Fluence and wavelength dependent ultrafast differential transmission dynamics in graphene

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Keywords: graphene, ultrafast pump-probe spectroscopy, differential transmission dynamics

Abstract
We performed degenerate pump-probe transmission measurements of graphene supported on glass for a range of pump fluences that enable us to observe both positive and negative differential transmission dynamics. Our results show that at an intermediate pump fluence, a transition from negative to positive response occurs, the differential transmission dynamics is an order of magnitude faster than at higher and lower pump fluences. This effect can be explained by equal contributions of inter- and intraband transitions with opposite signs to the transient optical conductivity of graphene at an intermediate pump fluence. Moreover, the intermediate threshold pump fluence is shown to increase with decreasing probe energy, which is in agreement with the theoretical model. Furthermore, we show that the relaxation time of the electronic temperature increases monotonically over the range of fluences studied. In perspective, this work is of importance to graphene-based opto-electronic applications such as light modulators.

1. Introduction
Graphene is a single layer material of carbon atoms with high carrier mobility and unique opto-electronic properties which make it useful for a wide range of applications [1–6]. The use of the advantageous properties of graphene in application to opto-electronic devices inevitably entails the generation of hot carriers with energies significantly exceeding the Fermi energy [1, 2, 4]. Many groups have studied graphene related relaxation dynamics using various techniques such as measurements of photocurrent, ultrafast pump-probe spectroscopy and time resolved Raman spectroscopy [7–9]. Ultrafast pump-probe spectroscopy has been particularly fruitful in providing valuable insights into electron-electron, electron-phonon and phonon-phonon interactions [9–16]. Typically, after electrons and holes are excited into a non-thermal distribution by an ultrafast laser pulse, they thermalize into a Fermi–Dirac distribution through Coulomb interactions in tens of femtoseconds [17]. The cooling of the hot thermal population of carriers occurs through the emission of optical phonons. When the temperatures of the electron and phonon systems equilibrate, the hot phonon bottleneck occurs, which significantly lessens the rate of cooling [14, 18, 19]. Subsequent cooling primarily arises from the hot optical phonons undergoing anharmonic decay into acoustic phonons. However, in the case of supported graphene, direct coupling of the charge carriers to surface phonons in polar substrates is a possible cooling channel [20–23]. As predicted by theory and measured in experiments, the time constant of hot optical phonon decay in graphene is of the order of a few picoseconds [8, 13–16, 18, 24].

Previous measurements of photo-induced ultrafast dynamics in graphene by pump-probe spectroscopy have yielded a diversity of differential transmission and reflection dynamics [25, 26]. It has been shown that differential transmission dynamics (DTD) depends on carrier scattering time, doping, Fermi level and probe energy [10, 25–28]. Several different regimes for DTD have been observed: (i) fully positive DTD [10, 14, 27, 29–34], (ii) fully
negative DTD [11, 35–37], and (iii) a complex DTD consisting of a positive peak near zero time delay followed by a negative slow recovery tail [10, 25–27, 29, 38, 39].

In this paper CVD graphene on a soda-lime glass substrate is studied using degenerate pump-probe spectroscopy at the wavelengths of 775, 800, 825 and 850 nm. In our measurements, we examine the transition from complex DTD to fully positive DTD as a function of pump fluence. At low pump fluences, after the initial positive spike arising from the bleaching of the interband transitions, the differential transmission crosses zero and its slow recovery tail is negative due to primarily intraband absorption processes. A paper by F. Kadi et al [40] explains the observation of transient negative differential transmission, i.e. a zero-crossing of the differential transmission at about 300 fs, using a microscopic treatment of intraband absorption in graphene. At high pump fluences, the slow recovery tail of the differential transmission is positive due to predominant interband transitions. In contrast, at intermediate fluences, we observed the characteristic relaxation time of differential transmission to be an order of magnitude smaller as compared to the results observed at low and high fluences. While such an effect could be intuitively inferred from known theoretical models (for example, see A. Tomadin et al [41]), no experimental demonstration, to the best of our knowledge, has been reported. This effect can be explained by equal magnitude and opposite sign contributions of intra- and interband transitions to the differential optical conductivity of graphene. Thus, this work provides a novel experimental observation of an order of magnitude faster relaxation of differential transmission at the intermediate pump fluences, where the intraband and interband contributions are equal in magnitude and opposite in sign. It is important to note, as shown later in this paper, that by fitting the experimental data with our model, we find the relaxation time of the electron temperature to increase monotonically over the range of pump fluences used in this study. Comparison with the measured relaxation times of the differential transmission implies that pump fluence greatly modifies time-dependent optical properties, while the electron and phonon relaxation processes remain unaffected.

