Analytical approach to semiconductor Bloch equations

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received 10 July 2009; accepted in final form 16 November 2009
published online 14 December 2009

PACS 71.35.-y – Excitons and related phenomena

Abstract – Although semiconductor Bloch equations have been widely used for decades to
tackle ultrafast optical phenomena in semiconductors, they have a few important drawbacks:
i) Coulomb terms between free electron-hole pairs require a Hartree-Fock treatment which, in its
usual form, preserves excitonic poles but loses biexcitonic resonances. ii) The resulting coupled
differential equations impose heavy numerics which completely hide the physics. This can be
completely avoided if, instead of free electron-hole pairs, we use correlated pairs, i.e., excitons.
Their interactions are easy to handle through the recently constructed composite-boson many-
body theory. This allows us to obtain the time evolution of the polarization induced by a laser pulse
analytically. Polarization is shown to come from Coulomb interactions between virtual excitons,
but also from Coulomb-free fermion exchanges, these being dominant at large detuning.

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Interaction of semiconductors with ultrafast laser pulses
has been widely studied for decades. Light-matter inter-
action leads to a large variety of nonlinear effects of
great interest in semiconductor technology. Fascinating
ones are those induced by unabsorbed photons through
their coupling to virtual excitons. These virtual excitons
interact with carriers present in the sample, not only
through the Coulomb potential, but mostly through the
Pauli exclusion principle. This Pauli blocking appears
in the many-body theory for composite bosons we have
recently constructed [1] by means of “Pauli scatterings”:
they correspond to fermion exchange between excitons
in the absence of fermion interaction, as nicely visual-
ized by the so-called Shiva diagrams [2]. These exchanges
control all nonlinear optical effects in semiconductors.
Among those induced by unabsorbed photons, we can cite
the exciton optical Stark shift [3], Faraday rotation [4]
and oscillation [5] in photoexcited semiconductors, preces-
sion [6] and teleportation [7–9] of trapped electron spin,
quintum [9–11] and classical optical computing [12] using
trapped qubits. Effects induced by these virtual excitons
decrease with photon detuning but increase with photon
number, i.e., pulse intensity.

Derivation of these effects faces a major problem. As
excitons play a key role, we must include the electron-hole
Coulomb interaction exactly, to get these bound states.
However, many-body effects can be approached through
perturbative expansion only. This is fine for interaction
between the exciton fermionic components, since excitons
are neutral objects. An exact treatment of the electron-
hole interaction is however required to get the excitonic
poles. The clean separation between electron-hole ladder
processes binding the exciton and those making excitons to
interact has stayed an open problem for decades [13]. Our
composite-exciton many-body theory [1] provides such a

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57007-p1
operators. These operators have an important drawback: excitons being composite bosons, their commutation relations are not strict. We can however overcome this difficulty through “Pauli scatterings” \( \lambda (\alpha i m n) \) which describe fermion exchanges in the absence of fermion interaction. They appear in
\[
[B_m, B_i^\dagger] = \delta_{m,i} - D_{mi},
\]
\[
[D_{mi}, B_j^\dagger] = \sum_n \left[ \lambda \left( \begin{array}{c} n \\ m \end{array} \right) + (m \leftrightarrow n) \right] B_n^\dagger,
\]
where \( B_i^\dagger \) creates an exciton in state \( i \), with orbital indices \((Q_i, \nu_i)\) and spin indices \((s_i, m_i)\).

Since the exciton operators \( B_i^\dagger \) contain the electron-hole part of the Coulomb interaction leading to excitonic poles in an exact way, the remaining part of Coulomb interaction can be treated perturbatively through Coulomb scatterings \( \xi \left( \begin{array}{c} n \\ m \end{array} \right) \). They appear in
\[
[H, B_i^\dagger] = E_i B_i^\dagger + V_i^\dagger,
\]
\[
[V_i^\dagger, B_j^\dagger] = \sum_{m,n} \xi \left( \begin{array}{c} n \\ m \end{array} \right) B_{m}^\dagger B_n^\dagger.
\]
These Pauli and Coulomb scatterings allow easy calculations of physical quantities dealing with excitons, with the Pauli exclusion principle treated in an exact way.

