THz-Radiation – a Probe in Solid State Physics

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Abstract (shortened version of talk at SICAST 2007)**:** THz-radiation is the natural energy probe to investigate physical mechanisms in solids, because the corresponding transition energies are typically centered around 10 meV. To distinguish, however, in solids between ionic and electronic transitions an external magnetic field as "filter" has to be applied. Whereas the magnetic field tunes the electronic energies, the ionic levels are not affected. A review of THz-magneto spectroscopy covering a total of three decades in energy around 1 THz in magnetic fields up to 1,000 Tesla is given.

Keywords: Magnetospectroscopy, FIR/THz, Semiconductor, Megagauss

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1. Introduction

THz-radiation covers a very special part of the spectrum for electromagnetic radiation: it is centered in a transition range between quantum and wave properties. Both aspects are present and determine essentially the experimental setup. THz-radiation can be generated by wave-typical resonance circuits (gyrotrons, carcinotrons etc.) as well as by quantum-transitions in lasers. In the experimental data the wave character may manifest by interference-effects in the spectrum and the quantum aspect by a definite absoprtion onset. It should be noted that the frequency of ν = 1 THz corresponds to an energy of 4.13 meV being equivalent to an thermal energy kT with a temperature of about T = 50 K. This means that temperature is an important parameter to vary the population of the energy levels involved in any THz-experiment. For the interested reader we give a global reference [1] where many specific references can be found.

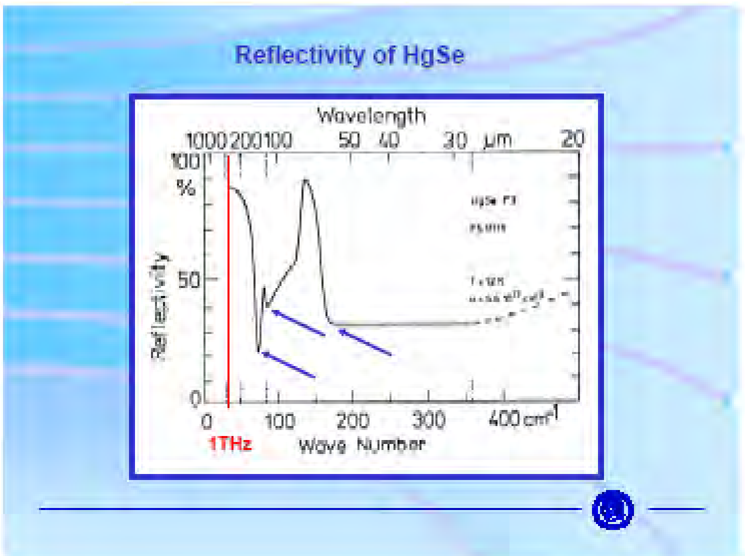

Fig. 1 Reflectivity spectrum of THz-radiation for HgSe.

THz-radiation as electromagnetic waves interact with all charge carriers in the solid, as are the ions of the lattice constituents and quasi-mobile carriers responsible for electric current. For a semiconductor physicist, who aims at the conductive properties of new semiconductor materials for high-tech application, only the electronic properties are interesting. In an ordinary optical spectrum in the THz-range, however, both, ions and electrons, manifest as seen in fig. 1. Here the normal reflectivity of the zero-gap semiconductor material HgSe is plotted as a function of the wavelength. To separate now the effects of ionic coupling to the THz-radiation from those of the electrons additional efforts have to be made.

For this purpose we consider the effect of an external magnetic field on both types of charge carriers in the solid, namely the ions and the electrons. Due to the extremely high mass ratio for the two types of charge carriers the effect on ionic motion is mostly negligible, whereas the electronic energy levels are easily tuned by the magnetic field. This fact is directly visualzed by the corresponding cyclotron frequencies $\omega_c = eB/m$. Here e is the elementary charge, m the mass of the charge carrier and B the external magnetic field. Also sensitive to the magnetic field is the Larmor frequency $\omega_L = g_* \mu_B B$, where g is the electronic g-factor and μ_B Bohr´s magneton. Both types of interaction are visualzed in fig. 2. In this way THz-*magneto* optics provides an excellent tool for the investigation of electronic properties of solids.

