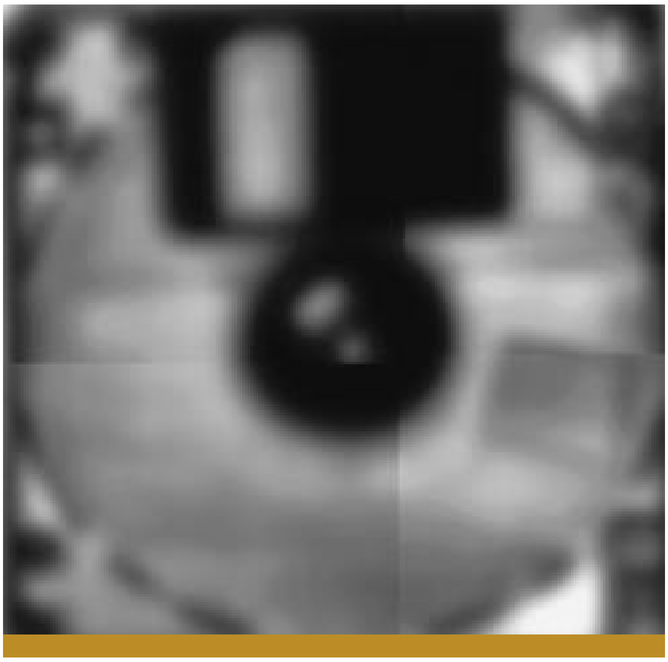
THz radiation: The no-man’s-land between optics and electronics



After James Clerk Maxwell had developed his famous equations in 1864, which predicted the existence of propagating electromagnetic waves, scientists started to explore the areas beyond the small spectral region humans can detect with their bare eyes. Heinrich Hertz was the first who demonstrated that radio-waves and light are the same kind of radiation only at different frequencies. And Heinrich Rubens wrote in a famous article about ‘Heat rays of great wave length’ in 1897: “since we become accustomed to think of electrical energy and light waves as forming component parts of a common spectrum, the attempt has often been made to extend our knowledge over the wide region that separates the two phenomena, and bring them closer together”. More than hundred years later, heat rays are called infrared radiation and electrical energy can be produced up to frequencies of several hundred GHz employing semiconductor devices; but still there is a small nearly untouched region between optics and electronics, the so called THz gap, sketched in **(01)**.

SUMMARY

Between the highest frequencies which can be generated by electronic devices and the lowest which can be handled with optics, there is a nearly untouched region, the so called THz-gap (300 GHz to 10 THz). Despite this problems of generating and detecting radiation at these frequencies, the THz range is rich in interesting scientific questions and potential applications for example in material science and medicine. THz-rays can penetrate matter similar to x-rays, but they are non-ionizing and therefore assumed to be harmless and they give specific spectroscopic information not accessible with other optical techniques in the visible or infrared, with x-rays or nuclear magnetic resonance. But broader applications and the development of new techniques in this interesting frequency range are hindered by the lack of compact, cheap and easy to handle THz-sources. Additionally for most imaging applications a good resolution is required. Unfortunately THz-radiation has a very long wavelength (1mm – 30 µm). Therefore the maximum resolution in the far-field is strongly limited by diffraction. Here we want to give on the one hand a short overview over the interesting scientific questions which can be investigated in the THz-range, on the other hand we want to show same application from material science and medicine.

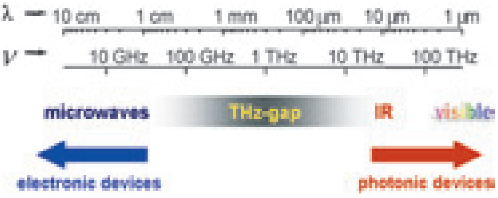
1. Introduction

In our days the limits of this THz range are generally taken to be 300 GHz – 10 THz or in wavelength between 1mm – 30 µm. On the low energy side (microwaves) of this region, cables and hollow metallic waveguides are utilized to transport the energy, but it is hard to built circuits capable to handle higher frequencies. On the high energy side (infrared) one has normal optics with lenses and mirrors to guide the freely traveling waves, but new optical components and sources have to be developed for larger wavelength. Experiments in the THz-region are still challenging largely due to a lack of compact and easy to handle sources and detectors.