Before calculating the polarization induced by a laser pulse within this new many-body formalism, we wish to stress that the semiconductor Bloch equations (SBE) [14-21] use free pairs \( a^\dagger_{k_s k_h} b^\dagger_{k_s k_h} \) instead of correlated pairs, \( i.e., \) excitons \( B_i^\dagger \). This just corresponds to a basis change
\[
B_i^\dagger = \sum_{k_s k_h} a^\dagger_{k_s k_h} b^\dagger_{k_s k_h} (k_s k_h | i),
\]
\[
a^\dagger_{k_s k_h} b^\dagger_{k_s k_h} = \sum_i B_{i}^\dagger (i | k_s k_h).
\]
Difficulties with SBE in fact come from using these free-pair operators in problems controlled by excitonic poles. By working with exciton operators, we can get \( \langle B_i^\dagger \rangle_t \) instead of \( \langle a^\dagger_{k} b^\dagger_{k} \rangle_t \) analytically, thereby avoiding heavy numerics coming from the standard SBE procedure. Moreover, the physical meaning of the various contributions to the polarization becomes transparent: one part of the polarization comes from Coulomb interaction, but the dominant part, at large photon detuning, comes from Pauli blocking between the exciton carriers.

**Proposed approach.** We look for the polarization induced by a single laser pulse starting from a nonexcited semiconductor. Extensions to semiconductors having carriers or to experiments with different photons, such as in four-wave mixing, will be presented elsewhere, the goal of this letter being to present the main steps of the new procedure we propose.

The time evolution of the semiconductor polarization can be written in terms of \( \langle \psi_t | B_i^\dagger | \psi_t \rangle = \langle B_i^\dagger \rangle_t \), where \( |\psi_t\rangle \) is the semiconductor state resulting from its coupling \( W_t \) to the photon field. This \( \psi \) exciton mean value obeys
\[
-i \frac{\partial}{\partial t} \langle B_i^\dagger \rangle_t = \langle [H_{sc} + W_t, B_i^\dagger] \rangle_t.
\]
\( H_{sc} \) is the semiconductor Hamiltonian, while \( W_t = U_t + U_t^\dagger \), with \( U_t = \sum_i \Omega_i(t) B_i \). The time dependence of the Rabi coupling \( \Omega_i(t) \) to exciton \( i \) follows the laser pulse. Since \( \langle B_i^\dagger, B_i^\dagger \rangle = 0 \), eqs. (1), (3) readily turn eq. (7) into
\[
-i \frac{\partial}{\partial t} \langle B_i^\dagger \rangle_t = \langle [H_{sc}, B_i^\dagger] \rangle_t + \sum_i \Omega_i(t) \langle D_{ip} \rangle_t + \langle V_i^\dagger \rangle_t.
\]

Besides a naïve linear term \( \langle \Omega_i(t) \rangle \) that contains contributions coming from the \( p \) exciton composite nature, through \( D_{ip} \), and from Coulomb interaction with the \( p \) exciton, through \( V_i^\dagger \).

If the photons with frequency \( \omega \) were introduced adiabatically with a 1/\( \epsilon \) risetime, \( \Omega(t) \) would read as \( \Omega \epsilon^{i\omega t} \epsilon^{-t} \). For a nonexcited semiconductor, \( |\psi_{\epsilon \rightarrow -}\rangle = |\psi_t\rangle \), the \( \Omega_i(t) \) term in eq. (8) then gives the free part of \( \langle B_i^\dagger \rangle_t \) as \( \Omega \epsilon^{i\omega t} \epsilon^{-t} / (\omega - E_p - i\epsilon) \), if we only keep resonant terms.

