AC transport properties of single and bilayer graphene

J.Z. Bernád, U. Zülicke, Klaus G. Ziegler

AC transport properties of single and bilayer graphene

J.Z. Bernád [a], U. Zülicke [a,b,*], K. Ziegler [c]

[a] Institute of Fundamental Sciences and MacDarmid Institute for Advanced Materials and Nanotechnology, Massey University (Manawatu Campus), Private Bag 11 222, Palmerston North 4442, New Zealand
[b] Centre for Theoretical Chemistry and Physics, Massey University (Albany Campus), Private Bag 102904, North Shore MSC, Auckland 0745, New Zealand
[c] Institut für Physik, Universität Augsburg, D-86135 Augsburg, Germany

1. Introduction

Graphene is a single sheet of carbon atoms tightly packed into a two-dimensional (2D) honeycomb lattice. This material has recently become available for experimental study [1,2], and its exotic electronic properties are attracting a lot of interest [3]. In particular, the conical shape of conduction and valence bands, together with the absence of a gap, near the **K** and **K′** points in the Brillouin zone renders graphene an intriguing type of quasi-relativistic condensed-matter system [4]. Recent experiments have verified that the band dispersion of charge carriers in graphene is indeed linear as expected for massless Dirac fermions [5,6]. A multitude of interesting physical effects arising in single-layer and bilayer graphene samples have been discussed [3], both theoretically and experimentally.

In this work, we focus on the ac electric transport properties of graphene, which have been the subject of numerous theoretical (mostly numerical) studies [7–28] and several recent experiments [29–34]. Measurements of quantities related to the optical conductivity are expected to give deeper insight into the electronic properties of graphene samples, making it possible to infer details of their morphology [33,28] and suitability for applications. It is thus important to obtain a clear understanding of the features exhibited in the frequency-dependent conductivity, in particular, their relation to microscopic

* Corresponding author at: Institute of Fundamental Sciences and MacDarmid Institute for Advanced Materials and Nanotechnology, Massey University (Manawatu Campus), Private Bag 11 222, Palmerston North 4442, New Zealand. Tel.: +64 6 356 9099x7259; fax: +64 6 350 5682.
  E-mail address: u.zuelicke@massey.ac.nz (U. Zülicke).

parameters and behavior at finite temperature T. Furthermore, it is advantageous to have mathematical expressions available that can be straightforwardly used for comparison with data. This is the motivation for our study. We have developed a formalism that lends itself for generalization to many situations, in particular, the treatment of inelastic scattering. Extrapolation to zero frequency will enable us to discuss the dc conductivity, shedding new light on the phenomenon of minimal conductivity in graphene. The full range of our results will be presented elsewhere; here we focus on discussing the method and present selected results.

2. Calculation of the conductivity: basic formalism

Our starting point is the familiar [35] Kubo formula

$$\sigma_{\mu\nu}(\omega) = \int_{-\infty}^{0} dt e^{i(\omega - i0^+)t} K_{\mu\nu}, \tag{1}$$

with the kernel

$$K_{\mu\nu} = \frac{ie}{\hbar} \text{Tr}\left\{ e^{-iHt/\hbar} j_\mu e^{iHt/\hbar} [r_\nu, \varrho] \right\}. \tag{2}$$

Here $j_\mu = -e\dot{r}_\mu \equiv (-e/\hbar)i[H, r_\mu]$ is the current operator, and ϱ the density matrix. An alternative expression for the kernel [35],

$$K_{\mu\nu} = \int_0^{1/k_B T} d\lambda \text{Tr}\left\{ e^{-iHt/\hbar} j_\mu e^{iHt/\hbar} e^{-\lambda H} j_\nu e^{-\lambda H} \varrho \right\}, \tag{3}$$

will become particularly useful to enable discussion of the effect of inelastic scattering. When quasiparticle interactions are neglected, it is possible [12] to express the conductivity in terms of matrix elements between eigenstates $|n\rangle$ of the single-particle

Hamiltonian having energy ε_n:

$$\sigma_{\mu\nu}(\omega) = \frac{e^2}{i\hbar} \sum_{n,n'} \frac{\langle n|[H,r_\mu]|n'\rangle \langle n'|[H,r_\nu]|n\rangle}{(\varepsilon_{n'}-\varepsilon_n)(\varepsilon_{n'}-\varepsilon_n+\hbar\omega-i0^+)}[f(\varepsilon_n)-f(\varepsilon_{n'})]. \tag{4}$$

Here $f(\varepsilon) = 1/(1+\exp\{[\varepsilon-\mu]/[k_BT]\}$ denotes the Fermi function, which depends on the chemical potential μ. In the following, we use the expression (4) to derive conductivity formulae applicable to graphene.