Despite these problems, the THz range is rich in interesting scientific questions and potential applications. THz-rays can penetrate matter similar to x-rays, but they are non-ionizing and therefore assumed to be harmless and they give specific spectroscopic information not accessible with other optical techniques in the visible or infrared, with x-rays or nuclear magnetic resonance. For example, THz-rays penetrate many visually opaque materials such as paper, plastics and clothes but are strongly absorbed by water and metals. Many specific excitations lie in the THz-range, like molecular vibrations and librations, or low-energy excitations in solids. The role of protein structure on molecular recognition had been investigated by X-rays and NMR spectroscopy since long. However, only recently the role of protein dynamics had been recognized. Due to the large 3-dimensional structure of proteins this dynamics lies mainly in the THz-range.

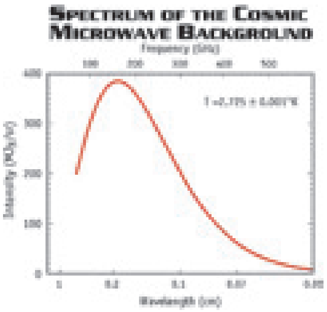
Even the echo of the Big Bang, the cosmic background radiation, peaks in the THz-region as displayed in **(02)**. Since the photon energy of a few meV is comparable to a few Kelvin in temperature and a few Tesla in magnetic field, the THz spectral range is ideally suited to study superconductivity and similar collective phenomena. Fundamental concepts of solid state physics can be tested, new states of matter can be investigated, and the scaling laws close to quantum phase transitions. As a consequence of the confinement also in Nanoscience many excitations lie in the THz-range.

Based on decades of experience and strong efforts, the 1. Physikalisches Institut at the Universität Stuttgart became a center of THz research on solids well recognized throughout the world. Using Fourier transform spectrometers with Helium cooled bolometers as detectors in the far infrared and coherent source THz spectrometers based on backward wave oscillators a huge variety of new materials can be investigated. Also novel approaches are surveyed to create tunable continuous-wave THz radiation, for instance by mixing two solid state lasers with the help of non-linear devices in order to obtain the difference frequency which is chosen to lie in the THz range. These compact and easy-to-use THz sources might become the heart of future spectrometers to discover the *terra incognita*. But also small parts, like lenses, beam splitters, polarizers, attenuators, etc. have to be developed and optimized. To this end, material properties have to be investigated, design studies to be made, numerous tests to be conducted. Photonic crystals and even materials with negative refractive index are among the possible novelties which are discussed for this particular spectral range. Only slowly a network of competent laboratories is



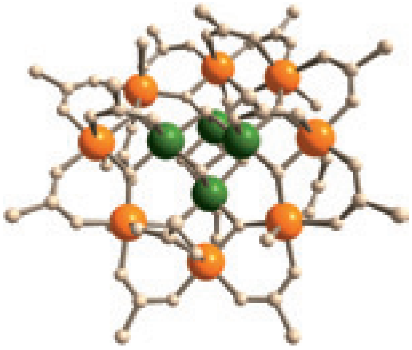
01

The electromagnetic spectrum still includes a gap between microwaves and infrared region which contains a wealth of interesting phenomena and potential applications, but which is explored only to a very small extend.

