

A. Frequently Used Quantities and Abbreviations

Quantities

b	Bose-Einstein distribution function; band index
b_Q	occupation number of phonon mode Q
c	vacuum light speed
ϵ	single-electron energy
f	Fermi-Dirac or classical distribution function
f_k	occupation number of electronic Bloch state k
k	electronic Bloch state
\mathbf{k}	wavevector
n	refractive index
D	electronic density of states
\mathbf{E}	electric field
\mathbf{P}	induced electric polarization
Q	phonon mode
T	temperature
ϵ	dielectric function
τ	temporal delay between pump and probe pulse
ω	angular frequency
$\omega/2\pi$	frequency

A. Frequently Used Quantities and Abbreviations

Abbreviations

BZ	Brillouin zone
DFG	difference-frequency generation
DFT	density-functional theory
DOT	direct optical transition
e	electron
eDOS	electronic density of states
HiPCO	high-pressure carbon monoxide
HOPG	highly oriented pyrolytic graphite
i	ion
imp	impurity
interDOT	direct optical interband transition
intraDOT	direct optical intraband transition
IOT	indirect optical transition
n	neutral particle
NC	nanocylinder, short straight segment of a NT
NT	single-wall carbon nanotube
ph	phonon
SCOP	strongly coupled optical phonon
SFG	sum-frequency generation
TRTS	time-resolved THz spectroscopy
TS	THz (time-domain) spectroscopy
2TM	2-temperature model
\mathbb{N}	set $\{1, 2, \dots\}$ of all positive integer numbers
\mathbb{Z}	set $\{0, \pm 1, \pm 2, \dots\}$ of all integer numbers

B. Mathematical Definitions and Theorems

Fourier Transformation

$$\begin{aligned}x(\omega) &= \mathcal{F}[x(t)](\omega) = \int_{-\infty}^{\infty} dt x(t) \frac{\exp(i\omega t)}{\sqrt{2\pi}} \\x(t) &= \mathcal{F}^{-1}[x(\omega)](t) = \int_{-\infty}^{\infty} d\omega x(\omega) \frac{\exp(-i\omega t)}{\sqrt{2\pi}}\end{aligned}\tag{B.1}$$

Convolution

$$(x * y)(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} dt' x(t - t') y(t')\tag{B.2}$$

Convolution Theorem

$$\begin{aligned}\mathcal{F}(x * y) &= \mathcal{F}(x)\mathcal{F}(y) \\ \mathcal{F}^{-1}(x * y) &= \mathcal{F}^{-1}(x)\mathcal{F}^{-1}(y)\end{aligned}$$

Comparison

$x \approx y$: x is approximately equal to y
 $x \sim y$: x and y are of the same order of magnitude

B. Mathematical Definitions and Theorems

C. Details of Numerical Calculations for Graphite

C.1. Direct Optical Transitions in Graphite

The contribution $\varepsilon^{\text{interDOT}}$ to the dielectric function of graphite is given by Eq. (1.19). The integration over \mathbf{k} can be reduced to the vicinity of the HKH line where 1 Fermi surface pocket is located. The other pocket around H'K'H' is accounted for by a factor of 2.

The eigenenergies ϵ_k required are obtained by diagonalizing the SWM Hamiltonian (4.4); the momentum matrix elements are obtained by applying the unitary transformation, which diagonalizes (4.4), to the 2nd term on the right-hand side of Eq. (4.4).

The diagonalization and integration are performed numerically for a mesh of frequencies ω and electronic temperatures T_e whereas $\varepsilon^{\text{interDOT}}$ for all other ω and T_e is obtained by 2-dimensional interpolation.

