Probing the momentum relaxation time of charge carriers in ultrathin semiconductor layers

S. Funk, G. Acuna, M. Handloser, and R. Kersting
Photonics and Optoelectronics Group & Center for NanoScience,
University of Munich, 80799 Munich, Germany
(Dated: August 6, 2009)

We report on a terahertz time-domain technique for measuring the momentum relaxation time of charge carriers in ultrathin semiconductor layers. The phase sensitive modulation technique directly provides the relaxation time. Time-resolved THz experiments were performed on n-doped GaAs and show precise agreement with data obtained by electrical characterization. The technique is well suited for studying novel materials where parameters such as the charge carriers’ effective mass or the carrier density are not known a priori.

PACS numbers: 87.50.U-, 78.47.J-, 73.50.-h, 78.66.Db, 78.20.-e

I. INTRODUCTION

Terahertz (THz) time-domain spectroscopy (TDS) has developed towards an alternative technique for characterizing electrical transport in condensed matter. Many studies on semiconductors demonstrate excellent agreement with the Drude model and characteristic properties such as the complex conductivity have been deduced [1, 2]. One of the most fundamental quantities which describe charge transport is the Drude momentum relaxation time \( \tau \). Deducing \( \tau \) from optical data demands knowledge of further parameters, such as of the carriers’ density \( N \), the effective mass \( m^* \), the background permittivity \( \varepsilon_{\infty} \), and the thickness \( d \) of the sample [1, 3, 4]. The latter can be deduced in time-resolved THz experiments when the optical thickness exceeds the wavelength of the radiation [3, 4]. However, the progress in material science provides many novel materials, which are extremely thin and have unknown charge carrier densities and effective masses. Recently, terahertz spectroscopy as well as far-infrared and microwave techniques are used for characterizing modern materials [7, 5, 6, 9]. But these approaches require precise knowledge of material parameters or use fitting procedures.

In this work, we present an optical method for probing the Drude relaxation time without using any further parameters. In our technique we modulate the optical properties of the sample and record the differential response signal at THz frequencies. We will show that the phase of this signal directly provides the relaxation time. It is worth pointing out, that our method is not restricted to the THz band. The method can be adapted to any spectroscopic technique, which provides the optical phase of transmitted or reflected radiation.

Small modulations of the sample’s optical properties can be achieved most conveniently by electromodulation techniques similar to the method described by Allen et al. [10]. The fact that the modulation of the charge carrier density resembles the operation of a field-effect transistor illustrates the application potential of the technique. Currently, numerous novel materials such as nanostructures or organic semiconductors have been developed for similar devices. Many of these materials, however, are inhomogeneous, include a high density of traps and grain boundaries, or have a broadened density of electronic states. Dispersive transport and hopping dominate electrical properties and mask the fundamental carrier dynamics [12, 13]. In such cases THz modulation spectroscopy may provide an unambiguous insight into the fundamental carrier relaxation dynamics.

II. METHOD

The concept of our method is illustrated in Fig. 1(a). We study a thin sheet of a semiconductor, which has the thickness \( d \) and contains mobile carriers. Within the small fraction \( \Delta d \) the charge carrier density can be modulated, e.g. the density can be reduced to zero. A fraction of an incident THz electric field \( \mathbf{E}_{\text{inc}} \) is transmitted through the doped layer, and generates a current density \( \mathbf{j} \) which is proportional to the complex conductivity \( \sigma \). In the following, it is convenient to discuss differential quantities such as the differential field \( \Delta \mathbf{E} = \mathbf{E}_1 - \mathbf{E}_0 \) where \( \mathbf{E}_1 \) and \( \mathbf{E}_0 \) are the transmitted electric fields in the case of an interaction across the entire layer and in the case the layer thickness is reduced by \( \Delta d \), respectively. The differential current density \( \Delta \mathbf{j} \) can be described by a change of the thickness

\[
\Delta j \approx \sigma E_1 \frac{\Delta d}{d},
\]

where we assumed the electric field to be virtually constant across the layer, which is a valid approximation for optically thin films. In order to deduce the impact of \( \Delta \mathbf{j} \) on the reflection and transmission of radiation, we
FIG. 1: a) Schematic diagram of a thin metallic layer of thickness d embedded in a dielectric with a refractive index n. The differential current $\Delta j$ driven by the THz field $E_1$ induces a differential magnetic field $\Delta H$, which in turn causes the differential field $\Delta E$ measured by THz-TDS. b) Complex diagram of the y-components of the electric fields $E_{1y}$, $E_{0y}$, and $\Delta E_y$.

