Ultra-fast carriers relaxation in bulk silicon following photo-excitation with a short and polarized laser pulse

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Abstract – We use an atomistic approach to provide a novel and ground-breaking interpretation of the ultra-fast carriers relaxation in a realistic material: bulk silicon. By comparing the results of ab initio simulations with recent two-photon photo-emission measurements we show that the description of the carrier relaxation in terms of \(L \rightarrow X\) inter-valley scattering is not correct. The ultra-fast dynamics measured experimentally is, instead, due to the scattering between degenerate \(L\) states that is activated by the non-symmetric population of the conduction bands induced by the laser field. This ultra-fast relaxation is, then, entirely due to the specific experimental setup and it can be interpreted by introducing a novel definition of the quasi-particle lifetimes in an out-of-equilibrium context.

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Introduction. – Silicon (Si) is a fundamental building block of semiconductors physics and microelectronics industry [1]. The miniaturization of Si-based devices to the nano-scale regime and the never-ending search for faster devices call for a deep understanding of the fundamental quantum-mechanical process that governs the ultra-short time dynamics of electrons and holes [2,3]. Most of the knowledge of the electronic and optical properties of Si remain, however, limited to the equilibrium regime. Only recently the development of ultra-fast laser pulses [4,5] has opened the opportunity to directly investigate the real-time dynamics in the non-equilibrium (NEQ) regime [6].

In real-time experiments the system is initially perturbed with a short laser pulse (the pump) followed by a second weaker pulse (the probe) that measures a specific physical observable like, for example, the absorption [7,8] or the photo-emission [9,10] spectra. The dynamics induced by the pump is, then, monitored by observing and analyzing the modifications induced in these observable by the presence of photo-excited carriers.

Despite the enormous experimental interest and the continuous development of more refined experimental techniques, the simulation methods are still based either on equilibrium first-principles approaches or on NEQ model Hamiltonians.

In the case of model Hamiltonians the relaxation paths can be calculated by using the NEQ Green’s function [2,11,12] or the Monte Carlo [13] methods. The advantage of these approaches is that the modifications induced by the presence of photo-excited charges are correctly taken into account in the evaluation of the scattering transitions. However \textit{ad hoc} parameters must be introduced to describe both the photo-excitation process and the specific material properties.

First-principles simulations are commonly performed by using time-dependent density-functional theory (DFT) [14] or equilibrium many-body perturbation theory [15]. In the first case the coupling with the laser pulse is described but the dissipative processes are neglected [16] or described in an empirical way [17]. In the second case the laser pulse is replaced by some \textit{ad hoc} initial guess of the carriers distribution and the scattering rates are derived from the equilibrium quasi-particle (QP) lifetimes [18].

In this letter we demonstrate that an \textit{ab initio} description of both the photo-excitation process and of the time dependence of the carrier scattering rates is essential for the correct interpretation of the experimental results. In particular we will reproduce the time evolution carriers in bulk Si, observed in a recent two-photons photo-emission
(2PPE) experiment [9,10], without relying on any parameter. We will highlight and discuss the different scattering channels created by the pump excitation showing the existence of two different decay regimes: an ultrafast regime due to transitions between energetically degenerated states, made possible by the symmetry breaking caused by the pump pulse; and a slower regime, where the carriers are taken to the minimum of the conduction band. In addition we will investigate the very fundamental problem of defining the lifetime of a photo-excited carrier. We will show that this definition differs from the equilibrium case, also at a very low carriers concentration. Moreover this lifetime time dependence will clearly mark the different regimes (fast and slow) of the carrier relaxation.

Method. – Our theoretical framework is based on a non-linear equation for the time-dependent occupation factors [19], \( f_i(t) \) (\( i \equiv \{n,k\} \)) represents a generic band, \( n \), and reciprocal space point, \( k \):

\[
\partial_t f_i(t) = \left| \partial_t f_i(t) \right|_{\text{pump}} + \left| \partial_t f_i(t) \right|_{\text{relax}}.
\] (1)

All ingredients of eq. (1) are calculated ab initio [20,21] by using DFT [22]. \( \partial_t f_i(t) \) \( \left| \text{relax} \right| \) could be derived by using a standard semi-classical Boltzmann approach [2]. Instead \( \partial_t f_i(t) \) \( \left| \text{pump} \right| \) requires an approach based on Green’s functions. This is, indeed, the why we use NEQ Green’s function, which describes the creation of carriers in presence of an a smeared out energy conservation, plus a coherent term approximation and ii) we use \( \gamma_{ij}^2(t) = 0 \) with \( \gamma_{ij}(t) = \eta \), a constant de-phasing of the polarization. such equation accounts for excitonic effects and, thus, ensures a very accurate description of the interaction with the laser pulse.

