Quantum-kinetic theory of photocurrent generation via direct and phonon-mediated optical transitions

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A quantum kinetic theory of direct and phonon mediated indirect optical transitions is developed within the framework of the non-equilibrium Green’s function formalism. After validation against the standard Fermi-Golden-Rule approach in the bulk case, it is used in the simulation of photocurrent generation in ultra-thin crystalline silicon p-i-n-junction devices.

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I. INTRODUCTION

Quantum effects in semiconductor nanostructures are widely exploited in optoelectronic devices such as light emitting diodes or lasers. With the increasing demand of renewable energy supply, high efficiency photovoltaic devices were proposed, which also make use of semiconductor nanostructures. However, these devices are not based on the standard III-V direct band gap materials conventionally used in optoelectronics, but on silicon, which is most common in solar cell applications. It was shown that even though the suppression of direct optical transitions is somewhat relaxed in low dimensional silicon structures, it is the phonon assisted indirect processes that dominate the optical response, and thus a suitable theoretical description of the photovoltaic behaviour needs to include these processes responsible for energy and momentum transfer, in addition to the general requirements of a quantum photovoltaic device, which were addressed in . As an extension of this previous work, the present paper presents a microscopic description of phonon-mediated optical transitions in indirect semiconductor nanostructures, based on the non-equilibrium Green’s function (NEGF) formalism and thus compatible with advanced quantum transport theories.

The paper is organized as follows. After the derivation of the general expressions for optoelectronic rates within the non-equilibrium Green’s function theory, these are applied to the case of a direct gap bulk semiconductor and compared to the standard Fermi-Golden-Rule (FGR) result. Next, the description of phonon-mediated indirect transitions within the NEGF formalism is discussed and the resulting expression for the generation rate again compared to the FGR, both in an analytical approximation and a numerical computation for a simple effective mass model of bulk silicon. Finally, the theory is extended to thin films in order to model photocurrent generation in ultrathin indirect gap semiconductor p-i-n junctions, which form important building blocks of future nano-photovoltaic devices.

II. NEGF THEORY

In order to be able to describe both optical transitions and inelastic quantum transport in semiconductor nanostructures within the same microscopic picture, a theoretical framework based on the NEGF formalism was developed in . Before considering the specific case of photogeneration in indirect semiconductors, the aspects of the theory related to interband transitions will be outlined below in a general formulation, which relies on the extensive work on similar systems investigated with focus on their light-emitting and lasing properties.

A. Interband generation and recombination rates

The macroscopic equation of motion for a photovoltaic system is the continuity equation for the charge carrier density

$$\partial_t \rho_c (\mathbf{r}) + \nabla \cdot \mathbf{j}_c (\mathbf{r}) = G_c (\mathbf{r}) - R_c (\mathbf{r}), \quad c = e, h$$ (1)

where $\rho_c$ and $j_c$ are charge carrier and particle current density, respectively, $G_c$ the generation rate and $R_c$ the recombination rate of carriers species $e,h$. The microscopic version of this equation in terms of NEGF corresponds to (1) in the form: $\mathcal{C} = \{ \{ r_1, t_1 \in \mathbb{C} \} \}$

$$\lim_{2 \to 1} \left\{ i \hbar \left( \frac{\partial}{\partial t_1} + \frac{\partial}{\partial t_2} \right) G(1, 2) + [H_0 (r_1) - H_0 (r_2)] G(1, 2) \right\}$$

$$\lim_{2 \to 1} \int_{\mathcal{C}} d3 \left[ \Sigma(1, 3) G(3, 2) - G(1, 3) \Sigma(3, 2) \right],$$ (2)

where $H_0$ is the Hamiltonian of the non-interacting electronic system, $G$ is the electronic non-equilibrium Green’s function defined on the Keldysh contour $\mathcal{C}$ and $\Sigma$ is the self-energy encoding the interaction of the electronic system with phonons, photons and with itself, i.e. the scattering processes which may give rise to intra- or interband transitions. In steady state, (1) becomes

$$\nabla \cdot \mathbf{j}_c (\mathbf{r}) = G_c (\mathbf{r}) - R_c (\mathbf{r}), \quad c = e, h.$$ (3)

In the microscopic theory, the divergence of the electron (particle) current corresponds to the limit $1 \to 2$ of the
RHS in Eq. (4)

\[
\nabla \cdot j(r) = - \frac{2}{V} \int \frac{dE}{2\pi\hbar} \int d^3r' \left[ \Sigma^R(r, r'; E) G^< (r', r; E) + \Sigma^< (r, r'; E) G^A(r', r; E) - G^R(r, r'; E) \times \Sigma^< (r', r; E) - G^< (r, r'; E) \Sigma^A(r', r; E) \right].
\]

If the energy integration is taken over the range of all bands connected by the interband scattering process, i.e. both valence and conduction bands, the resulting divergence should vanish, if the self-energies are chosen properly, ensuring the conservation of the overall current, which is nothing else than the corresponding formulation of the detailed balance requirement. If the integration is restricted to one of the bands (e.g. B=valence band or conduction band), the above equation corresponds to the microscopic version of Eq. (3), and provides on the RHS the total local interband scattering rate,

\[
R_{rad}(r) \equiv \mathcal{R}(r) - \mathcal{G}(r)
\]

\[
\quad = - \frac{2}{V} \int_{B(r)} \frac{dE}{2\pi\hbar} \int d^3r' \left[ \Sigma^R(r, r'; E) G^< (r', r; E) + \Sigma^< (r, r'; E) G^A(r', r; E) - G^R(r, r'; E) \times \Sigma^< (r', r; E) - G^< (r, r'; E) \Sigma^A(r', r; E) \right].
\]

Depending on the nature of the interaction described by \( \Sigma \), the scattering process may be highly non-local, in which case the above rate contains contributions from a large volume. The total interband current is found by integrating the divergence over the active volume, and is equivalent to the total global transition rate and, via the Gauss theorem, to the difference of the interband currents at the boundaries of the interacting region. Making use of the cyclic property of the trace, it can be expressed in the form

\[
R_{rad} = \frac{2}{V} \int d^3r \int_{B(r)} \frac{dE}{2\pi\hbar} \int d^3r' \left[ \Sigma^< (r, r'; E) G^> (r', r; E) - \Sigma^> (r, r'; E) G^< (r', r; E) \right].
\]

with units \([R_{rad}] = \text{s}^{-1}\). Within the general (basis-independent) NEGF picture, the RHS of Eq. (7) can be interpreted as follows: \( h^{-1}\Sigma^{<,>}(E) \) represents the rate at which charge carriers with energy \( E \) may leave (occupy) a state at that energy whereas \( G^{>,<}(E) \) quantifies the energy resolved probability that the system can accept (donate) a particle of energy \( E \), i.e. that there is an unoccupied (occupied) state at the right energy. The first term thus represents the total inscattering rate, while the second term provides the total outscattering rate.

