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Relativistic Doppler reflection as a probe for the initial relaxation of a non-equilibrium electron-hole plasma in silicon

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Abstract. This paper reviews the status of investigations of the relativistic Doppler reflection of a broadband terahertz pulse at a counter-propagating plasma front of photo-excited charge carriers in undoped silicon. When a THz pulse with 20-THz bandwidth impinges onto a moving plasma front with a carrier density in the range of $10^{19}$ cm$^{-3}$, one observes a spectral up-shift, which is, however, much less pronounced than expected from simulations assuming a Drude plasma characterized by a single carrier relaxation time $\tau$ of the order of 15-100 fs. Qualitative agreement between simulations and experiments can be achieved if $\tau$ is chosen to be less than 5 fs. In order to explore carrier relaxation in more detail, optical-pump/THz-probe experiments in the conventional co-propagation geometry were performed. If the pump-probe delay is long enough for monitoring of the equilibrium value of the scattering time, $\tau$ ranges from 200 fs at low carrier density to 20 fs in the $10^{19}$-cm$^{-3}$ density range. For small (sub-picosecond) pump-probe delay, the data reveal a significantly faster scattering, which slows down during energy relaxation of the charge carriers.

1. Introduction

Early on in the history of relativity, the Doppler frequency shift arising upon reflection of an electromagnetic wave at a fast-moving mirror was investigated as an illustrative example of relativistic effects [1]. If the mirror is a moving plasma front, the same frequency shift is expected independent of whether the charge carriers participate in the motion or are immobile [2, 3]. In the latter case, no energy can be transferred from the mirror to the wave, while a transfer of kinetic energy from the mirror occurs if the particles, which constitute the plasma, propagate with the front. A situation with a plasma front at relativistic speed and static or slow-moving charge carriers can be obtained by the optical inter-band excitation of a semiconductor material, which has to be a volume material in order to enable the propagation effect, while the carriers themselves can be spatially confined (e.g., in quantum structures) or unconfined.

In this contribution, we investigate the relativistic Doppler reflection of THz pulses at counter-propagating photo-ionization fronts in undoped silicon at room temperature (see Fig. 1). Unlike in gaseous media, an amplified laser pulse can readily generate a charge-carrier plasma in silicon which has the density and homogeneity to act as an effective mirror for a focused THz pulse. The relatively weak absorption of the pump light by the silicon sample resulting from the indirect character of the inter-band transitions enables a sufficient long interaction time of the plasma front and the THz pulse of about 140 fs [4]. The reflection coefficient strongly depends on the carrier density. Below a critical density $N_{crt}$ (under-dense plasma), only a small portion of the incoming wave is reflected. Good reflec-
tivity requires over-dense-plasma conditions, which are achieved if all angular frequencies \( \omega_i \) of the incoming THz pulse are below a critical angular frequency, i.e. \( \omega_i \leq \omega_{cr} \). The critical angular frequency is connected with the angular plasma frequency at the critical density according to \( \omega_{cr}^2 = \omega_{pl}^2 / \Gamma = (N_e e^2 / (\varepsilon_r \varepsilon_0 m_{eff} \Gamma) \). Here, \( \Gamma \) is the up-shift factor which determines how the angular frequency \( \omega_i \) of the input wave transforms upon reflection in the laboratory frame, \( \omega_r = \Gamma \omega_i \). For an estimate of the expected up-shift, one can assume a lossless (collisionless) plasma. In this case, \( \Gamma \) only depends on the ratio \( \beta = U/c \) of the velocity \( U \) of the ionization front and the velocity \( c \) of the wave in silicon propagating in the opposite direction: \( \Gamma = (1 + \beta) / (1 − \beta) \). For the material parameters of Si relevant for our experiments (photoexcitation at a wavelength of 775 nm), the up-shift factor has a value of about eight [4]. Absorption losses and pump-pulse depletion effects reduce the up-shift factor [4]. In the experiments, we achieve a plasma frequency \( \omega_{pl}/2\pi \) of up to 30 THz (half of this value in the experiments shown in Fig. 2) [5, 6], which suggests that over-dense conditions can be reached for frequencies \( \omega_i/2\pi \) of the incoming waves of up to 11 THz (5.5 THz in Fig. 2), and up-shifted signals should be observable above 80 THz (above 40 THz in the data of Fig. 2).

2. Measurement of the relativistic Doppler up-shift in highly photo-excited silicon

The Doppler up-shift is measured with optical pulses of a 1-kHz Ti:Al\(_2\)O\(_3\) amplifier laser whose 150-fs pulses are spectrally broadened in an Ar-gas-filled hollow-core fibre and compressed with chirped mirrors. Most of the 30-fs-long pulses is used to excite the silicon sample, the remainder is employed for the generation of THz pulses by two-colour photo-ionization of air [7-11], respectively their detection by sum-frequency generation in a 500-µm-thick ZnTe crystal (sensitivity up to 150 THz) [5, 6, 11, 12].

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**Figure 1.** Relativistic Doppler reflection in the over-dense case. A THz wave propagating to the right encounters an ionization front moving to the left. Upon interaction, a substantial part of the energy of the THz pulse is lost to the plasma by excitation of a stationary, so-called streaming magnetic field. Pump depletion and carrier relaxation (which would dampen the streaming magnetic field) are neglected. From [4].

**Figure 2.** Measured frequency up-shift occurring around time delay zero, when the THz pulse encounters a counter-propagating plasma front. The spectrum at negative time delay (pump pulse has not arrived yet) is that of the unperturbed THz pulse. For positive time delay beyond 100 fs, when the pump pulse is completely absorbed, the THz pulse is strongly attenuated by the static plasma. From [5].

