Anisotropic ultrafast optical response of terahertz pumped graphene

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We have measured the ultrafast anisotropic optical response of highly doped graphene to an intense single cycle terahertz pulse. The temporal shape of the signal is similar to that of the pump terahertz electric field modulus. It grows with increasing carrier density and demonstrates a specific nonlinear dependence on the electric field strength. To describe the signal, we have developed a theoretical model that is based on the energy and momentum balance equations and takes into account optical phonons of graphene and substrate. According to the theory, the anisotropic response is caused by the displacement of the electronic momentum distribution from zero momentum induced by the pump electric field in combination with polarization dependence of the matrix elements of interband optical transitions.

Due to the peculiar electronic band structure of graphene, the field-induced motion of electrons was predicted to be strongly nonlinear in this material. High nonlinearity together with the unique electronic and optical properties make graphene a prospective material for photonic and optoelectronic applications. In the light of this perspective, nonlinear optical phenomena in graphene are actively studied. Among them are harmonic generation, saturable absorption, self-phase modulation, and four-wave mixing. In the optical range the frequency of light is higher than the electron-electron scattering rate, so the resulting "coherent" electronic response is determined by the properties of single-electron band structure. In the THz range another limiting case is realized — the characteristic time of electron-electron scattering processes is shorter than the period of the light wave. The energy imparted by the electric field to electrons is quickly redistributed heating the electron gas, while the electron-phonon collisions cool and decelerate the gas. The concept of "incoherent" nonlinearity that appears due to the change of electron gas conductivity upon heating was employed recently to explain highly effective generation of THz harmonics in graphene.

The routine technique used to evaluate the optical nonlinearity of graphene is the spectral analysis of light transmitted through the sample in search of harmonics of the pump frequency radiated by the nonlinear current. In the present work we employ an alternative approach by using an optical probe to detect the transient THz field-induced shift of electron momentum distribution (note that such shift can induce the optical 2-n harmonic generation, as was recently observed). In graphene, due to the specific polarization dependence of matrix elements of interband transitions in combination with Pauli blocking, an anisotropy of electronic distribution implies an anisotropy of infrared optical conductivity, which can be measured by detecting depolarization of probe light reflected from the sample.

We measure the ultrafast anisotropic optical response of graphene to intense THz pulses and show that though the corresponding signal is rather weak, it can be reliably detected for heavily doped graphene and contains specific nonlinear features. To interpret the signal, we develop a model based on the Boltzmann kinetic equation solved in the hydrodynamic approximation.

The sample used in our experiments was a sheet of single-layer CVD graphene on the SiO2/Si substrate (the thickness of SiO2 was 300 nm). Four indium contacts were attached to the sample in order to apply gate voltage and to measure the resistance of the graphene layer. Nearly single-cycle THz pulses with a duration of about 1 ps were generated in a lithium niobate crystal in the process of optical rectification of femtosecond laser pulses with tilted fronts (see, e.g., Ref. 23 for details). The THz generation stage was fed by 50 fs laser pulses at 800 nm, 1.2 mJ per pulse at 1 kHz repetition rate. THz radiation was focused by a parabolic mirror so that the peak electric field of the THz pulses incident on the sample was ∼ 400 kV/cm (denoted below as Emax). The waveform of the pulses was characterized by means of electro-optic detection in a 0.15 mm thick (110)-cut ZnTe crystal. In the experiments we detected transient anisotropic changes of reflectance of the sample caused by the pump THz pulses. The probe 50 fs pulses at 800 nm were polarized before the sample at an angle of ∼ 7° relative to the vertical polarization of pump THz pulses. Both pump and probe beams were incident onto the sample at an angle of ∼ 7°. Upon reflection from the sample excited by THz radiation the probe pulses experienced a small rotation of polarization, which was detected by measuring the densities of two orthogonal polarization components of the reflected probe beam Ixy and Ixx using a Wollaston prism and a pair of photodiodes.

The quantity

\[ F = 1 - \frac{I_{xy}}{I_{xx}} \]  (1)

as a function of the probe pulse delay time is referred to as anisotropy signal or anisotropic response.