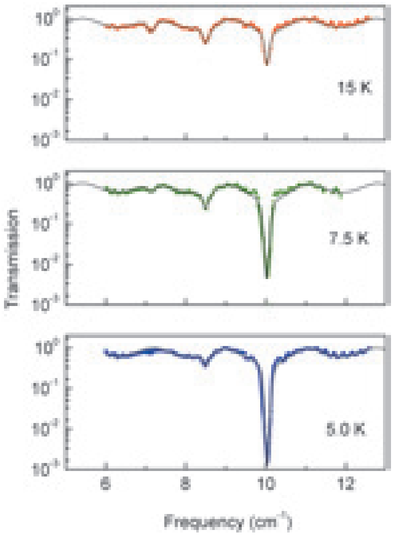


02

Spectral intensity of the cosmic background radiation. Tiny deviations from Planck's radiation law yield information on the origin of the universe.



03a



03b

03c

a *Nanomagnets are the smallest possible magnetic structures. $Mn_{12}ac$ is considered the prime example of this class of novel materials.*

b *Due to the coupling of the magnetic atoms in the molecule characteristic low energy excitations in the THz-range can be observed.*

c *The photograph shows the THz-spectrometer at the 1. Physikalisches Institut at the Universität Stuttgart.*

formed to enable the exchange of experience and know-how between the different scientific communities which participate in this unique endeavor.

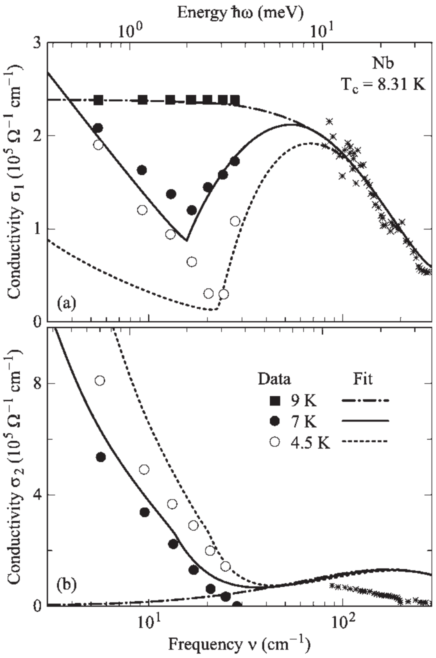
2. Basic research

Nanosized materials can be achieved by either structuring large samples for example by lithography (top-down approach) or by assembling small nanometer-sized building blocks, as single atoms or molecules, to larger units (bottom-up approach).

A new bottom-up approach in the design of magnetic materials for high-density data storage and quantum computation goes in the direction of single molecule magnets. They are exchange coupled clusters of four to about thirty paramagnetic ions, usually from the first period of the transition metals, that have large spin ground states, for example S = 10. They do not interact with each other. Some of these molecular magnets can be magnetized at low temperatures, and will remain magnetized even after removal of the external field. For this reason these systems have been dubbed “single molecule magnets”. Applications in data storage devices have been proposed. The maximum theoretical data density would be up to 10 000 times what is possible today, since every molecule can be considered as a bit of data. Beyond that even applications in spintronics and quantum computers could be possible. Apart from these promising perspectives, their physical properties are also hugely interesting, due to their mesoscopic size of the systems. Molecular magnets show many distinct quantum properties like quantum tunneling of the magnetization. In order to optimize these macromolecules and tailor the physical properties, a close collaboration between chemists, material scientists and physicists is required. To investigate the material parameter, we have developed a novel spectroscopic technique in the THz range of frequency, which can be described as Frequency Domain Magnetic Resonance Spectroscopy. As an example the spectrum of a nanomagnet recorded at our institute is shown in **(03)**. We nicely observe the transitions between the magnetic sublevels split by the crystal field.

Superconductivity is one of the most fascinating phenomena in solid state physics which did not lose any of its challenge and attraction to theory and experiment. Although we have a rough understanding how superconductivity works in most conventional metals, high-temperature superconductivity still remains a mystery. The superconducting electrons form pairs due some attractive interaction, usually lattice vibrations, but also magnetic interaction is discussed as a possibility. For typical superconductors like niobium it takes the energy of a few milli-electron-volts (1 meV corresponds to 8 cm^{-1} which corresponds to 240 GHz) to break up this pair and this can best be investigated by THz spectroscopy. In **(04)** the electrodynamic response is displayed in the real and imaginary parts of the conductivity. Clearly seen is the development of the gap-like feature around 20 cm^{-1} as the temperature drops below $T_c = 8.31$ K.