2. Results and discussion

2.1. Experimental data

Single layer CVD graphene (Graphene Supermarket) was transferred onto a soda-lime glass substrate by a wet transfer method (see Methods). The quality and uniformity of the transferred graphene layer was confirmed by Raman spectroscopy using a Thermo Scientific DXR Raman Microscope. Figure 1 shows Raman spectrum averaged over ten different points on the sample with positions of G and 2D peaks being 1591.6 ± 1.5 cm⁻¹ and 2681 ± 3 cm⁻¹, respectively. The Fermi level (n-type) was estimated to be 306 meV [42]. Ultrafast degenerate pump-probe transmission measurements were carried out using a Ti:Sapphire oscillator producing 120 fs pulses at a 76MHz repetition rate. Pump and probe wavelengths were initially both set to 800 nm (1.55 eV) and subsequently changed to 775 nm (1.60 eV), 825 nm (1.51 eV) and 850 nm (1.46 eV). Both beams were focused onto the specimen with spot diameters of 80 μm and 50 μm for pump and probe, respectively. The pump beam was chopped using a SR540 optical chopper operating at about 4 kHz. The differential transmission signal was detected using a Lock-In amplifier.

Figures 2(a) and 3(a), (c), (e) show the pump fluence dependent DTD at 800 nm, 775 nm, 825 nm and 850 nm, respectively. After the initial positive spike in the differential transmission due to Pauli blocking of the interband transitions upon photoexcitation, the slow recovery differential transmission dynamics can be either positive or negative depending on the pump fluence. At high fluences (above 20 μJ cm⁻², 25 μJ cm⁻², 33 μJ cm⁻² and 39 μJ cm⁻² for 775 nm, 800 nm, 825 nm and 850 nm, respectively), the differential transmission is fully positive due to predominant contribution of interband transitions [40], while for lower fluences (below 20 μJ cm⁻², 25 μJ cm⁻², 33 μJ cm⁻² and 39 μJ cm⁻² for 775 nm, 800 nm, 825 nm and 850 nm, respectively), the differential transmission is fully negative. This finding is in good agreement with the predictions of the theoretical models.
25 μJ cm$^{-2}$, 33 μJ cm$^{-2}$ and 39 μJ cm$^{-2}$ for 775 nm, 800 nm, 825 nm and 850 nm, respectively). The differential transmission becomes negative past ≈0.5 ps due to greater contribution of intraband transitions. At intermediate fluences (see data at the fluences of 20 μJ cm$^{-2}$ in figure 2, 25 μJ cm$^{-2}$ in figure 2b, 33 μJ cm$^{-2}$ in figure 3c and 39 μJ cm$^{-2}$ in figure 3e) for 775 nm, 800 nm, 825 nm and 850 nm, respectively), where a transition from intra- to interband dominated response occurs, much faster relaxation dynamics of DT is observed as compared to low and high fluences (indicated schematically by arrows in figures 2(b) and 3(a), (c), (e)).