consider that $\Delta j$ causes a differential magnetic field $\Delta H$, which can be calculated by Ampere’s law:

$$\int \Delta H \cdot d\mathbf{r} = \mathbf{\Delta j} \cdot dA .$$

(2)

In layers which are thin compared with the wavelength ($d \ll \lambda$), the x-components of $\Delta H$ dominate the above equation whereas the z-components can be neglected. We also consider that $\Delta d$ is smaller than the skin depth. In consequence, the modulated current density $\Delta j$ does not change significantly across the depleted layer. The magnetic field $\Delta H$ causes a differential electrical field given by $\Delta E = Z_0 \Delta H / n$, where $Z_0$ is the impedance of vacuum and n is the refractive index of the surrounding medium. Using $E_{1y}$ as reference provides:

$$S = \frac{\Delta E_y}{E_{1y}} \approx -\Delta z \frac{Z_0}{2n} .$$

(3)

In the Drude model the complex conductivity is

$$\sigma = \frac{Ne^2}{m^*} \frac{1 - i\omega\tau}{1 + \omega^2\tau^2} ,$$

(4)

where $N$ is the carrier density and $m^*$ is the carrier’s effective mass $[14, 15]$. Relating the imaginary part of $S$ to the real part yields the optical phase of the signal:

$$\tan \phi = -\omega\tau .$$

Thus, the momentum relaxation time can be directly deduced from $\phi$ without the use of further parameters. Figure(1b) displays the fields in the complex plane.

Alternatively, the amplitude or intensity of the signal $S$ can be analyzed in case that $\omega\tau \approx 1 \ [11]$. In the Hagen-Rubens range ($\omega\tau \ll 1$), however, the spectra become structureless because the amplitude is proportional to $1/\sqrt{1 + \omega^2\tau^2}$. In contrast, the phase is proportional to $\omega\tau$, which illustrates the strength of our technique when characterizing materials in which ultrafast scattering occurs.

III. EXPERIMENTAL

We tested the method on a GaAs structure grown by molecular beam epitaxy (MBE). It comprises an n-doped layer grown on a semi-insulating GaAs substrate. The layer is doped at $2 \times 10^{16}$ cm$^{-3}$ and has a thickness of 2 $\mu$m. For electronic control we fabricated a Schottky contact on top of the structure by depositing a 10 nm thick film of Cr. The second terminal is an Ohmic AuGe contact. The doped layer in the GaAs can be partially depleted from electrons by the application of a negative bias to the Schottky contact $[10]$. In our experiments the changes of the layer thicknesses are of the order of 100 nm. This is much smaller than the skin depth of the electron gas, which is about 20 $\mu$m at THz frequencies. Thus, the assumptions made in section[I] are justified. They can be applied to many other devices where conductive sheets are modulated, as for instance, in field-effect transistors.

In our experiments we apply time-resolved THz transmission spectroscopy. The sample is mounted in a cryostat, which allows for low-temperature measurements. A titanium-sapphire laser delivers pulses of 80 fs duration at 780 nm center wavelength and at a repetition rate of 80 MHz. About 700 mW of the laser power is used for the photoexcitation of an interdigitated THz emitter $[17, 18]$. Parabolic mirror optics with $N.A = 0.3$ focus the THz radiation onto the GaAs structure. The transmitted radiation is time-resolved by standard electro-optic sampling in a [110] ZnTe crystal of 1 mm thickness $[19]$. The setup has a bandwidth of 2.8 THz and the signal to noise ratio is about $10^5$ Hz$^{1/2}$. We measured the field strength of the THz pulses using the procedure described in Ref. $[20]$ and deduced a peak field of 70 V/cm. The upper curve in Fig. 2 shows a THz pulse transmitted through the GaAs structure. The lower curve displays the differential signal, which we obtained by modulating the structure with a bias of -5 V. This corresponds to a change of the depletion width of about 400 nm.
IV. RESULTS AND DISCUSSION

