

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1. Introduction

Modern research on the spectral range between light waves and microwaves, that is, between optics and electronics, really expanded since the beginning of 1950s with the development of far-infrared (FIR) grating spectrometer for which a weak incoherent thermal source was used. The grating spectrometer was gradually replaced by the Fourier transform spectrometer (FTS) since mid 1960s. It has some advantages over the grating spectrometer, but a weak incoherent thermal source was still in use. At almost the same time liquid-helium-cooled sensitive detectors were begun to be used. An important breakthrough, development of such gaseous lasers as HCN, H_2O, *etc.* was made around 1964, followed by the enthusiastic years of CO_2 laser-pumped gaseous molecular laser since 1970. As a result, coherent sources became available together with solid-state lasers. A lot of fundamental studies and applications were accomplished during that period [1]. After an additional 20 years, the advent of the solid Ti:Sapphire femtosecond laser changed this field from a special field of physics and chemistry to a field of electronics. This paper describes the progress of this field from the beginning of FIR research to recent activities based on terahertz (THz)-wave optoelectronics.

2. Times of the FIR Grating Spectrometer

In the beginning of 1950s, Prof. Yoshinaga started pioneering FIR studies constructing a mostly handmade spectrometer. They acquired primitive spectroscopic data, and published them in a journal in 1952 [2].

Around the same time, Prof. Oetjen and his staff at Ohio State University published a paper on a completely automatic large FIR vacuum grating spectrometer, which used advanced optical elements such as precisely machine-ruled gratings and the newly developed Golay cell [3]. It is justly said that this grating spectrometer opened the door to a new age of FIR spectroscopy.

Following the work at Ohio State University, Prof. Genzel and Eckhardt at Freiburg University constructed a high- performance FIR grating instrument [4].

Prof. Yoshinaga obtained a large amount of research budget and accepted a proposal of cooperation from Prof. Oetjen－who had looked at the first Yoshinaga's paper [2] and was deeply impressed by his strong resolution to open up new field. Through this cooperative research, they constructed a refined, fully-automatic FIR-grating spectrometer (Fig. 1) in 1958 [5]. In Japan, this FIR spectrometer established the way for a full-dressspectroscopic investigation in the FIR region. The spectral range covered by the grating spectrometer was nominally 17.5m to 1mm, but practically 17.5m to about 300m or 400m. By using this instrument, a lot of original works were achieved: the most famous results are the "Yoshinaga filter" [6] and "Reflection measurements of Restrahlen bands of ionic crystals" [7] cited in Kittel's world-famous book on solid-state physics.

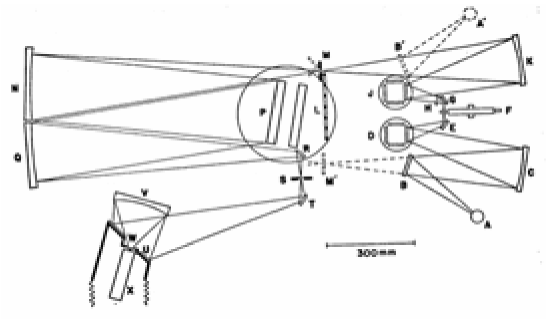

Fig.1 Optical arrangement of a Czerny-Turner-mount FIR-grating spectrometer at OsakaUniversity

3. Times of FIR Fourier-Transform Spectrometer

To extend the spectral coverage to longer wavelength regions, the author constructed Fourier-transform spectrometers, which became increasingly popular in the mid 1960s. He constructed a Michelson-type interferometer first [8], and then constructed a refined lamellar-grating-type spectrometer (Fig.2) [9] combined with an InSb hot-electron bolometer operated at 4.2 K [10].

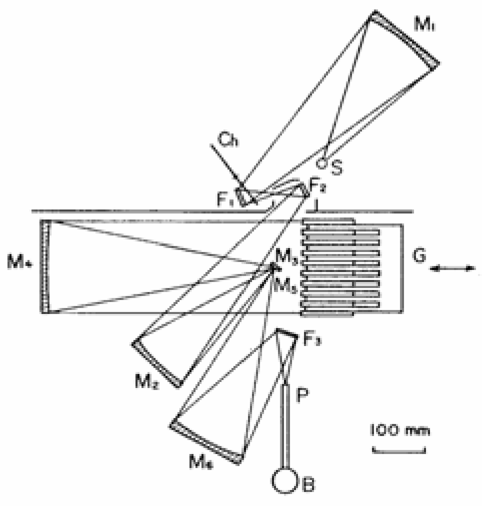

Fig.2 Optical arrangement of a lamellar-grating Fourier-transform spectrometer at Osaka University

