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Semiconductor Bloch Equations including Spin and Polarization Degrees of Freedom

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Abstract. The well-known concept of semiconductor Bloch equations is extended to include the spin degree of freedom in a six-level system comprising the spin-split conduction, heavy- and light-hole bands (or corresponding lowest subbands in quantum wells). These equations represent the frame to describe the response of semiconductors (bulk or quantum wells) under near band gap optical excitation with arbitrary polarization including effects of spin relaxation. We discuss selected examples.

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1. Introduction

Semiconductor Bloch equations (SBE) represent the basic concept to describe a semiconductor under near band gap excitation by an intense light source.[1, 2] SBE are a set of coupled equations of motion (e.o.m.) for the components of the density matrix for a two-level system driven by the electric field of the exciting light which include the interaction between the photo-generated carriers (electrons and holes). The single-particle part of these equations refers to a pair of conduction and valence band states at fixed $\mathbf{k}$. The interaction renormalizes the single-particle energies and the coupling to the light field but couples also the two-level systems for different $\mathbf{k}$ thus giving rise to inhomogeneous broadening. Relaxation due to scattering with impurities and phonons can be included by adding phenomenological damping terms. SBE can be considered under different assumptions [2] which allow to describe (i) coherent (*e.g.* Rabi oscillations, photon echo) or (ii) quasi-equilibrium phenomena (*e.g.* luminescence) in their characteristic time domains: the interband polarization - initially coherent with the exciting electromagnetic field - decays by transverse relaxation due to carrier-carrier interaction and the system evolves into quasi-equilibrium populations of electrons and holes, which decrease by radiative (or non-radiative) processes (longitudinal relaxation). Under low-excitation conditions the SBE can be reduced to the inhomogeneous Schrödinger equation for the Wannier-Mott exciton. The strength of the SBE concept has been demonstrated in numerous fundamental investigations and applications, among which semiconductor microcavities and lasers represent prominent examples.[3]

In spite of their great success, the original two-level SBE were not designed to account for the degrees of freedom connected with the carrier spin or with the transversality of the exciting light. Both aspects have recently attracted increasing attention: (1) Optical excitation with circular polarization is used to generate spin-polarized carrier populations (optical spin alignment) and to investigate their dynamics.[4] (2) The polarization instability of vertical

cavity surface emitting lasers (VCSEL) has been ascribed to coupling between two-level systems (active under left and right circularly polarized light) by spin-relaxation processes.[5] (3) The helicity of light can be converted into a stationary current (circular photogalvanic effect, CPGE [6]) in systems lacking inversion symmetry, connected with removed spin-degeneracy of the single-particle states. (4) The nonlinear absorption (proportional to the PGE signal) is different for linear and circular polarized excitation, thus allowing to measure spin relaxation times.[7] (5) The mechanisms of spin relaxation, known from intensive studies for semiconductor bulk material [8], are essentially the same for semiconductor quantum wells.[9] The dominant ones for most semiconductor structures under investigation are the D'yakonov-Perel' (DP) mechanism, related to the spin-splitting of the single-particle states, and the Bir-Aronov-Pikus (BAP) mechanism, caused by electron-hole exchange processes. Spin relaxation is described by e.o.m. for the spin-density matrix, formulated so far only for the electron system.[8, 10]

Here we present an extension of the SBE which includes the spin and polarization degrees of freedom and represents the theoretical frame for all these and related phenomena.

2. Coherent SBE for the two-level system

The SBE originally formulated by Koch and Lindberg [1] for the two-level system, a conduction and a valence band level at the same wave vector $\mathbf{k}$, driven by a scalar electric field $E(t)$, have been derived from the e.o.m. for the two-fermion operators $c^\dagger(\mathbf{k},t)c(\mathbf{k},t), v(\mathbf{k},t)v^\dagger(\mathbf{k},t)$, and $v^\dagger(\mathbf{k},t)c^\dagger(\mathbf{k},t)$ whose thermal expectation values form the 2×2 density matrix

$$\begin{pmatrix} \rho^e(\mathbf{k},t) & P(\mathbf{k},t) \\ P^\dagger(\mathbf{k},t) & 1-\rho^h(\mathbf{k},t) \end{pmatrix} \tag{1}$$