The anisotropic response of the sample induced by the pump THz pulse is shown in Fig. 1 for the peak values of the electric field of Emax and Emax/2. As soon as the signal from the regions of SiO2/Si substrate not covered by graphene was below the noise level, we concluded that the source of the observed anisotropy signal was the graphene layer itself. Fig. 1 also shows the temporal profile of the electric field of the pump THz pulse. In order to ensure linearity of the electrooptic detection in the ZnTe crystal we attenuated pump THz beam power by a factor of ∼ 400 using a variable metallic fil-

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To record the electric field profile the filter was “closed” so that the THz beam passed through the fused silica plate covered by the thickest metallic layer. The sample response at $E_{\text{max}}/2$ was measured with the “opened” filter as the pump THz pulses passed only through fused silica (the 2 mm thick fused silica plate reduces the THz field by a factor of $\approx 2$). Finally, in order to detect the anisotropic response induced by the strongest pump electric field available ($E_{\text{max}}$) we removed the variable filter so that the THz radiation traveled to the sample only through air. As soon as the fused silica plate causes a large additional retardation of THz pulses the anisotropic response measured at $E_{\text{max}}$ was time-shifted so that it matched the signal detected at $E_{\text{max}}/2$ in time domain. As follows from Fig. 1 the third peak in the signal measured at $E_{\text{max}}$ occurs earlier than the corresponding peaks in the signal detected at $E_{\text{max}}/2$ and in the electric field profile $|E(t)|$. This effect is due to group velocity dispersion in the fused silica plate that leads to a small lengthening of the pulse and of the signal.

To estimate the doping level of graphene, we measured the sample resistance as a function of gate voltage $V_g$ (shown in the inset to Fig. 2). For highly doped graphene this dependence can be approximated by the formula $R(V_g) \approx R_0 + A/V_g - V_{\text{CNP}} + B/V_g - V_{\text{CNP}}^{1/2}$, which takes into account short- and long-range impurities$^{2}$ (the first two terms), and the contribution of optical phonons of graphene$^{26}$ and substrate$^{27-29}$ (the 3-rd term). Based on the charge neutrality point location $V_{\text{CNP}}$, we estimated the hole density in our sample as $n \approx 2 \times 10^{11}$ cm$^{-2}$, which corresponds to the Fermi level $E_F \approx -500$ meV. (Below we assume positive $E_F$ for clarity, because our model is particle-hole symmetric). In order to investigate the dependence of the anisotropy signal on the doping level, we repeated the experiment with applied gate voltages of $V_g = \pm 30$ V, as shown in Fig. 2. The negative (positive) gate voltage in our case increases (decreases) the charge carrier concentration, which leads to an increase (decrease) of the anisotropy signal.

Time evolution of the electron gas in highly doped graphene under intense THz field $E(t)$ is dominated by its intraband dynamics$^{30,31}$, described in terms of two separate momentum distribution functions $f_\gamma(k,t)$ for electrons in conduction and valence bands. Time evolution of these functions is described by the semiclassical Boltzmann kinetic equation

$$\frac{\partial f_\gamma}{\partial t} = -\frac{eE}{\hbar} \cdot \frac{\partial f_\gamma}{\partial k} + \frac{(f_\gamma + n \cdot (nf_\gamma) - f_\gamma)}{\tau_{\text{imp}}(k)} + \Gamma_\gamma(1 - f_\gamma) - \Gamma_\gamma^{\text{out}} f_\gamma + \left(\frac{\partial f_\gamma}{\partial t}\right)_{\text{ee}}.$$  \hspace{1cm} (2)

The terms in the right hand side describe, respectively, electron acceleration by the applied electric field, elastic collisions with impurities$^{32}$ with momentum-dependent scattering time $\tau_{\text{imp}}(k)$, electron-phonon and electron-electron collisions. $\Gamma_\gamma^{\text{in}}(k,t)$ and $\Gamma_\gamma^{\text{out}}(k,t)$ are the rates of electron scattering into the $k\gamma$ state and out of this state$^{33-35}$.

Interband dynamics of the electron gas induced by the THz field in our case should be slow with respect to fast electron-electron collisions, which thermalize the electron gas on a time scale less than $30$ fs$^{36,37}$. Consequently, $f_\gamma(k,t)$ can be taken in the form of the “hydrodynamic” distribution function

$$f_\gamma^{\text{drift}}(k,t) = \left\{\exp\left[\frac{E_\gamma - \hbar k \cdot V(t) - \mu(t)}{T(t)}\right] + 1\right\}^{-1},$$  \hspace{1cm} (3)