Due to the severe experimental constrains and non-availability of commercial instruments, only very few laboratories worldwide are capable of performing these com-



04

Frequency dependence of the real and imaginary parts of the conductivity in niobium at various temperatures. The transmission through a 150 Å thick film on sapphire was measured by a Mach-Zehnder interferometer; the stars were obtained by reflection measurements. The solid lines correspond to the theoretical prediction. The inset shows basically shows the fraction of condensed charge carriers.

plicated experiments by now, Stuttgart certainly is among the leading ones.

3. Potential applications

Beside fundamental research there are also a large number of potential applications in the THz range ranging from so different fields as non-destructive testing and quality control, over homeland security to new medical imaging techniques and rapid screening for the development of new drugs. None of them is thoroughly explored nor well established, let alone commercially available. However, even if only a fraction of them materialize, the financial implications would be tremendous. All this gives strong motivation to develop new systems for generating, detecting and manipulating radiation at THz frequencies.

During the last five years worldwide activities in THz research can be observed and, in particular in the United States, the funding has multiplied and numerous laboratories and companies mushroomed.

Most real samples, biological as well as artificial, are spatial inhomogeneous. For their characterization imaging techniques and microspectroscopy are very important. Unfortunately the extremely long wavelengths of up to a millimeter in the THz range cause severe problems in the optical design due to diffraction and unintended interference (standing waves). In principle near-field techniques can overcome this problem. But sub-wavelength pinholes with radius r reduce the intensity proportional to r^6 and they show exponential signal decay with the probe-sample distance. Therefore it is often hard to find a good compromise between intensity and resolution and the probe-sample distance had to be controlled accurately. This is in principle also true for apertureless methods using small tips as near-field probes. With these methods superior resolution can be achieved, but they image only the sample surface, which especially in the field of life science is often not the relevant information. Another complication is, that in the THz-range biological samples show only little differences in their overall optical properties. On one side this requires a large dynamic range (high S/N ratio) on the other side the interpretation of the observed image contrast calls for additional spectroscopic information.

In the visible the development of solid-immersion lens (SIL) techniques has dramatically increased the capacity of optical memories. Due to the ionic polarisation the refractive index of all mate-

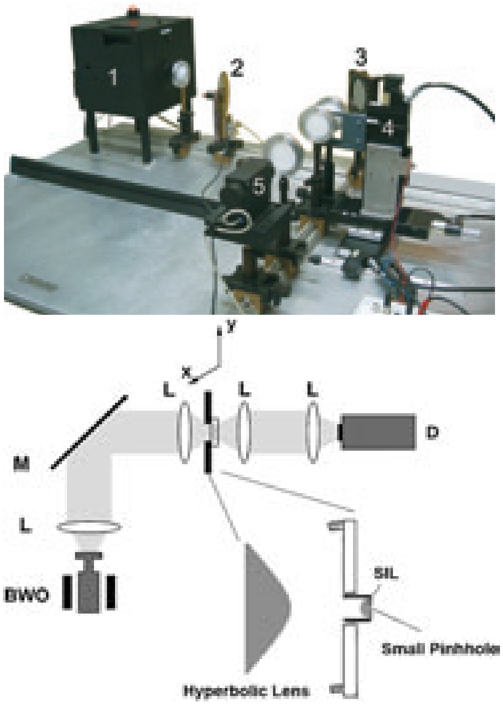
05

A silicon hemisphere serves as a solid immersion lens for the THz spectral range in order to increase the spatial resolution of imaging applications by $n = 3.5$.

06

Custom-designed and special-made hyperbolic lens of polyethylene suitable for the THz range of frequency.

