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Nonlinear Terahertz Spectroscopy in Solids with Single-Cycle Terahertz Pulses

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Abstract. We present novel generation methods of intense terahertz single cycle pulses. The Cherenkov scheme with tilted wave-front technique in the $LiNbO_3$ crystal gives us the maximum electric field larger than 1 MV/cm, which ponderomotive energy is as large as 10 eV. The ponderomotive energy is strong enough to ionize bound electronic states in solids such as donors and accepters and easy to induce nonlinear optical effects in solids.

1 Introduction

Thanks to developments in terahertz (THz) light sources and detection methods, especially with terahertz time-domain spectroscopy (THz-TDS), we can now easily obtain complex refractive indexes or dielectric constants of many different materials in the THz frequency region [1]. With photon energies of a few meV, THz wave may resonantly drive electronic or vibrational states in molecules, nanostructures, and condensed matters. The THz responses of these excitations reveal the dynamics of excitonic polarization or lattice vibrational anharmonicity. Since THz-TDS is based on femtosecond pulse laser technology, it is suitable for time-resolved measurements, and enables the measurement to investigate the dynamical interplay of photo-excited excitons and unbound electron-hole pairs, which would have been impossible ten years ago: Time-resolved THz-TDS has been applied successfully to clarify non-equilibrium dynamics by monitoring internal transitions of excitons and Drude dispersion of electron-hole plasma, both existing in the THz frequency region [2-4]. Recent development of strong THz pulse generation has a possibility to open a new category of non-linear phenomena and unexplored non-linear effects in semiconductors, where the THz pulse can be thought of as an ultrafast electric or magnetic-field pulse-switch operating at femtosecond to picosecond time-scale that is much faster than that achieved through conventional electronics.

2 Intense single-cycle THz pulse generation

The technological innovations of the last decade have made it possible for intense THz pulse (peak field strength: >1 MV/cm) to be generated by bench-top equipment. The most efficient generation method involves using a femtosecond laser to obtain Cherenkov radiation from lithium niobate (LN) [5-9]. LN has an intrinsically large second-order nonlinear susceptibility $\chi^{(2)}$, and is thus expected to generate intense THz pulse through the optical rectification (OR) process with broad-band femtosecond laser pulses. However, since the phase velocity of the THz frequency region differs significantly from the group velocity of the visible region, this causes a phase mismatch in the

coaxial alignment so that only non-coaxial Cherenkov light is generated, making it impossible to obtain an intense THz light source. Since Hebling *et al.* proposed generating THz pulse with high efficiency using the tilted-pulse-front scheme so that the nonlinear polarization phase matches the terahertz light phase along the direction of propagation of the THz pulse [5], the technique has been rapidly developing. Recently we have developed a system where THz maximum electric field strength reaches as high as 1.2 MV/cm with energy conversion efficiency of ~ 10^{-3} [9] (Fig.1).

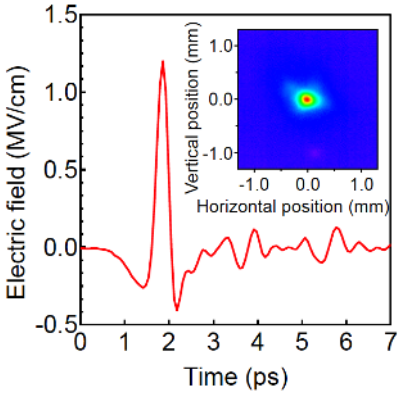

Fig. 1. Time profile of the electric field of the THz pulse generated by tilted wave-front technique with Ti: sapphire regenerative amplifier (4 mJ, 100 fs). The inset shows THz intensity image measured at the focused point after the last off-axis parabolic mirror (PM3 in Fig. 1) [7].

3 Terahertz nonlinear spectroscopy

3.1 Carrier multiplication in GaAs MQWs

We have applied the intense THz pulses to study carrier multiplication in nominally undoped GaAs multiple quantum wells (MQWs) with intense THz pulses. Carrier multiplication was induced by a nearly half−cycle THz pulse lasting 1 ps and having a maximum peak electric field amplitude of 1.05 MV/cm. The THz pulses were focused onto a GaAs MQWs sample that was mounted on the cold finger of a helium−flow cryostat. The electric field was perpendicular to the stacking direction and along the (100) direction of the sample. Under THz irradiation, we clearly observed near−infrared luminescence centered around 1.55 eV from MQWs at 10 K, even though the central photon energy of the pulse (~ 4 meV) is about 390 times lower than the luminescence photon energy, and the luminescence intensity drastically decreases as the electric field decreases. Since luminescence is proportional to the created e−h pair density, the generated carrier density can be estimated by comparing the luminescence measurement with optical pulse excitation measurements. The carrier density is extremely nonlinear and roughly proportional to the electric field to the eighth power. The minimum and maximum electric fields of 0.47 and 1.05 MV/cm give the carrier-density change over three orders of magnitude. This THz−field dependence is in agreement with phenomenological theory based on an impact ionization model including electron motion in k−space, suggesting that the carriers coherently driven by a strong electric field can efficiently gain enough kinetic energy to induce series of impact ionizations [10].

3.2 Nonlinear carrier transport in Graphene

Time-resolved high-field carrier transport in graphene is studied using terahertz-pump optical-probe technique. The sample is CVD growth graphene, which is transferred onto a SiO_2 substrate from the original metal substrate. Optical transmission of the graphene sample is 97% at 800 nm, which

shows single-layer nature of the sample. An intense THz pulse with air plasma method is irradiated on the sample so that we can apply a 300kV/cm pulse electric field within less than 200 fs. We used a 50-fs pulse duration 800-nm probe beam to measure the change of absorption that corresponds to the change of population occupation at the wavelength. Figure 2 shows the differential optical density as a function of time delay. The differential optical densities become negative right after the excitation due to the phase-space filling of electrons and holes accelerated by THz field, and go back to zero within 2 ps. To explain the experimental results, we performed Monte Carlo simulations with the semi-classical Boltzmann equation including carrier-carrier scattering, optical phonon scattering, impact ionization and Auger recombination. The experimental results show good agreement with our numerical results, suggesting nonlinear carrier transport at the initial stage of terahertz excitation and the possibility of higher harmonic generation.

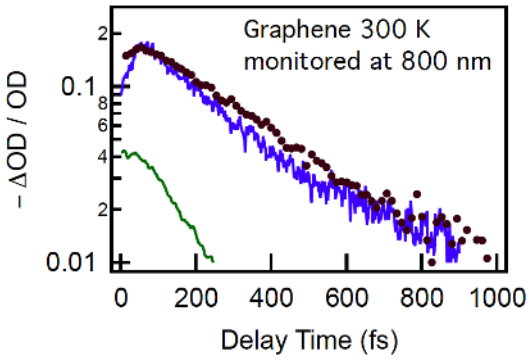

Fig. 2. Time-development of differential optical density induced by single-cycle THz pulse with peak electric field of 300 kV/cm. Experimental data is shown by dots. Simulation results are shown by blue and green lines: with and without impact ionization and Auger recombination, respectively.

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