If we are interested in the interband scattering rate, we can neglect in Eq. (7) the contributions to the self-energy from intraband scattering, e.g. via interaction with phonons, low energy photons (free carrier absorption) or ionized impurities, since they cancel upon energy integration over the band. However, if self-energies and Green’s functions are determined self-consistently as they should in order to guarantee current conservation, the Green’s functions are related to the scattering self-energies via the Dyson equation for the propagator,

\[
G^{\mathcal{A}}(r_1, r_1'; E) = G^R_0(r_1, r_1'; E) + \int d^3r_2 \int d^3r_3 G^R_0(r_1, r_2; E) \times \Sigma^{\mathcal{A}}(r_2, r_3; E) G^{\mathcal{A}}(r_3, r_1'; E),
\]

and the Keldysh equation for the correlation functions,

\[
G^\mathcal{K}(r_1, r_1'; E) = \int d^3r_2 \int d^3r_3 G^\mathcal{K}(r_1, r_2; E) \times \Sigma^\mathcal{K}(r_2, r_3; E) G^\mathcal{A}(r_3, r_1'; E),
\]

and will thus be modified due to the intraband scattering. This means that in the case of self-consistent solutions, it is in general not possible to completely separate the effects of the different scattering processes, nor to isolate coherent from incoherent transport.

In the remainder of the paper, the general theory of interband transitions outlined above will be applied to optical interband transitions in direct and indirect semiconductors. For computational purposes and to ease comparison with existing descriptions, the theory will be reformulated using a simple effective mass band basis for completely homogeneous bulk systems and for inhomogeneous thin film devices.

\section*{B. Bulk semiconductor}

For a homogeneous bulk system, the field operators for carriers in band \( b \) can be written in the Bloch state basis,

\[
\hat{\psi}_b(r, t) = \sum_k \psi_{bk}(r) \hat{c}_{bk}(t).
\]

The expression for the total radiative rate of carriers in band \( b \) is simplified by using the Fourier space representation of Green’s functions and self-energies, which reads (\( O = G, \Sigma \))

\[
O_{b,b'}(r, r'; E) = \sum_k \psi_{bk}(r) O_{b,b'}(k; E) \psi_{b'k}^*(r'),
\]

where \( O_{b,b'}(k; E) \) is the steady-state Fourier-transform of the real-time Keldysh components of the contour-ordered Green’s function

\[
O_{b,b'}(k; t - t') = \frac{1}{i\hbar} \langle \mathcal{T}_C \{ \hat{c}_{bk}(t) \hat{c}_{b'k}^+(t') \} \rangle.
\]
Inserting these expressions in (7), the band-resolved rates are obtained as

$$R_{rad,b} = \frac{2}{V} \int \frac{dE}{2\pi \hbar} \sum_{\mathbf{k}} \sum_{\mathbf{q}} \left[ \Sigma_{b,b}^{\text{c}}(\mathbf{k}; E)G_{b,b}^{\gamma}(\mathbf{k}; E) \right. $$

$$\left. - \sum_{b'}^{\text{c}}(\mathbf{k}; E)G_{b',b}^{\gamma}(\mathbf{k}; E) \right].$$

(13)

In the following, the off-diagonal terms will be neglected \((O_b \equiv O_{b,b'})\), which means that only incoherent interband and subband polarization is considered.

1. Direct interband transitions

Inserting the electron-photon self-energy for a two-band model of a direct semiconductor \((\text{App. A})\), and neglecting intraband processes (free-carrier absorption and emission), the expression for the interband absorption rate becomes (in the following, the energy integration is restricted to the conduction band)

$$R_{abs} = \frac{2}{V} \int \frac{dE}{2\pi \hbar} \sum_{\mathbf{k}} \sum_{\lambda, \mathbf{q}} |M_{\text{c}}(\mathbf{k}, \lambda, \mathbf{q})|^2 N_{\lambda, \mathbf{q}}^c$$

$$\times G_{\lambda, \mathbf{q}}^{\gamma}(\mathbf{k}; E - \hbar\omega_{\mathbf{q}})G_{\gamma}^{\lambda}(\mathbf{k}; E),$$

(14)

with \(M_{\text{c}}\) the optical matrix element \((\text{App. A})\) and \(N_{\lambda, \mathbf{q}}^c\) the occupation of the photon modes. The latter is obtained from the modal photon flux via \(N_{\lambda, \mathbf{q}}^c = \phi_{\lambda, \mathbf{q}} V / \bar{c}\), where \(\bar{c}\) is the speed of light in the active medium. The modal photon flux in turn is given by the modal intensity of the EM field as \(\phi_{\lambda, \mathbf{q}} = \gamma_{\lambda, \mathbf{q}}^c \omega_{\mathbf{q}} / (\hbar\omega_{\mathbf{q}})\). Similarly, stimulated interband emission reads

$$R_{em, \text{st}} = \frac{2}{V} \int \frac{dE}{2\pi \hbar} \sum_{\mathbf{k}} \sum_{\lambda, \mathbf{q}} |M_{\text{c}}(\mathbf{k}, \lambda, \mathbf{q})|^2 N_{\lambda, \mathbf{q}}^c$$

$$\times G_{\lambda, \mathbf{q}}^{\gamma}(\mathbf{k}; E - \hbar\omega_{\mathbf{q}})G_{\gamma}^{\lambda}(\mathbf{k}; E),$$

(15)

while spontaneous interband emission is expressed as

$$R_{em, \text{sp}} = \frac{2}{V} \int \frac{dE}{2\pi \hbar} \sum_{\mathbf{k}} \sum_{\lambda, \mathbf{q}} |M_{\text{c}}(\mathbf{k}, \lambda, \mathbf{q})|^2$$

$$\times G_{\lambda, \mathbf{q}}^{\gamma}(\mathbf{k}; E - \hbar\omega_{\mathbf{q}})G_{\gamma}^{\lambda}(\mathbf{k}; E).$$

(16)

The net absorption \(R_{abs, \text{net}} = R_{abs} - R_{em, \text{st}}\) of photons in mode \((\lambda, \mathbf{q})\) is thus given by

$$R_{abs, \text{net}}(\lambda, \mathbf{q}) = \frac{2}{V} \int \frac{dE}{2\pi \hbar} \sum_{\mathbf{k}} |M_{\text{c}}(\mathbf{k}, \lambda, \mathbf{q})|^2 N_{\lambda, \mathbf{q}}^c$$

$$\times \left[ G_{\lambda, \mathbf{q}}^{\gamma}(\mathbf{k}; E - \hbar\omega_{\mathbf{q}})G_{\gamma}^{\lambda}(\mathbf{k}; E) - G_{\lambda, \mathbf{q}}^{\gamma}(\mathbf{k}; E - \hbar\omega_{\mathbf{q}})G_{\gamma}^{\lambda}(\mathbf{k}; E) \right].$$

(17)

Conventionally, the rates for absorption and emission are calculated based on Fermi’s Golden Rule (FGR), corresponding to the Born approximation within first order perturbation theory. It is thus instructive to compare the above expressions with the FGR-rate for net direct interband absorption\(^{[17]}\)

$$R_{abs, \text{net}}^{\text{FGR}}(\lambda, \mathbf{q}) = \frac{2}{V} \sum_{\mathbf{k}} 2\pi |M_{b,b}^{\gamma}(\mathbf{k}, \lambda, \mathbf{q})|^2 N_{\lambda, \mathbf{q}}^c$$

$$\times \delta(\varepsilon_{\gamma}(\mathbf{k}) - \varepsilon_{\lambda}(\mathbf{k}) - \hbar\omega_{\mathbf{q}})[f_{\gamma}(\mathbf{k}) - f_{\lambda}(\mathbf{k})].$$

(18)

Here, the absorbing (bulk) material is described by the dispersion relations \(\varepsilon_{\lambda}(\mathbf{k})\), \(b = c, v\), and assumed to be in a quasi-equilibrium state with occupation described by the Fermi function \(f_{\lambda}(E) = (\exp[\beta \varepsilon_{\lambda}(\mathbf{k})] + 1)^{-1}\), \(\beta = (k_B T)^{-1}\), where \(\mu_b\) is assumed to be global quasi-Fermi levels.