This instrument extended the spectral coverage to a few mm inspite of using such a low-intensity thermal source as high- pressure mercury lamp. Some typical experimental results are shown in Figs.3 to 5. Figure 3 shows the pure rotational spectra of some symmetric-top molecules [11]. Figure 4 is the zero-field splitting of Fe^{3+} in a bio-molecule hemin [9]. Figure 5 shows the transmittance of inductive metal meshes, which have been recently used as samples of metallic photonic crystals or meta-materials[12].

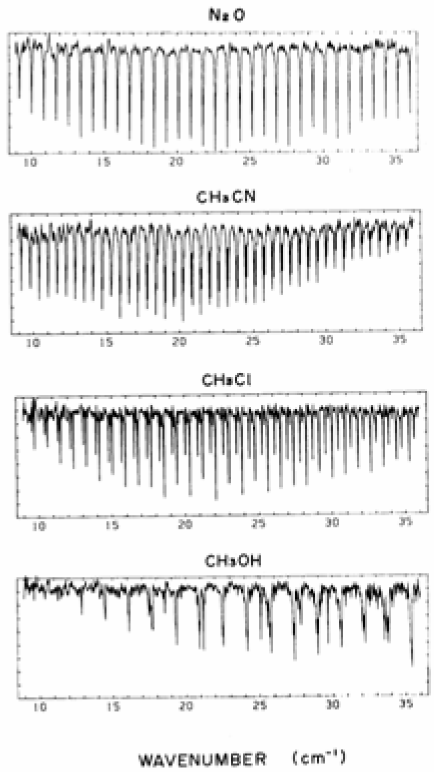

Fig. 3 Pure rotational spectra of some gaseous molecules measured with a resolution of 0.030 cm^{-1}.

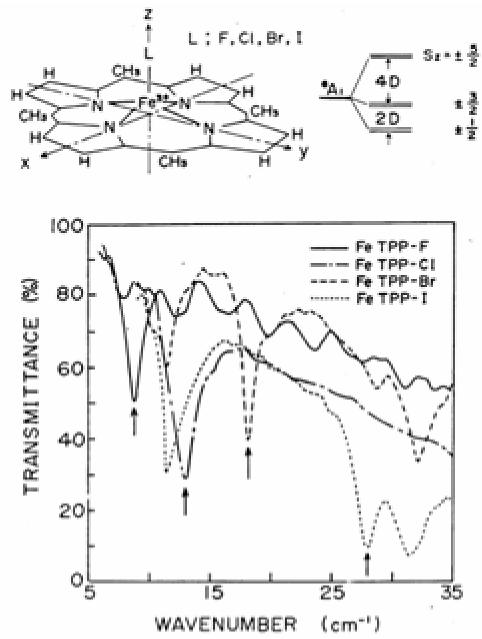

Fig. 4 FIR magnetic resonance of hemin. The ground state splits 2D is observed without applying the magnetic field.

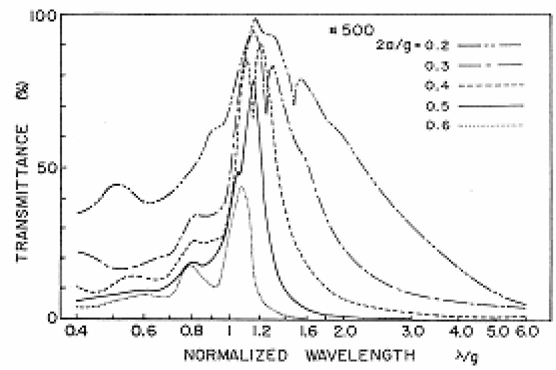

Fig. 5 Transmission curves obtained from inductive metal meshes with different line-width to grid-constant ratios.

A chapter on metal meshes was written by Sakai and Genzel in 1983 [13]. An invited review article on Fourier transform spectroscopy was also written by Genzel and Sakai in 1977 [14].

The author constructed a more sophisticated Michelson / Martin-Puplett type FTS by use of advanced technologies such as air-bearing and laser-interferometer sampling. This spectro-meter covered whole FIR region, *i.e.*, between20 μ m to 3 mm [15].