with the population functions $\rho^e(\mathbf{k},t)(\rho^h(\mathbf{k},t))$ for electrons (holes) and the interband polarization function $P(\mathbf{k},t)$. The coupled equations of motion for these quantities result from applying the HF truncation, by which four-operator terms appearing in the e.o.m. are replaced by products of thermal expectation values of two-fermion operators (a more complete treatment employs the dynamically controlled truncation [11]). By considering only terms diagonal in the wave vector, thus leaving out collision terms due to carrier-carrier scattering, one arrives at the coherent part of these equations which read [1]

$$i\hbar\partial_t\rho^e(\mathbf{k},t) = -2iIm(\Omega(\mathbf{k},t)P^*(\mathbf{k},t)) = i\hbar\partial_t\rho^h(\mathbf{k},t) \tag{2}$$

$$i\hbar\partial_t P(\mathbf{k},t) = (\varepsilon^c(\mathbf{k})-\varepsilon^v(\mathbf{k}))P(\mathbf{k},t) - (1-\rho^e(\mathbf{k},t)-\rho^h(\mathbf{k},t))\Omega(\mathbf{k},t)\,, \tag{3}$$

but they can also be written using the commutator $[A,B]=AB-BA$ (dependence on $\mathbf{k}$ and t understood)

$$i\hbar\partial_t \begin{pmatrix} \rho^e & P \\ P^* & 1-\rho^h \end{pmatrix} = \left[\begin{pmatrix} \varepsilon^c & -\Omega \\ -\Omega^* & \varepsilon^v \end{pmatrix}, \begin{pmatrix} \rho^e & P \\ P^* & 1-\rho^h \end{pmatrix} \right]. \tag{4}$$

Here $\varepsilon^\alpha(\mathbf{k}) = \varepsilon_0^\alpha(\mathbf{k}) + \Sigma^\alpha(\mathbf{k})(\alpha = c,v)$ are the single-particle energies with self-energy corrections

$$\Sigma^{c,v}(\mathbf{k},t) = -\sum_{\mathbf{q}} V(|\mathbf{q}|)\rho^{e,h}(\mathbf{k}-\mathbf{q},t) \tag{5}$$

due to carrier-carrier interaction ($V(q)$ is the interaction potential) and

$$\Omega(\mathbf{k},t) = d_{cv}^* E(t) + \sum_{\mathbf{q}\neq\mathbf{k}} V(|\mathbf{k}-\mathbf{q}|)P(\mathbf{q},t) \tag{6}$$

the renormalized interaction with the light field (d_{cv} is the dipole matrixelement between valence and conduction band). The second term of Eq. 6 is due to the optically generated electron-hole pairs and expresses together with the self-energy corrections the many-body character of the problem. Without these terms Eqs. 2 and 3 reduce (in the rotating wave approximation) to the coherent optical Bloch equations which are frequently used by including phenomenological damping terms characterized by the longitudinal relaxation time T_1 (for the decay of population inversion) and the transverse relaxation time T_2 (for the dephasing or decay of coherence).[2]

Equations 2 and 3 do not account for the realistic electronic structure at the band edges of semiconductors with degeneracy due to spin and angular momentum. The removal of these degeneracies by spin-orbit interaction, which at finite wave vector $\mathbf{k}$ can give rise to $\mathbf{k}$ linear terms in the dispersion of conduction and valence bands (or subbands in quantum well structures) is connected with the DP spin relaxation. In addition the two-level system coupled to the scalar field $E(t)$ does not allow to account for the possible different transverse polarizations of the exciting light in a real system.