In order to reproduce the FGR result from the more general expression in terms of Green’s functions, the unperturbed (i.e. non-interacting) equilibrium form of the latter needs to be used, which corresponds to the expression for free fermions in equilibrium,

$$G_{b,b}^{\gamma}(\mathbf{k}; E) = 2\pi i f_b(E) \delta(E - \varepsilon_b(\mathbf{k})),\quad (19)$$

$$G_{b,b}^{\gamma}(\mathbf{k}; E) = 2\pi i [f_b(E) - 1] \delta(E - \varepsilon_b(\mathbf{k})).\quad (20)$$

Introducing these expressions in (17) and carrying-out the energy integration reproduces the FGR expression \(^{[18]}\).

To estimate the deviation of the rate from the FGR result for self-energies beyond the first Born approximation, the FGR rate is first used to derive the standard expression for the bulk absorption coefficient of the two band effective mass model, which amounts to

$$\alpha(h\omega_{\gamma}) = \frac{R_{abs, \text{net}}(h\omega_{\gamma})}{S(h\omega_{\gamma})/h\omega_{\gamma}},$$

(21)

where \(S(h\omega_{\gamma})\) is the monochromatic energy flux density of the EM field \((i.e.\ the\ absolute\ value\ of\ the\ Poynting\ vector)\) given by

$$S(h\omega_{\gamma}) = \rho_{\gamma}(h\omega_{\gamma})\hbar\omega_{\gamma} \bar{c} \int \frac{d\Omega}{4\pi} \sum_{\lambda} N_{\lambda}^c(h\omega_{\gamma}, \Omega),$$

(22)

where

$$\rho_{\gamma}(h\omega_{\gamma}) = \frac{(h\omega_{\gamma})^2}{2\pi^2(h\bar{c})^3}$$

(23)

is the photonic density of states of an optically isotropic medium with refractive index \(n_0 = \sqrt{\varepsilon_0}\) and corresponding speed of light \(\bar{c} = c_0/n_0\). With the standard approximation of isotropic and momentum-independent optical matrix elements, i.e. \(|M_{\text{c}}(\mathbf{k}, \lambda, \mathbf{q})|^2 \approx M_{\text{c}}^2(h\omega_{\gamma})\), the absorption rate is rewritten as follows,

$$R_{abs}(h\omega_{\gamma}) = M_{\text{c}}^2(h\omega_{\gamma}) J_{\text{c}}(h\omega_{\gamma}) \rho_{\gamma}(h\omega_{\gamma})$$

$$\times \int \frac{d\Omega}{4\pi} \sum_{\lambda} N_{\lambda}^c(h\omega_{\gamma}, \Omega)$$

$$= M_{\text{c}}^2(h\omega_{\gamma}) J_{\text{c}}(h\omega_{\gamma}) S(h\omega_{\gamma})/(h\omega_{\gamma} \bar{c}),$$

(24)
which provides the bulk absorption coefficient

\[ \alpha(\hbar \omega_g) = \tilde{M}_{cv}^\gamma (\hbar \omega_g) J_{cv}(\hbar \omega_g), \]  

with \( \tilde{M}_{cv}^\gamma = M_{cv}^\gamma / \epsilon. \) The difference between FGR and NEGF approaches concerns the term \( J_{cv}(\hbar \omega_g) \), which in the FGR case takes the specific form (using the continuum approximation \( \int d^3k \)

\[ J_{cv}^{FGR}(\hbar \omega_g) = \frac{2\pi}{\hbar} J_{cv}(\hbar \omega_g) (f_v - f_c), \]  

\[ J_{cv}(\hbar \omega_g) = \frac{2}{(2\pi)^3} \int d^3k \delta(\varepsilon_c(k) - \varepsilon_v(k) - \hbar \omega_g). \]  

If the occupation depends only marginally on crystalline momentum, the above expressions related to the joint density of states \( J_{cv} \) with suitable occupation,

\[ J_{cv}^{FGR}(\hbar \omega_g) = \frac{2\pi}{\hbar} J_{cv}(\hbar \omega_g) (f_v - f_c), \]  

\[ J_{cv}(\hbar \omega_g) = \frac{2}{(2\pi)^3} \int d^3k \delta(\varepsilon_c(k) - \varepsilon_v(k) - \hbar \omega_g). \]  

In the NEGF case, joint density of states and occupation cannot be separated, but are both contained in the Keldysh Green’s functions

\[ J_{cv}^{GF}(\hbar \omega_g) = \frac{2}{(2\pi)^3} \int \frac{dE}{\hbar} \int \frac{d^3k}{2\pi^3} [G_v^<(k; E - \hbar \omega_g) G_c^>(k; E) - G_v^>(k; E - \hbar \omega_g) G_c^<(k; E)] \]  

\[ = \tilde{\mathcal{P}}_{cv,0}(0, \hbar \omega_g), \]  

which via \( \tilde{\mathcal{P}}_{cv,0} = \mathcal{P}_{cv,0}^> - \mathcal{P}_{cv,0}^< \) is related to the Keldysh components of the free-carrier interband polarization function

\[ \mathcal{P}_{cv,0}^<(\mathbf{q}, E) = \frac{2}{(2\pi)^3} \int \frac{d^3k}{2\pi^3} G_v^<(k; \mathbf{q} + \mathbf{k}, E) \times G_c^>(k; \mathbf{q} - \mathbf{k}, E - E). \]  

This expression is valid in any situation that can be described in terms of single-particle Green’s functions. The deviations from the FGR result are marginal in the quasi-equilibrium case and in absence of further interactions beyond electron-light coupling and of sources of non-equilibrium, which both modify the Green’s functions, causing them to differ from the expressions given in Eqs. (19) and (20). It is straightforward to show that inserting the latter expressions in (30) reproduces (27), and for the special case of spherical bands, the joint density of states is given by the well-known analytical expression

\[ J_{cv}^{dir}(\hbar \omega_g) = \frac{(2m^* e)^{3/2}}{\pi^2 \hbar^3} \sqrt{E_g - E}, \]  

where \( m^* e \) is the reduced effective mass and \( E_g \) is the direct band gap.