However, even if most papers consider an adiabatic establishment of the photon field, this is not realistic for ultrashort laser pulses with steep rise time and duration \( T_p \) long compared to the photon period \( \omega^{-1} \). Instead, \( \Omega(t) \) should be taken as \( \Omega \epsilon^{i\omega t} \) for \( 0 < t < T_p \) and zero otherwise, with \( |\psi_t\rangle = |\psi\rangle \) for \( t \leq 0 \) (sudden approximation). This procedure is the one adopted for NMR pulses [22]. When the pulse is on, \( i.e., \) for \( 0 \leq t \leq T_p \), the free part of \( \langle B_i^\dagger \rangle_t \), which now cancels for \( t = 0 \), reads as
\[
e^{-i\omega t} \langle B_i^\dagger \rangle_{t,\text{free}} = e^{-i\omega t} \Omega \Delta_t^{(1)}(\omega_p),
\]
where \( \omega_p = \omega - E_p \) is the photon detuning with respect to exciton \( p \), while \( \Delta_t^{(1)}(x) \) is the Fermi golden rule as long as the photon field is not switched off.

In addition to its physical relevance, the sudden rise of \( \Omega(t) \) allows us to eliminate \( t \) from the Hamiltonian through the so-called “rotating frame” unitary transformation that we can take\(^1\) as \( Z_t = \exp(-i\omega t \sum k a^\dagger_k a_k) \). This gives \( Z_t^{-1} a^\dagger_k Z_t = e^{i\omega t} a^\dagger_k \), while \( b^\dagger_k \) stays unchanged. The effective Hamiltonian \( \tilde{H} = \tilde{Z}_t^{-1} \tilde{H}_t Z_t - i \tilde{Z}_t^{-1} \tilde{Z}_t \), which rules the time evolution of \( |\psi_t\rangle = Z_t^{-1} |\psi\rangle \), reduces to \( \tilde{H} = \tilde{H}_{sc} + \tilde{W} \), where \( \tilde{H}_{sc} \) is just \( H_{sc} \) with all electron energies shifted by \( -\omega \), which amounts to replace \( E_0 \) by \( E_0 - \omega \). The coupling \( \tilde{W} \) then reads as \( \tilde{U} + \tilde{U}^\dagger \) where \( \tilde{U} = \sum_i \Omega_i B_i \) is now \( t \)-independent. The time evolution of \( \langle \psi_t | B_i^\dagger | \psi_t \rangle \) is given by eq. (8), with \( \langle \Omega_i(t), -E_p \rangle \) replaced by \( \langle \Omega(t), \omega_p \rangle \).

\(^1\)This transformation is usually taken symmetrical with respect to electron and hole operators but this is unnecessary.
The linear term $\Omega_p$ leads to $\langle \langle (B^\dagger_p) \rangle \rangle_{\text{free}}$ given in eq. (9). To get the Pauli and Coulomb contributions coming from $\langle \langle D_{ip} \rangle \rangle_t$ and $\langle \langle V^\dagger_p \rangle \rangle_t$, we first note that, for a time-independent Hamiltonian, $\langle \langle |\psi_t\rangle \rangle$ reduces to $e^{-iHt}|\psi_i\rangle$. We can then use the integral representation of the exponential, namely
\[ e^{-iHt} = \int_{-\infty}^{+\infty} \frac{dx}{(2\pi i)^t} e^{-i(x+i\sigma t_\eta)}t_H + \hat{H} \]
\[ = \sum_{t=0}^{\infty} \left( \frac{1}{a-H} \hat{W} \right)^n \frac{1}{a-H} \].
(11)
\[ \sigma_t \equiv (1+1) \text{ for } t \text{ positive}, \text{ as in the present calculation}, \text{ but } (1-t) \text{ for } t \text{ negative}, \text{ being an arbitrary positive constant. In a second step, we decouple interaction with photons from interactions between carriers, through}
\[ \frac{1}{a-H} = \frac{1}{a-H_{sc}} + \frac{1}{a-H} \hat{W} \frac{1}{a-H_{sc}} \]
\[ = \sum_{n=0}^{\infty} \left( \frac{1}{a-H_{sc}} \hat{W} \right)^n \frac{1}{a-H_{sc}} \].
(12)
\[ Since \hat{W} \text{ and } V^\dagger_p \text{ change the pair number by one, while } D_{ip} \text{ conserves it, } \langle \langle (D_{ip}) \rangle \rangle_t \text{ only has even terms in photon coupling, while } \langle \langle (V^\dagger_p) \rangle \rangle_t \text{ only has odd terms. The polarization given in eq. (8) thus has odd terms only in Rabi coupling } \Omega_i. \]