3. Extension to the six-level system

To overcome these limitations the two-level system has been replaced by the more realistic multi-band model.[12] We consider here for simplicity a six-level model comprising for each wave vector $\mathbf{k}$ the lowest conduction band ($c, m_c = \pm 1/2$) and the topmost valence band ($v, m_v = \pm 3/2, \pm 1/2$), where the quantum numbers m_α refer to the angular momentum representation of the band edge states with $\mathbf{k} = 0$. For finite $\mathbf{k}$ the states are mixed due to $\mathbf{k} \cdot \mathbf{p}$ interaction. The six-level model is applicable to bulk as well as to quantum wells and includes in the latter case the lowest electron, heavy- and light-hole subbands (each twofold due to spin). The density matrix for this system consists of diagonal 2×2 and 4×4 blocks for the electrons $\boldsymbol{\rho}^e(\mathbf{k}, t)$ and holes $\boldsymbol{\rho}^h(\mathbf{k}, t)$, respectively, and a 2×4 off-diagonal block (and its hermitian conjugate) for the interband polarization $\mathbf{P}(\mathbf{k}, t)$. This structure of the density matrix corresponds to that assumed for the system Hamiltonian: 2×2 and 4×4 diagonal blocks $\mathbf{H}^c(\mathbf{k})$ and $\mathbf{H}^v(\mathbf{k})$ for the conduction and valence band states, respectively, due to block diagonalization with respect to interband $\mathbf{k} \cdot \mathbf{p}$ coupling. We may split $\mathbf{H}^\alpha(\mathbf{k}) = \mathbf{H}_0^\alpha(\mathbf{k}) + \boldsymbol{\Sigma}^\alpha(\mathbf{k})$ into independent-particle part $\mathbf{H}_0^\alpha(\mathbf{k})$ and self-energy correction $\boldsymbol{\Sigma}^\alpha(\mathbf{k})$ as for the two-level system. Dipole selection rules determine the coupling $\boldsymbol{\Omega}$ between valence and conduction band states with the electric field

$$\mathbf{E}(t) = \mathbf{E}(\omega) e^{i\omega t} + \mathbf{E}^*(\omega) e^{-i\omega t} \tag{7}$$

with $\mathbf{E}^*(\omega) = \mathbf{E}(-\omega)$ to make $\mathbf{E}(t)$ real. For light propagating along z the two independent transverse polarizations are obtained for $\mathbf{E}_\pm^*(\omega) = E_0(\mathbf{e}_x + \mathbf{e}_y e^{\pm i\varphi})/2$ where $\pm$ indicates the helicity of light which for $\varphi = \pi/2$ corresponds to right and left circular polarization. Instead of d_{cv} we have a 2×4 matrix $\mathbf{d}_{m_c m_v}$ for the vector of the dipole operator and for $\mathbf{k} = 0$ an unrenormalized interaction matrix $\mathbf{E}(\omega) \cdot \mathbf{d}_{m_c m_v} (R = d_{cv}/\sqrt{2})$:

$m_c = +\frac{1}{2}$	$-\frac{E_0 R}{2}(1 + ie^{\pm i\varphi})$	0	$\frac{E_0 R}{2\sqrt{3}}(1 - ie^{\pm i\varphi})$	0
$m_c = -\frac{1}{2}$	0	$-\frac{E_0 R}{2\sqrt{3}}(1 + ie^{\pm i\varphi})$	0	$\frac{E_0 R}{2}(1 - ie^{\pm i\varphi})$
$m_v =$	$\frac{3}{2}$	$\frac{1}{2}$	$-\frac{1}{2}$	$-\frac{3}{2}$

The obvious compact form of the e.o.m. for the density matrix of the six-level system is that of Eq. 4 but with the scalar quantities replaced by the corresponding matrices:

$$i\hbar \partial_t \begin{pmatrix} \boldsymbol{\rho}^e & \mathbf{P} \\ \mathbf{P}^\dagger & \mathbf{1} - \boldsymbol{\rho}^h \end{pmatrix} = \left[\begin{pmatrix} \mathbf{H}^c & -\boldsymbol{\Omega} \\ -\boldsymbol{\Omega}^\dagger & \mathbf{H}^v \end{pmatrix}, \begin{pmatrix} \boldsymbol{\rho}^e & \mathbf{P} \\ \mathbf{P}^\dagger & \mathbf{1} - \boldsymbol{\rho}^h \end{pmatrix} \right]. \tag{8}$$

In order to identify the individual terms, in particular the expressions for the self-energy and the renormalized interaction matrix $\mathbf{\Omega}$, one has to go through the lengthy algebra of deriving the e.o.m. of the two-fermion operators as for the two-level system and to apply the HF truncation scheme. This has been done in the past and the resulting generalizations of Eqs. 2 and 3 can be found in the literature.[12] The present investigation, however, is an extension by including spin-splitting of the (sub)bands and electron-hole exchange interaction.[13]