In order to characterize the relaxation dynamics of the DT, we fit a bi-exponential decay convoluted with Gaussian pump and probe pulses to our data using the following expression [43]:

$$ D_1 \exp \left( -\frac{t - t_0}{\tau_1} \right) \left[ 1 - \text{erf} \left( \frac{w}{2\tau_1} - \frac{t - t_0}{w} \right) \right] + D_2 \exp \left( -\frac{t - t_0}{\tau_2} \right) \left[ 1 - \text{erf} \left( \frac{w}{2\tau_2} - \frac{t - t_0}{w} \right) \right] , $$

(1)

where $D_1$, $D_2$, $t_0$, $\tau_1$, $\tau_2$ are the fitting parameters. $w = \sigma \sqrt{2}$ is the width of the autocorrelation function with $\sigma$ being the width of the Gaussian pump and probe pulses. The time constants $\tau_1$ and $\tau_2$ refer to the fast and slow decay processes, respectively. We focus mostly on the slow relaxation time constant $\tau_2$, as it largely characterizes the total relaxation times of DT at most fluences.

The fitting results using expression (1) are shown in figures 4(a) and 3(b), (d), (f) for 800 nm, 775 nm, 825 nm and 850 nm, respectively. The amplitude of the slower exponent corresponding to the relaxation time
constant $\tau_2$ changes sign from negative to positive when going from lower to higher fluences, which explains the instability of the biexponential fit at the intermediate pump fluences. As the pump fluences approach the intermediate values, at which the aforementioned transition occurs, the double exponential character of the
response vanishes and instead becomes a single exponential due to the fact that the amplitude of the slower component is essentially zero at the intermediate fluences. As a result, the characteristic relaxation time of the differential transmission drops by an order of magnitude at the intermediate fluences. The onset of the single exponential behavior at the intermediate fluences is seen more clearly on the log scale in figures 2(c), (d). In the next section we show that an order of magnitude faster DTD observed at the intermediate fluences is explained well by a model that accounts for the contributions of both intra- and interband transitions to DTD in graphene, which depend on the temperature of the excited electrons.

2.2. Modelling
Applying the optical boundary conditions at the air/graphene/substrate interfaces, the optical transmission \( t(\omega) \) through the single layer graphene on a substrate normalized to the transmission through the substrate can be expressed as [44, 45]

\[
t(\omega) = \frac{1}{|1 + \sigma(\omega)Z_\text{o}/(1 + n_s)|^2},
\]

where \( \sigma(\omega) \) is the optical conductivity of graphene, \( Z_\text{o} \) is the vacuum impedance and \( n_s \) is the refraction index of the substrate. Since \( |\sigma(\omega)|Z_\text{o}/(1 + n_s) \ll 1 \), the contribution of the imaginary part of the optical conductivity is negligible.

The optical conductivity of single layer graphene is given by [46]

\[
\begin{align*}
\frac{\text{Re}(\sigma(\omega))}{\sigma_Q} &= \frac{4k_B T}{\hbar \pi} \left[ \ln \left(1 + e^{E_F/k_B T}\right) + \ln \left(1 + e^{E_F/k_B T}\right) \right] \frac{\tau}{\omega^2 + 1} \\
&\quad + \frac{1}{2} \left[ \tanh \left( \frac{h\omega - 2E_F}{4k_B T} \right) + \tanh \left( \frac{h\omega - 2E_F}{4k_B T} \right) \right],
\end{align*}
\]

where \( \sigma_Q = e^2/4\hbar \) is the universal quantum conductivity, \( k_B \) is Boltzmann constant, \( T \) is the carrier temperature, \( E_F (E_F^\text{h}) \) is the electron (hole) Fermi level, \( \tau \) is the momentum scattering time. The first term in equation (3) describes a Drude-like intraband contribution to the optical conductivity, whereas the second term arises from interband transitions. The measured differential transmission is proportional to \( -\Delta \left[ \text{Re}(\sigma(\omega))/\sigma_Q \right] \) according to equation (2). Considering high n-type doping \( (E_F^\text{h} \gg 2k_B T) \) and neglecting the contribution from minority carriers (holes), the optical conductivity can be written as

\[
\frac{\text{Re}(\sigma(\omega))}{\sigma_Q} = \frac{4E_F^\text{e}}{\pi \hbar} \frac{\tau}{\omega^2 + 1} + \frac{1}{2} \left[ \tanh \left( \frac{h\omega - 2E_F^\text{e}}{4k_B T} \right) \right].
\]