**Equation (12) generates the polarization as an expansion in photon coupling } \Omega_i. \text{ Such an expansion is a priori valid for small laser intensity, a limitation fully consistent with considering } \langle \langle (B^\dagger) \rangle \rangle_t \text{ for } N = 1 \text{ only. For large laser intensity, the } \hat{W} \text{ expansion is not valid anymore but the relevant operators in this case are not exciton operators, but polariton operators. In a near future, we will address these large photon fields, using our recent work on interacting polaritons [23,24], along similar ideas.**

**Pauli part.** — As shown in the appendix, the Pauli part of $\langle \langle (B^\dagger_p) \rangle \rangle_t$, which comes from the $D_{ip}$ term of eq. (8), has a third-order expression in photon coupling which reads
\[ \langle \langle (B^\dagger_p) \rangle \rangle_{\text{Pauli}} \sim 2e^{-i\omega_d \lambda_j m} \sum \Omega_i \Omega_j \Omega_k \lambda_{ijkp} \Delta_j^{(3)}(\omega_p, -\omega_j, -\omega_k), \]
(13)
\[ where 2\lambda_{ijkp} \equiv \lambda (\omega_j) + (i \leftrightarrow j) \text{ while the } \Delta_j^{(n)} \text{ functions are linked by}
\[ x_{n+1} \Delta_j^{(n+1)}(x_1, x_2, \ldots, x_{n+1}) = \Delta_j^{(n)}(x_1, x_2, \ldots, x_n), \]
(14)
\[ \text{Coulomb part.} — \text{ The Coulomb part of } \langle \langle (B^\dagger_p) \rangle \rangle_t, \text{ comes from the } V^\dagger_p \text{ term of eq. (8). In contrast with } \langle \langle (D_{ip}) \rangle \rangle_t, \text{ which only reads in terms of exciton and wave functions, the exact calculation of } \langle \langle (V^\dagger_p) \rangle \rangle_t \text{ at third order in photon coupling requires the knowledge of the whole two-pair eigenstate spectrum. It is however possible to approximate this Coulomb part in two limits:}

i) For materials having a well-separated biexciton, we find, for photons close to the biexciton resonance,
\[ \langle \langle (B^\dagger_p) \rangle \rangle_{\text{Coulomb}} \sim e^{-i\omega_d \lambda_j m} \sum \Omega_i \Omega_j \Omega_k \xi_{ijkp} \]
\[ \times \Delta_j^{(4)}(\omega_p, -\omega_j, -\omega_j, -\omega_k), \]
(15)
\[ \xi_{ijkp} \text{ can be seen as a Coulomb interaction between excitons mediated by the molecular biexciton state and energy, obtained through numerics only.}

ii) For large detuning, we can expand the Coulomb part of the polarization in Coulomb scattering divided by detuning, through
\[ \frac{1}{a-H_{sc}} (\chi^\dagger + \frac{1}{a-H_{sc}} B^\dagger_j + \frac{1}{a-H_{sc}+\omega_j}). \]
(17)
\[ As shown in the appendix, the first-order term of this expansion leads to\]
\[ \langle \langle (B^\dagger_p) \rangle \rangle_{\text{Coulomb}} \sim e^{-i\omega_d \lambda_j m} \sum \Omega_i \Omega_j \Omega_k \xi_{ijkp} \]
\[ \times \Delta_j^{(4)}(\omega_p, -\omega_j, -\omega_j, -\omega_k), \]
(18)
\[ where \xi \text{ is the mixed direct-exchange Coulomb scattering, standard for exciton time evolution [1],}
\[ 2\xi_{ijkp} = \left[ \xi \left( \frac{j}{k} \right) - \xi \left( \frac{i}{k} \right) \right] + (i \leftrightarrow j). \]
(19)
\[ The “in” Coulomb exchange part of this scattering is equal to \sum_{m,n} \lambda \left( \frac{j}{m} \right) \xi \left( \frac{n}{k} \right). \text{ Higher-order terms in Coulomb scattering divided by detuning can be obtained along the same line.}

