Exciton-Population Inversion and Terahertz Gain in Resonantly Excited Semiconductors

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The build-up of exciton populations in resonantly laser excited semiconductors is studied microscopically. For excitation at the 2s-exciton resonance, it is shown that polarization with a strict s-type radial symmetry can be efficiently converted into an incoherent p-type population. As a consequence, inversion between the 2p and 1s exciton states can be obtained leading to the appearance of significant terahertz gain.

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Resonant laser excitation of semiconductors induces a coherent interband polarization between conduction-band electron and valence-band hole states. Through interaction and scattering processes this optical polarization may be converted into incoherent populations of unbound or bound electron-hole pairs (excitons). Even though excitonic features of the coherent polarization are well understood [1, 2], the study of its decay into incoherent many-body states is an area of active research.

Several recent experiments have applied terahertz (THz) fields to directly probe the optically generated semiconductor quasi-particle excitations strongly in-  

situ with difference-frequency generation [8]. Since the two-particle correlations not only influence the population densities obey the semiconductor Bloch equations [2]

\[ i\hbar \frac{\partial}{\partial t} P_k = [\varepsilon_k + jkA] P_k - [1 - f_k^- - f_k^+] \Omega_k - i\Gamma_k \]

and similarly for \( f_k^+ \). Here, \( V_k \) is the Coulomb-matrix element whereas \( \varepsilon_k \) and \( \Omega_k \) denote the renormalized kinetic energy and Rabi frequency, respectively. The coupling to the THz field follows from the \( jA \) term which contains the vector potential \( \mathbf{e}_\sigma \) with direction \( \mathbf{e}_\sigma \) and the current-matrix element \( j_k = e \left( \frac{\mathbf{p}}{m_e} + \frac{\mathbf{p}^*}{m_h} \right) h \mathbf{k} \cdot \mathbf{e}_\sigma \) with the electron (hole) mass \( m_e, m_h \). The true two-particle correlations stem from \( \varepsilon_{q,v}', \varepsilon_{q,v}' \cdot \varepsilon_{q,v}' \Delta \equiv \Delta \langle a_{q,k}^\dagger a_{q,k} a_{q,k}' \rangle \), and \( D_{q,k}^\lambda, \mu \equiv \sum_{q,k} \mathcal{G}_{q,k} \Delta \langle (D_{q,k}^\lambda + D_{q,k}^\mu) \rangle a_{q,k}^\dagger a_{q,k} \) where \( \Delta \) and \( D \) are the bosonic phonon operators and \( \mathcal{G}_{q,k} \) is the phonon-matrix element. The notation \( \Delta \) indicates that the factorized single-particle contributions (subscript \( S \)) are removed, e.g.

\[ \Delta \langle a_{q,k}^\dagger a_{q,k} a_{q,k}' \rangle = \langle a_{q,k}^\dagger a_{q,k} a_{q,k}' \rangle - \langle a_{q,k}^\dagger a_{q,k} \rangle \langle a_{q,k}' \rangle \]

and similarly for \( f_k^+ \).

As a model system, we analyze quantum-wire structures but we show that the main results are equally valid for quantum-well systems. The electronic excitations are described by Fermion operators \( a_{q,v}(k) \) and \( a_{q,v}(k) \) related to an electron with carrier momentum \( k \) in the conduction (valence) band. We include the carrier-carrier Coulomb interaction as well as the couplings to light fields and phonons [2]. The dynamic evolution of single-particle quantities related to the microscopic polarization \( P_k \equiv \langle a_{q,k}^\dagger a_{q,k} \rangle \), electron \( f_k^- \equiv \langle a_{q,k}^\dagger a_{q,k} \rangle \), and hole \( f_k^+ \equiv \langle a_{q,k}^\dagger a_{q,k} \rangle \) densities obey the semiconductor Bloch equations [2]
$$D(\hat{X}) \prod_k a_{\epsilon,k}^\dagger \langle \Psi_0 \rangle$$ where $|\Psi_0\rangle$ is the unexcited semiconductor while $L_k^\dagger(t) = e^{i\varphi_X(t)} \sin\beta_k(t) a_{\epsilon,k}^\dagger + \cos\beta_k(t) a_{\epsilon,k}^\dagger$ is a fermion operator with $\sin^2\beta_k(t) = n_k(t)$ and $e^{i\varphi_X(t)} = P_k(t)/|P_k(t)|$.