The Fermi level \( E_F^\text{e} \) and temperature \( T \) of electrons in equation (4) are in general time dependent quantities.
The dynamics of the electron temperature $T$ is modelled by a bi-exponential decay (see Methods):

$$T(t) = T_0 + \frac{(T_1 - T_0)}{2}[(1 - k)e^{-t/\tau_{\text{ep}} - \rho} + (1 + k)e^{-t/\tau_{\text{op}}}], \quad (5)$$

where $T_0$ is the equilibrium lattice temperature, $T_1$ is the temperature upon photo-excitation, $k, \tau_{\text{ep}}, \tau_{\text{op}}$ serve as fitting parameters. For graphene, the heat capacity of phonons is much larger than the heat capacity of electrons. This indicates that $\tau_{\text{ep}}$ can be thought of as the electron-phonon thermalization time constant, while $\tau_{\text{op}}$ is the hot phonon relaxation time constant (see Methods).

The initial electron temperature $T_1$ and the Fermi level $E_F$ are estimated from energy conservation [37, 46]:

$$U_c(T_0, N) + \Delta Q = U_c(T, N + n) + U_b(T, p), \quad (6)$$

where

$$U_{c,h} = \frac{4k_B^2 T^3}{\pi^2 h^2} F_1 \frac{E_F^h}{k_B T}, \quad (7)$$

$$n, p = \frac{2k_B^2 T^2}{\pi^2 h^2} F_2 \frac{E_F^h}{k_B T} \quad (8)$$

$F_1$ and $F_2$ are the first- and second-order Fermi integrals, respectively, $v = 10^6$ m/s is the Fermi velocity in graphene, $N$ is the electron concentration at equilibrium. The time dynamics of the Fermi level $E_F$ is taken into account by solving equation (8) with carriers recombining exponentially $n = N + n_0 e^{-t/\tau_{\text{op}}}$. The effective recombination time $\tau_{\text{re}} = 1.3$ ps [37] was fixed for all fits.

2.3. Discussion

Figure 4(b) shows the pump fluence dependence of the phenomenological optical phonon relaxation time constant $\tau_{\text{op}}$. Clearly, $\tau_{\text{op}}$ does not show a negative peak at the intermediate pump fluences and increases monotonically. This indicates that the observed fast relaxation dynamics of DT is not due to a change in the physical relaxation mechanism, but due to equal magnitude contributions with opposite signs of inter- and intraband transitions to the differential optical conductivity of graphene at the intermediate pump fluences. The monotonic changes in $\tau_{\text{op}}$ as a function of pump fluence occur as a result of ignoring the temperature dependence of the electron-phonon coupling constant and the electron and phonon heat capacities. While microscopic theories describing ultrafast dynamics in graphene without invoking any fitting parameters have been developed [40, 47], the present approach is sufficient to demonstrate that an order of magnitude faster DTD at the intermediate pump fluences is well described by the model applied here accounting for intra- and interband transitions. Furthermore, an order of magnitude difference between the characteristic relaxation time of DT at the intermediate pump fluences $\tau_1$ and the characteristic relaxation time of the electron temperature $\tau_{\text{op}}$ indicates significant changes in the optical properties without altering the electron and phonon relaxation mechanisms.