2. Phonon assisted interband transitions

Even in the case of a direct semiconductor discussed above, the presence of phonons can have a considerable effect on the optical transition rates as new excitation paths become available, i.e. the phonons strongly increase the number of initial-final state pairs separated by a given transition energy. However, comparing to the direct transition, this enhancement of the joint density of states is overcompensated by the fact that the transitions become much more unlikely due to the need for coupling to a suitable phonon and the detour via the virtual state. As a consequence, phonon assisted transitions may be neglected in direct bulk semiconductors where crystalline momentum is conserved. Obviously, the situation is completely different in indirect bulk semiconductors, where no direct transitions are possible. There, at lowest nonvanishing order, four different excitation processes exist (Fig. 1):

\[ S_{1+} : 1. \text{ photon absorption, 2. photon absorption,} \]

\[ S_{1-} : 1. \text{ photon emission, 2. photon absorption,} \]

\[ S_{2+} : 1. \text{ photon absorption, 2. photon absorption,} \]

\[ S_{2-} : 1. \text{ photon absorption, 2. photon emission.} \]

![FIG. 1. Possible excitation pathways for optical interband transitions in indirect semiconductors on the example of bulk silicon.](image)

To exemplify the inclusion of the phonon scattering in the NEGF description of optical interband transitions, we will in the following focus on processes \( S_{2\pm}. \) The FGR transition rate for these processes are obtained from second order perturbation theory as

\[ R_{abs,\pm}^{FGR}(\lambda, \mathbf{q}) = \frac{2\pi}{\hbar} N^\gamma_\lambda \sum_{\mathbf{k}, \mathbf{k}', \mathbf{Q}} [M_{\lambda Q}^{\alpha\beta}(\mathbf{k}, \mathbf{k}')]^2 [M_{\lambda Q}^{\alpha\beta}(\mathbf{k}, \mathbf{k}')]^2 \times \delta(\varepsilon_c(\mathbf{k}) - \varepsilon_v(\mathbf{k}) - \hbar \omega_{\mathbf{q}}) \times \int \frac{d^3k}{2\pi^3} f_v(\mathbf{k}) [1 - f_c(\mathbf{k})]. \]  

(34)
Here, $M_{\Lambda,Q}^{ph}$ encodes the matrix element for coupling of electrons to the phonon mode $(\Lambda, Q)$ with energy $h\Omega_{\Lambda,Q}$ and occupation given by the Bose-Einstein distribution $N_{\Lambda,Q}^{ph} = (e^{h\Omega_{\Lambda,Q}} - 1)^{-1}$, $\beta = (k_B T)^{-1}$ at lattice temperature $T$. In analogy to the direct case, with the previously used approximations for the optical matrix elements and the additional assumptions

$$\left| \varepsilon_c(k_v) - \varepsilon_v(k_c) - h\omega_q \right| \approx |E_{g0} - h\omega_q|, \quad M_{\Lambda,Q}^{ph}(k_v, k_c) \approx M_{\Lambda}^{ph,\delta}(k_c \rightarrow k_v + Q)$$

we can write the (phonon-assisted) absorption coefficient

$$\alpha^{ind}(h\omega_{\gamma}) = \frac{N^{\gamma \gamma}(h\omega_{\gamma})^2}{|E_{g0} - h\omega_{\gamma}|^2} \sum_{\Lambda} M_{\Lambda}^{ph} \times \sum_{s=\pm} \left[ N_{\Lambda}^{ph} + \frac{1}{2} - s \frac{1}{2} \right] J^{ind}_{cv,\pm}(h\omega_{\gamma}),$$

but where now the joint density of states for indirect transitions is used in (27),

$$J^{ind}_{cv,\pm}(h\omega_{\gamma}) = \frac{2}{(2\pi)^3} \int d^3 k_1 \int d^3 k_2 \delta (\varepsilon_c(k_1) - \varepsilon_v(k_2) - h\omega_q \mp \hbar\Omega_{\Lambda,k} - k_2),$$

which for spherical bands and a single phonon frequency $\Omega$ may be simplified to

$$J^{ind}_{cv,\pm}(h\omega_{\gamma}) = \frac{(m^*_c m^*_v)^2}{(2\pi)^3} (h\omega_q - E_g \mp \hbar\Omega)^2,$$

where $E_g$ denotes the (indirect) band gap.

In the following, the phonon-assisted absorption rate shall be derived within the NEGF formalism, starting from the expression for the absorption rate, but where now the Green’s functions contain the contributions due to the electron-phonon scattering. Since the Green’s functions and interaction self-energies are evaluated in a self-consistency iteration process, all possible single phonon processes are included to all orders, i.e. the Green’s functions contain contributions from a number of scattering processes that corresponds to the number of self-consistency iteration steps. It is thus via this self-consistent computation that phonon assisted optical transitions are enabled, even though the self-energies themselves are only on the level of a first order self-consistent Born approximation, i.e. do not include the combination of both electron-phonon and electron-photon scattering at the same time. In Fig. 2, this procedure is shown for the technologically relevant example of indirect interband absorption of photons in silicon, where zone-boundary phonons provide the wave-vector difference in a $\Gamma - X$ inter-valley scattering process.

We start again from the general expression for the in-scattering rate, which in this case reads

$$R_{in} = \frac{2}{V} \int \frac{dE}{2\pi \hbar} \sum_k \sum_{\lambda,\sigma} \frac{\Delta G_{\sigma}(k; E) G_{\sigma}^\gamma(k; E)}{G_{\lambda X}^\gamma(k; E)}.$$  

(40)

Assuming a photon-first indirect process ($S_{2\pm}$), the corresponding expression for $\Gamma - X$-scattering is inserted for the self-energy (see Eq. (A28) in App. A, leading to the equivalent of Eq. (14) ($\sigma = L, T, O$; phonon mode),

$$R_{in} = \frac{2}{V} \int \frac{dE}{2\pi \hbar} \sum_k \sum_{\lambda,\sigma} |M_{\sigma}^{ph}(\Omega_{\sigma})|^2 \left[ N_{\sigma}^{ph} G_{\sigma}(k; E) - h\Omega_{\sigma} \right] + (N_{\sigma}^{ph} + 1) G_{\sigma}^\gamma(k; E) + \hbar\Omega_{\sigma} G_{\sigma}^\gamma(k; E).$$

(41)

In the next step, the Keldysh equation for electron-photon interaction is used to replace the lesser $\Gamma_{c,GF}$,

$$G_{\sigma}^{\beta}(k; E) = G_{\sigma}^{R}(k; E) \Sigma_{\sigma}^{\gamma\beta}(k; E) G_{\sigma}^{\Lambda}(k; E)$$

(42)

providing the modal absorption rate

$$R_{abs}(\lambda, q) = \frac{2}{V} \int \frac{dE}{2\pi \hbar} \sum_k \sum_{\lambda,\sigma} |M_{\sigma}^{ph}(\Omega_{\sigma})|^2 |M^{\gamma}(k, \lambda, q)|^2 \times N_{\lambda,q}^{\gamma} \left[ N_{\sigma}^{ph} G_{\sigma}^{R}(k; E) - h\Omega_{\sigma} \right] + G_{\sigma}^{R}(k; E) N_{\sigma}^{ph} G_{\sigma}^{R}(k; E) - h\Omega_{\sigma} - h\omega_q \right] \times G_{\sigma}^{\Lambda}(k; E) + \hbar\Omega_{\sigma} - \hbar\omega_q) G_{\sigma}^{\Lambda}(k; E) - h\Omega_{\sigma})$$