For our subsequent discussions, it is convenient to introduce an exciton operator $X_{\nu,q} = \sum_k \phi_\nu(k) c_{\nu,k}^\dagger a_{\epsilon,-k+q}$ with a center-of-mass momentum $q$ and $q_{\nu}(h) = m_{\nu}(h)/(m_e + m_h) q$. By choosing $X = \hat{X}_{\nu,0}$ and $\phi_\nu(k) = \beta_k e^{i\phi_k}$, $|\Psi_{\text{coh}}(t)\rangle$ follows from $D(\hat{X}) = e^{\hat{X}_{\nu}^\dagger - \hat{X}}$ acting on the full valence band. Since $D(\hat{X})$ is formally analogous to the coherent state generator of bosonic fields [9], we may interpret $|\Psi_{\text{coh}}(t)\rangle$ as a coherent exciton state even though $\hat{X}$ is not bosonic [10].

To study, how efficiently $|\Psi_{\text{coh}}(t)\rangle$ can be converted into incoherent excitons, we need to solve the full Eqs. [11–13]. For resonant excitation, both $c_{\nu,\lambda,c}$ and $D_{\nu,q}$ convert a coherent excitonic polarization into incoherent two-particle populations where $c_{\nu}^{q,k'} = c_{\nu,\lambda,c}^{q,k'} e^{i\varphi_\nu(k)}$ describes incoherent excitons. Using the cluster expansion [11], we derive equations for the two-particle correlations:

\begin{equation}
\frac{i\hbar}{\partial t} c_{\nu}^{q,k'} = (e^{i\varphi_k} + j_{k+q-k} A)c_{\nu}^{q,k'} + S^{q,k'}
\end{equation}

\begin{equation}
+ (1 - f_{\nu}^c - f_{\nu}^h) \sum_l V_{k-l} c_{\nu}^{l,k'}\end{equation}

\begin{equation}
- (1 - f_{\nu}^c - f_{\nu}^h) \sum_l V_{k-l} c_{\nu}^{l,k'} \end{equation}

\begin{equation}
+i G^{q,k'} + D^{q,k',c} + T^{q,k'}, \end{equation}

where $c_{\nu}^{q,k'}$ is the renormalized kinetic energy of the two-particle state and $S$ contains Coulomb induced in-and-out scattering of single-particle quantities. The Coulomb sums with the phase-space filling factor $(1 - f^e - f^h)$ describe the attractive interaction between electrons and holes, allowing them to become truly bound electron-hole pairs, i.e. incoherent excitons which can be probed via the THz induced $j \cdot A$ coupling. The $G$ term contains the same $c_{\nu,\lambda,c}$ and $D^{\nu,c}$ correlations as $\Gamma$ in Eq. [13], showing how coherent excitons are converted into incoherent ones. The remaining two-particle contributions are denoted as $D_{\text{rest}}$ while $T$ symbolizes the three-particle Coulomb and phonon terms treated here at the scattering level. This way, we fully include one- and two-particle correlations and obtain a closed set of equations providing a consistent description of optical as well as THz excitations in semiconductors.

The polarization to population conversion efficiency is determined from the density of incoherent $\nu$-excitons $\Delta n_\nu = \frac{1}{c^2} \sum_q \Delta \langle \hat{X}_{\nu,q}^\dagger \hat{X}_{\nu,q} \rangle = \sum_{k,q'} \phi_\nu^*(k) \phi_\nu(k) e^{i\phi_k} - a_{\epsilon,k+q} \rightarrow a_{\epsilon,k}$ where $L^d$ is the normalization volume. We evaluate the conversion efficiency by numerically integrating the complete set of equation for a planar arrangement of identical quantum wires. Later on, we estimate the conversion efficiency also for a quantum well. We choose standard GaAs-type parameters and the wire and well sizes are taken such that the energy separation between the two lowest exciton states is 5 meV. The lattice temperature is assumed to be 10 K such that it is sufficient to include only acoustic phonons [13]. To study the generation of incoherent excitons in their different quantum states, we assume 4 ps pulsed optical excitation resonant with either the 1s- or 2s-resonance. We repeat the computations for different pump intensities and evaluate the final quasi-stationary exciton fraction $\Delta n_\nu/n$ relative to the generated carrier density $n = \frac{1}{c^2} \sum_k f_k^{\text{inh}}$.

![FIG. 1:](image-url)

FIG. 1: (a) For excitation at the 1s exciton resonance with a 4 ps laser pulse (dot-dashed line), the temporal evolution of the induced optical polarization $|P|^2$ (shaded area), together with the generated incoherent 1s (dashed line) and 2p (solid line) exciton densities $[10^4 \text{ cm}^{-2}]$ are shown. The inset shows the pump (shaded area) and linear absorption (solid line) spectra; $E_{1s}$ is the 1s-exciton energy. (b) The polarization to population conversion efficiency for 1s (dashed line) and 2p excitons (solid line) is plotted as function of excitation density $n$. The arrow indicates the density at which the dynamics is shown in a). The shaded area represents the result obtained without the phonon scattering.