4. Applications of FTS and grating techniques to other fields

The technique of FTS was transferred to an industry to produce commercial instrument (FTIR), and furthermore the techniques of FTS and grating spectroscopy were transferred to the plasma diagnosis.

The author developed a rapid scanning FTS and a grating polychromator in collaboration with Institute of Plasma Physics in Nagoya University to measure the electron cyclotron emission (ECE) from a toroidal (JIPPT-II) plasma. Measurement of ECE became widely recognized as a powerful tool for diagnosing high temperature plasmas. When the plasma emits and absorbs the radiation as a black body, it is possible to determine the electron temperature profile and its temperature variation simply from the ECE measurements. It is also possible to obtain the total power radiated by ECE, information about non-thermal electrons. Figure 6 shows an optical layout of the rapid-scanning FTS. It is a double-pass Martin-Puplett polarizing inter- ferometer to increase the spectral resolution. It was verified experimentally that this system has 6.6 GHz spectral (4 cm spatial) resolution and 10 msec time resolution [16].

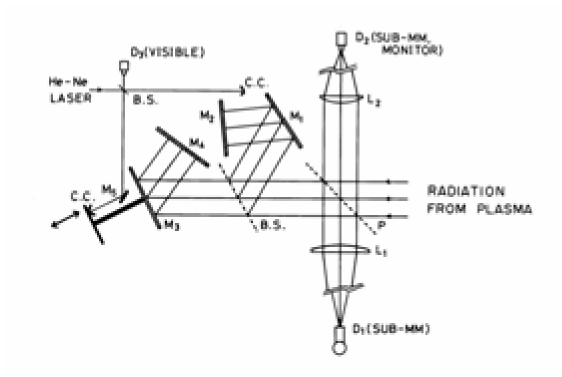

Fig. 6 Optical layout of double-pass rapid scanning Fourier transform spectrometer

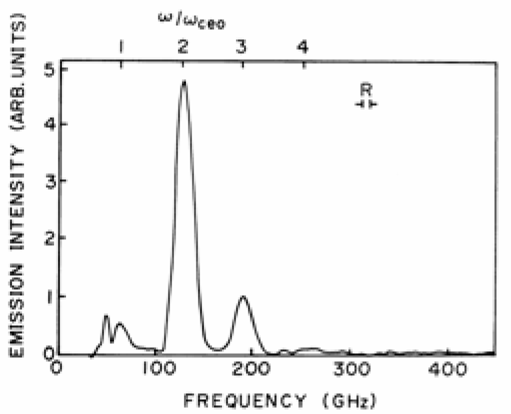

Fig. 7 Typical electron cyclotron emission spectra

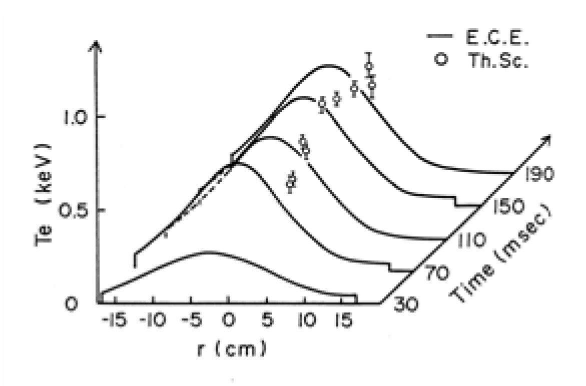

Fig. 8 Time behavior of radial electron temperature profile determined from 2nd harmonic electron cyclotron emission

Figure 7 shows a typical emission spectra at an electron density. Spectral peaks arising from ECE are seen around nf_{ceo} (n=1, 2, 3), where f_{ceo} is the electron cyclotron frequency at the center of the plasma. When the electron density is larger than ～2 x 10^{13} cm^{-3}, the 2nd harmonics is optically thick for the extra- ordinary mode and the plasma radiates and absorbs as a black body. Thus the electron temperature profile is obtained from the 2nd harmonic ECE measurements. Figure 8 is the time behavior of radial electron temperature profile measured with calibrated FTS, and Thomson scattering electron temperature [17].

Limiting the spectral coverage only to the 2nd harmonic ECE range, a grating polychromator is more advantageous over the FTS in respect to the spectral (spatial) resolution, time resolution and ease of operation due to the mechanically non-moving part during the operation. Figure 9 illustrates a 10 channel grating polychromator, with 3 GHz spectral (1.8 cm spatial) resolution having 10 InSb hot electron bolometers operated at 4.2 K. Figure 10 shows the time evolution of the electron temperature profile determined by the 2nd harmonic ECE. Since the time resolution is so high (<2s) that fluctuations of electron temperatures at 10 spatial points can be observed [18].