which is formed due to electron-electron collisions with conservation of total energy, momentum and particle number$^{32,38}$. Here $E_\gamma = \gamma \hbar \omega k$ are the single-particle energies, while temperature $T$, chemical potential $\mu$, and drift velocity $V$ are slowly varying functions of time. Fig. 3 shows examples of $f_\gamma^{\text{drift}}$ for $n$-doped graphene.
A nonzero drift velocity \( V_x \) (we take \( E \) and \( V \) along the \( x \) axis) makes the distribution functions\(^{\text{(3)}}\) angular anisotropic at the probe pulse wave vector modulus \( |k| = k_{\text{pr}} = \omega_{\text{pr}} / v_p \). In combination with the angular dependence of the matrix elements of interband transitions\(^{\text{(35)}}\), it leads to the anisotropy of the optical conductivity tensor at \( \omega = \omega_{\text{pr}} \):

\[
\begin{align*}
\sigma_{xx} / \sigma_{yy} &= \frac{e^2}{4\pi\hbar} \frac{2\pi}{\int_{0}^{\phi} \left( \sin^2 \phi / \cos^2 \phi \right) (f_- - f_+) |k = k_{\text{pr}} |. (4)
\end{align*}
\]

The small difference between \( \sigma_{xx} \) and \( \sigma_{yy} \) manifests itself in the reflectances \( R_{x,y} \) for the \( x \)- and \( y \)-polarized probe pulses at normal incidence. Defining the optical contrast of graphene\(^{\text{(3)}}\) as

\[
F = \frac{R_x - R_y}{R_x} \approx C = \frac{\sigma_{yy} - \sigma_{xx}}{e^2 / 4\hbar} \frac{2\pi}{\int_{0}^{\phi} \cos 2\phi (f_- - f_+) |k = k_{\text{pr}} |. (5)
\]

For graphene on Si covered by the 300nm-thick SiO\(_2\) layer, the optical contrast at \( \lambda_{\text{pr}} = 800\text{nm} \) is rather small and negative\(^{\text{(40)}}\). Calculating it using the transfer matrix method\(^{\text{41}}\) with taking the universal optical conductivity of graphene \( \sigma = e^2 / 4\hbar \), we get \( C \approx -0.0044 \). In principle, by adjusting the SiO\(_2\) layer thickness\(^{\text{(39)}}\) in order to enhance the visibility of graphene it is possible to increase the observed signal.

The physical origin of the optical anisotropy is illustrated by Fig.\(^{\text{(3)b)}}\). The interband transitions for the \( y \)-polarized light become suppressed with respect to those for the \( x \) polarization due to Pauli blocking, caused by the thermal tail of the displaced distribution function at \( V_x \neq 0 \). The resulting difference of the conductivities, \( \sigma_{yy} < \sigma_{xx} \), leads to a positive anisotropy signal\(^{\text{(5)}}\) since \( C \) is negative. This picture is symmetric when \( V_x \) changes sign, so in the limit of low drift velocity \( F \propto V_x^2 \). Unlike studies with linearly polarized optical pump\(^{\text{35,42-45}}\), where momentum distribution of the photoexcited electrons and holes is highly anisotropic (\( \sim \sin^2 \phi \)) from the very beginning in spite of the zero total momentum, in our case the anisotropy arises as the electron Fermi sphere is displaced from zero momentum by the strong THz field. In both cases the distribution functions acquire nonzero second angular harmonics (\( \sim \cos 2\phi \)), which is necessary for the anisotropy of the optical response.

We solve the Boltzmann equation\(^{\text{(2)}}\) in the hydrodynamic approximation\(^{\text{(3)}}\), using balance equations for the total energy, momentum and particle number of the electron gas similarly to the works on electron transport in graphene in stationary high electric fields\(^{\text{46,47}}\). In these equations, the energy and momentum time derivatives caused by phonons are calculated using full electron-phonon collision integrals. We consider 6 phonon modes: 4 modes of graphene \( \Gamma \) and \( K \) optical phonons\(^{\text{33-35,48}}\) and 2 modes of SiO\(_2\) surface polar phonons\(^{\text{27,29,49}}\). We assume polarization- and momentum-independent phonon occupation numbers \( n_{\mu} = \exp[\hbar \omega_{\mu} / T_{\text{ph}}] - 1 \)\(^{-1}\) dependent on two separate temperatures for graphene optical \( T_{\text{GO}} \) and surface polar \( T_{\text{SPP}} \) phonons. Since hot phonons play an important role in the electron gas dynamics in strong fields\(^{\text{29,35,48}}\), we calculate time evolution of \( T_{\text{GO}} \) and \( T_{\text{SPP}} \) from the energy balance for the phonon gas which exchanges energy with electrons and additionally loses energy via phonon decay with the characteristic times \( \tau_{\text{ph}} \approx 2 \mu s\(^{\text{36,47}}\) and \( \tau_{\text{SPP}} \approx 1 \mu s\(^{\text{49}}\)\). For the scattering time on long-range impurities, relevant for graphene on a SiO\(_2\) substrate\(^{\text{26}}\), we take \( \tau_{\text{imp}}(k) \approx s / k \), where \( s \) can be related to the low-field carrier mobility \( \mu_c = 2e\nu Fs / \hbar \), which is about 1000cm\(^2\)/V\( \cdot \)s in our sample. In numerical calculations we use the THz electric field strength of \( \sim 70\text{kV/cm} \) that is several times lower than the incident field \( E_{\text{max}} \). This reduction of the field acting on graphene electrons is caused by the destructive interference of the incident THz wave with the wave reflected from the underlying \( p \)-doped Si substrate with the reflectivity \( R_{\text{THz}} \approx 0.6 \div 0.7\(^{\text{42}}\).

