Novel approach to nonlinear susceptibility

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Abstract

The calculation of the third order susceptibility still is a long standing fundamental problem of particular importance in nonlinear nanooptics: Indeed, cancellation of size-dependent terms coming from uncorrelated excitations is expected, but up to now shown for very simple Hamiltonians only. Using a many-body theory recently developed to handle interacting close-to-bosons, we prove it here for arbitrary H. This new formalism actually provides the first clean way to calculate nonlinear susceptibilities, with results different from previous ones.

PACS.: 71.35.-y Excitons and related phenomena
Size enhancement effect in nonlinear optical processes is of great interest in nanoscience and technology as it provides an additional degree of freedom for the control of matter. Care is however necessary in the calculation of nonlinear susceptibilities due to possible cancellation among their various terms [1]. Indeed, for non-interacting bosons, the cancellation is so complete that all nonlinear susceptibilities vanish identically. This shows that optical nonlinearities only come from the non-bosonic nature of the excitations, and/or their possible interactions.

In the various terms of the third order susceptibility $\chi^{(3)}$, the ones with uncorrelated excitations increase linearly with the sample “volume” $L^D$. As the susceptibility is an intensive quantity, all these volume-linear terms have clearly to cancel. However, along with them, other terms in $L^0$ can also disappear. This is why, if this cancellation is not handled properly, the obtained $\chi^{(3)}$ may well be incorrect. The cancellation problem is thus crucial for nonlinear optics [2].

A lot of controversial arguments [3] have been raised to justify the cancellation of volume-linear terms. Some fundamental aspects were clarified through the study of a simple model consisting of non-interacting 1D Frenkel excitons with Pauli exclusion. It allows to analytically show [4] the cancellation of these volume-linear terms for arbitrary frequencies. Numerical evaluation of the remaining terms shows a size enhancement for sample sizes within the long wavelength approximation and a possible saturation when the size approaches the smaller of either, the coherence length due to non-radiative scatterings, or the relevant wavelength of light [5]. This model calculation, extended to higher dimension for excitons having some kind of interactions [6], again shows cancellation of the volume-linear terms.

Although these simple models give a reasonable picture of the problem, they are not sufficient to carry out reliable calculations in realistic situations. A general method for calculating nonlinear susceptibilities which does not suffer from this cancellation problem, was thus highly needed.

A new many-body theory has been recently developed [7-13] to handle interactions between close-to-bosons: It allows to extract from the quantities of physical interest, the parts coming from pure Pauli “interaction” and the parts coming from Coulomb interaction, either through direct or exchange processes. We use this “commutation technique”
here, to solve the cancellation problem of the third order susceptibility, by proving that the terms independent from exciton interactions cancel out exactly for any matter Hamiltonian. As expected, the remaining terms of $\chi^{(3)}$ depend on interactions only, pure Pauli or Coulomb dressed by exchange, our new formalism allowing to write them in a compact way.

The third order nonlinear susceptibility is the integral kernel relating the field vector potentials to the current density induced by the semiconductor-photon coupling taken at third order. For photons turned on adiabatically from $-\infty$ to $t_0$, this induced current density reads [14]

$$J = \int_{-\infty}^{t_0} dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_2} dt_3 \langle v | \left[ \left[ [0, I_1], I_2 \right], I_3 \right] | v \rangle \, ,$$

where $I_{j \neq 0} = -ie^{iHt_j}H^\text{int}_{t_j}e^{-iHt_j}$, while $I_0 = e^{iHt_0}J_r e^{-iHt_0}$. $H$ is the matter Hamiltonian, $J_r$ the induced current density operator and $H^\text{int}_t = e^{eitU + h.c.}$ the coupling between matter and $(\omega, Q)$ photons, if we consider one photon field only for simplicity, $\epsilon = 0_+$ being the adiabatic switching factor. $J_r$ and $H^\text{int}_t$ create or destroy one excitation in the sample. In terms of creation operators for the one-excitation eigenstates $B^\dagger_m$ defined as $(H - E_m)B^\dagger_m | v \rangle = 0$, they formally read

$$J_r = \sum_m J_{r,m} \, , \quad J_{r,m} = j_m(r) B_m + h.c. \, ,$$

$$U \simeq \sum_m \mu_m B_m \, ,$$

eq. (3) being valid within the rotating wave approximation. For Wannier excitons, $m$ stands for $(\nu_m, Q_m)$ with $Q_m$ being the exciton center of mass momentum and $\nu_m$ the relative motion index; $\mu_m = A \cdot G \delta_{Q_m, Q} \langle r = 0 | \nu_m \rangle L^{D/2}$, while $j_m(r) = -G e^{iQ_m \cdot r} \langle r = 0 | \nu_m \rangle L^{-D/2}$, with $A$ being the field vector potential and $G$ the valence-conduction Kane vector [15].

Due to eq. (2), $J$ thus appears as $\sum_m J_m$, where $J_m$ is given by eq. (1), with $I_0$ calculated using $J_{r,m}$, instead of $J_r$. By expanding the commutator of eq. (1) and by noting that $I^\dagger_{j \neq 0} = -I_j$ while $I^\dagger_0 = I_0$, $J_m$ actually reads $\tilde{J}_m + c.c.$, with $\tilde{J}_m = K_{0123} + K_{2103} + K_{3102} + K_{3201}$ and

$$K_{ijkl} = \int_{-\infty}^{t_0} dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_2} dt_3 \langle v | I_i I_j I_k I_l | v \rangle \, .$$

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The purpose of this letter is to show how the 8 terms of $J_m$ can be calculated formally, i.e., without knowing $H$, in order (i) to prove that the sum of their non-interacting contributions cancel out exactly, (ii) to write a compact expression of the remaining terms.