2. The Dielectric Function as Interface between Microscopic and Macroscopic Features of the Experiment

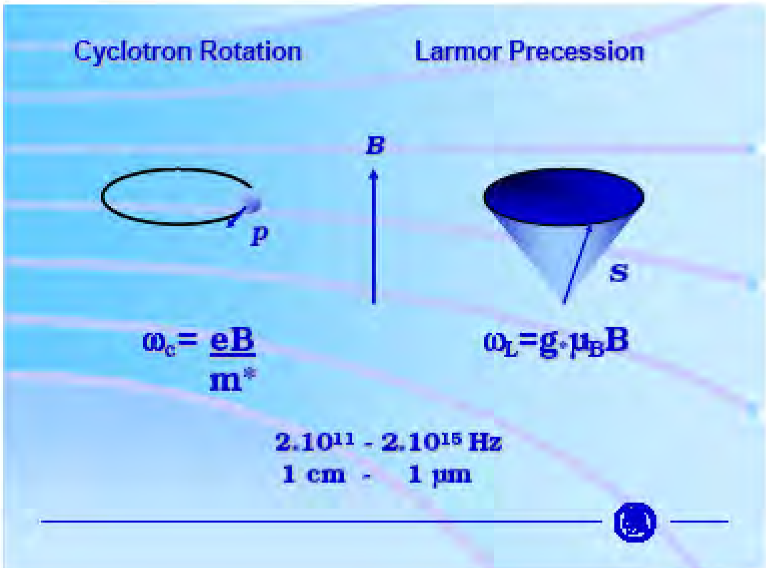

Fig 2 Schematic of the THz-interaction with the electron

The dielectric tensor function ε is the central interface between microscopic parameters of the charge carriers described either by a classical or quantum-mechanical model and the macroscopic measuring quantities determined by the accidental optical boundary conditions. The tensor components $\varepsilon_{\mu\nu}$ depend generally not only on the microscopic parameters of the charge carriers (m, τ) and radiation (ω, $\boldsymbol{k}$) but also on state parameters as are temperature T and an external magnetic field $\boldsymbol{B}$. So $\varepsilon_{\mu\nu} = \varepsilon_{\mu\nu}(\omega, \boldsymbol{k}, T, \boldsymbol{B},)$. The magnetic field dependence ensures that the electronic energy levels can be tuned in such a way that for given radiation frequency a resonance in the dielectric function can be enforced. In this case the radiation energy ħω equals the energy separation of the electronic levels E_{final} - $E_{initial}$ involved. In fig. 3 we have schematically plotted the dielectric function in dependence of both ω and B. The resonance is most easily observed as singularity in the imaginary part corresponding to the absorption. It should be noted that the the resonance can be recorded by two alternative methods: either for constant magnetic field B_{fix} as a function of ω (ω-cut) or for constant frequency ω_{fix} and varying B (B-cut). This is shown schematically in fig. 3. Whereas the ω-cut requires a continuous radiation source in connection with a spectrometer, for the B-cut only discrete radiation frequencies as provided by lasers, gyrotrons etc. are necessary.

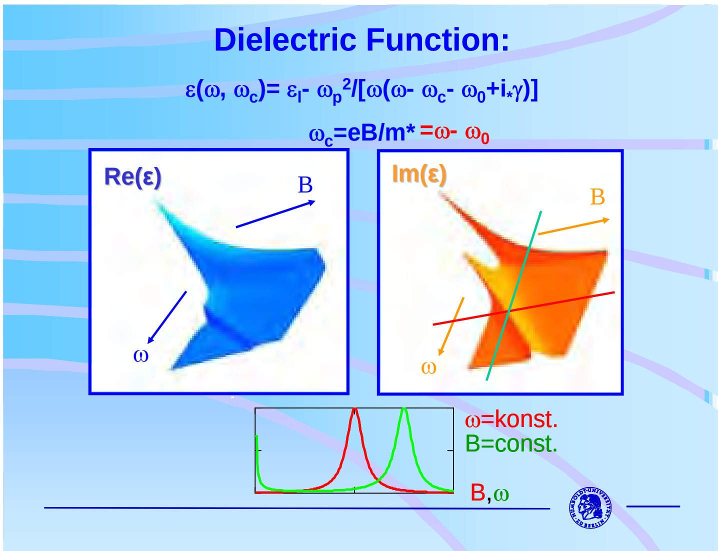

Fig. 3 The dielectric function in the magnetic field.

3. Experimental Setup

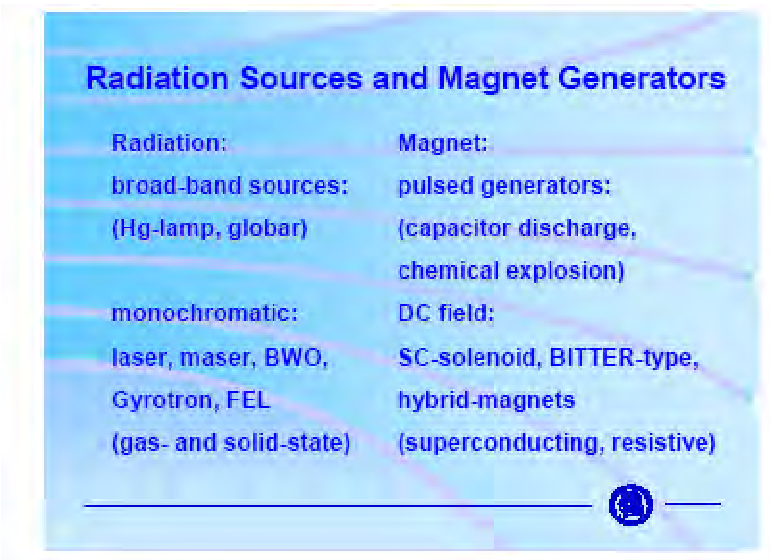

Fig. 4 Necessary experimental components.

Besides of the sample two components for the performance of an THz-magneto optical experiment are necessary: Radiation source with detector and magnetic field generator. For the purpose of changing the thermodynamical population of the energy levels involved an controllable cryogenic environment for the sample is necessary. In fig. 4 some of those necessary components are listed. Whereas the generators for THz-radiation are mostly straight-forward setups, the magnetic field generators for megagauss fields are unique experimental installations available only at few places over the globe.