As mentioned already, spin-splitting of the electron or hole (sub)bands is a prerequisite for the DP mechanism of spin-relaxation. Therefore, $\mathbf{H}_0^\alpha(\mathbf{k})$ should contain besides the bilinear terms in the components of the wave vector also odd-power terms which arise from $\mathbf{k}\cdot\mathbf{p}$ decoupling in systems (bulk or QW) without inversion symmetry. Thus, in the basis of eigenstates of $\mathbf{H}_0^\alpha(\mathbf{k})$ the independent-particle energies $\varepsilon^\alpha_{m_\alpha}(\mathbf{k})$ can depend on the angular (or spin) quantum number m_α and, as a consequence, one finds *e.g.*

$$[\mathbf{H}_0^\alpha(\mathbf{k}),\boldsymbol{\rho}^\alpha(\mathbf{k})]_{m_\alpha m'_\alpha} = \left(\varepsilon^\alpha_{0,m_\alpha}(\mathbf{k}) - \varepsilon^\alpha_{0,m'_\alpha}(\mathbf{k})\right)\rho^\alpha_{m_\alpha m'_\alpha}(\mathbf{k}). \tag{9}$$

Note, that this commutator appears in the e.o.m. for the spin-density matrix of electrons for the DP spin relaxation.[8, 10, 18]

In the present context the Coulomb interaction ($\mathbf{x}$ refers to space and spin coordinates)

$$H_{Coul} = \frac{1}{2}\int d\mathbf{x}\int d\mathbf{x}'\Psi^\dagger(\mathbf{x})\Psi^\dagger(\mathbf{x}')V(|\mathbf{r}-\mathbf{r}'|)\Psi(\mathbf{x}')\Psi(\mathbf{x}) \tag{10}$$

is formulated by expanding the field operators $\Psi(\mathbf{x})$ in a series of electron and hole single-particle functions (time-inverted electron functions $\tilde{\psi}$ are used for holes)

$$\Psi(\mathbf{x}) = \sum_{m_c} c_{m_c}(\mathbf{k},t)\psi_{cm_c\mathbf{k}}(\mathbf{x}) + \sum_{m_v} v^\dagger_{m_v}(\mathbf{k},t)\tilde{\psi}_{vm_v\mathbf{k}}(\mathbf{x}) \tag{11}$$

the expansion coefficients being fermion operators for electrons and holes. H_{Coul} falls into four different contributions (*e-e, h-h,* direct *e-h,* and *e-h* exchange) of which the *e-h* exchange term reads

$$H^X_{eh} = \sum_{\substack{m_c m'_c \mathbf{k}\\ m_v m'_v \mathbf{k}'\mathbf{q}}} \mathcal{V}^{eh,X}_{\substack{m_c m_v\\ m'_c m'_v}}(\mathbf{k},\mathbf{k}',\mathbf{q})c^\dagger_{m_c}(-\mathbf{k}+\mathbf{q})c_{m'_c}(-\mathbf{k}'+\mathbf{q})v^\dagger_{m'_v}(\mathbf{k})v_{m_v}(\mathbf{k}') \tag{12}$$

with

$$\mathcal{V}^{eh,X}_{\substack{m_c m_v\\ m'_c m'_v}}(\mathbf{k},\mathbf{k}',\mathbf{q}) = \int d\mathbf{x}\int d\mathbf{x}'\psi^\dagger_{cm_c-\mathbf{k}+\mathbf{q}}(\mathbf{x})\tilde{\psi}^\dagger_{vm_v\mathbf{k}'}(\mathbf{x}') V(|\mathbf{r}-\mathbf{r}'|)\psi_{cm'_c-\mathbf{k}'+\mathbf{q}}(\mathbf{x}')\tilde{\psi}_{vm'_v\mathbf{k}}(\mathbf{x})\,. \tag{13}$$

