Microspectroscopy and Imaging in the THz Range Using Coherent CW Radiation

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A novel THz near-field spectrometer is presented which allows to perform biological and medical studies with high spectral resolution combined with a spatial resolution down to \( \lambda/100 \). In the setup an aperture much smaller than the used wavelength is placed in the beam very close to the sample. The sample is probed by the evanescent wave behind the aperture. The distance is measured extremely accurate by a confocal microscope. We use monochromatic sources which provide powerful coherent cw radiation tuneable from 50 GHz up to 1.5 THz. Transmission and reflection experiments can be performed which enable us to study solids and molecules in aqueous solution. Examples for spectroscopic investigations on biological tissues are presented.

I. INTRODUCTION

During recent years there have been many efforts to understand in detail how cells function. To cope with this complex problem, it is necessary to examine the components of the system, like DNA, bio-molecules, proteins, cell-membrans. Due to the large entities and the high density of electronic, vibrational and rotational states in these bio-molecules, spectroscopic investigations in the millimeter, submillimeter, and far-infrared spectral range (often called THz range) seem to be an appropriate method for this task. However, the dimensions of the samples under study are rather small, often well below Abbe’s diffraction limit, implying that in the millimeter to far-infrared spectral range it is not possible to perform measurements by standard optical methods. In addition to the scientific point of view, the THz range of frequency attracts considerable attention because of possible applications, in particular with the aim of THz imaging. While a number of groups try to peruse time domain technique utilizing short laser pulses in order to generate a broad spectrum terahertz radiation \[ \omega \rightarrow 0 \] following Planck’s law of black body radiation; (ii) the spot size to which the light can be focused for measurements is diffraction limited to approximately the wavelength. Here we present a new approach to overcome this limitations by utilizing monochromatic but tunable backward wave oscillators as powerful radiation sources and a near-field microscope in order to perform microspectrometry in the THz range.

II. COHERENT SOURCE SPECTROMETER

The frequency range between microwaves and infrared has proven to be particularly challenging to experimentalists since it falls right between the range of guided waves (coaxial and microwave technique) and optical techniques (free space propagation). Microwave oscillators are in general not tunable over a larger range of frequency and/or deliver only a very small output power; hence they basically cannot be employed above 100 GHz. Coming from optical methods, standard infrared spectroscopy using Fourier transform interferometer has to cope with two problems by trying to go to lower frequencies: (i) the output power of the thermal radiation sources vanishes for \( \omega \rightarrow 0 \) following Planck’s law of black body radiation; (ii) the spot size to which the light can be focused for measurements is diffraction limited to approximately the wavelength. Here we present a new approach to overcome this limitations by utilizing monochromatic but tunable backward wave oscillators as powerful radiation sources and a near-field microscope in order to perform microspectrometry in the THz range.

FIG. 1. Mach-Zehnder type interferometer used for quasi-optical transmission measurements in the THz range of frequency. The coherent radiation generated by backward wave oscillators is split by wire grids. The length of the interference arm can be adjusted by moving mirror 2. After the sample is introduced, the interferometer is readjusted in order to obtain the phase difference.

The use of backward wave oscillators as radiation sources for spectroscopic measurement in the frequency range from 1 cm\(^{-1}\) to 50 cm\(^{-1}\) (\( f = 30 \text{ GHz to 1.5 THz}; \lambda = 0.2 \text{ mm to 10 mm} \)) was developed in the group of G. Kozlov during the last twenty years \[ \cite{3,4} \]. With a
cw output power of 1 to 200 mW reliable optical experiments can be performed with a very short data acquisition time. The radiation can be tuned in frequency with an accuracy and stability of $10^{-6}$ allowing to map very narrow absorption lines. The radiation is detected either by a Golay cell or by a silicon bolometer, which is cooled down to 1.2 K. The spectrometer has a dynamical range of up to $10^7$. Experiments can be performed either in reflection or transmission, where a Mach-Zehnder interferometer (Fig. 1) also allows to probe the change in phase and thus to evaluate the real and imaginary part of the electrodynamic response without performing a Kramers-Kronig analysis. Since a quasi-optical arrangement is limited by diffraction and standing-wave problems, samples smaller than approximately three times the wavelength cannot be investigated.

III. NEAR-FIELD SETUP

We have developed a THz near-field microscope which allows us to perform reflection measurement on spot sizes down to $\lambda/100$. As displayed in Fig. 2, the setup consists of a near-field unit mounted rotatable by 90 degrees (1); one axis is used for a quasi-optical spectrometer (2), and the second by a confocal microscope (3) for distance control.

![Diagram](image.png)

FIG. 2. Schematic of the THz near-field spectrometer. The sample is placed in the near-field unit (1); the spectroscopic measurements are done by a coherent source spectrometer (2); the distance control and measurement is done by a confocal microscope (3).

The spectroscopic investigations are done with a coherent source spectrometer similar to the one discribed above in Sec. I just arranged for reflection measurements. The cw beam generated by backward wave oscillators is mechanically chopped and focused on the pinhole by teflon leses. Within a few seconds it can be electronically tuned over the full range of the oscillator. The reflected beam is guided to the bolometer by a wire-grid beam splitter.