Figure 2 displays the key result of the reflection measurements. If the arrival of the THz pulse, which has measureable signal up to 25 THz (see Fig. 2 for delay times below -100 fs), is properly timed such that it encounters the moving plasma front, one observes newly created frequency components up to 28 THz. Compared with Drude model calculations [4, 5], this degree of up-shift and the intensity of the up-shifted signal are lower than expected, even if one assumes a value of the
scattering time \( \tau \) as low as 15 fs [5]. A reduction of the up-shift factor and of the signal intensity can be qualitatively explained with a scattering time of less than 5 fs (data to be published in [13]).

In the Drude model, \( \tau \) is a momentum relaxation time to which elastic and inelastic scattering events of the charge carriers contribute. Phonon emission in silicon is not effective [6], hence carrier-carrier scattering is the main candidate for momentum relaxation. In the literature, little is found on Drude scattering in silicon at the relevant excitation densities and for non-equilibrium conditions. It is worth remarking that the scattering time as estimated above for our single-crystalline sample appears to approach that of amorphous silicon [14]. The experiments described in the following are carried out in order to shed more light on the carrier dynamics.

3. Measurement of the equilibrium Drude scattering times of highly photo-excited silicon

The measurements are performed as optical-pump/THz-probe measurements, but now in co-propagation geometry (see Fig. 3). The data are again analyzed with a Drude ansatz. High-quality fits with the Drude model necessitate that the Drude response has been measured over a broad frequency range around \( \omega = \omega_{pl} \), the region where the spectral changes of the Drude response are most pronounced. For this reason, we employ in this study white-light THz/mid-IR pulses with a bandwidth of more than 150 THz combined with a suitable sum-frequency detection technique, which via analysis of the spectra allows us to reliably extract scattering times even shorter than 10 fs [6, 7].

Our study reveals that \( \tau \) exhibits a time evolution during the initial energy relaxation of the photo-excited charge carriers. Equilibrium values are measured if the time delay between the maxima of the pump and probe pulses is 1 ps and longer. Figure 4 displays THz/mid-IR reflection data for a delay time of 1 ps. The spectra of the reflected pulses can be reproduced very well by a Drude analysis if care is taken to treat the depth-distributed character of the reflection process properly [6]. For an excitation density ranging from 0.35 to \( 2.1 \times 10^{19} \) electron/hole pairs per cm\(^3\), and a plasma frequency ranging from 15 to 31 THz (effective reduced mass of Si: 0.15 m\(_0\)), the scattering time is found to decrease from 31 fs at the lowest density to 20 fs at the highest.

Figure 4 shows these data (open circles) together with additional results (full circles) for low excitation densities obtained by us with conventional time-domain THz spectroscopy (0.1-3 THz). The experiments cover the density regime up to \( 3.3 \times 10^{17} \) cm\(^{-3}\), the pump-probe delay in these measurement is 5 ps [6].\( \tau \) is found to decrease from 200 fs at low excitation density to 90 fs at \( 3.3 \times 10^{17} \) cm\(^{-3}\).

The curves in Fig. 4 are the results of generalized Drude model calculations based on the theoretical determination of scattering rates in a thermalized plasma as outlined in [6, 15]. The solid curve traces the excitation density dependence of the total scattering rate. The theoretical prediction reproduces the measured data both with regard to the density trend and the absolute values well. According to theory, carrier momentum scattering at low densities is dominated by energy relaxation of the carriers via deformation-potential interaction with acoustic phonons. Interaction with optical phonons is weak as expected. With increasing density, the acoustic relaxation channel becomes still more effective because of the increasing carrier velocity, before phase-space constraints reduce its effectiveness. At a density of \( 5 \times 10^{17} \) cm\(^{-3}\) (mean carrier distance of about 10-15 nm), a cross-over occurs above which electron-hole scattering is the dominant momentum relaxation process.
carrier momentum relaxation of less than 5 fs at our excitation densities in the 10^14 photo-excited charge carriers in silicon. The degree of up-shift is strongly limited by extremely fast.

We have demonstrated a frequency up-shift of THz pulses reflected off a counter-propagating front of

lines show the contributions from various relaxation channels. From [6].

Figure 4. Measured (symbols) and calculated (full line) Drude scattering time of a thermalized electron-hole plasma as a function of electron/hole pair density. The symbols in colours other than blue are from the literature, the reader is referred to [6]. The broken lines show the contributions from various relaxation channels. From [6].

4. Subpicosecond evolution of the Drude scattering time for highly photo-excited silicon

If one extends the THz/mid-IR reflection measurements to shorter delay times, and performs a Drude-model analysis of the resultant reflection spectra, one obtains the data shown in Fig. 5. Compared with the build-up of the carrier density, τ exhibits a delayed rise with a time constant of 450 fs. This value is close to that attributed to the initial energy relaxation of the charge carriers [16].

5. Conclusion

We have demonstrated a frequency up-shift of THz pulses reflected off a counter-propagating front of photo-excited charge carriers in silicon. The degree of up-shift is strongly limited by extremely fast carrier momentum relaxation of less than 5 fs at our excitation densities in the 10^{19}-cm^{-3} range. Additional measurements show that the scattering time gradually increases to a density-dependent value of 20-30 fs as the charge carriers thermalize first among each other and then with the lattice. Practical exploitation of the Doppler up-shift will require much reduced electron-hole scattering.

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