For the contribution $\varepsilon^{\text{intraDOT}}$, the calculation of the plasma frequency (1.22) is sufficient which requires the knowledge of the velocity-weighted eDOS (1.23). For this purpose, the ϵ axis was divided in intervals $[\epsilon_j, \epsilon_j + \Delta\epsilon]$. Scanning the \mathbf{k} space and the band structure $\epsilon_{\mathbf{k}b}$ point by point, the resulting band velocity $\mathbf{v}_{\mathbf{k}b}$ was added to the j th interval fulfilling $\epsilon_{\mathbf{k}b} \in [\epsilon_j, \epsilon_j + \Delta\epsilon]$. The ordinary eDOS (1.7) was calculated similarly.

C.2. 2-Temperature Model for Graphite

The temporal dynamics of the electron temperature T_e is obtained from that of the total electronic energy E_e by

$$C_e(T_e) \frac{\partial T_e}{\partial t} = \frac{\partial E_e}{\partial t}.$$

The electronic heat capacity C_e is taken from Ref. [Nih03], and $\partial E_e / \partial t$ is given by Eq. (1.6) which requires the evaluation of the Eliashberg function

$$H(\epsilon, \epsilon', \Omega) = \sum_{kk'r} |M_{k'r}^r|^2 \delta(\epsilon - \epsilon_k) \delta(\epsilon' - \epsilon_{k'}) \delta(\Omega - \Omega_Q). \quad (\text{C.1})$$

In graphite, the usual approximation $\epsilon = \epsilon' = \epsilon_F$ [All87] is not justified because of the strongly varying eDOS at the Fermi edge. To avoid the 6-dimensional integration over

C. Details of Numerical Calculations for Graphite

transition $(\mathbf{k}_\perp, b) \longrightarrow (\mathbf{k}'_\perp, b')$	$r = \text{LO}$	$r = \text{TO}$
$(\overrightarrow{\Gamma\text{K}} + \boldsymbol{\kappa}, \pi) \longrightarrow (\overrightarrow{\Gamma\text{K}} + \boldsymbol{\kappa}', \pi^*)$	$g_\Gamma^r [1 - \cos(\alpha - \alpha')]$	$g_\Gamma^r [1 + \cos(\alpha - \alpha')]$
$(\overrightarrow{\Gamma\text{K}} + \boldsymbol{\kappa}, \pi^*) \longrightarrow (\overrightarrow{\Gamma\text{K}} + \boldsymbol{\kappa}', \pi^*)$	$g_\Gamma^r [1 + \cos(\alpha - \alpha')]$	$g_\Gamma^r [1 - \cos(\alpha - \alpha')]$
$(\overrightarrow{\Gamma\text{K}'} + \boldsymbol{\kappa}, \pi) \longrightarrow (\overrightarrow{\Gamma\text{K}'} + \boldsymbol{\kappa}', \pi^*)$	≈ 0	$g_\text{K}^r [1 + \cos(\alpha - \alpha' - 60^\circ)]$
$(\overrightarrow{\Gamma\text{K}} + \boldsymbol{\kappa}, \pi^*) \longrightarrow (\overrightarrow{\Gamma\text{K}'} + \boldsymbol{\kappa}', \pi^*)$	≈ 0	$g_\text{K}^r [1 - \cos(\alpha - \alpha' - 60^\circ)]$

Table C.1.: Electron-phonon matrix elements for the relevant electronic transitions in graphene taken from Ref. [Pis04]. The k_z component can change arbitrarily. Transitions within the π band are assumed to have the same matrix element as those within the π^* band.

\mathbf{k} space some simplifications are introduced: We assume that the e-ph coupling and the phonon dispersion of the SCOPs is the same like in graphene. Moreover, we neglect trigonal warping of the electronic π and π^* bands. It is important to take care of all prefactors since the absolute values of H are required.