**Discussion.** — The above derivation shows that $\langle \langle (B^\dagger_p) \rangle \rangle_t$ contains two cubic terms in photon coupling, given in eqs. (13), (18) — or eq. (15) when the molecular biexciton plays a key role. One term comes from fermion exchange with exciton p without Coulomb process, the other from Coulomb interactions with exciton p with or without fermion exchange. They have the same structure except that, Coulomb scatterings $\xi_{ijkp}$ being energylike while Pauli scatterings $\lambda_{ijkp}$ are dimensionless, the Coulomb term has one more detuning in the denominator than the Pauli term $\Delta_j^{(3)}$ has $\omega_j$, while $\Delta_j^{(4)}$ has $\omega_j\omega_k$. This shows that, as usual, $\langle \langle (B^\dagger_p) \rangle \rangle_t$ is fully controlled at large detuning by the Pauli exclusion principle in the absence of Coulomb process.

We can then note that $\Omega_i$ depends on $i$ through $r(=0|m_i)$, where $r|\mu\rangle$ is the i exciton relative motion wave function. This makes photons dominantly coupled to the s exciton ground state, labelled 0. Moreover, $\langle \langle (B^\dagger_p) \rangle \rangle_t$ decreases when the photon detuning increases. Therefore,
a sizeable $\langle B_{p}^{(3)} \rangle$, implies photons with small detuning to exciton 0. For such photons, the $\omega_{ij,k} \omega_{jk}$ denominator of $\Delta_{t}^{(4)} \omega_{p}$ in eq. (18) leads us to only keep $(i,j,k) = 0$ in the sum. Similarly, due to the $\omega_{jk} \omega_{kj}$ denominator of $\Delta_{t}^{(3)} \omega_{p}$ in eq. (13), we only keep $(j,k) = 0$. The cubic term of $\langle B_{p}^{(3)} \rangle$ then reduces to

$$\langle B_{p}^{(3)} \rangle \approx e^{iE_{p}t} \Omega_{0} | \Omega_{0} |^{2} \left[ 2 \lambda_{000p} \Delta_{t}^{(3)} \omega_{p}, - \omega_{0}, \omega_{0} \right] - \xi_{000p} \Delta_{t}^{(4)} \omega_{p}, - \omega_{0}, - \omega_{0}, \omega_{0} \right],$$

(20)

where $\Omega_{0} \lambda_{i00p} = \sum_{i} \Omega_{i} \lambda_{i00p}$, the summation over $i$ being performed through closure relation.

The above result is valid during the pulse duration, $0 < t < T_{p}$, and for $\omega_{0} = \omega - E_{0}$ small compared to $\omega_{p} \neq 0$ but still large enough to possibly keep one term only in the $\xi / \omega_{0}$ expansion of $\langle B_{p}^{(3)} \rangle_{\text{Coulomb}}$. For $p$ being a bound state, dimensional arguments give $\lambda_{000p} = \lambda_{D} (\alpha_{X} / L)^{D}$ and $\xi_{000p} = \xi_{D} R_{X} (\alpha_{X} / L)^{D}$, where $\alpha_{X}$ and $R_{X}$ are the exciton Bohr radius and Rydberg energy, $D$ the space dimension, $L$ the sample size, and $(\lambda_{D}, \xi_{D})$ numerical prefactors of the order of 1. Their analytical values for $p = 0$ can be found in refs. [1,23].