Figure 5 shows the dependence of the threshold pump fluence on the wavelength as measured in our experiments. At the threshold pump fluence, a perfect balance between intra- and interband transitions leads to an order of magnitude faster DT relaxation. Beyond this fluence interband transitions dominate the signal. As seen from figure 5, the threshold pump fluence as well as the corresponding excited carrier density are greater for lower probe energies. This behavior can be qualitatively understood based on equation (4) describing the contributions of intra- and interband transitions to the optical conductivity of graphene. The first Drude-like intraband term increases with decreasing probe energies. In other words, intraband absorption is more significant at lower probe energies. The only way to make the interband term have greater contribution is to reach higher electron temperatures so that the changes in the optical conductivity due to that interband term become greater. Hence, one needs to excite more electrons in order to reach higher temperatures and, consequently, overcome intraband absorption.

3. Conclusion

We have investigated ultrafast differential transmission dynamics in supported graphene on a soda-lime glass substrate using transmission pump-probe spectroscopy. The range of applied pump fluences distinctly shows different regimes of the DTD. The main result is an experimental measurement of an order of magnitude faster relaxation of the DT at the intermediate pump fluences as compared to low and high fluences. Such unconventional differential transmission relaxation times at the intermediate pump fluences are explained well within the model of intra- and interband transitions contributing with opposite signs to the differential optical conductivity of graphene. Fitting the measured differential transmission spectra with our model, which is generally based on previous models [16, 37, 39, 46], reveals
that $\tau_{op}$ characterizing the relaxation of the electronic temperature does not show a negative peak with respect to the pump fluence, while the differential transmission relaxation time has a minimum at the intermediate pump fluences. Approximately equal contributions in magnitude and opposite in sign of inter- and intraband processes to the differential optical conductivity at the intermediate pump fluences result in an order of magnitude faster DTD as compared to high and low fluences. The value of the threshold pump fluence and the critical excited electron density, at which the faster relaxations are observed, increase with decreasing probe energies as the intraband absorption contributes more at lower probe energies. In perspective, this work demonstrates an order of magnitude faster DTD as well as the diversity of the differential transmission dynamics that can be controlled by pump fluence, which is of interest to graphene-based opto-electronic devices such as light modulators.

**Acknowledgments**

The authors acknowledge the ARO for financial support under Contract No. W911NF-14-1-0290. Portions of this work were completed using the shared resources of the Vanderbilt Institute of Nanoscale Science and Engineering (VINSE) core laboratories.

**Appendix. Methods**

**A.1. Sample preparation**

Single layer CVD graphene on a copper foil was obtained from Graphene Supermarket. Graphene was transferred onto a soda-lime glass substrate using the wet transfer method. First, poly-methyl methacrylate (PMMA) was spun coated on top of the graphene/copper sample at 4500 rpm. Then, the sample was left in Iron Chloride ($\text{FeCl}_3$) for 24 hours to etch away copper. After the copper foil is etched, the PMMA/graphene layer was cleaned of the copper etchant residue by consequently dipping it in Deionized Water (DI) and then leaving it in DI for 24 hours. Next, the PMMA/graphene sample was mechanically transferred onto a soda-lime glass substrate. At the last step, we removed PMMA layer by dissolving it in acetone followed by an isopropanol (IPA) rinse.

**A.2. Ultrafast pump-probe spectroscopy**

Ultrafast pump-probe spectroscopy measurements were carried out in transmission geometry using Ti:Sapphire Coherent Mira 900 oscillator producing 120 fs pulses at 76 MHz repetition rate. Pump and probe wavelengths were set to 800 nm (1.55 eV). Both beams were focused onto the specimen with spot diameters of 80 $\mu$m and 50 $\mu$m for pump and probe, respectively. Pump and probe beams were cross-polarized for improved detection. The pump beam was chopped using a SR540 optical chopper operating at 4 kHz, which served as a reference the lock-in amplifier.

