High Power Terahertz Sources for Nonlinear Spectroscopy of Direct Bandgap Semiconductors

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Abstract: Nonlinear dynamics of free-carriers in direct bandgap semiconductors at terahertz (THz) frequencies is studied using high power, few-cycle pulses. The physical mechanisms that give rise to such dynamics will be discussed in details.

Ultrafast nonlinear processes have been extensively explored in the visible and near infrared frequency range, thanks to the availability of ultrashort pulses delivered by mode-locked lasers. Here, the combination of high excitation intensities together with a very fine temporal resolution have shed new light on diverse aspects of condensed-matter dynamics [1]. On the other hand, this kind of phenomena has remained relatively unexplored in the THz spectral region (typically 0.1-10 THz), mainly because of the lack of sources delivering high-energy, few-cycle THz pulses. Nowadays, this kind of sources is becoming available [2,3], thus opening the route towards the understanding of new aspects of radiation-matter interaction.

Nonlinear interactions at THz frequencies possess interesting properties and peculiarities: on one side, in this spectral range one can observe an intermediate regime in which both electronic and ionic motions contribute to the nonlinear dielectric function of a material. On the other side, the very low energy per photon associated to this radiation allows to neglect multiphoton interactions in semiconductors, thus opening up the possibility of observing drift-velocity-based nonlinearities owing to free carriers in this type of systems.

While these processes were studied in the past using THz pulses with a duration of several tens of nanoseconds [4,5], the new-generation few-cycle THz sources allow now to explore their ultrafast dynamics in the picosecond domain [6].

In our experiments, high-energy THz pulses are generated by optical rectification in a large aperture (75 mm diameter) ZnTe single crystal wafer [2]. The Ti:sapphire laser beam lines of the Advanced Laser Light Source (ALLS, INRS) used in these experiments operates at a repetition rate of 100 Hz and provides 800 nm, ~ 40 fs laser pulses with energies as high as 70 mJ per pulse.

The experimental setup comprises three main parts: (i) a THz generation chamber held under vacuum ($<10^{-6}$ torr), (ii) an 800-nm probe line in air, and (iii) a dry-nitrogen-purged THz propagation line. The THz emitter consists of a 1-mm-thick (110) ZnTe single crystal wafer with a diameter of 75 mm. To minimize saturation and to avoid damage to the crystal surface, the 800nm beam is spatially expanded to about 36 cm^2 at the surface of the ZnTe emitter. Any remaining 800nm light transmitted through the ZnTe wafer is blocked using a black polyethylene absorber, which is transparent to THz radiation.

Five off-axis parabolic mirrors are used after the THz generating crystal to redirect the THz wave to the detector, with 3 focus positions: (i) at the exit of the vacuum chamber, (ii) at the sample position for spectroscopy, and (iii) at the ZnTe detector crystal for EO sampling. A chopper positioned at the first focus allows modulation of the THz beam for lock-in detection. Two 4” diameter wire-grid polarizers (Microtech Instruments model G30L) are used to control the intensity and the polarization of the THz beam.

To detect the THz pulse waveform, we use free-space electro-optic (EO) sampling in a second, 0.5mm thick, (110) ZnTe crystal [7]. A lock-in amplifier connected to the output of a pair of photodiodes and referenced to the chopper is used to acquire the THz waveforms.

The source provides picosecond THz pulses in the frequency range of 0.1–3 THz with μJ-level energies. These pulses are focused by an off-axis parabolic mirror down to a spot size of 1.6 mm (full-width at $1/e^2$ of the maximum) at the second focal position (i.e. the sample position). We used a $BaSrTiO_3$ (BST) pyroelectric infrared camera (Electrophysics model PV320-L2V) to image the THz spot at the focus. This camera operates with an internal 10Hz chopper and has a 320x240 pixel imaging array with a pixel spacing of 48.5 μm. The THz beam profile is found to be well fitted by a Gaussian shape. For measuring the THz energy, we used a calibrated pyroelectric energy detector (Coherent Molectron J4–05).

Using the experimental set-up presented above, we performed an open aperture Z-scan measurement [8]. Whenever nonlinear absorption takes place, the intensity of the transmitted THz beam depends on the spot size of the beam at the sample position. Clearly, THz intensity is maximum at the focus and is progressively reduced as the sample is moved away from the focus (in both directions). This method is widely used in

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multiphoton absorption studies [9,10], and has proven to be effective even for the characterization of saturable absorbers.

