

## Ultrafast two-dimensional THz spectroscopy of graphene

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**Abstract.** With two-dimensional THz spectroscopy the dynamics of low-energy carriers in graphene is determined. Both intra- and interband absorption contribute to the observed ultrafast pump-probe signals.

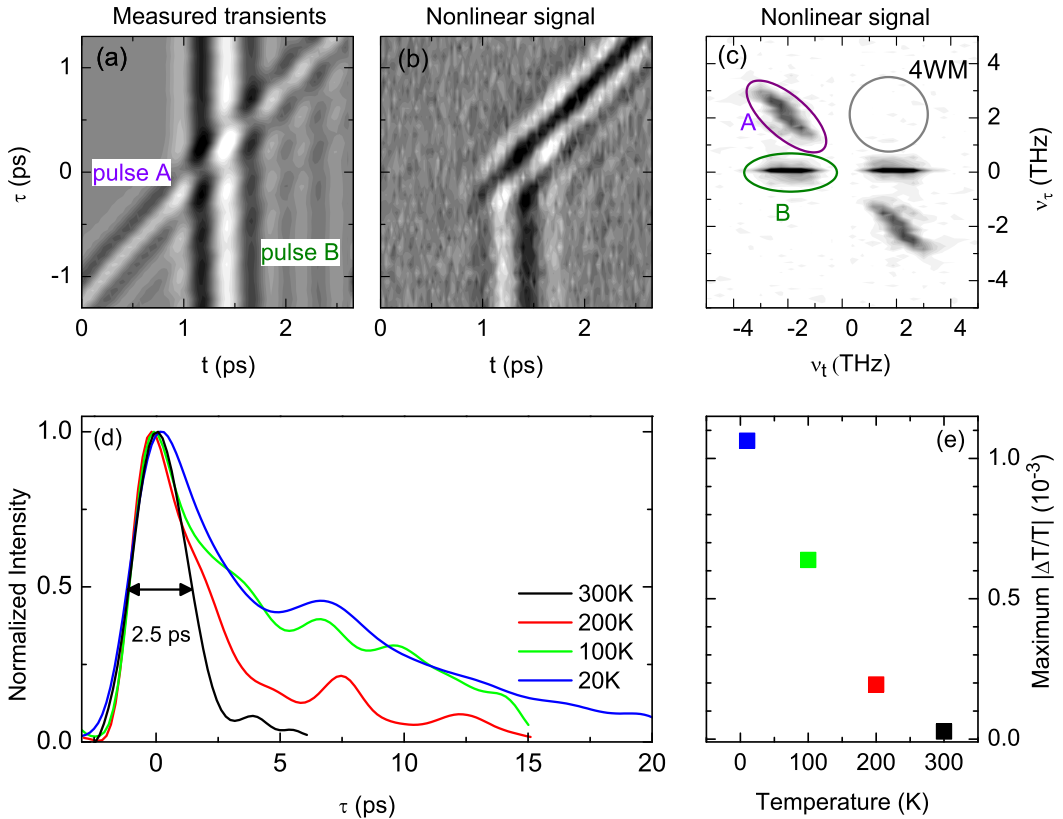
Graphene, or a single layer of graphite, has a peculiar bandstructure, which allows for the study of massless charge carriers with a velocity independent of their energy. Charge transport in graphene has been widely explored but there is still much to be learned about carrier dynamics on the ultrafast time scale, in particular at very small energies close to the K and K' (Dirac) points in k-space. So far, there have been ultrafast pump-probe studies of graphene using higher energies to excite off-equilibrium carriers [1,2]. Thermalization occurred within 250 fs due to Coulomb and optical phonon scattering. Later cooling occurs from interactions with optical and finally with acoustic phonons. Carrier dynamics at energies close to the Dirac point ( $E = 0$ ), well below the optical phonon energy, should be quite different, as shown by recent mid-infrared measurements [3].

Here we present the first nonlinear THz study of graphene. We do this using amplitude- and phase-resolved two-dimensional spectroscopy at 2 THz[4]. We demonstrate that the third-order response is dominated by pump-probe signals. These reveal an induced absorption that decays with a time constant as short as 2.5 ps depending on the sample temperature. At lower temperatures an additional slower component ( $\approx 10$  ps) is present. We attribute this to a combination of intra- and interband absorption followed by thermalization. The absence of any other nonlinear signal points to a very short decoherence time of THz excitations in graphene.

The 2D THz measurements are based on the method presented in Ref. [4]. A phase-locked pair of pulses centred at 2 THz propagate parallel to one another and are focused onto the graphene sample where they spatially overlap. The total transmitted electric field is measured in (real) time  $t$  with electro-optic sampling, for each delay  $\tau$  between the two pulses [Fig. 1(a)]. Using two choppers, the two THz pulses are also measured individually, so that they can be subtracted from the measured total signal to isolate the nonlinear part (Fig. 1(b)). The measured electric field is proportional to the current in the sample. As explained in Ref. [4], the various contributions to the nonlinear signal can be separated in two-dimensional frequency space, and then transformed back to the time domain. We used a 40 layer C-face epitaxial graphene sample (from Graphene Works).

We measured the 2 THz pump-probe signals from the graphene sample at  $T = 20$  K. Pulse A had a field strength of 5 kV/cm and pulse B was 8 kV/cm. The pump-probe signal was  $\pi$  phase shifted with respect to the driving electric field (pulse B) indicating an induced transient absorption. The enhanced absorption as a function of pump-probe delay  $\tau$  was measured at several temperatures (pump-probe signal B, Fig. 1d). With increasing temperature, the picosecond decay of the enhanced absorption becomes faster and the maximum in the differential transmission decreases drastically from 0.1 to 0.003%, see Fig. 1(e).

Both the induced absorption and the absence of the photon-echo signals are explained by a model using a pseudopotential bandstructure for graphene including the electron-light coupling via the vector potential. The model further includes radiative coupling. This model reproduces very well our exper-



**Fig. 1.** (a) The total electric field transmitted through the graphene sample is plotted as a function of the (real) time  $t$  and the delay  $\tau$  between the two THz pulses. (b) The total nonlinear THz signal. (c) The 2D Fourier transform of the nonlinear signal allows us to separate out the various nonlinear (third order) contributions. Only pump-probe signals are present, and no four-wave mixing (4WM). (d) Measured pump-probe signals (amplitude averaged over time) at different sample temperatures. (e) Temperature dependence of the amplitude of the pump-probe signal.

imental results without having to introduce extremely fast decoherence processes or other scattering mechanisms. Both observed effects have their origin in the huge transition dipole moments. Because of this, multi-photon transitions, which lead to induced absorption, are stronger than one-photon transitions which would lead to bleaching. Since the Rabi frequencies are different at each point in  $k$ -space, the superposition of all photon echo components leads to a negligible total signal.

## References

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