The term \( \partial_t f_i(t) \) \( \left| \text{pump} \right| \) originates from the commutator on the r.h.s. of eq. (2) and describes the creation of carriers by the interaction between the polarization and the external field. Thus, the equation for \( f_i(t) \) is coupled to the one for \( n_{ij} \), i.e. the polarization \( P(t) = -e \sum_{ij} r_{ij} n_{ij}(t) \), and the two must be propagated together.

The term \( \partial_t f_i(t) \) \( \left| \text{relax} \right| \) originates from the last two terms on the r.h.s. of (2). It describes the relaxation and dissipation of the photo-excited carriers. It ensures that the carriers dissipate energy and scatter with phonons and electrons in such a way to relax towards the lowest energy states and has the form

\[
\partial_t f_i(t) \left| \text{relax} \right| = -\gamma_i^{(e)}(t) f_i^{(e)}(t) + \gamma_i^{(h)}(t) f_i^{(h)}(t),
\] (3)

with \( f_i^{(e)} = f_i \) and \( f_i^{(h)} = 1 - f_i \) the electron and the hole occupations. In this case the dynamics is fully dictated by the electron and hole lifetimes, \( \gamma_i^{(e)} \) and \( \gamma_i^{(h)} \), include both an electron-phonon (e-p) and an electron-electron (e-e) contribution: \( \gamma_i^{(e)} = \gamma_i^{(e/h)}_{\text{e-e}} + \gamma_i^{(e/h)}_{\text{e-p}} \).

In the e-p channel we have

\[
\gamma_i^{(e/h)}_{\text{e-p}} \propto \sum_{j \neq k} N^2_{j} \left| g_{\lambda_{j} \rightarrow \lambda_{k}} \right|^2 \times P(\omega_{\lambda} \pm \Delta_{ij}) f_j^{(h/e)}(t).
\] (4)

while in the e-e channel

\[
\gamma_i^{(e/h)}_{\text{e-e}} \propto \sum_{j \neq k} |W_{i \rightarrow j}^{(e)}|^2 P(\Delta_{ij} - \Delta_{kl}) \times f_j^{(h/e)}(t) f_k^{(h/e)}(t) f_i^{(h/e)}(t).
\] (5)

Here \( \Delta_{ij} = \xi_{ij}^{QP} - \xi_{ij}^{QP} \) and \( \lambda \) represents a generic phonon mode with momentum \( q \) and branch \( \eta \). \( N_{\lambda}^{\pm} = (1 + N_{\lambda}(T)/2) \) and \( N_{\lambda}^{\pm} = N_{\lambda}(T)/2 \), with \( N_{\lambda}(T) \) the Bose distribution function at energy \( \omega_{\lambda} \) and temperature \( T \). \( W \) is the statically screened Coulomb interaction. The \( P \) functions represent a smeared energy conservation condition. Finally, \( g_{\lambda}^{\pm} \) are the screened ionic potential derivatives, calculated within density-functional perturbation theory [25,26]. We have verified that, in the present work, the e-e scattering channel have only a negligible effect on the carrier dynamics due to the very low density of the photo-excited electrons. This is in agreement with the results of recent simulations [18,27,28].

Equations (3)–(5) make clear the different role played by \( \gamma_i^{(e)} \) and \( \gamma_i^{(h)} \). They describe the elemental process where an initial electron (hole) is scattered in another electron (hole) emitting or absorbing an electron-hole pair (e-e channel) [3] or a phonon [3,29,30]. In the e-p case the energy is transferred back and forth from the electronic to the phonon sub-systems until a thermal equilibrium is
reached [2]. Thus, eq. (3) describes both relaxation and dissipation. In the (e) channel the term $\gamma^{(e)} f^{(e)}$ describes the removal of electrons from the state (i) and gives a negative contribution to $\partial_t f^{(e)}(t)$, while $\gamma^{(b)} f^{(b)}$ describes removal of holes, thus the filling of the state (i), and gives a positive contribution.