It is characteristic for the *e-h* exchange term that the integration is over products of electron and hole wave functions at different $\mathbf{k}$. Their periodic parts can be Fourier expanded with plane waves of the reciprocal lattice vectors $\mathbf{G}$. As it turns out there is no contribution for $\mathbf{G}=0$ (because of the orthogonality of the periodic parts of Bloch functions at the same $\mathbf{k}$) and, therefore, *e-h* exchange terms are usually not considered in the context of SBE (an exception is Ref. [17]). On the other hand *e-h* exchange manifests itself in the exciton finestructure in bulk [14, 15] and quantum well systems [16] and is also responsible for (exciton) spin relaxation (BAP mechanism) [9, 10]. In our extension of the SBE for the six-level system we take *e-h* exchange into account and discuss its consequences with respect to spin relaxation. In this contribution we do so by taking the electron self-energy as an example and refer for a more detailed presentation to [13].

The electron self-energy takes the form

$$\begin{aligned}\Sigma^c_{m_c m'_c}(\mathbf{k}) = & -\sum_{\mathbf{q}}\Big\{\sum_{\tilde{m}_c\tilde{m}'_c}\mathcal{V}^{ee}_{m'_c\tilde{m}_c m_c\tilde{m}'_c}(\mathbf{k}-\mathbf{q},\mathbf{k},\mathbf{q})\rho^e_{\tilde{m}_c\tilde{m}'_c}(\mathbf{k}-\mathbf{q})\\ & -\sum_{m_v m'_v}\mathcal{V}^{eh,X}_{m'_c-m_v m_c-m'_v}(-\mathbf{k}+\mathbf{q},-\mathbf{k}+\mathbf{q},\mathbf{q})\rho^h_{m'_v m_v}(\mathbf{k}-\mathbf{q})\Big\}\,,\end{aligned} \tag{14}$$

where the first term derives from the *e-e* interaction while the second term is due to the *e-h* exchange interaction. The interaction matrixelements of the Coulomb interaction $\mathcal{V}^{ee}$ are modified due to the $\mathbf{k}\cdot\mathbf{p}$ mixing of the single-particle wave functions but reduce without this mixing according to $\mathcal{V}^{ee}_{m_c'\tilde{m}_c m_c\tilde{m}_c'}(\mathbf{k}-\mathbf{q},\mathbf{k},\mathbf{q}) \rightarrow V(|\mathbf{q}|)\delta_{m_c'\tilde{m}_c'}\delta_{m_c\tilde{m}_c}$ and we recover the self-energy of Eq. 5 for the two-level system. The second term, due to *e-h* exchange interaction, contributes only for finite hole population $\boldsymbol{\rho}^h$, as is typical for the BAP spin relaxation mechanism. In general, the off-diagonal terms of $\Sigma^c_{m_c m_c'}(\mathbf{k})$ describe spin-flip processes which can be due to band-mixing or *e-h* exchange interaction.

4. Special cases and discussion

Let us consider now the special case of two two-level systems, consisting of electron ($m_c = \pm 1/2$, here abbreviated as ± 1) and heavy-hole states ($m_v = \pm 3/2$, here abbreviated as ± 3), under excitation with circularly polarized light. For this system (including cavity effects) San Miguel *et al.* [5] formulated the Maxwell-Bloch equations in order to study the polarization instability of VCSEL. These equations (except for cavity effects) can be obtained from our six-level SBE by neglecting the light hole states, using the nonvanishing interaction terms $\Omega_{\pm 1\pm 3}$ for circularly polarized light, and adding phenomenological damping terms. The e.o.m. for the population inversion $D = \frac{1}{2}\{\rho^e_{11} + \rho^e_{-1-1} - (2 - \rho^h_{33} - \rho^h_{-3-3})\}$ and for the spin polarization $d = \frac{1}{2}\{\rho^e_{11} + \rho^h_{33} - \rho^e_{-1-1} - \rho^h_{-3-3}\}$ take the forms

$$\partial_t D = -\frac{1}{\tau_{||}} D + \frac{2}{\hbar}\mathrm{Im}(\Omega_{13}P^*_{13} + \Omega_{-1-3}P^*_{-1-3}) \tag{15}$$

and

$$\begin{aligned}\partial_t d = & -\frac{1}{\tau_{||}} d + \frac{2}{\hbar}\mathrm{Im}(\Omega_{13}P^*_{13} - \Omega_{-1-3}P^*_{-1-3}) \\ & + \frac{1}{i\hbar}\Big(\Sigma^c_{1-1}\rho^e_{-11} - \rho^e_{1-1}\Sigma^c_{-11} - \rho^h_{3-3}\Sigma^v_{3-3} + \Sigma^v_{-33}\rho^h_{-33}\Big)\,.\end{aligned} \tag{16}$$