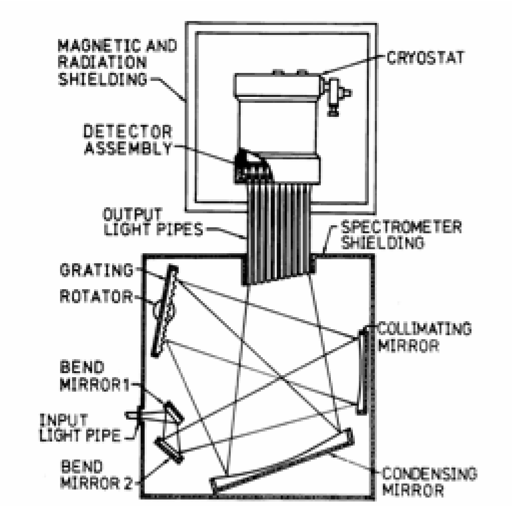

Fig. 9 Optical layout of a 10 channel grating polychromator for ECE measurement

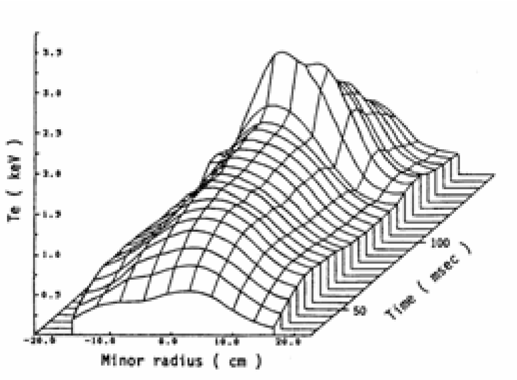

Fig.10 Time evolution of the electron temperature profile determined by the 2nd harmonic ECE

5. Times of Terahertz Pulse Techniques

5.1 Introduction

A remarkable progress of terahertz (THz) pulse technique has been brought by the development of the ultrashort optical pulse lasers and the development of semiconductor growth and microfabrication techniques. A key point of the THz pulse technique is an ultrafast optical switch called "Auston switch". The author was fortunate to meet Prof. Auston at a symposium held in Kobe when he was going to start this kind of research at Kansai (*present* Kobe) Advanced Research Center (KARC) of the Communications Research Laboratory (CRL: *present* National Institute of Information and Communications Technology NICT) in early 1990s. This field of research has expanded to spectroscopy and imaging.

Historical background and variety of activities made until recently mainly by author's group are summarized in a book [19].

5.2 Spectroscopy

When the THz pulse technique is used in spectroscopy, it is called THz time-domain spectroscopy (THz-TDS). Figure 11 shows a typical optical layout of the THz-TDS and THz imaging. The system uses photoconductive (PC) antennas (based on Auston switch) for the emitter and the detector. It gives rise to many advantages over the other spectroscopic techniques.

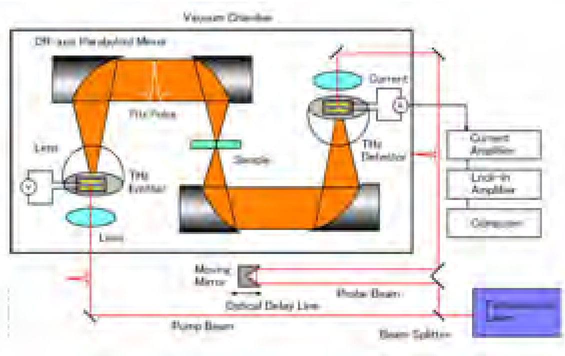

Fig. 11 Optical layout of a THz-TDS and THz imaging

Two examples characteristic of THz–TDS are explained in the following. In Fig.12 the observed LO phonon-plasmon coupling modes in an InSb film are shown [20]. The upper spectrum shows time-domain waveforms of the THz radiation detected in (a) reflection and (b) transmission geometry. The lower one shows the Fourier-transformed spectra of the data. The coupling modes (L_- and L_+) are shown clearly. Figure 13 is another example, showing the amplitude and phase-shift spectra of a pseudo-simple-cubic photonic crystal fabricated in Si [21]. The transmission spectra and the dispersion relation are in good agreement with the calculations based on transfer matrix. These results indicate that THz-TDS is a useful tool for studying such novel materials as photonic crystal.