For $r$ small, i.e., for $\omega r \ll 1$, we do have $\Delta_{t}^{(n)} \approx (it)^{n} / n$, so that $\langle B_{p}^{(3)} \rangle$ rises as

$$\langle B_{p}^{(3)} \rangle \approx e^{iE_{p}t} \Omega_{0} | \Omega_{0} |^{2} \left[ 2 \lambda_{000p} (it)^{3} / 3 - \xi_{000p} (it)^{4} / 4 \right].$$

(21)

More generally, the recursion relation between $\Delta_{t}^{(n)} \omega_{p}$ gives the $t$-dependence of the Pauli part of $\langle B_{p}^{(3)} \rangle$ as

$$\Delta_{t}^{(3)} \omega_{p}, - \omega_{0}, \omega_{0} = \omega_{0}^{-2} \left[ \Delta_{t}^{(1)} \omega_{p} + \omega_{0} \right] + \Delta_{t}^{(1)} \omega_{p} - \omega_{0} \right] - 2 \Delta_{t}^{(1)} \omega_{p},$$

(22)

while for the Coulomb part, it reads

$$\Delta_{t}^{(4)} \omega_{p}, - \omega_{0}, - \omega_{0}, \omega_{0} = \omega_{0}^{-3} \left[ 3 \Delta_{t}^{(1)} \omega_{p} - \omega_{0} \right] + \Delta_{t}^{(1)} \omega_{p} + \omega_{0} \right] - 3 \Delta_{t}^{(1)} \omega_{p},$$

(23)

$\Delta_{t}^{(1)} \omega_{p}$ is essentially a delta function in the large $\omega t$ limit which is currently fulfilled within usual pulse duration.

Figure 1 shows the real parts of $\Delta_{t}^{(n)}$ for $n = (1, 3, 4)$, when $p = 0$.

For $t > T_{p}$, the time evolution of the polarization is a priori simpler because it is free, i.e., not driven by the photon field. This free time evolution, also performed by using eq. (11) with a different initial state, will be given in an extended version of this work.

**State of the art.** – The semiconductor polarization is usually derived within the free-pair basis by calculating $p_{k} = (a_{k} b_{-k})_{t}$, through $-i \partial_{t} p_{k} = \{ [H_{sc} + W_{f}], a_{k} b_{-k} \}_{t}$. The Coulomb part of $H_{sc}$ generates expectation values like $(a^{\dagger}_{i} a_{j} a_{j})_{t}$, $(b^{\dagger}_{j} b_{i} b_{i})_{t}$, and $(a^{\dagger}_{i} b_{j} b_{i})_{t}$. These are then cut by a Hartree-Fock (HF) procedure to get a closed set of differential equations between $p_{k}$ and $f^{(c)}_{k} = (a^{\dagger}_{k} a_{k})_{t}$, $f^{(h)}_{k} = (b^{\dagger}_{k} b_{k})_{t}$. The resulting equations, which are quite heavy, are then solved numerically using a 4th-order Runge-Kutta scheme.

One unpleasant drawback of this SBE procedure [14], is a divergence in the carrier-carrier scattering rate, coming from the HF approximation, which has to be cut “by hand”. Standard HF approximation generates the excitonic poles [14], but higher-order Coulomb correlations are out of reach. These, including interband electron-hole exchange, can be included, in the case of equilibrium, through the Bethe-Salpeter equation [25–29], or nonequilibrium Green functions [30,31]. Both techniques again are numerically quite demanding [29,32].

It has recently been found that single-exciton SBE based on time-dependent density functional theory (TDDFT) provide a simpler approach [33]. However, they cannot yet account for two-exciton correlations. In particular, dark excitons with spin $\pm 2$ are not reached [21] while, evidently, such dark excitons are produced by carrier exchange between two opposite spin bright excitons [34,35]. Furthermore, the biexcitonic bound state cannot be reached. Many important dynamical effects are thus completely lost in these equations. Possible improvement can come from dynamics-controlled truncation (DCT) at the two-exciton level [14,17,18] in order to better obtain biexcitonic correlation. This DCT method is similar to the BBKY (Bogoliubov-Born-Green-Kirkwood-Yvon) hierarchy in statistical mechanics for the time evolution.
of a many-body system. Schumacher et al. [36] made recently use of this DCT scheme.

In these works, the physical understanding of the results, given through numerical curves, is quite difficult to grasp because the equations are supposed to contain “everything” at once. In particular, these methods are rather inappropriate to separate effects induced by Coulomb interaction from those coming from carrier exchanges between excitons, while our coboson procedure makes this analysis quite transparent.