2.1. Clean limit: plane-wave representation

Using continuum-model descriptions of the band structure near the **K**- points, single-particle eigenstates of clean graphene systems can be written as a direct product of a plane wave in real space and a $2N$- spinor (the latter subsuming the two sub-lattice and N layer-index degrees of freedom): $|n\rangle = |\mathbf{k}\rangle \otimes |\sigma\rangle_{\mathbf{k}}$. In single-layer graphene, σ distinguishes the two (electron and hole) bands. Note that the spinor wave function depends on wave vector **k**. The current operator j_μ or, equivalently, the commutator $[H,r_\mu]$, is diagonal in the real-space part $|\mathbf{k}\rangle$ while having off-diagonal matrix elements in pseudospin space. It is then straightforward to specialize Eq. (4) to this case, finding

$$\sigma_{\mu\nu} = \frac{e^2}{i\hbar} \sum_{\sigma,\sigma'}^{2N} \int \frac{d^2k}{(2\pi)^2} \frac{\langle\sigma|w_\mu(\mathbf{k})|\sigma'\rangle_{\mathbf{k}} \langle\sigma'|w_\nu(\mathbf{k})|\sigma\rangle_{\mathbf{k}}}{\varepsilon_{\mathbf{k}\sigma'}-\varepsilon_{\mathbf{k}\sigma}+\hbar\omega-i0^+} \frac{f(\varepsilon_{\mathbf{k}\sigma})-f(\varepsilon_{\mathbf{k}\sigma'})}{\varepsilon_{\mathbf{k}\sigma'}-\varepsilon_{\mathbf{k}\sigma}}. \tag{5}$$

Here $w_\mu(\mathbf{k})$ is found from $[H,r_\mu]|\mathbf{k}\rangle = w_\mu(\mathbf{k})|\mathbf{k}\rangle$. Two contributions to the conductivity can be distinguished, arising from terms with $\sigma = \sigma'$ and $\sigma \neq \sigma'$, respectively. It is customary to call these the *intra*-band and *inter*-band contributions. Defining $w_\mu^{\sigma\sigma'}(\mathbf{k}) = \langle\sigma|w_\mu(\mathbf{k})|\sigma'\rangle_{\mathbf{k}}$, we find

$$\frac{\sigma_{\mu\nu}^{(\text{intra})}}{\sigma_0} = \frac{\delta(\hbar\omega)}{2} \sum_\sigma^{2N} \int d^2k\, w_\mu^{(\sigma\sigma)}(\mathbf{k})w_\nu^{(\sigma\sigma)}(\mathbf{k})f'(\varepsilon_{\mathbf{k}\sigma}), \tag{6}$$

$$\frac{\sigma_{\mu\nu}^{(\text{inter})}}{\sigma_0} = \frac{\sinh\left(\frac{\hbar\omega}{2k_BT}\right)}{2\hbar\omega} \sum_{\sigma\neq\sigma'} \int d^2k\, \delta(\hbar\omega-[\varepsilon_{\mathbf{k}\sigma}-\varepsilon_{\mathbf{k}\sigma'}])$$
$$\times \frac{-w_\mu^{(\sigma\sigma')}(\mathbf{k})w_\nu^{(\sigma'\sigma)}(\mathbf{k})}{\cosh\left(\frac{\hbar\omega}{2k_BT}\right)+\cosh\left(\frac{\varepsilon_{\mathbf{k}\sigma}+\varepsilon_{\mathbf{k}\sigma'}-2\mu}{2k_BT}\right)}. \tag{7}$$

For brevity, we use the scale factor $\sigma_0 = ge^2/(2\pi\hbar)$, where $g = 4$ has been introduced to account for the quasiparticle degeneracy (real spin and valley) in graphene. The intra-band term (6) is the usual dc Drude conductivity, which depends on states in the vicinity of the Fermi surface where the derivative f' of the Fermi function is peaked. It vanishes at the neutrality point ($\mu = 0$) in the zero-temperature limit. The inter-band contribution (7) is the interesting part for finite ω. It is calculated straightforwardly using continuum-model descriptions of single-layer and bilayer graphene. The expression (7) given here is well-suited for obtaining analytical results for the dependence on temperature but cannot be used to go beyond the clean limit. To discuss the effect of disorder, a more general formula is needed that will be given in the next subsection.