Depending now on the physical variable to be measured THz-magneto optics can be realized in different experiments involving transmission, Faraday-rotation, reflectivity or in a special multi-reflection experiment the strip-line transmission, cavity detuning, photoresponse in photoconductivity or thermomodulation etc.

We are going to present now some setups for demonstration.

3.1. ω-cut for Fourier-Transform Spectroscopy with SC-Solenoid

The ω-cut needs a continuous variation of the radiation frequency for constant magnetic field. This is most easily realized using a Fourier-transform spectrometer in the constant field of a superconducting solenoid. The cryogenic environment can also be used for cooling the sample. The schematic arrangement ist shown in fig. 5 and the data obtained by this setup on a HgSe-sample in fig. 6. At zero-magnetic field the reflectivity spectrum shows clearly the plasma edge, which shows with increasing magnetic field a splitting equal to the cyclotron energy $\hbar\omega_c$ = eB/m. This means that this THz-experiment can be considered as a “electron balance” providing directly a value for the effective mass of the electrons involved.

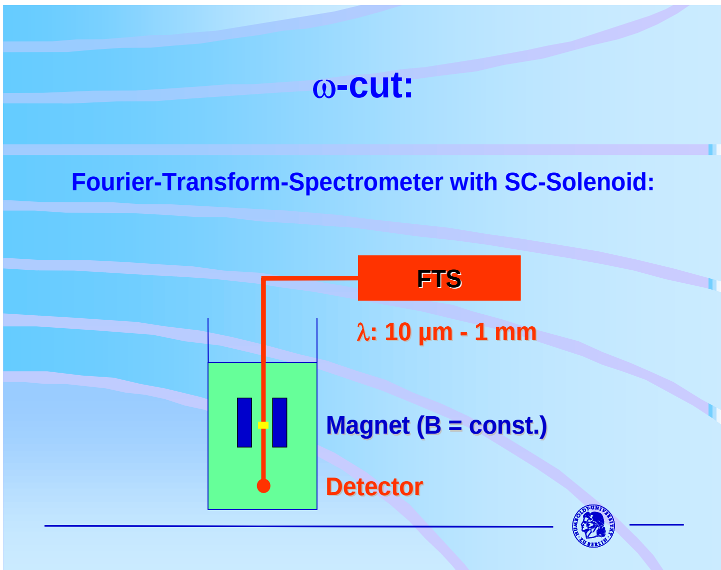

Fig 5 The ω-cut setup.

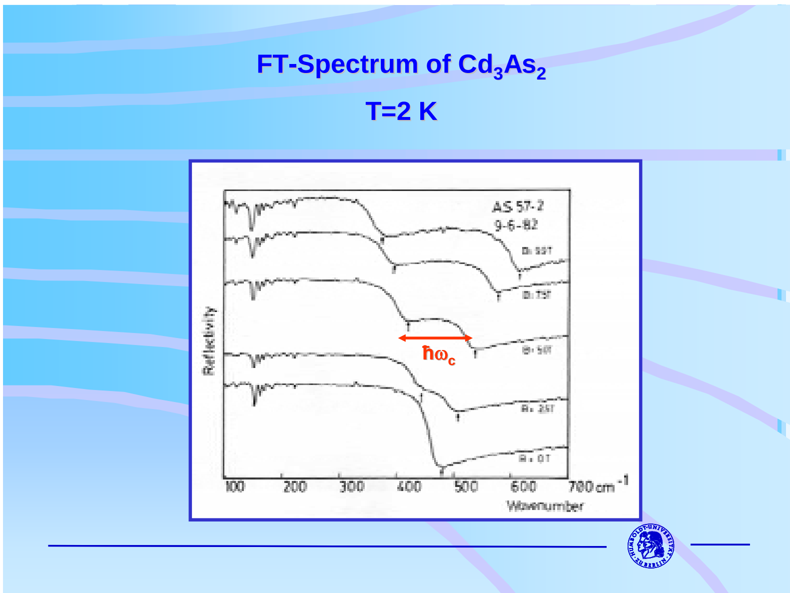

Fig. 6 Magneto-reflectivitiy on HgSe.

3.2. B-cut for THz-Laser Radiation with SC-Solenoid

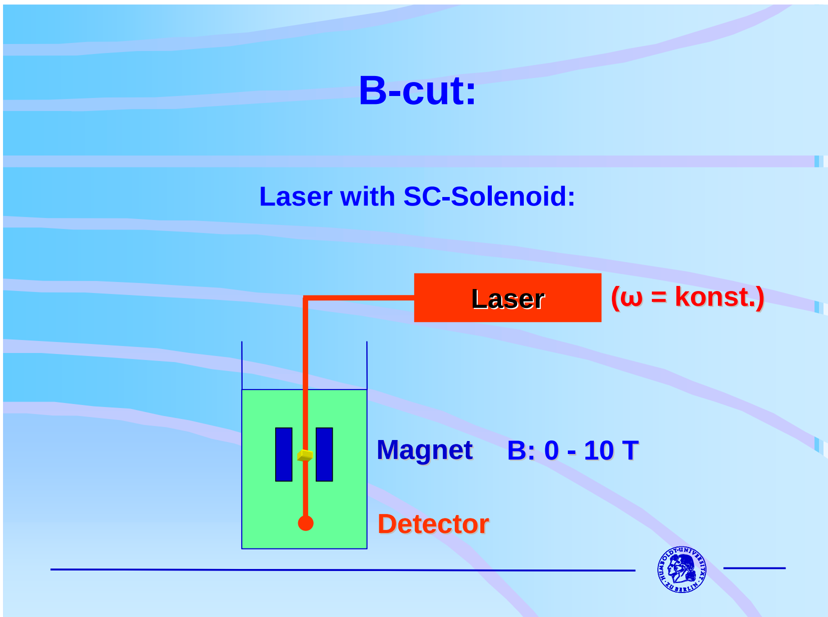

Fig. 7 Experimental setup for the B-cut.