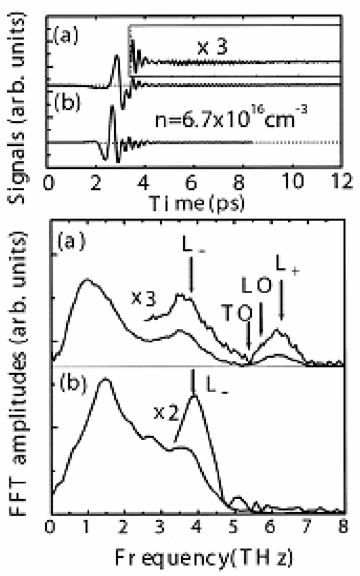

Fig. 12 Observation of LO phonon-plasmon coupling modes in an InSb film: upper spectrum shows time domain, lower shows Fourier-transformed spectra.

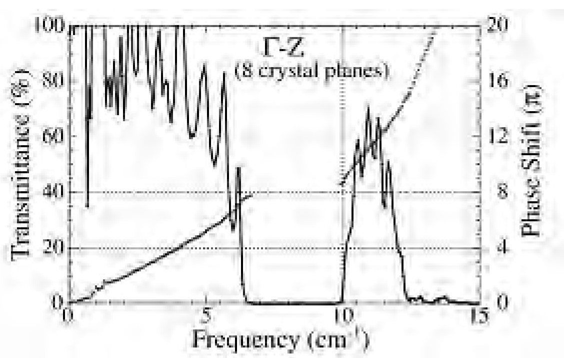

Fig. 13 Transmittance and phase-shift spectra of a pseudo-simple-cubic photonic crystal.

5.3 Imaging

The THz-TDS provides another attraction ,*i.e.*, THz imaging. Such attraction comes from that the THz-TDS is operated in a pulse mode. It is generally said that the THz imaging suits soft materials rather than such hard materials as the metal *etc*. imaged by X-ray. First example is a leaf concealed in an opaque teflon sheet, yielding a sub-mm spatial resolution (Fig. 14). The contrast is due to its water content.

The second is the images of silicon samples. Since the plasma frequency of silicon and other semiconductors is in the THz frequency range, THz waves are specially suited for probing these properties. The substrate consists of n-type silicon with a resistivity of 5 cm. The resulting images are shown in Fig. 15 [22]. The upper half of each silicon sample was boron-ion-implanted with the implantation dosages of 5×10^{13}, 5×10^{14}, 5×10^{15} cm^{-2} resulting in 0.5μm thin strongly p-doped layers that in spite of their thinness can effectively block THz waves depending on carrier concentration and mobility.

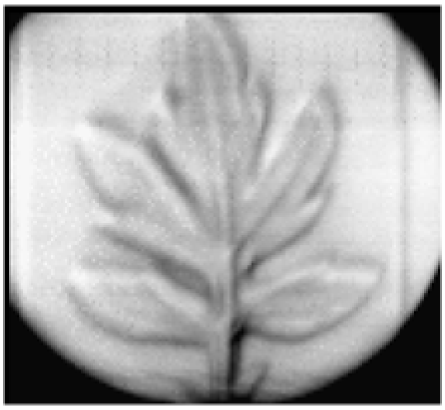

Fig. 14 THz imaging of a leaf concealed in a opaque Teflon sheet

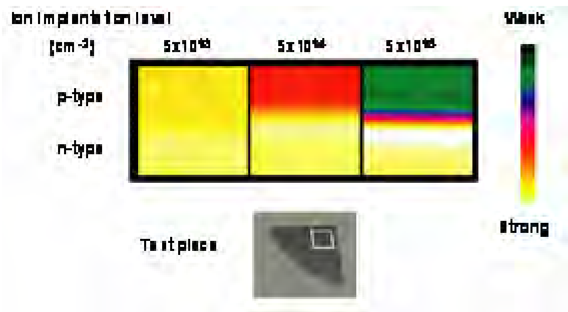

Fig. 15 THz images of n-type Si samples with doped thin p-type layers in respective upper half

Furthermore THz waves have both optical and radio-wave natures. They are more transparent than optical waves and less diffractive than radiowaves. We applied THz imaging to objects hidden in powders. Glass, eggshell and various plastics have been imaged in powders such as wheat flour, talc and sugar. Special interest was directed to imaging plastic objects in powders. Figure 16 shows images of three objects of two kinds of plastics, polyoxy-methylene (POM) and acrylnitrile- butadiene-styrole copolymer (ABS) in wheat powder. Two POM specimens with different thicknesses are included. With this kind of imaging, we have introduced a material parameter, $Q=k/n$, that exclude sample thickness in order to distinguish materials [23].