The last term in Eq. 16, consisting of products of off-diagonal terms of the electron and hole spin-density matrices and self energies, is a contribution to the decay of the spin polarization d. In fact, when identifying this term as $-2d/\tau_s$ with a spin-relaxation time τ_s one recovers the corresponding phenomenological e.o.m. of Ref. [5].

Spin relaxation of electrons due to spin-splitting caused by spin-orbit coupling combined with momentum scattering (DP mechanism) is described usually starting from an equation of motion for the electron spin-density matrix $\boldsymbol{\rho}^e(\mathbf{k},t)$.[8, 10] Such an equation is obtained from Eq. 8 by considering only the 2×2 electron block and identifying the driving term $\mathrm{Im}(\boldsymbol{\Omega}\mathbf{P}^*)$ as generation matrix $\mathbf{G}(\mathbf{k},t)$ (see also Ref. [18]):

$$\begin{aligned}\partial_t\boldsymbol{\rho}^e(\mathbf{k},t) - & \frac{1}{i\hbar}[\mathbf{H}^c_0(\mathbf{k}) + \boldsymbol{\Sigma}^c(\mathbf{k}), \boldsymbol{\rho}^e(\mathbf{k},t)] \\ & + \sum_{\mathbf{k}'} W(\mathbf{k},\mathbf{k}')\Big(\boldsymbol{\rho}^e(\mathbf{k},t) - \boldsymbol{\rho}^e(\mathbf{k}',t)\Big) = \mathbf{G}(\mathbf{k},t)\,,\end{aligned} \tag{17}$$

where we have added the last term on the *l.h.s.* to account for momentum scattering with phonons and (nonmagnetic) impurities. Equation 17 is a generalized form of equations used in the context of spin relaxation combining the single-particle spin-relaxation mechanism due to spin-splitting (DP) considered in $\mathbf{H}^c_0(\mathbf{k})$ with the many-body mechanisms described by the electron self energy $\boldsymbol{\Sigma}^c(\mathbf{k})$, in particular the one caused by the electron-hole exchange interaction (BAP). To the best of our knowledge such a unified formulation of both

mechanisms does not yet exist in the literature. A similar equation for the 4×4 hole-density matrix $\boldsymbol{\rho}^h(\mathbf{k}, t)$ could be used to calculate the hole-spin relaxation.

The response of a semiconductor system under intense excitation can be described by the thermal expectation value of the corresponding observable. Let us consider here as an example the photogalvanic effects (PGE) [6, 7] which are detected as a stationary current due to non-equilibrium populations of spin-split states under nonlinear excitations. This current can be written as

$$\mathbf{j} = e\mathrm{Tr}(\hat{\boldsymbol{\rho}}\hat{\mathbf{v}}) \tag{18}$$

with the operators $\hat{\boldsymbol{\rho}}$ and $\hat{\mathbf{v}}$ of the density matrix and of the velocity, respectively. As discussed in the literature the evaluation of Eq. 18 has to include the off-diagonal elements of $\hat{\boldsymbol{\rho}}$ and $\hat{\mathbf{v}}$, where the density matrix contains the nonlinear dependence on the electric field amplitude of the exciting light. The theory developed here does not directly apply to the situation of Ref. [6] with intersubband excitation by far-infrared laser light, but can be adapted to account for this case. On the other hand investigations of the PGE with optical excitation (from valence to conduction band states), to which the concept presented here applies, are conceivable. An aspect of particular interest of PGE measurements is the dependence of the saturation behavior on the light polarization, which provides as an alternative to time-resolved experiments the possibility to detect the spin-relaxation time.[7] Future work has to show how this dichroism of the PGE saturation can be described in the frame of the concept presented here.

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