The sample we used for this experiment was a thin film of a heavily doped direct bandgap semiconductor, namely Indium Gallium Arsenide. Specifically, it consists of a 500nm thick n-type $In_{0.53}Ga_{0.47}As$ epilayer with a doping concentration of about 2×10^{18} cm^{-3} grown by metal-oxide chemical vapor deposition on a lattice-matched, 0.5 mm thick semi-insulating InP substrate. At low excitation levels, the sample transmits about 3% of the incident THz energy. We have confirmed that this strong THz absorption is mainly due to the high conductivity of the epilayer, since measurements of the InP substrate alone have shown an overall THz transmission (including absorption and reflection losses) of about 60%.

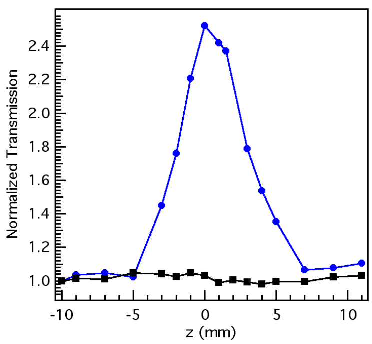

Figure 1. Energy Z-scan traces of the InGaAs epilayer on an InP substrate (blue curve), and of the substrate alone (black curve).

In particular, Fig. 1 shows how the energy transmission through the sample varies as a function of the z position along the scan, with an incident THz pulse energy of 0.8 μJ (peak THz field ~ 200 kV/cm). A clear bleaching of the absorption is evidenced at the focus of the terahertz beam. The same scan performed on the substrate alone (black curve in Fig. 1) does not return any evidence of the bleaching.

This remarkable phenomenon is attributed to terahertz-electric-field-driven scattering of electrons into satellite valleys of the conduction band. In these satellite valleys, electrons acquire a significantly higher effective mass, reducing the conductivity of the sample and thus increasing the terahertz transmission.

In order to confirm and better understand the results above, we have set up a more advanced THz-pump / THz-probe characterization experiment. The experimental setup in this case includes an additional THz probe beam, which is generated by a 10×10×0.5 mm ZnTe crystal placed after the gold off-axis parabolic mirror prior to the sample position (second THz focus). This THz probe beam was superimposed on to the THz pump beam at its focus. An additional black polyethylene sheet, which is transparent to the THz radiation, was placed before the sample to block any remaining 800-nm light transmitted through the ZnTe crystal used to generate the THz probe. In order to avoid crosstalk between the two THz beams, a chopper was inserted in the THz probe line, and the two beams transmitted through the sample were geometrically separated. In addition, we placed a metallic mask to block the remaining THz pump beam after the sample position.

Fig. 2 shows the normalized transmission of the main peak of the THz probe pulse as a function of the pump-probe delay time. The two curves show the transmission change for a probing polarization perpendicular to the pump beam and the same measurement performed for the parallel configuration, when both beams have the same polarization.

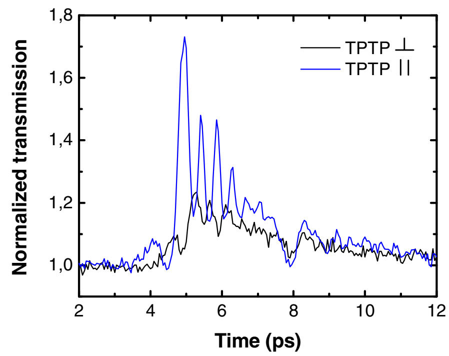

Figure 2. Normalized transmission of the peak THz probe electric field for horizontal THz pump polarization – horizontal THz probe polarization (parallel TPTP, ||) and for horizontal THz pump polarization – vertical THz probe polarization (perpendicular TPTP, ⊥).

If one performs instead the same experiment using parallel polarizations (blue curve in Fig. 2), the transmission enhancement is more evident and fast oscillations appear, which seem to follow the shape of the THz pump waveform. By performing the experiment with crossed polarizations, a significant enhancement in the transmission is observed. The dynamics of this process is in excellent agreement with our previous Z-scan results and can be attributed once again to electric-field-driven intervalley scattering, as we will elucidate through our theoretical considerations.

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