The techniques of THz-TDS and THz imaging were transferred to an industry ; we have collaborated with Tochigi Nikon Corporation. Instruments and some results will be presented at the conference.

5.4 Improvement of the spectral coverage

In general, either the photoconductive (PC) antenna or the nonlinear (NL) crystal (or electrooptic (EO) crystal) is most frequently used for THz pulse emission and detection until about a decade ago. The spectral coverage of THz-TDS was limited to less than several THz. An effort was made to extend the spectral coverage to much higher frequency (ω) region. THz pulse emission due to optical rectification with the NL crystal and THz pulse detection with the EO crystal have preceded in this respect [24-26] and soon the PC emission and detection have come to the same level [27,28]. According to author's estimation, the NL emitter has an advantage over the PC emitter in terms of bandwidth by a factor of ~. Accordingly NL crystals such as ZnTe or GaSe have been used effectively. On the other hand, it was estimated by a simple calculation and was verified experimentally that both the EO crystal and PC antenna have the same bandwidth extending over 100 THz (Fig. 17).

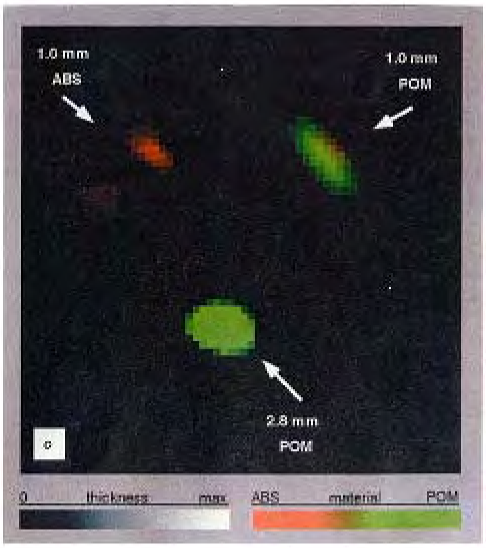

Fig. 16 Three objects in wheat powder POM: polyoxy-methylene ABS: acrylnitrile-butadiene- styrole copolymer

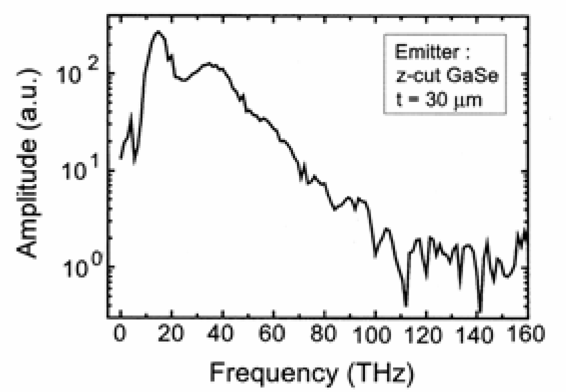

Fig. 17 Ultra-broadband detection of a THz pulse with a PC antenna

6. Summary

The trend of the FIR and THz researches have been reviewed since the beginning of 1950s. First, pioneering works in FIR spectroscopy made at Osaka University were introduced ; namely, the construction of a FIR grating spectrometer and some original results with this spectrometer were described. In the second, an extension of spectral coverage to a few millimeters by means of the FTS was explained with such a typical experimental result as FIR magnetic resonance of the bio-molecule hemin. In succession, applications of the FTS and the grating techniques to the plasma diagnosis were described. It was shown that such techniques were effective for the measurement of time behavior of radial electron temperature profile.

As an extension of the FIR studies, the author have made researches on THz pulse technology, in particular, the THz-TDS and THz imaging have been investigated. A novel applications of THz –TDS to a photonic crystal and THz imaging to plastics hidden in powders, and an innovative work, *i.e.*, ultra-broadband detection by means of a PC antenna were shown. It is recommended that readers refer to the book "Terahertz Optoelectronics" for precise explanation of the techniques described in this paper and for more informations from the author's research group and other group in Japan [19].

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