Development of time domain techniques in microwave spectroscopy

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Introduction and motivations

The field of solid state physics has changed our world. The fundamental understanding of the materials we employ made possible the technologies that we now consider essential for our society. High speed, small size electronics, clean, renewable energy sources, cheap, and easy communication between people across continents, new imaging techniques and implanted devices in the medical sciences, none of these would be possible without the work of scientists who endeavor to explain the microscopic structure and behaviour of the new substances that we learn to produce each day. If we just think of semiconductors in general and silicon in particular, it is very clear that fundamental research will be the first step for the technological revolutions yet to come.

There are several tools available to us as scientists to further explore solid state physics. Theoretical physicist work to understand new materials, but theories must always be confirmed or disproven by experiments. For an experimental physicist to be able to perform measurements, he/she must understand the theory, samples of the materials under scrutiny must be synthesized, and the instruments for the measurement must be available. Since we as physicists are always working on the edge of the understanding of humanity, we must understand and improve the techniques that we use for the investigations we wish to perform. We must learn from experts in the other fields of science and engineering to develop new techniques for our work. Often a new technology makes it possible to widen the scope of our explorations. An example of this was the invention of atomic force microscopy, where a few simple ideas led to a device which we now consider a basic tool for our work. We must always be willing to learn from the ideas of other scientists and engineers, and incorporate them into our work.

Research topic and its motivation

In many fields of science and engineering, the trend of replacing frequency-domain measurement techniques with time-domain solutions can be observed. This opens the possibility of performing measurements with higher sensitivity in shorter times, in some cases enabling entirely new fields of research.

The use of time-domain methods revolutionized nuclear magnetic resonance (NMR) spectroscopy. In earlier techniques, continuous wave (CW – stable or slowly changing frequency signal) sources were used to measure narrow resonance lines. This was replaced by the measurement of the time-domain response of the nuclei to short RF pulses and for which the Nobel prize in Chemistry in 1991 was awarded to R. R. Ernst. This improved the sensitivity, resolution, and precision of these measurements. These improvements led to the widespread use of magnetic resonance imaging techniques in the biological sciences, medicine, and solid state spectroscopy.

Objectives

Based on the ideas mentioned beforehand, my goal was to develop a method to measure the properties of microwave resonators, which is a task with wide ranging applications. Besides industrial processes (e.g. in heating applications), resonator measurements are employed in the so-called cavity perturbation method, which is a widely used technique in experimental solid state physics to perform contactless measurements of material properties including electrical permittivity, magnetic permeability, and electrical conductivity.

Using classical methods, cavity parameters are measured using frequency sweeps or modulation. During my PhD work, I investigated the transient behavior of microwave resonators and developed a method to employ the transients for high sensitivity measurements. I further developed the novel method to form a so-called feedback resonator setup to further improve the technique, increasing stability, sensitivity, and decreasing setup complexity. I investigated the performance of the new methods and compared them with techniques found in the literature. The novel methods have shown significant improvement over the conventional techniques in terms of signal to noise ratio, time resolution, and stability.

I present two applications of the novel methods. I used the novel methods to perform investigations on single wall carbon nanotubes, and managed to explain the origin of an anomalous low-temperature effect that was a topic of research in the past few years. I also performed microwave detected photoconductivity measurements on intrinsic and doped silicon wafers, making use of the high sensitivity and time resolution of the novel methods, to investigate samples that were in the past not analyzable with similar methods. This latter is a clear demonstration of the capabilities of the method.

Contents of the thesis

The structure of the thesis is as follows: after an introduction in Chapter 1, in Chapter 2, I present the theoretical background of my work. I start with the relevant parts of transmission line theory and its applications for the electric field propagation in media. The concept of the surface impedance is introduced. A connection of radio frequency wave propagation and optics is made. I explain the theoretical description of RF resonators, including their transient behaviour, which is essential for my work. The chapter continues with a description of cavity perturbation methods. After a discussion of the sources and representations of noise in measurement systems, I present the the general advantages of time-domain over frequency domain spectroscopy. The chapter concludes with a discussion of the photoconducting properties of silicon.

In Chapter 3, I present experimental techniques employed in my work. I explain the technique of down-mixing of measurement signals to lower frequencies, and the basic mathematics of discrete Fourier transformation. Finally, I present the methods to measure microwave detected photoconductive decay.

In Chapter 4, I present the results of my work, namely the development of a time-domain method to measure microwave resonator parameters. I continue with an additional improvement of this technique incorporating a feedback resonator. I discuss the sources of noise in my methods and define a figure of merit for a comparison of several techniques. Finally, I present the investigations performed used the novel methods, explaining the anomalous non-linear microwave absorption in single wall carbon nanotubes and the detection of photoconductive decay using the transients of microwave cavities.

In Chapter 5, I summarize the PhD thesis and list the thesis points.