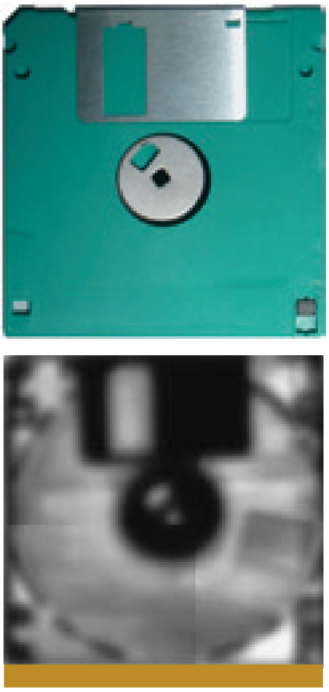
07



Simple setup utilized to obtain THz images in transmission geometry: (1) Backward wave oscillators in a permanent magnet serve as powerful, tunable and coherent radiation sources; (2) chopper connected to lock-in amplifier enables the operating even at low signal-to-noise ratio; (3) mirror or beamsplitter to guide, polarize and attenuate the radiation; (4) computer-controlled xyz-translation unit; (5) Golay cell radiation detector. The optical layout is sketched in the lower part. SIL: Solid immersion lens.

ZUSAMMENFASSUNG

Zwischen den höchsten Frequenzen, die elektronisch erzeugt werden können und den tiefsten, die optisch nutzbar sind, befindet sich ein bis jetzt wenig erforschter Bereich des elektromagnetischen Spektrums, der THz-Bereich (etwa 300 GHz bis 10 THz). Obwohl technologisch schwer zugänglich, ist dieser Frequenzbereich doch reich an interessanten wissenschaftlichen Fragestellungen und es wird ein großes Anwendungspotenzial in der Medizin, Pharmazie und in den Materialwissenschaften gesehen. THz-Strahlen durchdringen Materie ähnlich wie Röntgenstrahlen. Sie sind aber nicht-ionisierend und liefern spezifische spektroskopische Informationen, die mit anderen Techniken im Sichtbaren, mit Röntgenstrahlen oder NMR nicht zugänglich sind. Einer breiteren Anwendung und der Entwicklung neuer Messverfahren in diesem interessanten Frequenzbereich steht aber ein Mangel an kompakten, billigen und leicht zu handhabenden Quellen entgegen. Außerdem benötigt man für alle Abbildungsverfahren eine gute Auflösung. Die Wellenlänge von THz-Strahlung ist aber sehr lang (1mm – 30 µm). Deshalb ist die maximal erzielbare Auflösung im Fernfeld stark beugungsbegrenzt. Der Artikel soll auf der einen Seite einen kurzen Überblick geben über die wissenschaftlichen Fragestellungen, die mit THz-Strahlung bearbeitet werden können, und auf der anderen Seite einen Einblick in Anwendungsmöglichkeiten in der Materialprüfung und der Medizin geben.

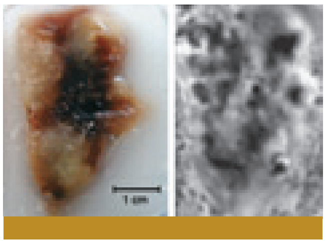


08

Floppy disk non-destructively tested by THz radiation. The THz image was taken at $\lambda = 1$ mm corresponding to 290 GHz. The plastic parts are more or less transparent for THz-rays, whereas metal is opaque.

09

Electronic security tag with fine antenna structure. Only the THz radiation can penetrate the plastic housing and reveal the interior. Solid immersion lenses allow a resolution well below the used wavelength.



