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Ultrafast nonlinear carrier dynamics in doped semiconductors in high THz fields

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Abstract: THz frequency saturable absorption and intervalley carrier scattering in n-type semiconductors were observed using intensity-dependent transmission experiments as well as THz-pump—THz probe spectroscopy with ultrabroadband probe pulses.

Time-resolved optical and near-IR spectroscopy has provided extensive information about carrier dynamics in semiconductors. Recently, the availability of strong picosecond sources with field strengths of several hundred kV/cm has opened the way to the study of ultrafast carrier dynamics in the THz range [1, 2]. New applications may arise from these studies, in which the elementary scattering processes in high fields can be observed in the absence of detrimental effects of above-bandgap laser excitations.

A direct consequence of high field transport is THz saturable absorption in n-type bulk semiconductors with moderate doping. Here, carrier mobility is modulated by nonlinear electron transport caused by the THz electric field, thus affecting the conductivity of the sample. The application of an external (THz) electric field leads to acceleration of carriers in the lowest-energy valley of the conduction band of an n-type semiconductor. At high-momentum states the valley nonparabolicity becomes pronounced [3], which leads to an increase in the effective mass and thus to a reduction of the mobility μ_e, consequently leading to a reduced dielectric loss in the THz range. At high enough electric fields, intervalley scattering is possible, leading to electron transfer into the satellite valleys with reduced curvature [4]. This results in an even smaller electron mobility compared to that of the conduction band minimum, and consequently lower THz dielectric loss. Since the absorption from free carrier absorption in n-type semiconductors is typically strongest at THz frequencies, the mobility change leads to a drastic increase in transmission, that may even exceed 100 percent in strong fields.

In our experiments, we generated intense single-cycle THz pulses by tilted pulse-front optical rectification [6] in a lithium niobate based on of 80 fs, 800 nm, 3 mJ laser pulses produced by a 1-kHz repetition rate Ti:Sapphire amplifier. The THz pulses were collimated and then refocused onto a sample point using a set of off-axis paraboloidal mirrors. A pair of broadband wiregrid polarizers was used to controllably attenuate the THz pulses. After propagation through the sample point, the transmitted THz pulses were detected coherently using standard free-space electro-optic sampling. Nonlinear THz transmission through n-type semiconductors GaAs, GaP, and Ge with different thickness and doping levels was studied at room temperature.

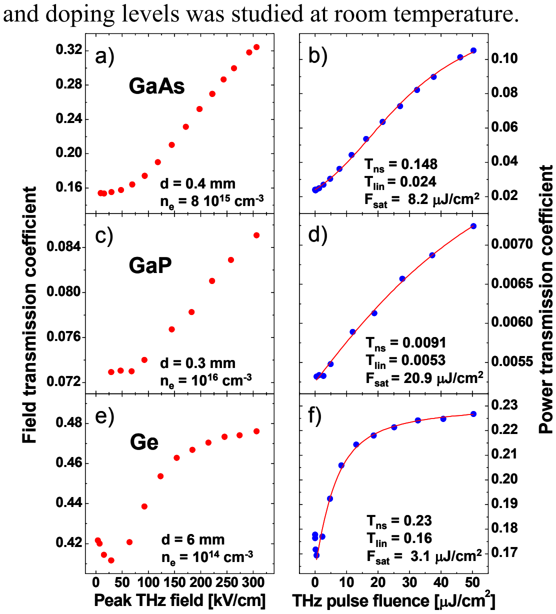

Fig. 1. Symbols: field transmission coefficient as a function of peak THz pulse field, and power transmission coefficient as a function of THz pulse fluence for GaAs (a-b), GaP (c-d), and Ge (e-f). Solid lines: fit with saturable transmission function Eq. (1).