(43)

Again, this is to be compared to the FGR result, which, for the same electron-photon and electron-phonon interaction Hamiltonian terms, follows from second-order perturbation theory to

$$R_{FGR}^{abs}(\lambda, q) = \int \frac{2\pi}{\hbar} \sum_{k_{c,\lambda}} \sum_{\sigma} |M_{\sigma}^{ph}(\Omega_{\sigma})|^2 |M^{\gamma}(k_{c}, \lambda, q)|^2 N_{\lambda,q}^{\gamma}$$

$$\times \left[ N_{\sigma}^{ph} \delta (\varepsilon_{\sigma}(k_{c}) - \varepsilon_{\sigma}(k_{c}) - h\omega_q - h\Omega_{\sigma}) + (N_{\sigma}^{ph} + 1) \delta (\varepsilon_{\sigma}(k_{c}) - \varepsilon_{\sigma}(k_{c}) - h\omega_q + h\Omega_{\sigma}) \right]$$

$$\times f_{\sigma}(k_{c}) (1 - f_{\sigma}(k_{c})),$$

(44)

Now, inserting the non-interacting equilibrium expressions for the lesser, greater and retarded GF in (43) provides the expression.
\[ R_{abs}(\lambda, q) = \frac{2}{V} \int \frac{dE}{2\pi \hbar} \sum_{k} \sum_{Q, \sigma} [M^p_{\sigma}(\Omega_{Q})]^2 [M^T(\kappa, \lambda, q)]^2 N_{\chi}^\gamma \left\{ \frac{N^p_{\sigma} i f_{\Gamma, \gamma}(E - \hbar \Omega_{\sigma} - \hbar \omega_{Q}) 2\pi \delta(E - h\Omega_{\sigma} - \hbar \omega_{Q} - \varepsilon_{\Gamma, \gamma}(Q))}{|E - h\Omega_{\sigma} - \hbar \omega_{Q} - \varepsilon_{\Gamma, \gamma}(Q) + i\eta_{\theta}|^2} \right\} i\delta(E - \varepsilon_{\chi, \kappa}(k)) \right\}, \tag{45} \]

which, upon performing the energy integration and with \( Q = k_x, k = k_z \), reproduces again the FGR result given in \( (44) \).

For the numerical implementation, a simple three-band effective mass model for the electronic structure of silicon is used. The \( X \) electrons are described by an multi-valley picture with identical values for transverse and longitudinal effective mass, and the \( \Gamma_v \) holes as well as the virtual \( \Gamma_c \) states used in the indirect transitions are modelled by single effective mass bands. The holes are modelled by a single effective masses corresponding to heavy and light holes. The band structure and interaction parameters used in the numerical examples are listed in Tab. I.

Fig. 3 displays the close agreement between NEGF and FGR for equilibrium bulk absorption close to the indirect band edge and the characteristic fingerprints of the involved TO and LA phonon modes.

### C. Thin film devices

In the case of ultra-thin-film and especially hetero-multi-layer devices, translational invariance does no longer apply in growth direction. For such a system, the appropriate representation of the field operators has the form

\[ \hat{\Psi}_b(\mathbf{r}, t) = \sum_{\mathbf{k}, i} \psi_{ib\mathbf{k}, i}(\mathbf{r}) \hat{c}_{ib\mathbf{k}, i}(t), \tag{46} \]

with the basis functions

\[ \psi_{in\mathbf{k}, i}(\mathbf{r}) = \varphi_{in\mathbf{k}, i}(\mathbf{r}) u_{n\mathbf{k}_0}(\mathbf{r}), \tag{47} \]

where \( \varphi_{in\mathbf{k}, i}(\mathbf{r}) \) is the envelope basis function for discrete spatial (layer) index \( i \) (longitudinal) and transverse momentum \( \mathbf{k}_i \), \( u_{n\mathbf{k}_0} \) is the Bloch function of bulk band \( n \), centered on \( \mathbf{k}_0 \). In the case of a system with large transverse extension, the envelope basis function can be written as

\[ \varphi_{in\mathbf{k}, i}(\mathbf{r}) = \frac{e^{i\mathbf{k}_i \cdot \mathbf{r}}}{\sqrt{A}} \chi_i(z), \tag{48} \]

where \( \mathbf{r} = (x, y), A \) is the cross sectional area and \( \chi_i \) is the longitudinal envelope basis function. For the latter, finite element shape functions are a popular choice. Here, we will use a simple finite difference basis equivalent to a separate single band tight-binding approach for each band. \( (49) \)

\[ \chi_i(z) = [\theta(z - z_i) - \theta(z - z_{i+1})] / \sqrt{A}. \tag{49} \]

In the above basis, the Green’s functions and self-energies have the potentially non-local representation (\( O = G, \Sigma \))

\[ O_{b, b'}(\mathbf{r}, \mathbf{r}'; E) = \sum_{\mathbf{k}, i} \sum_{i, j} \psi_{ib\mathbf{k}, i}(\mathbf{r}) O_{b, j\mathbf{k}, i}(\mathbf{r}; E) \psi_{jb\mathbf{k}, j}(\mathbf{r}'), \tag{50} \]

where the contour-ordered steady-state Green’s functions are now defined as

\[ O_{b, j\mathbf{k}, i}(\mathbf{r}; t - t') = \frac{1}{i\hbar} \left\langle \hat{T}_c \left\{ \hat{c}_{ib\mathbf{k}, i}(t) \hat{c}_{j\mathbf{k}, i}^\dagger(t') \right\} \right\rangle. \tag{51} \]

---

**TABLE I. Material parameters used in simulations**

| \( m_i^c \) | \( m_i^x \) | \( m_i^v \) | \( E_{g, \Gamma-X} \) | \( E_{g, \Gamma-\Gamma} \) | \( E_{c, \sigma}^2 / m_0 \) | \( \sigma \) | \( \hbar \Omega_{\sigma} \) | \( D_v K_{\sigma} \) |
|---|---|---|---|---|---|---|---|---|
| 0.3 | 0.3 | 0.54 | 3.1 eV | 1.17 eV | 4 eV |
| \( (\Gamma-X)_1 \) LA | 18.4 meV | 2.45 \times 10^8 eV/cm |
| \( (\Gamma-X)_2 \) TO | 57.6 meV | 0.8 \times 10^8 eV/cm |