10

Photograph and THz image of cancer cells: oesophagus (gullet) carcinoma of a horse embedded in paraffin. Taking THz spectra of the bright spots reveals distinct differences between healthy and cancerous tissue which go beyond the water content.

rials is much lower in the visible than in the THz-range, where values of up to 10 are possible. Recently we have shown that THz-radiation can be focused beyond the Abbe diffraction limit with a hemispherical SIL **(05)**. A photograph of the THz micro-spectrometer using a solid immersion lens for resolution enhancement is reproduced in **(07)**; also shown is a sketch of the optical layout. As THz-sources we use backward-wave oscillators (BWO), which supply highly monochromatic ($\Delta\nu/\nu \approx 10^{-6}$) and coherent radiation with an output power of up to 300 mW. The emitted radiation is collimated by a plan/convex polyethylene (PE) lens to a parallel beam with a diameter of about 23mm and afterwards focused to a diffraction limited spot on the microscope stage by a hyperbolic lens. The advantage of plano/hyperbolic PE lenses **(06)** is, that they produce, comparable to mirrors, aberration-free foci but allow much higher numerical apertures. In this frequency range they show no dispersion and can be easily produced by a computer-controlled lathe with high optical quality (roughness better than $\lambda/10$). The microscope stage (enlarged drawing in **(07)**) consists of a hemispherical silicon SIL ($n = 3.5$) with 8mm diameter clued on a thin (80µm) copper foil with a pinhole in the diameter of the reduced wavelength (λ/n). The sample is scanned in close contact to the pinhole and the transmitted intensity is collected by a confocal arrangement and than focused on the detector. SIL imaging is not really a near-field technique, which means, that beside the possibility to improve the resolution without cost of intensity, the probe-sample distance can be much larger than in normal near-field setups without a significant decrease of resolution.

As an example of non-destructive testing in **(08)** a Floppy disk is imaged by THz radiation. The image was taken at $\lambda = 1$mm (290 GHz) with low resolution in the far field. The plastic parts are more or less transparent for THz-rays, whereas metal is opaque. In **(09)** as an example of a high resolution image recorded with a SIL a security tag encapsulated in a plastic housing is shown; the THz transmission image clearly shows the fine antenna structure with subwavelength resolution.

(10) represents a photograph and a THz-image recorded at 0.48 THz cm^{-1} of oesophagus cancer from a horse. To stabilize the tissue it was embedded in wax. Whereas in the photograph the metastases are bright they appear as dark regions in the THz-image. Due to the fact that THz-radiation can penetrate tissue up to several millimetres, the THz-image shows much more details of the metastases than the photo. Details form inside the sample down to about 100µm can be resolved.

In order to go further beyond the Abbe's diffraction limit real near-field techniques had to be used. Although no radiation can propagate through an aperture considerably smaller than the wavelength, it is possible to utilize the evanescent waves to probe matter which is placed very closely behind the pinhole. This so-called near-field approach is well established for imaging in the near-infrared and visible spectral range. Very recently we were able to realize a near-field spectrometer in the THz range of frequency which allows us to take images with a spatial resolution of $\lambda/10$ or even $\lambda/100$ and at the same time obtain complete spectral information at each pixel in a reasonable period of time.

Although many visions might not be realistic and things are more complicated than anticipated, for sure the THz gap is about to be closed even when it always remains a difficult territory to work in. For basic solid state research this spectral range is already indispensable, and now more and more scientist form other disciplines, as biology, chemistry, geology, medicine, pharmacy, just to name a few, step into this unknown region and find new and exiting questions and applications in their fields. •

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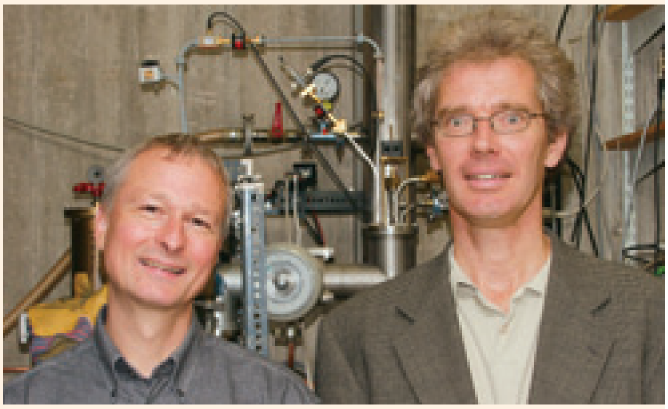

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