The B-cut needs a continuous magnetic field variation in connection with a unifrequent radiation. Here the radiation is provided by a molecular gas laser, as shown schematically in fig. 7. The transmission data obtained with 0.89 THz-laser radiation on a p-type Te-sample are reproduced in fig. 8 for the temperatures of T = 4.2 K and T = 14 K. At low temperature in addition to the p-type cyclotron resonance line (CR) several impurity transition are observed, because holes have populated at low temperatures the impurity levels. At elevated temperatures the carriers are excited into the quasi-free band states. This is a demonstration of

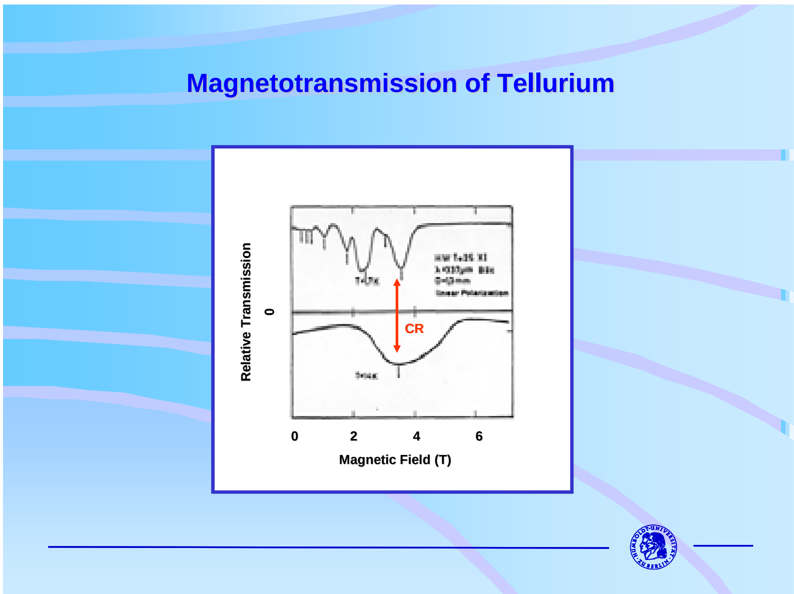

Fig. 8 Transmission of Te using 0.89 THz-radiation for different temperatures.

the usefulness of the temperature (thermal energy comparable to the radiation energy!) as additional parameter in a THz-magneto optical experiment.

It should be noted that the resonance structures observed in the measurement of the radiation intensity, namely the transmission, can also be detected in the physical properties of the sample itself as demonstrated in fig. 9 by the photoresponse in the upper part of the figure, whereas in the lower part the transmissionm is reproduced.

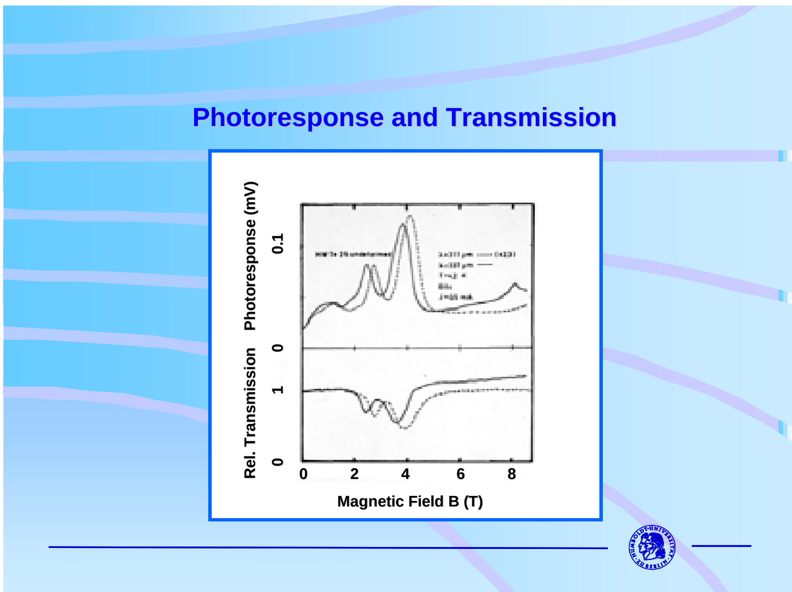

Fig. 9 Photoresponse in Te in comparison with transmission for 0.89 and 0.96 THz-radiation.

A very special measuring quantity is the *strip-line transmission*, which can be understood as the wave-optical limit of a classical multireflection experiment, as schematically indicated in fig. 10. If the distance D between mirror and sample is less twice the wavelength of the radiation the “transmitted” intensity is determined by the wave-guide attenuation.