Oestreich et al. [37] proposed a conceptually different method to derive the perturbative expansion of the polarization in the photon field. By formally introducing Hubbard operators associated to the N-pair eigenstates of the semiconductor Hamiltonian, they get an exact expression of the third-order polarization induced by a t-dependent photon field, which they assume to be established adiabatically (see eq. (2.31) of ref. [37]). Their work mainly deals with the part describing correlation effects between two excitons. However, since the two-pair states necessary to get the third-order Coulomb term of the polarization are not analytically known, this correlation term is ultimately computed numerically, within a 1D Hubbard model—which strongly simplifies the physics.

The procedure we present here is far more transparent since it allows us to find analytical expressions for the time evolution of the polarization—here given explicitly when the laser pulse is on—up to any arbitrary order in Coulomb processes and photon field. This photon field expansion is fully consistent with considering exciton operators instead of polariton operators. Our new procedure has the huge advantage to trace back all dynamical effects to these expansion terms. It is in particular possible to distinguish effects arising from Pauli blocking, from effects arising from Coulomb interaction and show that, once more, Pauli blocking dominates at large detuning.

Since our theory includes carrier-exchange interaction between two excitons, the effect of dark excitons on the polarization can also be directly analyzed, although not explicitly done here. This is of particular importance for the precise investigation of Bose-Einstein condensate since condensation is going to be formed within the dark exciton gas [34,35].

Conclusion. – We here present an alternative approach to semiconductor Bloch equations (SBE) which gives the time evolution of the polarization induced by a laser pulse analytically. This approach avoids the heavy numerics associated with SBE as well as spurious singularities coming from necessary truncations.

This analytical solution relies upon using correlated pairs, i.e., excitons, instead of free pairs, their interaction, responsible for the polarization, being handled through the composite-exciton many-body theory [1]. This approach makes the two physical channels producing the polarization completely transparent: fermion exchange and fermion interaction, the former being dominant at large photon detuning. Our approach can deal with biexcitonic resonance in a simple way, this resonance being uneasy to reach within the usual SBE formalism. It can also deal with the formation of dark excitons by carrier exchange between bright excitons.

Appendix

For readers interested in technical aspects, let us give the main steps leading to eqs. (13), (15), (18).

a) The Pauli part of $\langle \langle B^4 \rangle \rangle$ comes from $\langle \langle \psi_t | D_{ip} | \psi_t \rangle \rangle$. By using eqs. (10), (11), this mean value can be written as

$$
\langle \langle D_{ip} \rangle \rangle_t = \int_{-\infty}^{+\infty} \frac{dx'}{2\pi} \int_{-\infty}^{+\infty} \frac{dx}{2\pi} e^{i(x'-a_{0})t} e^{-i(x+a_{0})t} \times D_{ip}(x'-i0_+, x+i0_+),
$$

(A.1)

where, since $D_{ip}|v\rangle = 0$ while $\tilde{H}_{sc}|v\rangle = 0$, we do have

$$
D_{ip}(a', a) = \frac{1}{a'} \sum_{n,n'=0}^{\infty} \langle \phi_n(a') | D_{ip} | \phi_n(a) \rangle, \quad \text{(A.2)}
$$

$$
|\phi_n(a)\rangle = \left( \frac{1}{a - \tilde{H}_{sc}} \right)^n \frac{1}{a - \tilde{U}^\dagger} |v\rangle. \quad \text{(A.3)}
$$

Due to eqs. (1), (2), $\langle v | B_j D_{ip} B_l^\dagger | v \rangle$ is simply equal to $2\lambda_{ijkp}$. This gives the quadratic term in photon coupling of $D_{ip}(a', a)$, which comes from $n = n' = 0$ in eq. (A.2), as

$$
D_{ip}^{(2)}(a', a) = \frac{2}{a'} \sum_{jk} \frac{\Omega_{ij}\Omega_{k}^{*}}{|a' + \omega_j| |a + \omega_k|} \lambda_{ijkp}. \quad \text{(A.4)}
$$

$$
\langle \langle D_{ip} \rangle \rangle_t^{(2)}
$$

follows from the above equations inserted in eq. (A.1). Equation (8) then leads to eq. (13) for the cubic contribution to $\langle \langle B^4 \rangle \rangle_t$ induced by Pauli blocking.