**A.3. Model of electron temperature**

Upon photo-excitation, systems of electrons and optical phonons obey the rate equations, where the changes in the temperature of acoustic phonons is neglected.
\[ T = \alpha (T_{op} - T), \quad T(0) = T_0, \]
\[ T_{op} = \beta (T_{op} + \gamma (T_0 - T_{op})), \quad T_{op}(0) = T_0, \]
\[ T_{ac} = 0, \quad T_{ac}(0) = T_0, \]

where \( \alpha = G_{el,op}/G_{el} \), \( \beta = G_{el,op}/G_{op} \), \( \gamma = G_{op,ac}/G_{op} \) with G and C denoting coupling constants and heat capacities of electrons and phonons. Assuming the coefficients are constant with respect to temperature, this system of differential equations (A.1) can be solved analytically. The solution for the electron temperature is

\[ T(t) = T_0 + \frac{(T_0 - T_0)}{2} \left[ (1 - k)e^{-t/\tau_{ep}} + (1 + k)e^{-t/\tau_{ph}} \right], \]

where \( k = (-\alpha + \beta + \gamma)\sqrt{(-\alpha + \beta + \gamma)^2 + 4\alpha\beta} \), \( \tau_{ep-ph} = 2/\left[ ((\alpha + \beta + \gamma)^2 - 4\alpha\gamma) \right] \) and \( \tau_{ep} = 2/\left[ ((\alpha + \beta + \gamma)^2 - 4\alpha\gamma) \right] \). It is worth noting that since \( \alpha \gg \beta, \gamma \) (due to \( C_{el} \ll C_{op} \)), \( \tau_{ep-ph} \approx 1/\alpha \) and \( \tau_{ep} \approx 1/\gamma \). This indicates that \( \tau_{ep-ph} \) can be thought of as the electron-phonon thermalization time constant, while \( \tau_{ep} \) is hot phonon relaxation time constant.