2.2. General conductivity formula in terms of Greens functions

Mathematical manipulation of Eq. (4) yields the conductivity expressed in terms of single-particle Greens functions [12,13,22]. It has the general form

$$\frac{\sigma_{\mu\nu}(\omega)}{\sigma_0} = \int_{-\infty}^{\infty} d\varepsilon\, \mathcal{T}_{\mu\nu}(\varepsilon,\omega)\left[f\left(\varepsilon+\frac{\hbar\omega}{2}\right)-f\left(\varepsilon-\frac{\hbar\omega}{2}\right)\right]. \tag{8}$$

We have derived a new and, for our purposes, more convenient expression for the diagonal part of the frequency-dependent transmission function,

$$\mathcal{T}_{\mu\mu}(\varepsilon,\omega) = F_\mu\left(\frac{\hbar\omega}{2}-\varepsilon-i0^+, -\frac{\hbar\omega}{2}-\varepsilon-i0^+\right)$$
$$- F_\mu\left(\frac{\hbar\omega}{2}-\varepsilon-i0^+, -\frac{\hbar\omega}{2}-\varepsilon+i0^+\right) + \text{c.c.}, \tag{9}$$

given here in terms of functions

$$F_\mu(z,z') = \frac{\hbar\omega}{4} \sum_{\mathbf{r}} r_\mu^2 \text{Tr}_{\text{sl}}\{G_{\mathbf{r}}(z)G_{-\mathbf{r}}(z')\}. \tag{10}$$

$G(\mathbf{r},\mathbf{r}';z) \equiv G(\mathbf{r}-\mathbf{r}',0;z) =: G_{\mathbf{r}-\mathbf{r}'}(z)$ is the real-space representation of the single-particle Greens function in a translationally invariant system, and the trace Tr_{sl} is performed only over sublattice and layer degrees of freedom.

It is straightforward to specialize the general conductivity formula obtained in this subsection to the clean limit. Performing a Fourier transformation and using the fact that the single-particle Hamiltonian $H \to H_{\mathbf{k}}$ becomes diagonal in real space, we find

$$F_\mu(z,z') = \frac{\hbar\omega}{16\pi^2} \int d^2k\, \text{Tr}_{\text{sl}}\left\{G_{\mathbf{k}}(z)\left[\frac{\partial^2 H_{\mathbf{k}}}{\partial k_\mu^2} - 2\frac{\partial H_{\mathbf{k}}}{\partial k_\mu}G_{\mathbf{k}}(z)\frac{\partial H_{\mathbf{k}}}{\partial k_\mu}\right]G_{\mathbf{k}}(z)G_{\mathbf{k}}(z')\right\}. \tag{11}$$

Here $G_{\mathbf{k}}(z) = (H_{\mathbf{k}}-z)^{-1}$ is the single-particle Greens function in reciprocal-space (plane-wave) representation. Application to graphene yields the same results as obtained more easily using formulae from the previous subsection. However, Eq. (8) turns out to be very useful beyond the clean limit.

3. Single-layer graphene in the clean limit

We apply the continuum-model description of a single sheet of graphene to evaluate the conductivity formula (7). The single-particle Hamiltonian in plane-wave representation is given by [36,37]

$$H_{\text{sg}} = \hbar v(k_x\sigma_x+k_y\sigma_y)+\tau[(k_y^2-k_x^2)\sigma_x+2k_xk_y\sigma_y], \tag{12}$$

where v is the Dirac-fermion velocity characterizing the dispersion near the **K** point, and the term proportional to τ is a trigonal-warping correction to the band structure. Straightforward diagonalization of H_{sg} yields eigenvalues $\varepsilon_{\mathbf{k}\sigma}^{(\text{sg})}$, where $\sigma = \pm$ distinguishes the electron and hole bands. As $\varepsilon_{\mathbf{k}\sigma}^{(\text{sg})} \equiv -\varepsilon_{\mathbf{k},-\sigma}^{(\text{sg})}$, the dependence on temperature and chemical potential is universal, i.e., independent of the values of v and τ. The remaining dimensionless prefactor is only a function of ω, v, τ and, by simple dimensional analysis, can therefore only depend on these through the combination $\omega\tau/(\hbar v^2)$. The expression for the conductivity reads then