Typical calculation results for \( F \) are shown in Fig.\(^{\text{(1)}}\). We take the doping level \( \mu = 500\text{meV} \) and the values 70kV/cm and 35kV/cm for the peak strength of the electric field acting on graphene electrons. One can see that our numerical model reproduces the magnitude of \( F \) and the general similarity of \( F(t) \) and \( |E(t)| \) relatively well at realistic parameters. Fig.\(^{\text{(2)}}\) shows the examples of how the calculated \( F \) depends on the doping level: as in the experiment, \( F \) grows with the carrier concentration, although the theory predicts this growth to be highly nonlinear, especially at strong THz fields.

The detected anisotropic response of graphene, however, contains specific features, that our model is not able to reproduce. First, the third peak behaves differently with respect to the first two ones: its growth upon doubling the electric field is considerably higher (\( \sim 4.5 \)) than for the first two peaks (\( \sim 2.5 \)), and is underestimated by the theory. This anomalous behavior at the end of the THz pulse can be caused by the heating of the electron gas in graphene, the temperature of which is expected to be maximal after the action of the peak electric field, as shown by the calculated profiles of \( T(t) \) in Fig.\(^{\text{(3)a)}}\).
It should also be noted that the rise time of the third peak is the shortest of all three peaks (~ 50 fs) and is comparable with the characteristic time of electron-electron interactions in graphene, so in this regime our hydrodynamic approximation \[ \text{[3]} \] can miss some features of the coherent collisionless dynamics of electrons driven by the high electric field.

One more interesting property of the anisotropic response is the sharp bend or kink observed in the signal at \( \sim \) 40 fs, after the THz electric field has reached the maximum and just began to decrease. It is visible in the signals recorded at both field strengths and is marked by arrows in the inset to Fig. 1. One can see that due to this kink the form of the anisotropy signal differs considerably from the THz waveform. The latter evolves smoothly similar to a sine wave, while the former signal differs considerably from the THz waveform. The latter evolves smoothly similar to a sine wave, while the former signal differs considerably from the THz waveform. The latter evolves smoothly similar to a sine wave, while the former signal differs considerably from the THz waveform.

Finally, in view of the long-standing search of the nonlinear current response of graphene in the THz range\(^{3,15,16}\), we calculate the electric current density \( j \) (Fig. 3b). The electric current demonstrates strong nonlinearities: first, its peak values change insignificantly when the electric field is doubled, that can be considered as a manifestation of the electric current saturation\(^{29,47}\), and, second, \( j \) becomes lower at the same field strength near the end of the pulse, which can be attributed to the influence of electron gas heating.

In conclusion, we have measured the ultrafast anisotropic optical response of highly doped graphene under intense THz excitation and developed the model of temporal dynamics of the momentum distribution functions based on the Boltzmann equation, solved in the hydrodynamic approximation. Theoretical calculations provide good description of the general shape and magnitude of the anisotropy signal at realistic parameters, and also predict strong nonlinearities of the THz-field induced electric current. We demonstrate that the anisotropic optical response measured with subcycle temporal resolution contains information on the ultrafast dynamics of the electron gas, its heating, isotropization and concomitant nonlinearities. Our work links the areas of nonlinear THz electrodynamics of graphene, ultrafast pseudospin dynamics of Dirac electrons\(^{29-42}\), and strong-current graphene physics\(^{42}\), thereby providing a new tool for studying high-field phenomena in graphene in the far-IR and THz range.

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