---

**FIG. 3.** Photon absorption rate for indirect optical transitions in intrinsic bulk silicon: FGR and NEGF rates are in close agreement, since the correction fo the conduction band DOS due to the phonon-mediated \( \Gamma - X \) scattering is weak.
The conservation law corresponding to Eq. (4) for the divergence of the electron charge current between model layers $i-1$ and $i$ becomes

$$\frac{J_i - J_{i-1}}{\Delta} = -\frac{2e}{\hbar A \Delta} \sum_k \int dE \left[ \Sigma^R G^< - G^R \Sigma^< + \Sigma^< G^A - G^< \Sigma^A \right]_{i,i} (52)$$

$$\equiv R_{rad}(z_i), \quad (53)$$

where $R_{rad}(z)$ is again the local radiative rate. Making use of the cyclic property of the trace, the total radiative rate follows as

$$R_{rad} = \frac{2}{\hbar A} \int_B dE \frac{2\pi}{2\pi} \text{Tr} \left\{ \sum_k \left[ \Sigma_{\gamma,\gamma'}^<(k; E) G^{>}(k; E) - \Sigma_{\gamma,\gamma'}^>(k; E) G^{<}(k; E) \right] \right\} \equiv \Delta \sum_i R_{rad}(z_i) = \sum_i (J_i^B - J_i^{B-1}) \quad (54)$$

$$\equiv J_N^B - J_1^B \equiv \left\{ \begin{array}{l} J_N, \quad B = CB, \\ J_1, \quad B = VB. \end{array} \right. \quad (55)$$

In the case of the indirect gap material the total radiative rate is

$$R_{\gamma \gamma'} = \frac{2}{\hbar A} \int_{E_{\gamma'}} dE \frac{2\pi}{2\pi} \text{Tr} \left\{ \sum_{k_{\gamma'}} \left[ \Sigma_{\gamma,\gamma'}^{<}(k_{\gamma'}; E) G_{\gamma'}^{>}(k_{\gamma'}; E) - \Sigma_{\gamma,\gamma'}^{>}(k_{\gamma'}; E) G_{\gamma'}^{<}(k_{\gamma'}; E) \right] \right\}, \quad (57)$$

and the inter-valley phonon scattering rate reads

$$R_{ep,\Gamma-X} = \frac{2}{\hbar A} \int_{E_{\Gamma-X}} dE \frac{2\pi}{2\pi} \text{Tr} \left\{ \sum_{k_{\gamma}} \left[ \Sigma_{\gamma,\gamma'}^{<}(k_{\gamma}; E) - \Sigma_{\gamma,\gamma'}^{>}(k_{\gamma}; E) G_{\gamma'}^{<}(k_{\gamma}; E) \right] \right\} \times G_{\Gamma-x}^{>}(k_{\gamma}; E) \quad (58)$$

The optical rate and interband current as a function of self-consistency iteration steps is shown in Fig. 4. The convergence is fast due to the low carrier density in the absorbing quasi-intrinsic region.

Fig. 5 shows the indirect optical absorption for an ultra-thin bipolar junction with high doping, the global absorption coefficient starts to deviate from the bulk value due to the strong internal field. This so-called Franz-Keldysh effect is displayed in Fig. 6. Fig. 7 shows the energy resolved local generation rate in the virtual $\Gamma$-states and resulting local photocurrent spectrum in the X-valley band at a photon energy $E_{\text{photon}} = 1.17$ eV and very low internal field $F = 2$ kV/cm. The generation is relatively uniform throughout the device, as well as the increase of electron and hole current components towards the respective contacts. Fig. 8 shows the corresponding situation for the diode with strong internal field $F = 169$ kV/cm. In this case, the photogeneration and current contributions are distributed over a large energy range, the latter due to the absence of efficient relaxation mechanisms that would confine current flow closer to the band edge. In the vicinity of the $n$-contact, the high electron concentration results in large electron-phonon intraband scattering current contributions (Fig. 9), which due to the opposite signs of in- and outscattering components cancel upon energy integration over the band. The net current can thus be attributed exclusively to interband transitions, and the local sum of electron and hole contributions is perfectly conserved, as shown in Fig. 9.
Fig. 6. (a) Band profile of a silicon p-i-n junction from the self-consistent solution of the NEGF-Poisson equations with the indicated doping levels from $N_d = 10^{16}$ cm$^{-3}$ to $N_d = 10^{18}$ cm$^{-3}$. The corresponding fields in the intrinsic region vary from $F = 2$ kV/cm to $F = 169$ kV/cm. The equilibrium Fermi level lies at 1.1 eV. (b) Near band gap optical absorption rate in the intrinsic region of a silicon p-i-n junction, for various values of the built-in field corresponding to different doping levels.

Since the photogeneration rate is intimately related to the $\Gamma - X$ intervalley electron-phonon scattering rate via the self-consistent computation of Green’s functions and self-energies, and the photocurrent results from photogenerated excess charge that is transferred via the scattering process with phonons to the extended $X$-valley states, the rates of this phonon-mediated charge transfer are expected to show the signature of the photogeneration process. Indeed, if the rate of electron scattering out of (into) the virtual $\Gamma_c$-states into (out of) the $X$-states is considered, there is a large net scattering from $\Gamma_c$ to $X$ under illumination, while in- and outscattering rates are balanced in the dark. This is shown in Fig. 10 for the case of the low-doping diode where the intrinsic region

Fig. 7. (Color online) Energy resolved local generation rate in the virtual $\Gamma_c$-state and resulting local photocurrent spectrum in the $X$-valley band at very low internal field $F = 2$ kV/cm. The grey lines indicate the band edges ($X$-valley for electrons).

Fig. 8. (Color online) Same as Fig. 7, but for a strong internal field of $F = 169$ kV/cm. The large deviation close to the $n$-contact is due to the electron-phonon intraband scattering current that vanishes upon energy integration over the band (white=negative, black=large positive, see Fig. 9a).

Fig. 9. (Color online) (a) Current spectrum of $X$-valley electrons close to the $n$-contact. (b) Integrated net current for electrons (el) and holes (hl) in the high-field device. The total current is perfectly conserved over the whole device.
FIG. 10. (Color online) Phonon mediated electron transfer rate between virtual (Γ\textsubscript{c}) and extended (X) conduction band states (a) in the dark and (b) under illumination with \(E_{\text{phot}} = 1.17\) eV, for the low-doping diode, where the intrinsic region extends over the whole device. While in- and out-scattering are balanced in the dark, there is a strong net charge transfer from Γ\textsubscript{c} to X under illumination, and the inscattering rate reflects the spectral pattern of the photogeneration.

FIG. 11. (Color online) Phonon mediated electron transfer rate between virtual and extended conduction band states (a) in the dark and (b) under illumination with \(E_{\text{phot}} = 1.17\) eV in the center of the short intrinsic region of the high-doping diode. Closer to the contacts, the scattering rate contributed by the photogeneration is masked by the large intraband relaxation rate.

extends over the entire device. In the case of strong doping, the electron-phonon scattering rate in the dark is much stronger close to the contacts, such that the effect becomes only visible in the central, intrinsic region of the junction. There, the phonon-related charge transfer rate again reflects the local photogeneration rate, as displayed in Fig. 11.

III. CONCLUSIONS

We have presented a novel theoretical approach to the description of phonon-mediated photogeneration in indirect semiconductors. While compatible with the standard Fermi-Golden-Rule approach in the quasi-equilibrium bulk limit, its range of validity extends to quantum transport in open systems involving arbitrary heterostructure states far from equilibrium and the effects of non-locality in the scattering processes, which are important aspects of advanced photovoltaic and light emitting devices, where often spatial and spectral resolution are required to gain access to a deeper understanding of the device characteristics. Thanks to this versatility, the theoretical framework lends itself to the modelling of indirect semiconductor based nanostructures with potential applications in a multitude of optoelectronic devices, such as silicon-based quantum well solar cells.