The two-photon photo-emission experiment. – In the 2PPE experiment we aim at describing [9,10], a Si wafer, oriented both along the [111] and the [100] surface directions, is excited with a laser pulse at room temperature. The photo-excited sample is, then, probed with a second laser pulse that photo-emits in the continuum the excited carriers. The photo-emitted current of electrons is measured as a function of the time delay between the pump and the probe. The final result is a measure of the time-dependent occupation of the valence bands (represented by the dots in the main frame of fig. 1). More specifically we consider the population of carriers near the point $L_1$, i.e. at $E \approx 1.6$ eV above the Fermi level\(^1\).

We thus consider the electronic real-time dynamics in bulk Si, under the action of a laser pulse, whose parameters are taken directly from the 2PPE experiment [9,10]. The pulse is centered at around 3.4 eV, with duration (the full width at half-maximum) of $\sigma = 110$ fs and intensity of $10^4$ kW/cm\(^2\) corresponding to an electric-field intensity of $6 \times 10^8$ V/m. The total fluence is $4 \times 10^{-8}$ KJ/cm\(^2\), which means that the pump field creates a carrier density of about $4.5 \times 10^{18}$ el/cm\(^3\), $\approx 1.8 \times 10^{-4}$ el/$\Omega$ where $\Omega$ is the unit cell. As the laser transferred momentum is negligible, on the scale of the solid unit cell size, all pumped carriers are excited vertically from the valence to the conduction bands along the $\Gamma$-L line.

In fig. 1 the experimental occupation of the $L_1$ state (dots) is compared with the solution of eq. (1) (continuous line). The agreement between theory and experiment is excellent. Both the gradual filling and emptying of the $L_1\prime$ state follows quite nicely the experimental curve. The theoretical results correctly describe the ultra-fast decay in the experiment [9,10], as a bulk property.

![Fig. 1: (Color online) The time-dependent occupations of the $L_1$ (green continuous line) and $L_1\prime$ (green dashed line) levels are compared with experimental data (black dots) from ref. [9]. The envelope of the laser pulse is also represented (orange shadow). In the inset (a) the carriers population (electrons in blue, holes in red) is superimposed on the band structure at $t = 0$. The blue arrow indicates the direction of the ultra-fast $L_1 \rightarrow L_1\prime$ scattering process. In the inset (b) the dynamics with a shorter laser pulse ($\sigma = 50$ fs) is compared with a gedanken experiment (black dot-dashed line) where the same density of carriers is placed by hand at $t = 0$ in the $\Gamma_{15}$ state. With the shorter pulse, the difference between the fast $L_1 \rightarrow L_1\prime$ scattering and the slower $L_1 \rightarrow X_1$ transitions becomes evident.

shows that the level $L_1\prime$ is not populated and most of the carriers are injected in the $L_1$ level.

The symmetry breaking mechanism. – This symmetry breaking mechanism is made possible by the external field ($U(t)$ operator in eq. (1)) which, in the 2PPE experiment, is polarized along the crystallographic [111] direction. This breaks the $L \leftrightarrow L\prime$ symmetry as the operation that moves $L$ in $L\prime$, although being a symmetry of the unperturbed system, does not leave the [111] direction unchanged (see also fig. 2). In practice this means that eq. (1) does not respect this symmetry anymore and k-points connected by a rotation that does not leave the pumping field unchanged are populated in a different way [31].

![Image](431x669 to 530x754)

Electrons are injected in the conduction band along the $\Gamma$-L line but not, for symmetry reasons, along the $\Gamma$-L\' line. This is clearly shown in fig. 1 where the population of the $L_1\prime$ state is represented with a dashed line. The $L_1\prime$ state is gradually filled while $L_1$ is depleted revealing that the real source of the ultra-fast decay observed experimentally is the $L_1 \rightarrow L_1\prime$ scattering.

This scattering is faster than any other scattering as it involves states with the same energy. Indeed fig. 1 shows

\(^1\)The time-dependent occupation of the L-points is interpreted, in the experiment [9,10], as a bulk property.
The surface states close in energy may exist. This explains the small deviation from the measured behavior which, however, corresponds.