FIG. 3. Schematic of the near-field unit, consisting of pinhole (1a), piezo bender (1b), piezo motors (1c), sample (1d), and sample holder (1e).

The core part of the near-field spectrometer is a small pinhole (1) which is drilled in a copper foil (thickness 1 to 10 $\mu$m) with diameter as small as 1 $\mu$m and which generates the evanescent submillimeter wave the sample is probed with (Fig. 3). Behind the pinhole sits the sample (1d) which can be translated relative to the pinhole by three linear piezo-motors (1c). These motors are based on stick and slip motion with a step size of 40 to 400 nm. There is a fourth motor for moving the sample and pinhole together, which is required for the distance control via the confocal microscope. The distance between pinhole and sample is modulated up to $\pm 70$ $\mu$m with high frequencies (about 1 kHz) by an additional piezo-electric bender (1b) in order to using look-in technique for data acquisition. The near-field unit is installed on a rotatable frame for performing THz measurements on the one side and the distance control on the other side.

An accurate distance control is necessary because of the strong dependence of the signal on the distance between pinhole and sample. It is performed by a image-processing confocal microscope [Fig. 3 (3)], which was developed by Tiziani et al. Therefore the light of an infrared LED (3a) is focused on the sample via a rotating microlens array (3d). The reflected light is measured by a CCD camera (3h). Because of the low depth of focus, which is produced by the microlens array, the reflection has a maximum if the sample sits exactly in the focus of the last lens (3f). By moving pinhole and sample relatively to this lens a three dimensional quantitative image originates, from which the distance between pinhole and sample can be measured with a precision of 100 nm within an adequate time.
Fig. 4 demonstrates the resolution of the near-field spectrometer. The pinhole was scanned over an interface between a microwave absorbing film and a silver film, both deposited on a glass substrate. The increase of reflectivity within a width of 200 \( \mu \text{m} \), which corresponds to the diameter of the pinhole, can clearly be seen. With an used frequency of 180 GHz (\( \lambda = 1.7 \text{ mm} \)) the near-field ratio \( \lambda/d \) is about 10. Additional mathematical treatment (deconvolution) can be used to improve the spatial resolution considerably. Details of the experimental setup will be discussed in [7].

**IV. SPECTROSCOPIC MEASUREMENTS**

Due to the strong dependence of the oscillators’ output power on the frequency and due to the frequency-dependent absorption of the components of the optical spectrometer (lenses, grids, ...), measurements of absolute value of the reflectivity require to perform a reference measurement on an ideal reflecting sample (e.g. a silver mirror). If only spectroscopic features have to be identified, difference spectra are sufficient which were taken of two materials, at two different sample positions, or two states different in other respect.

Employing this near-field reflection spectrometer, we have performed investigations on a variety of biological and medical samples. As an example Fig. 5a exhibits the reflectivity of the meat part of Black Forest Ham relative to the fat part in the frequency range from 5.5 cm\(^{-1}\) to 8.5 cm\(^{-1}\). The circular pinhole used was 200 \( \mu \text{m} \) in diameter in a 10 \( \mu \text{m} \) thick copper foil. The distance between pinhole and sample was 3.0 \( \mu \text{m} \). To rule out inhomogeneities of the sample we performed three measurements on different the meat part as well as on the fat part and averaged them. The reproducibility of the main features and the homogeneity of the sample are demonstrated by dividing data sets taken at different positions, ideally leading to a 100% line (dotted line). The spectroscopic distinction between meat and fat become obvious when the ratio of the spectra is plotted (solid line).

Similar experiments have been performed on chicken bone which has a much lower water content and thus a smaller reflection coefficient. Fig. 5b shows the ratio of the the reflected power of the bone and of the cartilage. Again the spatial resolution is less than 200 \( \mu \text{m} \).
V. SPECTROSCOPIC IMAGING

In both spectra we find unexpectedly sharp and strong features; in certain ranges the reflected power is 5 times as high in cartilage compared to the chicken bone, for instance. This indicates strong absorption lines which may serve as a spectroscopic signature of the material. While at presence we cannot reliably assign the lines, we already want to note the possibility of using these large differences for imaging. Differential images may be a very sensitive tool to probe the spatial distribution of certain molecules with a resolution in the micrometer range. For this purpose only two distinct frequencies have to be measured which is easily possible by our spectrometer and which reduces the measurements time by orders of magnitude. This technique allows to selectively map out the composition of samples.

VI. CONCLUSION

We have developed a new microspectrometer to perform spectroscopic experiments in the THz range with a large frequency resolution of $10^7$. The use of near-field technique enables us to obtain a spatial resolution down to several micrometers. With a reflection setup we can investigate biological samples in aqueous solution. The instrument also provides spectro-images which show the spatial distribution of biological molecules with a high spatial resolution. This novel technique has an enormous potential for a variety of biological and medical applications.

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