$$\frac{\sigma_{\mu\nu}^{(\text{inter})}}{\sigma_0} = g\left(\frac{\hbar\omega}{2k_BT},\frac{\mu}{k_BT}\right)\Upsilon_{\mu\nu}\left(\frac{\omega\tau}{\hbar v^2}\right)\Theta(\omega), \tag{13}$$

where Θ is the Heaviside step function, and we introduced the abbreviation

$$g(\xi,\eta) = \frac{\sinh\xi}{\cosh\xi+\cosh\eta}. \tag{14}$$

It is found that $\Upsilon_{xy} = \Upsilon_{yx} = 0$ and

$$\left.\begin{array}{c}\Upsilon_{xx}(\xi)\\\Upsilon_{yy}(\xi)\end{array}\right\} = 2\int d^2\kappa\left\{\frac{\kappa_y^2(1-2\xi[\kappa_x+\xi\kappa^2])^2}{(1+2\xi\kappa_x)^2(\kappa_x-\xi\kappa^2)^2}\right\}\delta\left(1-2\sqrt{\kappa^2-2\xi\kappa_x[\kappa_x^2-3\kappa_y^2]+\xi^2\kappa^4}\right). \tag{15}$$

We find numerically that $\Upsilon_{xx}(\xi) = \Upsilon_{yy}(\xi) \equiv \Upsilon(\xi)$ and show this universal function in Fig. 1. In the limit $\tau \to 0$, the well-known
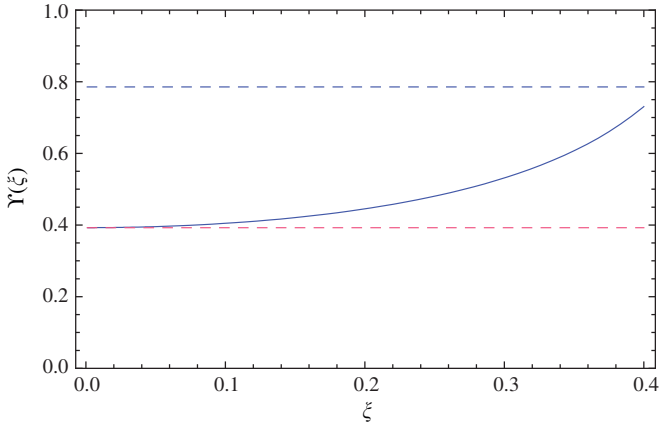
Fig. 1. Universal function capturing the effect of trigonal warping on the optical conductivity of a single graphene sheet. The dashed lines indicate the values of $\pi/8$ and $\pi/4$, respectively.

[13,20,27] universal conductivity of single-layer graphene is found: $\Upsilon(0) = \pi/8$. In the (for graphene unphysical) limit of large ξ, a different saturation value is realized: $\Upsilon(\infty) = \pi/4$.

4. Bilayer graphene in the clean limit

To describe bilayer graphene, we use the 4×4 continuum-model Hamiltonian [16] in plane-wave representation given by

$$
H_{\mathrm{bl}} = \begin{pmatrix} eV/2 & \hbar v(k_x+ik_y) & t_\perp & 0 \\ \hbar v(k_x-ik_y) & eV/2 & 0 & \hbar v_3(k_x+ik_y) \\ t_\perp & 0 & -eV/2 & \hbar v(k_x-ik_y) \\ 0 & \hbar v_3(k_x-ik_y) & \hbar v(k_x+ik_y) & -eV/2 \end{pmatrix}.
$$
(16)

Here $t_\perp$ and V parameterize the strongest inter-layer hopping and a potential difference applied between the layers, respectively. v_3 measures the strength of an additional inter-layer hopping that gives rise to trigonal warping. Straightforward diagonalization of H_{bl} yields the set of energy eigenvalues $\varepsilon_{\mathbf{k}\sigma}^{(\mathrm{bl})}$, with $\sigma = 1, 2, 3, 4$. These eigenvalues can be grouped into two pairs that add up to zero. Assuming without loss of generality that $\varepsilon_{\mathbf{k}1}^{(\mathrm{bl})} < \varepsilon_{\mathbf{k}2}^{(\mathrm{bl})} \leq \varepsilon_{\mathbf{k}3}^{(\mathrm{bl})} < \varepsilon_{\mathbf{k}4}^{(\mathrm{bl})}$, we have $\varepsilon_{\mathbf{k}1}^{(\mathrm{bl})} = -\varepsilon_{\mathbf{k}4}^{(\mathrm{bl})}$ and $\varepsilon_{\mathbf{k}2}^{(\mathrm{bl})} = -\varepsilon_{\mathbf{k}3}^{(\mathrm{bl})}$. This means that there are *two* contributions of the type encountered in the single-layer case. Depending on the situation, the remaining four contributions can be simplified as well. Here we just give the analytical result obtained for the case with zero inter-layer bias and trigonal warping neglected ($V = 0$ and $v_3 = 0$). We find $\sigma_{xx}^{(\mathrm{inter})} = \sigma_{yy}^{(\mathrm{inter})} \equiv \sigma_1 + \sigma_2 + \sigma_3$, where