A gedanken experiment. – To better disentangle the populations of the $L_1$ and $L'_1$ states which reach the same value at $t \approx 220$ fs. After this point both states decay simultaneously by using the slower $L \rightarrow X$ channel towards the conduction band minimum (CBM)\(^2\). After $t \approx 500$ fs the electrons (holes) can be already described by two Fermi distributions around the CBM VBM (= valence band maximum) with very high temperatures ($T_e \approx 9000$ K for electrons and $T_h \approx 2350$ K for hole). Once the Fermi distributions are created, the relaxation process is mostly dissipative and phonons are emitted in order to cool the carriers temperatures (after $t = 1$ ps, for example we obtain, $T_h \approx 550$ K and $T_e \approx 1900$ K).

A gedanken experiment. – To better disentangle the $L_1 \rightarrow L'_1$ process from the slower $L \rightarrow X$ channel we consider a shorter laser pulse with $\sigma = 50$ fs. We also consider a gedanken experiment where electrons are manually excited. One of the approximations most widely used in the literature is to mimic the effect of the laser pulse with some, ad hoc, initial population of carriers in the valence bands. This approximation corresponds to put $U = 0$ in eq. (2) defining some initial arbitrary population $f_i(t = t_0)$. Here we have chosen an initial population around the $\Gamma_{15}$ state, with a carrier density equal to the one measured experimentally.

We then show (fig. 1(b)) the population of the $L_1$ state in the gedanken experiment (dot-dashed line), and when the photo-excitation is performed with the shorter pulse for both $L_1$ (continuous line) and $L'_1$ (dashed line). By comparing the results we notice that the decay of the $L_1$ state, when the carriers are manually excited is much slower compared to the case when the carriers are photo-excited. This is because the dynamics following

\[ f_i(t_{rel}) = \frac{\gamma_i^{(h)}(t_{rel})}{\gamma_i^{(h)}(t_{rel}) + \gamma_i^{(e)}(t_{rel})}, \]

where we have used the fact that at $t = t_{rel}$ the pump field is switched off and $\partial_t f_i(t_{rel})|_{pump} = 0$.

In the equilibrium regime the dependence of the $\gamma_i^{(e/h)}$ lifetimes on the electronic energies is well known. Indeed for conduction bands $\gamma_i^{(h)}(t_{rel}) = 0$ while for valence states $\gamma_i^{(e)}(t_{rel}) = 0$. This means that, from eq. (6), the totally relaxed occupations would be zero for any conduction band.

This is the reason why in the RTA [18] $f_i^{\infty}$ is added as an adjustable parameter in the simulation. It is commonly parametrized as a Fermi distribution with a given temperature and chemical potential. But our simulations reveal that, in general, electrons and holes are distributed with two different Fermi distributions. This means that two different chemical potentials and temperatures. In our approach $f_i^{\infty}$ is a by-product of the simulation and it must not be provided at the beginning.

\[ \text{Carrier lifetimes: an out-of-equilibrium concept.} \]

The most time-consuming part of solving eq. (1) is the update of the $\gamma^{(e/h)}_i(t)$ functions whose dependence on the occupations must be re-calculated at each time step. A very tempting possibility would be to keep $\gamma_i^{(e/h)}_i(t)$ constant. This is indeed the main ingredient of the relaxation time approximation (RTA) that is based on the assumption that $\gamma_i^{(e/h)}_i(t) = \gamma_{i,eq}^{(e/h)}$, with $\gamma_{i,eq}^{(e/h)}$ the equilibrium lifetimes, calculated without the presence of any external field.\(^3\). This approach has been recently used in ref. [18] to describe the carrier relaxation in Si excited by the weak sunlight.

In order to investigate further the meaning of the $\gamma_i^{(e/h)}(t)$ time dependence and their crucial role in describing the experimental results, let us introduce the totally relaxed occupations, $f_i^{\infty}$, defined as the occupations at the time $t_{rel}$ such that $\partial_t f_i(t)|_{t=t_{rel}} = 0$. From eq. (1) and eq. (3) it follows that

\[ f_i^{\infty} \equiv f_i(t_{rel}) = \frac{\gamma_i^{(h)}(t_{rel})}{\gamma_i^{(h)}(t_{rel}) + \gamma_i^{(e)}(t_{rel})}, \]