Thesis points

1. I developed a novel method to determine the resonance frequency and quality factor of a microwave resonator, which is faster, more stable, and conceptually simpler than the earlier existing techniques. The resonator is irradiated with a microwave frequency far from its resonant frequency and after switch-off of the excitation signal, radiates a decaying signal at its eigenfrequency. This signal is down-mixed and digitized. Fourier transformation directly yields the

resonance curve from which the resonator parameters can be calculated [T1].

I defined a novel figure of merit for the methods which measure the resonator quality factor and eigenfrequency. The figure of merit was motivated by the need for a quality factor independent assessment of the resonator measurements. Studies on ultra-high Q resonators repeatedly return excessive performance values, however when the novel figure of merit is used, it turns out the different technical realizations return similar performance, which is independent on the resonator Q [T1].

2. I further improved the time-domain measurement of resonator parameters. The microwave resonator is placed in a feedback resonator setup, where the output of an amplifier is connected to its own input with the resonator as a band pass filter. This results in oscillations whose frequency is tuned automatically by any small changes to the resonator parameters in the presence of a sample. After reaching steady-state oscillation, the feedback circuit is disrupted by a fast microwave switch and the transient signal, which emanates from the resonator, is detected using down-conversion. I used the Fourier transform of the resulting time-dependent signal to directly obtain the resonance profile of the resonator [T2].

   I also assessed the performance of the time-domain methods and I showed that it significantly (by up to two orders of magnitude) outperforms the presently existing techniques [T2]. I have identified the origin of the noise in the time-domain technique and I have determined the ultimate noise limit [T2].

3. I studied the anomalous non-linear microwave absorption in single wall carbon nanotubes at low temperatures (below 20 K) with the time-domain method. It was debated for long whether this arises from a true electronic behavior or from a heating effect. I found that the anomaly has an extremely slow dynamics on a few hundred second timescale. This strongly suggested that the anomaly is not caused by an intrinsic electronic effect and that it is rather due to a slow heat exchange between the sample and the environment [T3].

4. I developed a novel method to detect photo-induced conductivity in semiconductors using microwave resonators. Earlier techniques using microwave resonators have yielded material parameters after involved modeling or with slow time dynamics (beyond a few ms-second). The new approach yields directly the resonator parameters, which are in turn related to the material parameters. It is based on the detection of the transient response of a microwave cavity. While the method encompasses all the known benefits of resonators in terms of sensitivity and accuracy,

its ultimate time resolution is the resonator time constant which can be as low as a few ns [T4].

The publications related to the thesis points are as follows:
[T1] B. Gyüre, B. G. Márkus, B. Bernáth, F. Murányi, and F. Simon: *A time domain based method for the accurate measurement of Q-factor and resonance frequency at microwave frequencies*, Review of Scientific Instruments **86**, 094702 (2015).
[T2] B. Gyüre-Garami, O. Sági, B. G. Márkus, and F. Simon: *A highly accurate measurement of resonator Q-factor and resonance frequency*, Review of Scientific Instruments **89**, 113903 (2018).
[T3] B. G. Márkus, B. Gyüre-Garmi, O. Sági, G. Csősz, F. Márkus, and F. Simon: *Heating causes non-linear microwave absorption anomaly in single wall carbon nanotubes*, Physica Status Solidi B **255**, 1800258, (2019).
[T4] B. Gyüre-Garami, B. Blum, O. Sági, A. Bojtor, S. Kollarics, G. Csősz, B. G. Márkus, J. Volk, and F. Simon: *Ultrafast sensing of photoconductive decay using microwave resonators*, Journal of Applied Physics **126**, 235702 (2019).

Other published works which are related to the thesis but are not included in the thesis points:
[5] S. Dzsaber, M. Negyedi, B. Bernáth, B. Gyüre, T. Fehér, C. Kramberger, T. Pichler, F. Simon, *A Fourier transform Raman spectrometer with visible laser excitation*, Journal of Raman Spectroscopy **46** 327 (2015).

[6] M. Negyedi, J. Palotás, B. Gyüre, S. Dzsaber, S. Kollarics, P. Rohringer, T. Pichler, F. Simon: *An optically detected magnetic resonance spectrometer with tunable laser excitation and wavelength resolved infrared detection*, Review of Scientific Instruments **88**, 013902 (2017).
[7] I. Gresits, Gy. Thuróczy, O. Sági, B. Gyüre-Garami, B. G. Márkus, F. Simon: *Non-calorimetric determination of absorbed power during magnetic nanoparticle based hyperthermia*, Scientific Reports **8**, 12667 (2018).
[8] B. G. Márkus, G. Csősz, O. Sági, B. Gyüre-Garami, V. Lloret, S. Wild, G. Abellán, N. M. Nemes, G. Klupp, K. Kamarás, A. Hirsch, F. Hauke, F. Simon: *Electronic properties of air-sensitive nanomaterials probed with microwave impedance measurements*, Physica Status Solidi B **255**, 1800250, (2018).