$$
\frac{\sigma_1}{\sigma_0} = \frac{\pi}{8} g\left(\frac{\hbar\omega}{2k_BT}, \frac{\mu}{k_BT}\right) \left[\frac{\hbar\omega+2t_\perp}{\hbar\omega+t_\perp}\Theta(\omega) + \frac{\hbar\omega-2t_\perp}{\hbar\omega-t_\perp}\Theta(\omega-2t_\perp/\hbar)\right],
$$
(17)

$$
\frac{\sigma_2}{\sigma_0} = \frac{\pi}{8}\left(\frac{t_\perp}{\hbar\omega}\right)^2 \Theta(\omega-t_\perp/\hbar)\left[g\left(\frac{\hbar\omega}{2k_BT}, \frac{2\mu-t_\perp}{2k_BT}\right) + g\left(\frac{\hbar\omega}{2k_BT}, \frac{2\mu+t_\perp}{2k_BT}\right)\right],
$$
(18)

$$
\frac{\sigma_3}{\sigma_0} = \frac{t_\perp}{\hbar}\delta(\omega-t_\perp/\hbar)\int_{t_\perp/2k_BT}^\infty \frac{d\kappa}{\kappa}\left[g\left(\frac{t_\perp}{2k_BT}, \frac{\mu}{k_BT}-\kappa\right) + g\left(\frac{t_\perp}{2k_BT}, \frac{\mu}{k_BT}+\kappa\right)\right].
$$
(19)

This result generalizes a previous expression [23] obtained for the zero-temperature limit. In Fig. 2, we show the effect of a finite inter-layer bias V.
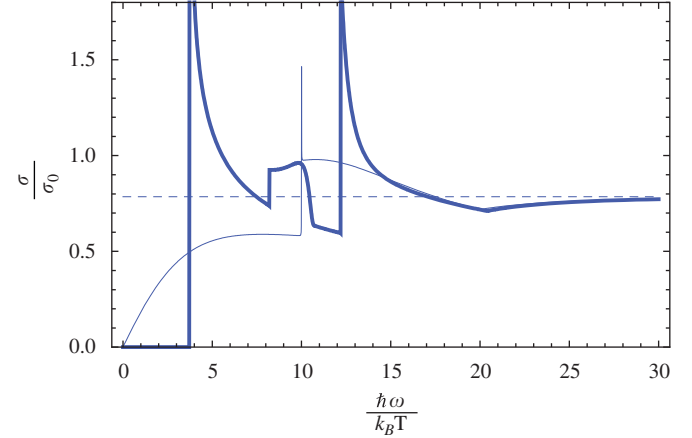

Fig. 2. Interband contribution to the conductivity of clean bilayer graphene ($t_\perp = 10k_BT$), with chemical potential at the symmetry point and a finite bias voltage $V = 2k_BT/e$ between the layers (thick solid curve). The zero-bias case is shown as the thin solid curve. The dashed line indicates the value $\pi/4$.

5. Conclusions

We have studied the ac conductivity of single and bilayer graphene. Analytical results were obtained for finite temperature and with trigonal warping included. Our expressions should be useful to facilitate detailed comparison with experiment and enable extraction of electronic-structure parameters from conductivity measurements.