\(^3\)The equilibrium lifetimes are calculated within the GW approximation [32] for the e-e channel and within the Fan approximation [33] for the e-p channel. The non-equilibrium lifetimes are obtained by extending to the non-equilibrium regime the GW and Fan approximations as described in ref. [19].
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Fig. 3: (Color online) The equilibrium lifetime, $\gamma_L^{EQ}$ (blue dot-dashed line), is compared with the time-dependent out-of-equilibrium lifetimes defined in eq. (7) for the $L_1$ and $L'_1$ states. Due to the symmetry breaking induced by the laser pulse (the orange shadow represents its Gaussian envelope) we have two in-equivalent lifetimes at $L_1$ (green line) and $L'_1$ (green dashed line). Their relative intensity defines the ultra-fast ($\gamma_L \gg \gamma_{L'}$) and the slow ($\gamma_L \approx \gamma_{L'}$) time regimes.

Moreover, the present scheme allows to go well beyond the RTA by formally defining a NEQ carrier lifetime, $\gamma_i^{(e/h)}(t)$, such that occupation functions satisfy the simple equation $\partial_t f_i^{(i)} = -\gamma_i^{(i)} f_i^{(i)}$ ($i = e/h$) with

$$\gamma_i^{(e/h)}(t) = \pm \left[ \frac{\gamma_i^{(e)} f_i^{(e)}(t) - \gamma_i^{(h)} f_i^{(h)}(t)}{f_i^{(e/h)}(t)} \right]. \quad (7)$$

In eq. (7) the + (−) corresponds to electrons (holes). Equation (7) demonstrates that a true instantaneous NEQ carrier lifetime includes contributions from both the electron ($\gamma_i^{(e)}$) and the hole ($\gamma_i^{(h)}$) lifetimes.

The deviation of $\pi_i^{(e/h)}(t)$ from $\gamma_i^{(e/h)}(t)$ is, indeed, strictly connected with the symmetry breaking mechanism that explains the experimental result. In the $L_1/L'_1$ case, indeed, $\gamma_{L_1}^{eq} = \gamma_{L'_1}^{eq} \approx 20 \text{ meV}$. From fig. 3 we see that, instead, both $\gamma_i^{(e)}$ and $\gamma_i^{(h)}$ changes during the simulation, by an order of magnitude. The reason is that, in eq. (7), the $\gamma_i^{(e)} f_i^{(e)}$ and $\gamma_i^{(h)} f_i^{(h)}$ factors are of the same order and their balance measures the difference of population between the $L_1$ and $L'_1$ states. We can, therefore, easily recognize in fig. 3 two well-defined regimes: when $\gamma_L^{(e)} \gg \gamma_{L'_1}^{(e)}$ the ultra-fast $L_1 \rightarrow L'_1$ scattering channel is active. When $\gamma_L^{(e)} \approx \gamma_{L'_1}^{(e)}$, instead, the relative $L_1$ and $L'_1$ populations are balanced and the dynamics is dictated by the slower $L \rightarrow X$ channel.

A global view of the real-time dynamics. – To further illustrate the results of our simulations we also show,

in fig. 4, an overview of the carriers dynamics by four snapshots. The carriers distribution is represented here on the band structure of Si. Added (removed) carriers are in blue (red). At $t = 0$ fs (panel (a)), corresponding to the laser pulse maximum, at $t = 200$ fs (panel (b)), when the $L_1$ and the $L'_1$ populations are almost the same, at $t \approx 500$ fs (panel (c)) when both the $L_1$ and $L'_1$ states are almost empty and finally after more than 1 ps (panel (d)) when the electrons have reached the CBM. The dominant process at each time considered is schematically represented by the arrows.

Conclusions. – In conclusion we have presented a fully ab initio simulation of the carrier dynamics in Si. The present scheme, based on the merging of DFT with NEQ Green’s function theory, successfully describes the ultra-fast decay of the $L_1$ carrier population measured in a recent 2PPE experiment. We have also highlighted that the microscopic mechanism that drives this ultra-fast decay is not a standard inter-valley scattering but it is due to an ultra-fast (as fast as 90 fs) $L_1 \rightarrow L'_1$ scattering channel activated by the specific polarization of the pump laser. This physical interpretation is, also, supported by introducing a novel definition of the non-equilibrium carrier lifetime that provides an intuitive picture of the physical processes activated by the initial photo-excitation.

The occupations and the energies on the band structure are fitted from the occupation and the energies evaluated on the double mesh used for the simulation.
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