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**References**

[1] Castro Neto A H, Guinea F, Peres N M R, Novoselov K S and Geim A K 2009 The electronic properties of graphene Rev. Mod. Phys. 81 109–62
[2] Bonaccorso F, Sun Z, Hasan T and Ferrari A C 2010 Graphene photonics and optoelectronics Nat. Photonics 4 611–22
[3] Peres N M R 2010 Colloquium: the transport properties of graphene: an introduction Rev. Mod. Phys. 82 2673–700
[4] Gabor M, Song J C W, Ma Q, Nair N L, Taychatanapat T, Watanabe K, Taniguchi T, Levitov I S and Jarillo-Herrero P 2011 Hot carrier-assisted intrinsic photoresponse in graphene Science 334 648–52
[5] Sun Z, Hasan T, Terris F, Popa D, Privitera G, Wang F, Bonaccorso F, Basko D M and Ferrari A C 2010 Graphene mode-locked ultrafast laser ACS Nano 4 803–10
[6] Liu M, Yin X, Ulín-Avila E, Geng B, Zentgraf T, Ju L, Wang F and Zhang X 2011 A graphene-based broadband optical modulator Nature 474 64–7
[7] Park J, Ahn Y H and Ruiz-Vargas C 2009 Imaging of photocurrent generation and collection in single-layer graphene Nanotechnology 20 1742–6
[8] Kang K, Abdula D, Cahill D G and Shim M 2010 Lifetimes of optical phonons in graphene and graphite by time-resolved incoherent anti-Stokes Raman scattering Phys. Rev. B 81 164505
[9] Breussing M, Kuehn S, Winzer T, Malic E, Milde F, Severin N, Rabe J P, Ropers C, Knorr A and Elsaesser T 2011 Ultrafast nonequilibrium carrier dynamics in a single graphene layer Phys. Rev. B 83 153410
[10] Sun D, Wu Z-K, Devin C, Li X, Berger C, de Heer W A, First P N and Norris T B 2008 Ultrafast relaxation of excited dirac fermions in epitaxial graphene using optical differential transmission spectroscopy Phys. Rev. Lett. 101 157402
[11] Newsom R W, Dean J, Schmidt B and Van Driel H M 2009 Ultrafast carrier kinetics in exfoliated graphene and thin graphite films Opt. Express 17 3236–33
[12] Ruzicka B A, Wang S, Liu J, Loh K P, Wu I J and Zhao H 2012 Spatially resolved pump–probe study of single-layer graphene produced by chemical vapor deposition Opt. Mater. Express 2 708–16
[13] Lin K-C, Li M-Y, Li L J, Ling D C, Chi C C and Chen J-C 2013 Ultrafast dynamics of hot electrons and phonons in chemical vapor deposited graphene J. Appl. Phys. 113 133511
[14] Wang H et al 2010 Ultrafast relaxation dynamics of hot optical phonons in graphene Appl. Phys. Lett. 96 081917
[15] Golla D, Brasington A, LeRoy B J and Sandhu A 2017 Ultrafast relaxation of hot phonons in graphene–hBN heterostructures APL Mater. 5 056101
[16] Malard I M, Mak K F, Neto A H C, Peres N M R and Heinz T F 2013 Observation of intra- and inter-band transitions in the transient optical response of graphene New J. Phys. 15 015009
[17] Johannsen J C et al 2013 Direct view of hot carrier dynamics in graphene Phys. Rev. Lett. 111 027403
[18] Iglesias J M, Martin M J, Pascual E and Rengel R 2016 Hot carrier and hot phonon coupling during ultrafast relaxation of photoexcited electrons in graphene Appl. Phys. Lett. 108 043105
[19] Gao B, Hartland G, Fang T, Kelly M, Jena D, Xing H G and Huang L 2011 Studies of intrinsic hot phonon dynamics in suspended graphene by transient absorption microscopy Nano Lett. 11 3184–9 pMID: 21696177
[20] Low T, Perebeinos V, Kim R, Freitag M and Avouris P 2012 Cooling of photoexcited carriers in graphene by internal and substrate phonons Phys. Rev. B 86 045413
[21] Hwang E H and Das Sarma S 2013 Surface polar optical phonon interaction induced many-body effects and hot-electron relaxation in graphene Phys. Rev. B 87 115432
[22] Ahn S, Hwang E H and Min H 2014 Inelastic carrier lifetime in a coupled graphene/electron-phonon system: role of plasmon–phonon coupling Phys. Rev. B 90 245436
[23] Tielrooij K-J et al 2018 Out-of-plane heat transfer in van der waals stacks through electron–hyperbolic phonon coupling Nat. Nanotechnol. 13 41
[24] Bonini N, Lazzieri M, Marzari N and Mauri F 2007 Phonon anharmonicities in graphite and graphene Phys. Rev. Lett. 99 176802
[25] Shen K, Li H, Ma L-P, Ren W, Chung T-F, Cheng H-M, Chen Y-P and Lai T 2014 Diversity of ultrafast hot-carrier-induced dynamics and striking sub-femtosecond hot-carrier scattering times in graphene Carbon 72 402–9