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Appendix A: Interaction self-energies

The self-energies as defined by the Dyson equations\(^\text{(3)}\) and\(^\text{(4)}\) encode the renormalization of the charge carrier Green’s functions due to the interactions with photons and phonons, i.e., generation, recombination and relaxation processes. The self-energies due to interactions can be evaluated from the perturbation expansion of the non-equilibrium statistical average defining the interacting NEGF, using either Wick’s theorem or Feynman diagrams. In the first case, the (contour ordered) self-energy \(\Sigma\) is derived from a perturbation expansion of the relaxation processes. The self-energies due to interactions can be evaluated from the perturbation expansion of the exponential in the definition of the contour ordered self-energy \(\Sigma\) is derived from a perturbation expansion of the relaxation processes. For numerical evaluation, the Hamiltonian is reformulated in a suitable representation using the field operators\(^\text{(10)}\) and used in the derivation of the corresponding self-energy expectation value of single-carrier operators,

\[
G_{\alpha,\alpha'}(k; t, t') = -\frac{i}{\hbar} \left\langle \mathcal{T}_C \left\{ e^{-\frac{i}{\hbar} \int_C ds H'(s) \hat{c}_{\alpha,k}(t) \hat{c}_{\alpha',k}(t') } \right\} \right\rangle, \tag{A1}
\]

with \(\alpha = n\) and \(k = (k_x, k_y, k_z)\) (bulk) or \(\alpha = i,n\) and \(k = (k_x, k_y) \equiv k_{\parallel}\) (thin film). In the following, the Hamiltonian terms for the perturbative interaction of electrons with photons and phonons shall be discussed and used in the derivation of the corresponding self-energy expressions for bulk and thin-film systems.

1. Interaction Hamiltonian

a. Electron-photon interaction For the electron-photon interaction, the perturbation Hamiltonian is given via the linear coupling to the vector potential operator of the electromagnetic field \(\hat{A}\),

\[
\hat{H}_{ep} = -\frac{e}{m_0} \hat{A} \cdot \hat{p}, \tag{A2}
\]

with \(\hat{p}\) the momentum operator and

\[
\hat{A}(r, t) = \sum_{\lambda, q} A_0(\lambda, q) b_{\lambda, q}(t) A_0^\dagger(\lambda, -q) \tilde{b}_{\lambda, -q}(t), \tag{A3}
\]

\[
A_0(\lambda, q) = \frac{\hbar}{\sqrt{2\epsilon_0 V \omega_q}} e_{\lambda q}, \tag{A4}
\]

where \(e_{\lambda q}\) is the polarization of the photon with wave vector \(q\) and energy \(\hbar \omega_q\) added to or removed from photon mode \((\lambda, q)\) by the bosonic creation and annihilation operators

\[
\hat{b}_{\lambda, q}(t) = \tilde{b}^\dagger_{\lambda, q} e^{i \omega_q t}, \quad \hat{b}_{\lambda, q}(t) = \tilde{b}_{\lambda, q} e^{-i \omega_q t}, \tag{A5}
\]

and \(V\) is the absorbing volume.

For numerical evaluation, the Hamiltonian is reformulated in a suitable representation using the field operators given in\(^\text{(10)}\) (bulk) and\(^\text{(40)}\) (thin film), respectively,

\[
\mathcal{H}_{ep}(t) = \int d^3 r \hat{\Psi}^\dagger(r, t) \hat{H}_{ep} \hat{\Psi}(r, t). \tag{A6}
\]

In the bulk case, the above expression results in\(^\text{(23)}\)

\[
\mathcal{H}_{ep}(t) = \sum_{q, \lambda} \sum_{m,n} \sum_{k,k'} \mathcal{M}^\gamma_{n,m}(k, k', q, \lambda) \times \hat{\epsilon}_{nk}(t) \hat{c}_{mk} \left( b_{\lambda, q} e^{i \omega_q t} + \tilde{b}^\dagger_{\lambda, -q} e^{-i \omega_q t} \right), \tag{A7}
\]

where the matrix element for interband transitions \((n \neq m)\) is obtained from a \(\textbf{k} \cdot \textbf{p}\) - type approximation\(^\text{[11]}\)

\[
\mathcal{M}^\gamma_{n,m}(k, k', \lambda, q) = -\frac{e}{m_0} A_0(\lambda, q) \times \int d^3 r \psi_{\lambda k}(r) (\epsilon_{\lambda q} \cdot \hat{p}) \psi_{\lambda k'}(r) \tag{A8}
\]

\[
\approx -\frac{e}{m_0} A_0(\lambda, q) \delta(k' + q - k) (\epsilon_{\lambda q} \cdot \textbf{p}_{nm}), \tag{A9}
\]

with the Bloch function momentum matrix element

\[
\textbf{p}_{nm} = \int_{\Omega} d^3 \tilde{r} \tilde{u}_{nk}(\tilde{r}) \tilde{u}_{nm}(\tilde{r}), \tag{A10}
\]

where \(\Omega\) denotes the unit-cell volume. This gives the final bulk expression

\[
\mathcal{H}_{ep}(t) = \sum_{q, \lambda} \sum_{m,n} \sum_{k} \mathcal{M}^\gamma_{n,m}(k, q, \lambda) \times \hat{\epsilon}_{nk}(t) \hat{c}_{mk} \left( b_{\lambda, q} e^{i \omega_q t} + \tilde{b}^\dagger_{\lambda, -q} e^{-i \omega_q t} \right). \tag{A11}
\]

For devices with broken translational invariance in the transport dimension, the representation of the electron-photon Hamiltonian\(^\text{(A2)}\) in the real-space effective mass basis\(^\text{(17)}\) acquires the similar from

\[
\mathcal{H}_{ep}(t) = \sum_{q, \lambda} \sum_{m,n} \sum_{k_{\parallel}} \mathcal{M}^\gamma_{in, jm}(k_{\parallel}, q, \lambda) \times \hat{\epsilon}_{nk}(t) \hat{c}_{mk} \left( b_{\lambda, q} e^{i \omega_q t} + \tilde{b}^\dagger_{\lambda, -q} e^{-i \omega_q t} \right), \tag{A12}
\]

where

\[
\mathcal{M}^\gamma_{in, jm}(k_{\parallel}, q, \lambda) \approx -\frac{e}{m_0} A_0(\lambda, q) M_{ij}(q_{\parallel}) (\epsilon_{\lambda q} \cdot \textbf{p}_{nm}), \tag{A13}
\]

with

\[
M_{ij}(q_{\parallel}) = \int d z \chi_{i}^* (z) e^{i q_{\parallel} z} \chi_j(z) = e^{i q_{\parallel} z_i} \delta_{ij}. \tag{A14}
\]

b. Electron-phonon interaction The vibrational degrees of freedom of the system are described in terms of the coupling of the force field of the electron-ion potential \(V_{ei}\) to the quantized field \(\hat{U}\) of the ionic displacement\(^\text{[23]}\),

\[
\hat{H}_{ep}(r, t) = \sum_{L, \kappa} \hat{U}( \textbf{L} + \kappa, t) \cdot \nabla V_{ei}(r - (\textbf{L} + \kappa)), \tag{A15}
\]
with the displacement field given by the Fourier expansion
\[ \hat{u}_\alpha(L\kappa, t) = \sum_{\Lambda, Q} U_{\alpha \kappa}(\Lambda, Q) e^{iQ \cdot (L + \kappa)} \{ \hat{a}_{\Lambda, Q}(t) + \hat{a}_{\Lambda, Q}^\dagger(t) \}, \]
(\alpha = x, y, z),