References

[1] K.S. Novoselov, A.K. Geim, S.V. Morozov, D. Jiang, Y. Zhang, S.V. Dubonos, I.V. Grigorieva, A.A. Firsov, Science 306 (2004) 666.
[2] K.S. Novoselov, D. Jiang, F. Schedin, T.J. Booth, V.V. Khotkevich, S.V. Morozov, A.K. Geim, PNAS 102 (2005) 10451.
[3] A.H. Castro Neto, F. Guinea, N.M.R. Peres, K.S. Novoselov, A.K. Geim, Rev. Modern Phys. 81 (2009) 109.
[4] M. Katsnelson, K. Novoselov, Solid State Comm. 143 (2007) 3.
[5] K.S. Novoselov, A.K. Geim, S.V. Morozov, D. Jiang, M.I. Katsnelson, I.V. Grigorieva, S.V. Dubonos, A.A. Firsov, Nature (London) 438 (2005) 197.
[6] Y. Zhang, Y. Tan, H.L. Stormer, P. Kim, Nature (London) 438 (2005) 201.
[7] M.I. Katsnelson, European Phys. J. B 51 (2006) 157.
[8] V.P. Gusynin, S.G. Sharapov, J.P. Carbotte, Phys. Rev. Lett. 96 (2006) 256802.
[9] V.P. Gusynin, S.G. Sharapov, Phys. Rev. B 73 (2006) 245411.
[10] V.P. Gusynin, S.G. Sharapov, J.P. Carbotte, Phys. Rev. Lett. 98 (2007) 157402.
[11] V.P. Gusynin, S.G. Sharapov, J.P. Carbotte, Phys. Rev. B 75 (2007) 165407.
[12] K. Ziegler, Phys. Rev. Lett. 97 (2006) 266802.
[13] K. Ziegler, Phys. Rev. B 75 (2007) 233407.
[14] N.M.R. Peres, F. Guinea, A.H.C. Neto, Phys. Rev. B 73 (2006) 125411.
[15] J. Nilsson, A.H. Castro Neto, F. Guinea, N.M.R. Peres, Phys. Rev. Lett. 97 (2006) 266801.
[16] J. Nilsson, A.H.C. Neto, F. Guinea, N.M.R. Peres, Phys. Rev. B 78 (2008) 045405.
[17] D.S.L. Abergel, V.I. Fal'ko, Phys. Rev. B 75 (2007) 155430.
[18] J. Cserti, Phys. Rev. B 75 (2007) 033405.
[19] J. Cserti, A. Csordás, G. Dávid, Phys. Rev. Lett. 99 (2007) 066802.
[20] L.A. Falkovsky, A.A. Varlamov, European Phys. J. B 56 (2007) 281.
[21] I. Snyman, C.W.J. Beenakker, Phys. Rev. B 75 (2007) 045322.
[22] S. Ryu, C. Mudry, A. Furusaki, A.W.W. Ludwig, Phys. Rev. B 75 (2007) 205344.
[23] E.J. Nicol, J.P. Carbotte, Phys. Rev. B 77 (2008) 155409.
[24] T. Stauber, N.M.R. Peres, A.K. Geim, Phys. Rev. B 78 (2008) 085432.
[25] N.M.R. Peres, T. Stauber, Internat. J. Modern Phys. B 22 (2008) 2529.
[26] N.M.R. Peres, T. Stauber, A.H. Castro Neto, Europhys. Lett. 84 (2008) 38002.
[27] L.A. Falkovsky, arXiv:0806.3663.
[28] H. Min, A.H. MacDonald, Phys. Rev. Lett. 103 (2009) 067402.
[29] F. Wang, Y. Zhang, C. Tian, C. Girit, A. Zettl, M. Crommie, Y.R. Shen, Science 320 (2008) 206.
[30] R.R. Nair, P. Blake, A.N. Grigorenko, K.S. Novoselov, T.J. Booth, T. Stauber, N.M.R. Peres, A.K. Geim, Science 320 (2008) 308.

758

[31] Z.Q. Li, E.A. Henriksen, Z. Jiang, Z. Hao, M.C. Martin, P. Kim, H.L. Stormer, D.N. Basov, Nat. Phys. 4 (2008) 532.

[32] K.F. Mak, M.Y. Sfeir, Y. Wu, C.H. Lui, J.A. Misewich, T.F. Heinz, Phys. Rev. Lett. 101 (2008) 196405.

[33] L.M. Zhang, Z.Q. Li, D.N. Basov, M.M. Fogler, Z. Hao, M.C. Martin, Phys. Rev. B 78 (2008) 235408.

[34] Z.Q. Li, E.A. Henriksen, Z. Jiang, Z. Hao, M.C. Martin, P. Kim, H.L. Stormer, D.N. Basov, Phys. Rev. Lett. 102 (2009) 037403.

[35] O. Madelung, Introduction to Solid-State Theory, Springer, Berlin, 1978.

[36] J.C. Slonczewski, P.R. Weiss, Band structure of graphite, Phys. Rev. 109 (1958) 272.

[37] R. Saito, G. Dresselhaus, M.S. Dresselhaus, Physical Properties of Carbon Nanotubes, Imperial College, London, 1998.