[26] Guo M, Ma L-P, Ren W and Lai T 2020 Control of the ultrafast photo-electronic dynamics of a chemical-vapor-deposited-grown graphene by ozone oxidation Photonics Res. 8 17–23
Shang J, Luo Z, Cong C, Lin J, Yu T and Gurzadyan G G 2010 Femtosecond uv-pump/visible-probe measurements of carrier dynamics in stacked graphene films Appl. Phys. Lett. 97 163103
Shang J, Yu T, Lin J and Gurzadyan G G 2011 Ultrafast electron-optical phonon scattering and quasiparticle lifetime in CVD-grown graphene ACS Nano 5 3278–83
Shang J, Yu T, Lin J and Gurzadyan G G 2011 Ultrafast electron-optical phonon scattering and quasiparticle lifetime in CVD-grown graphene ACS Nano 5 3278–83 pMID: 21391596
Huang L, Gao B, Hartland G, Kelly M and Xing H 2011 Ultrafast relaxation of hot optical phonons in monolayer and multilayer graphene on different substrates Surf. Sci. 605 1657–61 Graphene Surfaces and Interfaces.
Tani S, Blanchard F and Tanaka K 2012 Ultrafast carrier dynamics in graphene under a high electric field Phys. Rev. Lett. 109 166603
Zhao X, Liu Z-B, Yan W-B, Wu Y, Zhang X-L, Chen Y and Tian J-G 2011 Ultrafast carrier dynamics and saturable absorption of solution-processable few-layered graphene oxide Appl. Phys. Lett. 98 121905
Ruzicka B A, Werake L K, Zhao H, Wang S and Loh K P 2010 Femtosecond pump-probe studies of reduced graphene oxide thin films Appl. Phys. Lett. 96 173106
Dawlaty J M, Shivaraman S, Chandrashekhar M, Rana F and Spencer M G 2008 Measurement of ultrafast carrier dynamics in epitaxial graphene Appl. Phys. Lett. 92 042116
Huang L, Hartland G V, Chu L-Q, Feenstra R M, Lian C, Taby K and Xing H 2010 Ultrafast transient absorption microscopy studies of carrier dynamics in epitaxial graphene Nano Lett. 10 1308–13
George P A, Strait J, Dawlaty J, Shivaraman S, Chandrashekhar M, Rana F and Spencer M G 2008 Ultrafast optical-pump terahertz-probe spectroscopy of the carrier relaxation and recombination dynamics in epitaxial graphene Nano Lett. 8 4248–51
Choi H, Borondics F, Siegel D A, Zhou S Y, Martin M C, Lanzara A and Kaindl R A 2009 Broadband electromagnetic response and ultrafast dynamics of few-layer epitaxial graphene Appl. Phys. Lett. 94 172102
Breusing M, Kuehn S, Winzer T, Malić E, Milde F, Severin N, Rabe J, Ropers C, Knorr A and Elsaesser T 2011 Ultrafast nonequilibrium carrier dynamics in a single graphene layer Phys. Rev. B 83 153410
Chen K et al 2016 Non-destructive measurement of photoexcited carrier transport in graphene with ultrafast grating imaging technique Carbon 107 233–9
Kadi F, Winzer T, Malić E, Knorr A, Göttfert F, Mittendorff M, Winnerl S and Helm M 2014 Microscopic description of intraband absorption in graphene: The occurrence of transient negative differential transmission Phys. Rev. Lett. 113 035502
Tomadin A et al 2018 The ultrafast dynamics and conductivity of photoexcited graphene at different Fermi energies Sci. Adv. 4 eaar5313
Lee J E, Ahn G, Shin J, Lee Y and Ryu S 2012 Optical separation of mechanical strain from charge doping in graphene Nat. Commun. 3 1024
Prasankumar R P and Taylor A J 2016 Optical Techniques for Solid-State Materials Characterization (Boca Raton, FL: CRC Press)
Nuss M C, Goossen K W, Gordon J P, Mankiewich P M, O’Malley M L and Bhattacharjee M 1991 Terahertz time-domain measurement of the conductivity and superconducting band gap in niobium J. Appl. Phys. 70 2238–41
Dawlaty J M, Shivaraman S, Strait J, George P, Chandrashekhar M, Rana F, Spencer M G, Veksler D and Chen Y 2008 Measurement of the optical absorption spectra of epitaxial graphene from terahertz to visible Appl. Phys. Lett. 93 131905
Dani K M, Lee J, Sharma R, Mohite A D, Galande C M, Ajayan P M, Dattelbaum A M, Htoon H, Taylor A J and Prasankumar R P 2012 Intraband conductivity response in graphene observed using ultrafast infrared-pump visible-probe spectroscopy Phys. Rev. B 86 125403
Mihnev M T et al 2016 Microscopic origins of the terahertz carrier relaxation and cooling dynamics in graphene Nat. Commun. 7 11617