According to Section 4.4, the small Fermi surface of graphite allows only for wavevector changes $\Delta\mathbf{k}_\perp$ close to the Γ or K point. In Eq. (C.1), this is enforced by the first two δ functions together with $\epsilon \approx \epsilon' \approx \epsilon_\text{F}$. In other words, the electron either stays in the pocket around K or K' , or it jumps into the other pocket. We choose the electron BZ (eBZ) and phonon BZ (phBZ) differently such that umklapp processes do not occur for scattering from the K to the K' pocket. For reasons of symmetry, transitions from the K' to the K pocket give identical results and will be taken into account by a factor of 2. The eBZ used is the alternative eBZ in Fig. 4.6(a); the phBZ should be chosen such that Γ and K point are far away from the BZ boundary. Then, the sum in (C.1) is split up by using

$$\mathbf{k} = \mathbf{k}_c + \mathbf{k}_\perp = \mathbf{k}_c + \overrightarrow{\Gamma\text{X}} + \boldsymbol{\kappa} \quad \text{with} \quad \text{X} \in \{\text{K}, \text{K}'\}$$

where $\mathbf{k}_c = (0, 0, k_c)$ and \mathbf{k}_\perp are parallel and perpendicular to the c axis, respectively. The small translation $\boldsymbol{\kappa}$ is expressed in cylindrical coordinates as

$$\boldsymbol{\kappa} = \kappa \begin{pmatrix} \cos \alpha \\ \sin \alpha \\ 0 \end{pmatrix}$$

where α is the angle between $\boldsymbol{\kappa}$ and $\overrightarrow{\Gamma\text{K}}$. The definitions for k' are analog. This yields

$$H = \frac{1}{A^2} \sum_{bb'} \sum_{jj'} \sum_{k_z k'_z} \iiint d\kappa d\kappa' \kappa \kappa' d\alpha d\alpha' \underbrace{\delta(\epsilon - \epsilon_k) \delta(\epsilon' - \epsilon_{k'})}_{(i)} \sum_r \underbrace{\delta(\Omega - \Omega_Q)}_{(ii)} \underbrace{|M_{kk'}^r|^2}_{(iii)}, \quad (\text{C.2})$$

where the summation over the $\boldsymbol{\kappa}$ mesh has been approximated by an integration with A being the volume of a cell in the 2-dimensional $\boldsymbol{\kappa}$ space.

We now make the following assumptions for the labeled terms in Eq. (C.2): (i) We neglect the α -dependence of the electron bands which is reasonable since trigonal warping is weak

C.3. Indirect Optical Transitions in Graphite

at the energies considered here. (ii) We neglect phonon dispersion along q_z and around Γ and K which implies $\mathbf{q}_\perp \approx \overrightarrow{X'X} \in \{\Gamma, K\}$. (iii) The matrix elements $|M_{kk'}^r|^2$ are those of graphene and listed in Table C.1. This implies their independence of k_z .

Consequently, the angular integration in (C.2) affects only the e-ph matrix elements. Together with \sum_r , this yields $4\pi^2 g_\Gamma^r$ for the Γ phonons since the angular dependence of LO and TO branch cancel each other, whereas the K phonons yield $4\pi^2 g_K^r$ and 0 for the LO and TO branch, respectively. Going back to Cartesian coordinates gives

$$H = 2 \sum_{bb'r} [g_\Gamma^r \delta(\Omega - \Omega_\Gamma^r) + g_K^r \delta(\Omega - \Omega_K^r)] \sum_{\kappa\kappa'} \delta(\epsilon - \epsilon_\kappa) \delta(\epsilon' - \epsilon'_\kappa)$$

where

$$g_\Gamma^{\text{LO}} = g_\Gamma^{\text{TO}} = 0.0405 \text{ eV}^2, \quad g_K^{\text{TO}} = 0.0994 \text{ eV}^2, \quad \text{and } g_K^{\text{LO}} \approx 0$$

have been introduced [Pis04]. Since these quantities do not depend on the electron band indices b and b' we finally obtain

$$H(\epsilon, \epsilon', \Omega) = \frac{1}{8} \sum_r [g_\Gamma^r \delta(\Omega - \Omega_\Gamma^r) + g_K^r \delta(\Omega - \Omega_K^r)] D(\epsilon) D(\epsilon')$$

where we have exploited $\sum_\kappa \delta(\epsilon - \epsilon_\kappa) = D(\epsilon)/4$; the factor $\frac{1}{4}$ is due to the spin degeneracy and summation over only 1 of the 2 Fermi surface pockets.