In Figs. [11] and [12] we present numerical results for pumping at the 1s- and 2s-resonances, respectively. The insets show the spectral excitation conditions. In Fig. [11(a)], we plot the temporal evolution of the pump pulse, of the in-
duced optical polarization, and of the generated 1s and 2p exciton density. Figure 1(b) presents the relative percentage of excitons in the different quantum states showing, not surprisingly, that for 1s excitation the optical polarization is mainly converted into incoherent 1s excitons; \( \Delta n_{1s}/n \) is well above 90% for low densities. This large conversion fraction is expected since coherent and incoherent 1s excitons have an excellent energetic match. However, the generated exciton population drops well below 40% already at moderate densities above \( 10^5 \text{ cm}^{-1} \) where the phase space filling factor \((1 - f^e - f^h)\) peaks around 0.5. For even higher densities, \( \Delta n_{p}/n_{ch} \) vanishes since excitons start to ionize. A computation, where phonon scattering \( D \) is omitted, indicates that Coulomb scattering alone would lead to only roughly 15% exciton population.

The 2s-excitation results are presented in Fig. 2 where Fig. 2(b) shows \( \Delta n_{2s}/n \), \( \Delta n_{2p}/n \), and \( \Delta n_{1s}/n \). For not too high excitation densities, we observe that the 2s-polarization is converted into a mix of 2s and 2p populations. Whereas the amount of 2s population decreases monotonously with increasing excitation, the 2p population first increases up to 40% before it also decreases at higher densities where formation of 1s-excitons gradually becomes relevant.

Before we analyze the physical mechanism responsible for the significant formation of a 2p-exciton population, we study its signatures assuming a THz probe. For this purpose we evaluate the \( j \cdot A \) terms in Eqs. and to compute the generated THz current \( J_{THz} = \sum_{k,\lambda} j_{\lambda}(k) f_k^A \) with both coherent and incoherent contributions. We determine the linear THz gain \( g(\omega) = -\text{Im} \left[ J_{THz}(\omega)/\omega A(\omega) \right] \) assuming a 150 fs THz probe pulse capable of resolving temporal snapshots of \( g(\omega) \) during the exciton formation process. Figure 3 shows that for 1s-pumping the corresponding \( g(\omega) \) is always negative, i.e. we find THz absorption peaked around the 1s-2p transition. These results, as well as the asymmetric line shape due to transitions from the 1s to energetically higher bound and unbound states agree well with experimental findings reported in Ref. [5].

The same analysis is repeated for the 2s excitation (Fig. 3(b)) showing that \( g(\omega) \) rapidly changes from absorption to gain. At very early times, the system consists mainly of coherent 2s excitons such that only absorptive transitions from the 2s to higher states are present. However, as incoherent 2p excitons are generated, pronounced THz gain develops at the 2p to 1s transition as a consequence of the population inversion between these states.

Now, after we have presented examples of the fully numerical evaluation of our many-body theory, we want to analyze the relevant physical mechanisms. At first sight the significant generation of \( p \)-type excitons might be unexpected since it involves a symmetry change of the optically generated \( s \)-type polarization. To identify the microscopic origin of this process, we numerically study the relative importance of different contributions to the

![FIG. 2: Same as Fig.1 but for excitation at the 2s exciton resonance (inset). (a) Dynamics of optical polarization \(|P|^2\) (dot-dashed line) and incoherent densities of 2s (shaded area) and 2p (solid line) excitons \([10^4 \text{ cm}^{-1}]\). (b) Conversion efficiency for 1s (dashed line), 2p (solid line), and 2s (shaded area) excitons as function of excitation density \( n \).](image-url)
full theory. We find that for 2s pumping a switch off of phonon-induced scattering leaves the generated exciton fraction practically unchanged. Hence, in contrast to 1s pumping, the conversion of a 2s polarization predominantly results from Coulomb scattering.

In order to understand how the Coulomb interaction induces symmetry changes in the polarization conversion we now investigate the scattering (Γ) and conversion (G) mechanisms which stem from the same fermionic correlation \( c_{\nu,\lambda,\lambda,c} = \Delta(a^\dagger_{\lambda} a^\dagger_{\lambda} a^\dagger_{\nu} a^\dagger_{\nu}) \) between polarization and fermionic density.