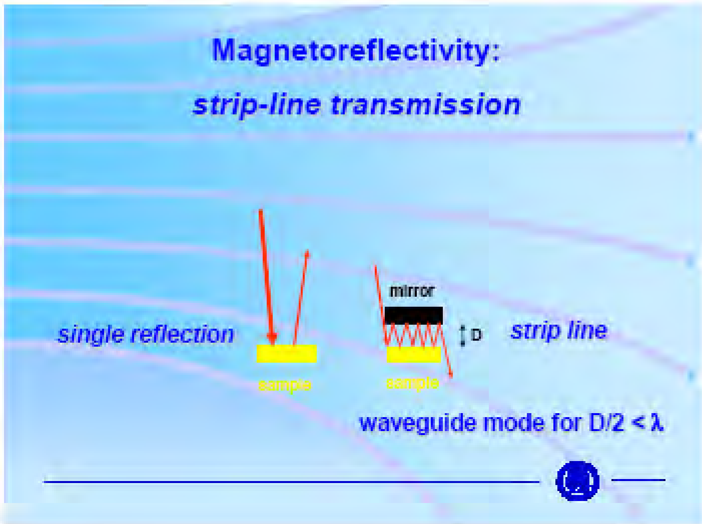

Fig. 10 Schematic of the *strip-line technique*.

This quantity is extremely sensitive to very small changes in the refractive index. As example we have plotted in fig. 11 the strip-line transmission of 2.54 THz-radiation on a a magnetic Eu-compound. This material has a complicated, temperature dependent magnetic phase diagram manifesting – including pronounced hysteresis effects – in the plots of fig. 11.

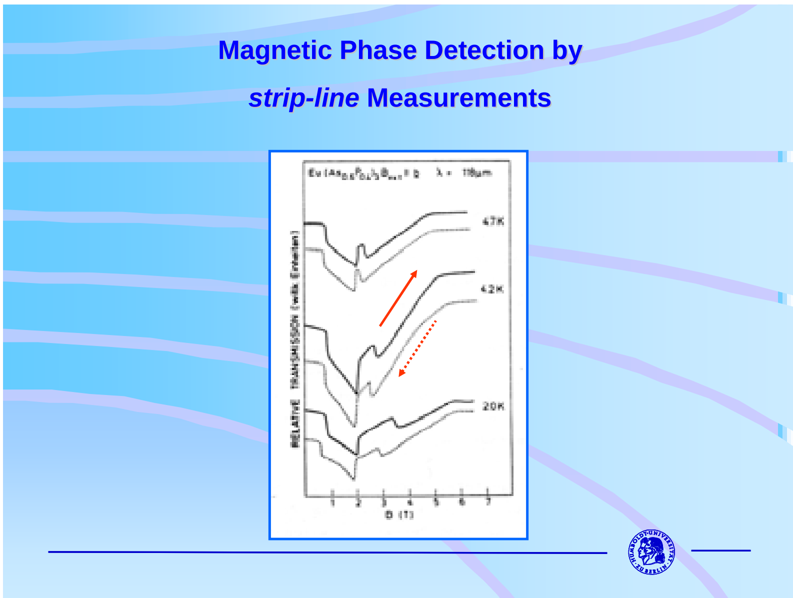

Fig. 11 The strip-line transmission on a magnetic Eu-compound using 2.54 THz-radiation**.**

4. THz-Magneto Optics in Megagauss Fields

The combination of THz-radiation and megagauss fields pushes the experimentalist to the frontiers of physics, to the limits of possibilities in experimental techniques, where quite unexpected and novel results are expected. The mission of the *Humboldt High Magnetic Field Center* in Berlin was the generation and application of megagauss fields for the investigation of electronic levels in solids. The generation of megagauss fields is still a challenge in experimental physics because a magnetic field coil producing 100 T erperiences a Maxwell pressure of 40 tons/cm^2 ! No material so far can withstand such pressure, so that dynamical forces – either inertia a chemical explosion forces - have to be used to perform such experiments.

4.1. THz-Magneto Optics using the *single-turn-coil* Technique

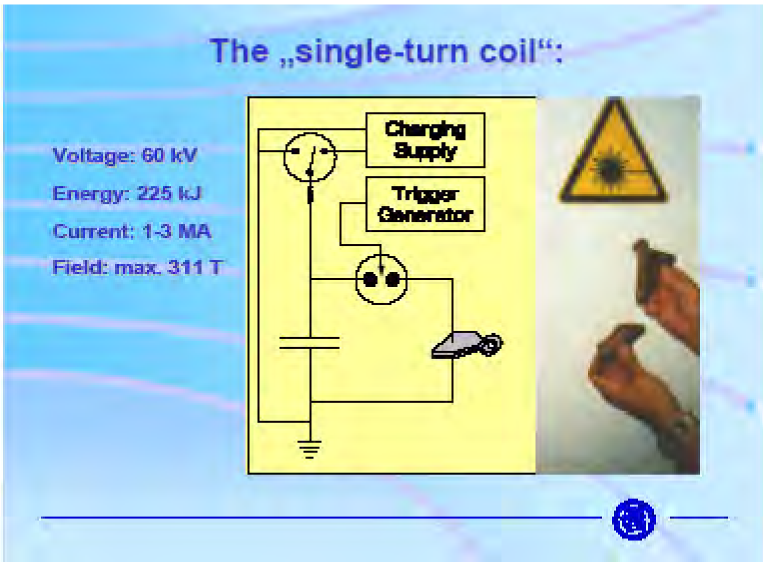

Fig. 12 Schematic of the *single-turn coil*.