In order to prove the concept of our technique we measured the differential transmission through the GaAs structure at room temperature. The electron gas was modulated by switching the bias to the Schottky contact between 0 V and -1 V. This corresponds to a modulated layer thickness of 120 nm or to a modulated sheet density of \( n_{2D} = 2.4 \cdot 10^{11} \text{ cm}^{-2} \). We recorded the THz signals \( E_1 \) and \( E_0 \) [21]. After Fourier transform of the differential THz signal \( \Delta E = E_1 - E_0 \), the optical phase was obtained as described above. The data depicted in Fig. 3 show the expected dependence of the phase on the frequency. Using a relaxation time of \( 198 \pm 2 \text{ fs} \) provides an excellent agreement with eq. (5). Additionally, the data indicate a phase of \( 180^\circ \) for zero frequency, which is characteristic of the Drude model.

In section II we assumed that only the lateral components of \( \Delta H \) contribute to Ampere’s law whereas the perpendicular components can be neglected. We also assumed that the exciting THz field is virtually constant across the modulated layer. We validate these assumptions by measuring the THz signals in dependence on the modulation depth \( \Delta d \) by increasing the modulation voltage \( V \). The results are summarized in Fig. 4. The measured phase shows no dependence on \( V \), which proves the validity of the assumptions. In contrast, the amplitude follows a square root dependence as expected for the increase of the depletion width with voltage [16]. The calculation of the amplitude shows deviations for higher voltages. We attribute these differences to imperfections of the Schottky contact and to inhomogeneities of the doping profile. This illustrates the difficulties that arise in deducing transport properties from amplitude data and highlights the reliability of phase measurements.

In the case of our well-defined MBE grown structure, parameters such as the doping density and effective carrier mass are well known. Thus in this particular case, the momentum relaxation time can be extracted from four-point measurements [22]. Figure 5 shows temperature resolved data. Electrical and phase measurement differ by

![Figure 2](image1.png)

**FIG. 2:** a) Time-resolved few-cycle THz pulse after transmission through the metal-semiconductor structure. b) Differential signal obtained by switching the GaAs between equilibrium and partial depletion. The modulation bias of -5 V increased the depletion zone by 400 nm.

![Figure 3](image2.png)

**FIG. 3:** Frequency dependence of the tangent of the phase angle \( \phi \) obtained on the GaAs structure at room temperature (symbols). The solid line shows a calculation for \( \tau = 198 \text{ fs} \). The dashed line indicates \( \omega \tau = 1 \).

![Figure 4](image3.png)

**FIG. 4:** a) Dependence of the phase angle \( \phi \) on the modulation bias for room temperature. The data show the phase for frequencies of 1 and 2 THz, respectively. b) Dependence of the amplitude of the electromodulation signal on the modulation bias at 1 and at 2 THz, respectively. The solid lines depict calculations that take into account the depletion underneath the Schottky contacts with increasing bias [16].
about 3%, which illustrates that our technique provides precise data. At low temperatures, however, freeze-out of the donors may lead to a reduced electrical conductivity, which explains the differences at about 100 K. We emphasize that our approach is not advantageous in the case of homogeneous semiconductors with well known properties. In this case electrical characterization certainly is less elaborate. Besides the fact that many modern materials exhibit unknown charge carrier densities and effective masses, most of them are inhomogeneous. In such systems grain boundaries may limit electrical transport and in consequence extremely low mobilities are observed by electrical characterization. Examples are polycrystalline semiconductors, percolated networks of carbon nanotubes, and many organic substances where hopping transport is the most limiting factor. Here, THz phase analysis may open up new opportunities for measuring the Drude momentum relaxation rate.

V. CONCLUSIONS

We developed a technique for directly measuring the momentum relaxation time of charge carriers using the optical phase of a differential THz signal. The modulation of the structure under investigation provides a phase signal which is proportional to $\tau$. In contrast to other spectroscopical approaches, our technique is advantageous particularly in the Hagen-Rubens range. Thus, we are confident that our technique will provide new insights into many modern materials where scattering occurs on ultrafast time scales and electronic properties are not directly accessible by electronic means.

Acknowledgments

This work is partially supported by the Deutsche Forschungsgemeinschaft (DFG) through the Nanosystems Initiative Munich (NIM), and by the Deutsche Forschungsgemeinschaft (DFG), contract Ke 516/1-1. The authors acknowledge technical support by A. Guggenmos and S. Niedermayer.

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