(1) In a first step, we make the lowest level approximation by replacing the microscopic \( \Gamma_k \) by a phenomenological decay: \( \Gamma_k = -\Gamma P_k \). In this case, \( G_{q,k,k'} = +2P_k^* \Gamma P_{k'} \delta_{q,0} \) and \( [P_k^* \Gamma P_{k'} + \sum_q c^k_q \chi^k_q] \) is a constant of motion with respect to the scattering. Therefore, this simple dephasing model only converts s-type polarization to s-like exciton populations, in contrast to our microscopic results.

(2) Looking at the process of excitation induced dephasing [13] we see that Coulomb induced dephasing is actually a diffusive redistribution of the microscopic polarizations since \( \sum_k \Gamma_k = 0 \) and \( \sum_k q_{k,q} G_{n,k,q} = 0 \). At the same time, Eqs. 4 and 5 impose a strict microscopic connection between \( \Gamma \) and \( G \). In order to analyze the consequences of these fundamental restrictions, we use a somewhat reduced model by utilizing a simplified structural form of the second-Born solution of \( c_{\nu,\lambda,\lambda,c} \) [17] via \( \sum_{\lambda} c^q_{\lambda,\lambda,c} = (\sum_{\lambda} c^q_{\lambda,\lambda,c})^* = iF(f,P)_{q,k}(P_{k-q} - P_q) \) where \( F(f,P) \) is a real-valued, nonlinear functional of \( f \) and \( P \) containing the energy and momentum conservation aspects of the Coulomb scattering. The reduced model implies \( \Gamma^\text{red}_k = -\sum_q U_q (P_k - P_{k-q}) \) and \( C^q_{\text{red},k,k'} = (P_k^* - P_{k-k'}^*) U_q (P_{k+q} - P_{k}) \), where \( U_q = 2V_q \sum_k F(f,P)_{k,q} \). As a result, \( \Gamma^\text{red}_k \) removes polarization from the state \( P_k \) and redistributes it to \( P_{k-q} \) while the rate of conversion to the exciton state \( \nu \) becomes \( C^q_{\text{red},\nu} = |M_{\nu,q}|^2 U_q \). Here, the scattering matrix element is \( M_{\nu,q} = \sum_k \phi^\dagger_{\nu}(k)[P_{k+q} - P_{k}] \) indicating that Coulomb scattering leads to the generation of excitons with finite momenta whereas no population in the \( q = 0 \) state is produced. For low to moderate 2s excitation, we may use the approximation \( P_k \propto \phi_{2s}(k) \). With the help of the symmetries \( \phi_{2s}(-k) = \phi_{2s}(k) \) and \( \phi_{2p}(-k) = -\phi_{2p}(k) \), we find \( M_{2p,q} \propto \sum_{\lambda} \phi^\dagger_{2p}(k) \phi_{2s}(k+q) \) which is clearly nonzero for \( \phi_{2p}(k) \neq 0 \).

For 2s pumping, the energy conservation aspects of \( U_q \) are practically the same for 2s and 2p since these states are nearly degenerate. As a result, the overlap of the wavefunctions with shifted arguments in \( M_{\nu,q} \) determines the conversion rate such that \( |M_{\nu,q}|^2 \) can be used to estimate the ratio of generated 2s and 2p populations. Using the low-density exciton wavefunctions, we obtain a ratio of 1.36 of 2s over 2p population for the quantum wire, which is close to the numerical result in Fig. 2b. Repeating the same calculations for the quantum well, we get a ratio 0.99 showing that the generation of p-like states is strong and qualitatively similar for quantum wells and wires.

Since the Coulomb interaction conserves the angular momentum, one may ask how this conservation law is fulfilled when a 2s polarization is converted into 2p excitons. Without THz fields, Eq. 6 implies correlations with a functional dependence \( c_{q,p-k-q,p+k-q} = c(|q||k|,|k|,\cos\phi_{k,q},\cos\phi_{k,q}) \) where \( \phi_{k,q} \) is the angle between \( k \) and \( q \). Consequently, the generated excitons have an angular dependency \( \cos^m\phi_{k,q} = (e^{i\phi_{k,q}} + e^{-i\phi_{k,q}})^m/2^m \) such that the eigenfunctions \( e^{i\mu \phi\nu} \) and \( e^{-i\mu \phi\nu} \) of the \( L_z \) state are generated with equal probability. As a result, the total \( \langle L_z \rangle \) always vanishes such that the total angular momentum is fully conserved even when 2p excitons are generated.

In summary, our microscopic study predicts significant formation of excitons in 2p states for excitation at the 2s resonance of the absorption spectrum. As a consequence, an exciton population inversion between the 2p and 1s states is realized leading to gain for the corresponding THz frequency. Using different semiconductor materials this scheme may become useful for THz amplification in a wide spectral range.

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