With the *single-turn-coil* technique magnetic fields slightly higher than 300 T can be generated in short pulses of about 5 µsec length. The coil of 10 mm diameter and 10 mm height consists of one single turn of 3 mm copper conductor and explodes violently during the field generation process. The trick is now, that all the electric energy has to be transformed before the coil has considerably expanded due to the Maxwell pressure. So the pulse length is limited to the order of µsec. On the other hand side the current has to be of the order of some 10^6 A. To energize the coil a low capacitance with high charging voltage has to be used. In the *Humboldt High Magnetic Field Center* in Berlin a capacitor bank with an energy of 225 kJ at 60 kV charging voltage is applied and generates 5 µsec long current pulses up to 3 MA and a field up to 311 T. The schematic is seen in fig. 12. The challenge for the THz-magneto optics experimentalist is, that inside the 10 mm diameter coil the optical setup including sample and cryogenics has to be installed. The special feature of the experiment is, that in about 90% of the experiments the equipment inside the coil survives, since the coil explodes radially in outward direction due to the Lorentz forces.

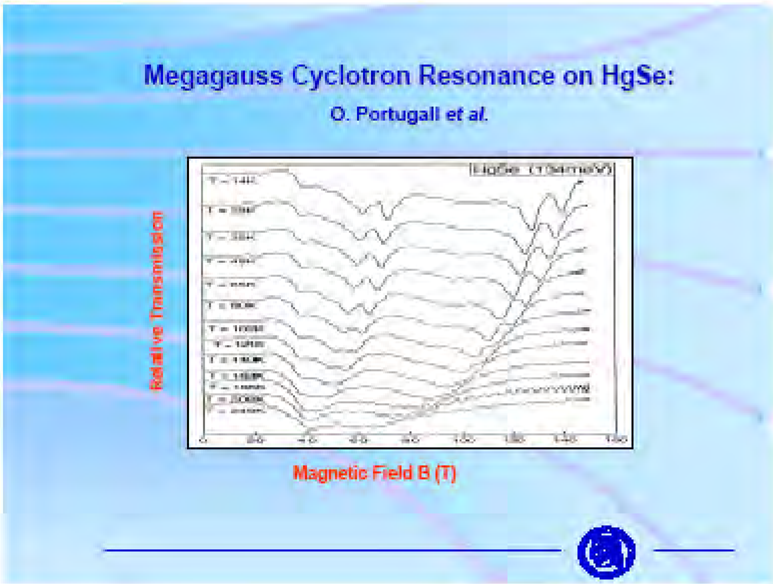

Fig. 13 The transmission of 32 THz-radiation through HgSe.

In fig. 13 we have reproduced the transmission data of 32 THz-radiation through a HgSe sample for different temperatures. These data were directly recorded without any smoothing sampling technique and demonstrate the excellent signal/noise ratio of the equipment. We had to apply for this high concentration material such high radiation frequency, because we had to shift the radiation frequency beyond the plasma edge for sufficient transmission.

Special interest have the nanostructured modifications of common semiconductor materials. So we have also investigated quantum-dot samples of the material HgSe and compare in fig. 14 the cyclotron resonance of quantum well (red curve) and quantum dot (blue curve) of HgSe:Fe. It should be noted that the data for the quantum dot sample reveal a 50% mass increase in comparison with those of the quantum well. This effect is due to strain effects in the lattice part for the quantum dot.

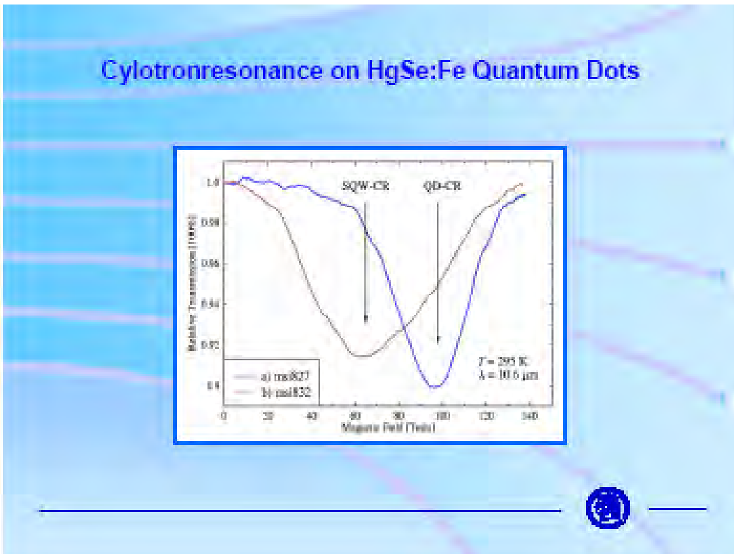

Fig.14 The cyclotron resonance of quantum dots (blue curve) in direct comparison with the cyclotron resonance in a quantum well (red curve) in HgSe:Fe.