This analytic formula for the Eliashberg function may have applications in all areas where the coupling between SCOPs and electrons in graphite is important. In particular, the 2TM (1.6) now reduces to an integration over 1 energy ϵ only which can be performed numerically without further ado.

C.3. Indirect Optical Transitions in Graphite

The IOT contribution of e-ph coupling to the dielectric function is, according to Eqs. (1.24) and (1.25),

$$\text{Im } \varepsilon_{\beta\beta}^{\text{IOT}} = 2 \frac{(2\pi\hbar e)^2}{(\hbar\omega)^4 V} \sum_{kk'r} \sum_{\pm\oplus} S_{\pm\oplus}(k, k', Q) |M_{k'k}^r|^2 (v_{k\beta} - v_{k'\beta})^2$$

and can be transformed into

$$\text{Im } \varepsilon_{\alpha\alpha}^{\text{IOT}} = 2 \frac{(2\pi\hbar e)^2}{(\hbar\omega)^4 V} \sum_{\pm\oplus} \iiint d\epsilon d\epsilon' d\Omega S_{\pm\oplus}(\epsilon, \epsilon', \Omega) H_{\beta\beta}^{\text{tr}}(\epsilon, \epsilon', \Omega).$$

Here, \oplus is related to photon absorption/emission and \pm to phonon emission/annihilation, respectively, and the factor

$$S_{\pm\oplus}(\epsilon, \epsilon', \Omega) = [b(\Omega) + \frac{1}{2} \pm \frac{1}{2}] f(\epsilon) \cdot [1 - f(\epsilon')] \cdot [\oplus\delta(\epsilon' - \epsilon \pm \hbar\Omega\oplus\hbar\omega)]$$

C. Details of Numerical Calculations for Graphite

embraces all electronic and phononic occupation numbers. The auxiliary function

$$H_{\beta\beta}^{\text{tr}}(\epsilon, \epsilon', \Omega) = \sum_{kk'r} |M_{kk'}^r|^2 (v_{k\beta} - v_{k'\beta})^2 \delta(\epsilon - \epsilon_k) \delta(\epsilon' - \epsilon_{k'}) \delta(\Omega - \Omega_Q)$$

is proportional to the Eliashberg function $\alpha_{\text{tr}}^2 F(\epsilon, \epsilon', \Omega)$ for transport which contains all information on electron-phonon-photon interaction [Gri81, All71]. Its evaluation is very similar to that of H in Appendix C.2 and results in

$$H_{\beta\beta}^{\text{tr}}(\epsilon, \epsilon', \Omega) = \frac{1}{8} \sum_{r,X} g_X^r \delta(\Omega - \Omega_X^r) [(v_\beta^2 D)(\epsilon) D(\epsilon') + D(\epsilon) (v_\beta^2 D)(\epsilon') + \Pi_X (u_\beta D)(\epsilon) (u_\beta D)(\epsilon')].$$

Here, $v_\beta^2 D(\epsilon)$ is the velocity-weighted eDOS (1.23) in β direction with $\beta \in \{x, y, z\}$, and $u_\beta D(\epsilon) := \sum_k u_{k\beta} \delta(\epsilon - \epsilon_k)$. In the last expressions one has $u_{k\beta} := v_{k\beta} / \cos \alpha$. The latter term only contributes for $\Pi_X = \frac{1}{2}$ which happens if $X = K$. Since the THz probe pulse is polarized perpendicularly to the graphite c axis only the x direction is relevant.

Now the summation over \mathbf{k} space is reduced to an integral over an energy which can be performed numerically without further ado.