(A16)

where the ion equilibrium position is \( L + \kappa \), with \( L \) the lattice position and \( \kappa \) the relative position of a specific basis atom at this lattice site, and \( \hat{a}_{\Lambda, Q}, \hat{a}_{\Lambda, Q}^\dagger \) are the bosonic creation and annihilation operators for a (bulk) phonon mode with polarization \( \Lambda \) and wave vector \( Q \) in the first Brillouin zone. The potential felt by electrons in heterostructure states due to coupling to bulk phonons can thus be written as
\[ \hat{H}_{ep}(r, t) = \frac{1}{\sqrt{V}} \sum_{\Lambda, Q} U_{\alpha \kappa}(\Lambda, Q) e^{iQ \cdot r} \{ \hat{a}_{\Lambda, Q}(t) + \hat{a}_{\Lambda, Q}^\dagger(t) \}, \]
(A17)

where \( r \) is the electron coordinate, and \( U_{\alpha \kappa} \) are related to the Fourier coefficients of the electron-ion potential.

The effective-mass Hamiltonian for electron-phonon interaction is obtained from (A17) in analogy to the electron-photon interaction, with the bulk result
\[ \hat{H}_{ep}(t) = \sum_{Q, \Lambda, n, k, i, j} \mathcal{M}_{ep}(Q, \Lambda) \hat{c}_{i, k, j}^\dagger(t) \hat{c}_{i, k, j} - \hat{Q}(t) \times \{ \hat{a}_{\Lambda, Q} e^{-i\Omega_{\Lambda, Q} t} + \hat{a}_{\Lambda, Q}^\dagger e^{i\Omega_{\Lambda, Q} t} \}, \]
(A18)

with
\[ \mathcal{M}_{ep}(Q, \Lambda) = \frac{U_{\alpha \kappa}}{\sqrt{V}}, \]
(A19)

and the thin-film expression
\[ \hat{H}_{ep}(t) = \sum_{Q, \Lambda, n, k_1, i, j} \mathcal{M}_{ep}(Q, \Lambda) \hat{c}_{i, k_1, j}^\dagger(t) \hat{c}_{i, k_1, j} - \hat{Q}(t) \times \{ \hat{a}_{\Lambda, Q} e^{-i\Omega_{\Lambda, Q} t} + \hat{a}_{\Lambda, Q}^\dagger e^{i\Omega_{\Lambda, Q} t} \}, \]
(A20)

where
\[ \mathcal{M}_{ep}(Q, \Lambda) = \frac{U_{\alpha \kappa}}{\sqrt{V}} e^{iQ \cdot z_1}. \]
(A21)

For the \( \Gamma - X \) intervalley scattering considered in the present discussion, the coupling reads
\[ |U_{\alpha \kappa}|^2 = \frac{\hbar (D_i r_i K_i)^2}{2 \rho \Omega_\sigma}, \]
(A22)

where \( \sigma \) labels the phonon mode, \( D_i \) is the associated deformation potential and \( K_i \) denotes the momentum transfer required for the scattering between two valleys.

2. Self-energy

At this stage, any renormalizing effect of the electronic system on the photons and phonons is neglected, i.e.

the coupling to the bosons corresponds to the connection to corresponding equilibrium reservoirs. While this treatment is generally a good approximation in the case of phonons, it is valid for the coupling to the photonic systems only in the case of low absorption, i.e. weak coupling or very short absorber length. The equilibrium propagators for non-interacting photons and phonons in isotropic media have the common form \((\alpha = \gamma, p)\)
\[ D^{\alpha,\gamma}(q; E) = -2\pi i \left[ N_{\lambda, q}^\alpha \delta(E + \hbar \omega_q) + (N_{\lambda, q}^\alpha + 1) \delta(E - \hbar \omega_q) \right], \]
(A23)

\[ D^{\alpha,R/A}(q; E) = \frac{1}{E - \hbar \omega_q + i\eta} - \frac{1}{E + \hbar \omega_q + i\eta}. \]
(A24)

In the above expressions, \( N_{\lambda, q}^\alpha \) denotes the occupation of the respective equilibrium boson modes, with the photon occupation given by the Bose-Einstein distribution at lattice temperature \( T \),
\[ N_{\lambda, Q}^\gamma = (e^{\beta \hbar \Omega_{\lambda, Q} - 1})^{-1}, \quad \beta = (k_B T)^{-1}, \]
(A25)

and the photon occupation \( N_{\lambda, Q}^\gamma \) introduced in Sec. III B 1. Inserting these propagators the Fock-term of the a general electron-boson self-energy in the first self-consistent Born approximation provides the steady-state components \((\alpha = \gamma, p)\)
\[ \Sigma^{\gamma\gamma}_{\alpha \\alpha}(k; E) = \sum_{\lambda, \gamma} \mathcal{M}^{\gamma\alpha}(k, \lambda, \gamma) \left[ N_{\lambda, q}^\alpha G^{\gamma\gamma}(k; E + \hbar \omega_{\lambda, q}) + (N_{\lambda, q}^\alpha + 1) G^{\gamma\gamma}(k; E - \hbar \omega_{\lambda, q}) \right] \times \mathcal{M}^{\gamma\alpha}(k, q, \gamma), \]
(A26)

and
\[ \Sigma^{\gamma\gamma}_{\alpha \\alpha}(k; E) = i \int \frac{dE'}{2\pi} \left[ \Sigma^{\gamma\gamma}_{\alpha \\alpha}(k; E') - \Sigma^{\gamma\gamma}_{\alpha \\alpha}(k; E') \right] \]
(A27)

with the same definitions of indices and momentum as in (A11). Using a constant mode specific coupling strength, the intervalley bulk (thin-film) electron-phonon scattering self-energy is further simplified to the diagonal form used in the simulations,
\[ \Sigma^{\gamma\gamma}_{\alpha \alpha}(k; E) = \sum_{\sigma, b' \neq b} \left[ \frac{h (D_i r_i K_i)^2}{2 \rho \Omega_\sigma} f_{b'}(\delta_{i, j}) \right] \times \int \frac{d\Omega}{4\pi^2} \left[ N_{\sigma}^p G^{\gamma\gamma}(k; E + \hbar \Omega_\sigma) \right] \left[ N_{\sigma}^p + 1 \right] G^{\gamma\gamma}(k; E - \hbar \Omega_\sigma), \]
(A28)

where \( b, b' \in \{\Gamma_c, X\} \).
The dimensions are that of a volume rate, $[G, R] = m^{-3} s^{-1}$.