4.2. Time-Dependent Spectroscopy using the *single-turn--coil* Technique

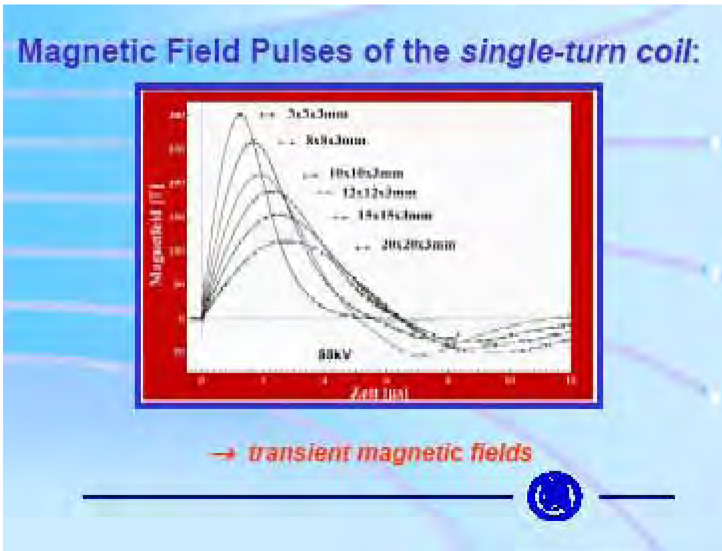

Fig. 15 The pulse-shape of the magnetic field pulses using the *single-turn* generator.

In fig. 15 we have plotted the magnetic field pulse of single-turn coils of different dimensions (diameter in mm x height in mm x thickness of copper in mm). The smaller the diameter of the coil the lower is the inductance and hence the ringing frequency of the L/C circuit increased. By variation of both the coil diameter and the maximum field we can tune the quantity dB/dt. In fig. 16 we demonstrate the existence of time-dependent hysteresis effects in indiumantimonide by use of low-field application of single-turn-coil pulses. The transmission anomalies are clearly detectable only for increasing field independent of the field direction.

5. THz-Spectroscopy in Explosive Flux Compression

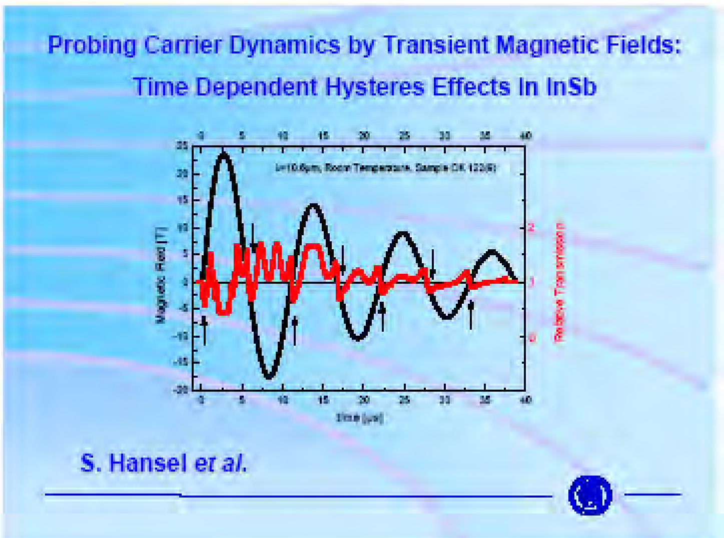

Fig. 16 Time dependent hysteresis effects in indiumantimonide

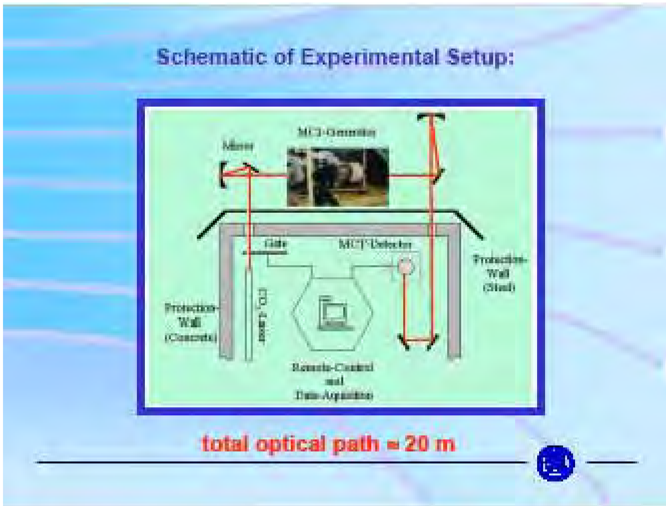

Fig. 17 The schematic arrangement of the explosive flux compression of the VNIIEF in Sarow, Russia