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Fig. 1 shows the field and power transmission coefficients as a function of THz pump field and fluence respectively in GaAs (d = 0.4 mm, $n_e = 8\times10^{15}$ cm^{-3}), GaP (d = 0.3 mm, $n_e = 10^{16}$cm^{-3}) and Ge (d = 6 mm, $n_e = 10^{14}$ cm^{-3}). The field and power transmission coefficients were obtained by integrating either the modulus or the square, respectively, of the THz fields transmitted through the sample, and dividing them by reference values recorded without the sample in the beam path. In all our samples we observed increased transmission at higher pump fluences. In particular, we observed a nearly five-fold increase in power transmission coefficient for GaAs sample in the full THz pulse fluence range of our experiments. The solid lines in Figs. 2 (b,d,f) are fits to measured data using a phenomenological saturable power transmission function, defined after Ref. [5] as

$$T(F_p) = T_{ns}\frac{\ln\left[1 + T_{lin}/T_{ns}(e^{F_p/F_{sat}} - 1)\right]}{F_p/F_{sat}} \qquad (1)$$

where T_{lin} and T_{ns} are linear and non-saturable power transmission coefficients, F_p is the pump fluence, and F_{sat} is the saturation fluence. Using fits with Eq. 1 we were able to extract the saturable absorber parameters for our semiconductor samples. These parameters, namely linear and non-saturable transmission, and saturation fluence are indicated in Fig. 1. In particular, the saturation fluence F_{sat} was found to be 8.2 µJ/cm^2 for GaAs, 20.9 µJ/cm^2 for GaP, and 3.1 µJ/cm^2 for Ge, which is of the same order of magnitude as values usually found for SESAMs at optical wavelengths [7, 8].

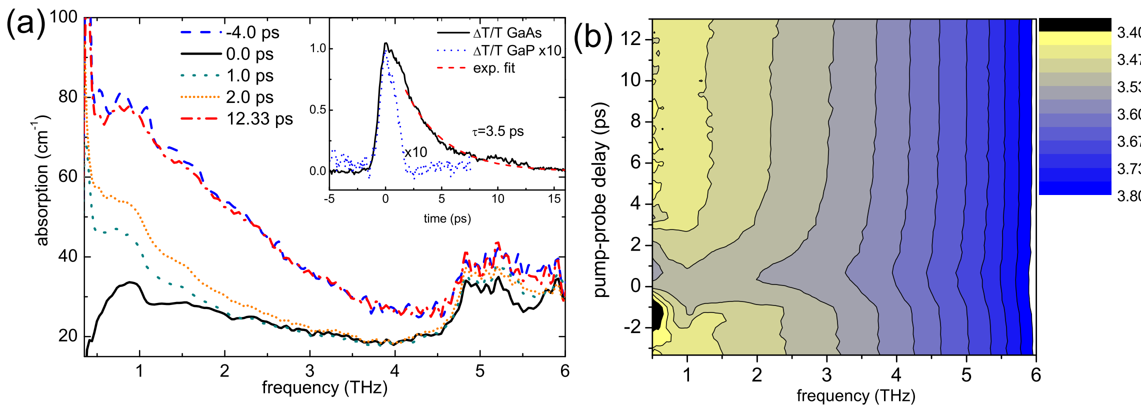

Fig. 2. (a) Absorption spectra of n-type GaAs at 100kV/cm pump for varying time delays. Inset: peak transmission change for n-type GaAs and GaP. (b) time resolved measurement of the GaAs index of refraction as function of frequency and time delay

In order to study the dynamics of the saturation process, we carried out THz-pump—THz-probe experiments, where a time-delayed second THz pulse with broad bandwidth was used as a probe. This pulse was generated by two-color mixing of a 25 fs laser pulse in air plasma [9]. Our experimental technique allows us to spectrally resolve the free-carrier absorption peak up to the onset of the LO phonon signature in the probe spectrum. Figure 2a shows the absorption spectrum of the probe pulses after propagating through the n-type GaAs sample at different times before and after the excitation. At temporal overlap a strong bleaching of the dominating Drude absorption in the range between 1 and 3 THz was observed. The absorption recovers with a time constant of 3.5 ps. In case of GaP, a 10 times smaller modulation was observed with an instantaneous absorption recovery essentially lasting only for the duration of the pump pulse. Further, we observe a 5% increase in refractive index in GaAs excited by a THz pulse (Fig 2b). This leads to complex propagation dynamics in the bulk material for strong field propagation, resulting in THz pulse shortening [10].

Optimized THz SESAMs based on nonlinear carrier effects could potentially be used with the THz sources like quantum cascade lasers or quasi-cw free electron lasers, enabling mode-locking in order to obtain high-energy ultrashort THz pulses.

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