b) The Coulomb part of $\langle \langle B^4 \rangle \rangle_t$ comes from $\langle \langle V_p^4 \rangle \rangle_t$. It reads as $\langle \langle D_{ip} \rangle \rangle_t$ in eqs. (A.1), (A.2) with $D_{ip}$ replaced by $V_p$. Since $V_p$ creates a pair while $V_p^4$ creates a pair, the lowest-order term in photon coupling, is obtained for $n = n' = 1$, instead of $n = n' = 0$ in eq. (A.4), as

$$
V_p^{(3)}(a', a) = \frac{1}{a'a'} \sum_{ijk} \frac{\Omega_{ij}\Omega_{k}^{*}}{|a' + \omega_j| |a + \omega_k|} C_{ijkp}(a'). \quad \text{(A.5)}
$$

Equation (4) allows us to write the Coulomb prefactor as

$$
C_{ijkp}(a') = \langle v | B_i B_j \frac{1}{a' - \tilde{H}_{sc}} V_k B_k^\dagger | v \rangle
$$

$$
= \sum_{m,n} M_{ijmn}(a') \xi (m, k). \quad \text{(A.6)}
$$

The knowledge of the whole two-pair eigenstate spectrum is required to calculate

$$
M_{ijmn}(a') = \langle v | B_i B_j \frac{1}{a' - \tilde{H}_{sc}} B_m^\dagger B_n^\dagger | v \rangle. \quad \text{(A.7)}
$$

i) When the molecular biexciton is well separated, it controls $M_{ijmn}(a')$ for photons close to the biexciton resonance. This $XX$ biexciton state then gives in
eq. (A.7)
\[ M_{ijmn}(a') \simeq \frac{\langle v|B_i B_j XX|XX B^\dagger_m B^\dagger_n|v \rangle}{a' + 2\omega - E_{XX}}, \]
so that eq. (A.6) gives
\[ C_{ijkp}(a') \simeq \frac{2\xi_{ijkp}(XX)}{a' + 2\omega - E_{XX}}, \]
with \( \xi_{ijkp}(XX) \) given by eq. (16). By inserting it into eq. (A.5), we get \( \langle \langle V_p \rangle \rangle_t \) through an equation similar to eq. (A.1), from which the Coulomb part of the polarization given in eq. (15) follows easily.

ii) We can also get \( M_{ijmn}(a') \) by iterating eq. (17),
\[ (a' + \omega_i + \omega_j)M_{ijmn}(a') = \langle v|B_i B_j B^\dagger_m B^\dagger_n|v \rangle + \sum_{p,q} \xi_{ijqp} M_{pqmn}(a'). \]
Equations (1), (2) give the first term as \( \delta_{i,n}\delta_{j,m} - \lambda \frac{j}{i} \frac{m}{n} + (m \leftrightarrow n) \). By keeping it only in eq. (A.6), we get
\[ C_{ijkp}(a') \simeq \frac{2\xi_{ijkp}}{a' + \omega_i + \omega_j}, \]
with \( \xi_{ijkp} \) defined in eq. (19). The contribution to \( \langle \langle V_p \rangle \rangle_t \) which is third order in photon coupling and first order in Coulomb scattering divided by detuning, given by eq. (18), then follows from eq. (A.5). Higher-order terms in Coulomb scattering divided by detuning can be obtained by iterating eq. (A.10).

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MC acknowledges a one-month invitation by the University of Central Florida at Orlando, where this work started. MNL acknowledges a one-month invitation by the University Pierre et Marie Curie in Paris, as well as support from NSF ECCS-0725514, DARPA/MTO HR0011-08-1-0059, NSF ECCS-0901784, and AFOSR FA9550-09-1-0450.

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