Magnetic fields beyond the 300 T-limit can only be generated by flux-compression techniques. Whereas at present the electromagnetic flux-compression generates fields up to 622 T, the explosive flux-compression generates peak fields of 2,800 T. For real experiments, however the upper limit is about at 1,000 T. The flux-compression makes use of the fact, that a magnetic flux Φ within a well-conducting closed loop remains constant independent of the actual cross-area F of the loop. So by squeezing of the loop the magnetic field $B = \Phi/F$ increases. For explosive flux compression the squeezing procedure is achieved by a concentric blast of 16 kg high explosive charge consisting of a mixture of TNT and hexogen with a total energy of 64 MJ. Part of this chemical energy is transformel into magnetic field energy. Because of the violent blast this kind of experiment can only be performed in open-air. A schematic of the installation at VNIIEF in Sarow, Russia is reproduced in fig. 17. For optical experiments a very long optical path of about 20 m has to be considered, because valuable equipment as laser, detector, and data recorders has to be protected by a bunker, as shown schematically in fig. 17. The magnetic field is measured in the experiment by a series of one-turn pick-up coils distributed over the crossection. The field increases rather exponentially during few µsec. The time correlation between field and optical data is critical for correct gauging of the field. In fig. 18 the data obtained on a GaAs-sample is reproduced. Using 32 THz-radiation the cyclotron resonance at the Γ-point CR at lower fields had been measured before on the same sample in the *single-turn coil*. These data are represented by the black dotted curve. The pronounced decrease in transmitted intensity near 1000 T is due to blocking of the optical path by fragments of the explosion. The completely novel feature, however, is the transmission minimum at about 550 T. This absorption is explained by the cyclotron resonance of electrons at the L-point of the Brilouin zone, which is populated in high magnetic fields. The corresponding Landau levels are plotted in fig. 19. The lowest Landau level at the Γ-point gets at high magnetic fields close to the lowest level at the L-point, so that population change occurs and absorption at the L-point becomes possible.

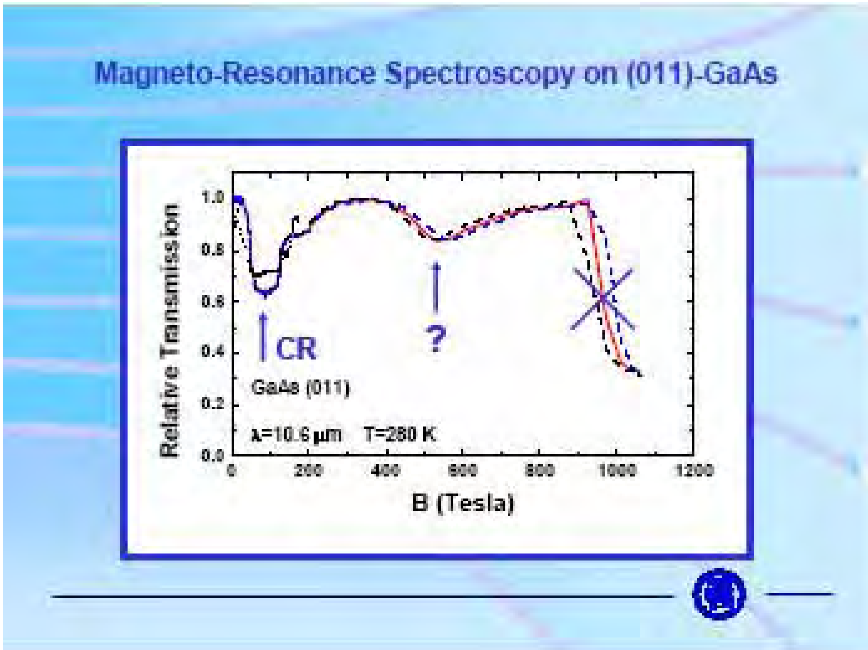

Fig. 18 The transmission of 32 THz-radiation trough GaAs for magnetic fields up to 1000 T.

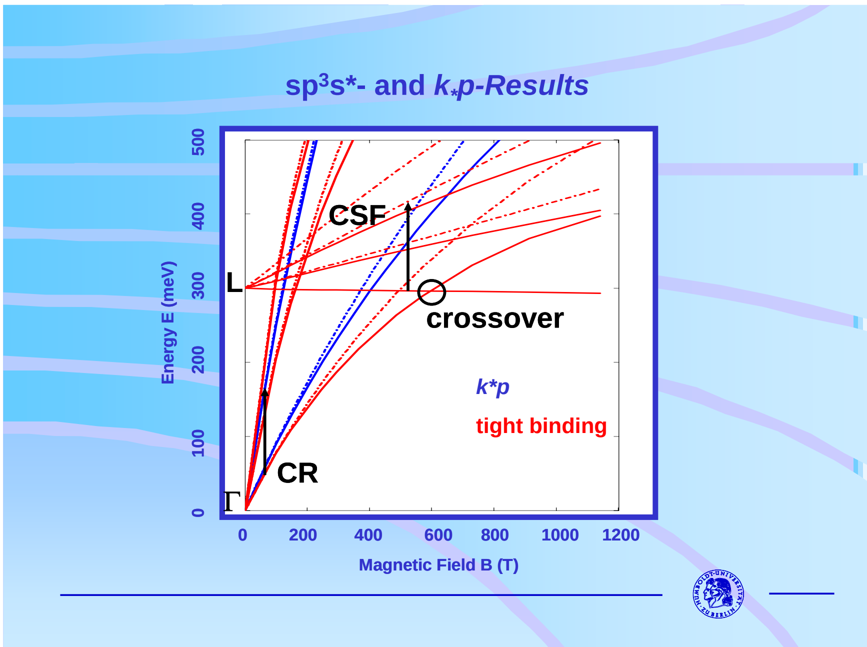

Fig. 19 The schematic of Landau levels in GaAs shows the level crossing, so that the ground state of the system switches from the Γ-point to the L-point for fields higher than 550 T.

5. Summary

Terahertz Technologies in combination with strong magnetic fields provide an extremely efficient tool not only for the investigation of electronic energy levels in solids but also due to the transient character of those fields for the study of the dynamic processes involving these levels.

Acknowledgments

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