As explicitly shown below, the $\langle v|I_i I_j I_k I_l|v\rangle$’s of eq. (4) split into “easy” terms in which the middle state is the vacuum — so that they can be calculated exactly as they depend on one-excitations only — and “tricky” terms in which the middle state contains two excitations. As the two-excitation eigenstates are usually unknown, it has long seemed hopeless to prove that, whatever $H$ is, these “tricky” terms contain parts which exactly cancel the “easy” terms.

Using the “commutation technique”[8,9] we recently developed to handle many-body effects between close-to-bosons, we can perform commutations between $B^\dagger_m$ and the $H$-dependent operators of these “tricky” terms, to rewrite them as a sum of “non-interacting”, “Pauli” and “Coulomb” contributions. The “tricky” terms of the $K_{ijkl\neq0123}$’s of $\tilde{J}_m$ then read $j^*_m(r, t_0) (\beta_{ijkl} + \gamma_{ijkl} + \delta_{ijkl})$, with $j^*_m(r, t_0) = j_m^*(r) e^{(3\epsilon + i\omega)t_0}$, the prefactors $\beta_{ijkl}, \gamma_{ijkl}, \delta_{ijkl}$ coming from non-interacting, Pauli and Coulomb processes, respectively. $K^{(tricky)}_{0123}$ has a similar form, with $j^*_m(r, t_0)$ replaced by $j_m(r, t_0)$, while the “easy” terms of all the $K_{ijkl}$’s of $\tilde{J}_m$ read $j_m(r, t_0) \alpha_{ijkl}$.

From the formal expressions of these $\alpha$’s and $\beta$’s, which appear as $H$ matrix elements, it is then possible to show the far from obvious relations, $\alpha_{0123} + \alpha_{2103} + \beta^*_{3102} = 0$, $\alpha_{3201} + \alpha_{3102} + \beta^*_{2103} = 0$ and $\beta_{0123} + \beta_{3201} = 0$, whatever $H$ is [16]. Consequently, only remains in the induced current density, the $\gamma$ and $\delta$ parts coming from Pauli and Coulomb interactions between two excitations, as expected. It ultimately reads

$$J_m = j^*_m(r) e^{i\omega t_0} \frac{2}{E_m} \sum_{p,q,n} \frac{\mu_p \mu_q \mu_n^*}{E_p' E_q' E_n'} \times \left[ \lambda_{pqmn} + \frac{\xi_{pqmn}^{\text{dir}} - \xi_{pqmn}^{\text{in}}}{E_p' + E_q'} + \cdots \right] + c.c.,$$

(5)

$E'_i = E_i - \omega$ being the detuned exciton energy, the next term having one more $\xi$ divided by detuning.

$\lambda_{pqmn}, \xi_{pqmn}^{\text{dir}}$ and $\xi_{pqmn}^{\text{in}}$ are the exchange, direct Coulomb and “in” exchange Coulomb scatterings of the “commutation technique”. Defined through [8,9]

$$[B_m, B^\dagger_l] = \delta_{mi} - D_{mi}, \quad [D_{mi}, B^\dagger_l] = 2 \sum_n \lambda_{mnij} B^\dagger_n,$$

(6)
\begin{equation}
[H, B^\dagger_i] = E_i B^\dagger_i + V^\dagger_i,
\quad [V^\dagger_i, B^\dagger_j] = \sum_{mn} \xi_{mnij} B^\dagger_m B^\dagger_n,
\end{equation}

and \( \xi_{mnij} = \sum_{rs} \lambda_{mnr} \xi_{rrij} \), they read in terms of the one-excitation eigenwavefunctions of \( H \) (see eqs. (26,28) of ref. [8]). The “deviation-from-boson” operator \( D_{mi} \) and the \( \lambda_{mnij} \)'s physically come from Pauli exclusion which makes the excitons close-to-bosons only, while the “creation potential” \( V^\dagger_i \) and the \( \xi_{mnij} \)'s come from Coulomb interactions with the \( i \) exciton.

To calculate \( J_m \), we have also used [10]

\begin{equation}
\frac{1}{a - H} B^\dagger_m = \left( B^\dagger_m + \frac{1}{a - H} V^\dagger_m \right) \frac{1}{a - H - E_m},
\end{equation}

which follows from eq. (7) and

\begin{equation}
e^{-iH\tau} B^\dagger_m = B^\dagger_m e^{-i(H+E_m)\tau} + W^\dagger_m(\tau),
\end{equation}

\begin{equation}
W^\dagger_m(\tau) = \int_{-\infty}^{+\infty} \frac{dx}{2i\pi} \frac{e^{-i(x-i\eta)\tau}}{x - H - i\eta} V^\dagger_m \frac{1}{x - H - E_m - i\eta},
\end{equation}

which follows from eq. (8) and the integral representation of \( e^{-iH\tau} \) which, for \( \tau < 0 \) and \( \eta > 0 \), reads [17]

\begin{equation}
e^{-iH\tau} = \int_{-\infty}^{+\infty} \frac{dx}{2i\pi} \frac{e^{-i(x-i\eta)\tau}}{x - H - i\eta}.
\end{equation}

To show how this “commutation technique” works and to grasp the physics which controls the \( \alpha, \beta, \gamma, \delta \) prefactors, let us consider a \( K_{ijkl} \) particularly complicated to calculate, namely \( K_{2103} \), because in addition to the fact that its middle state has two excitations, the times \( t_i \) are “shaken up” compared to \( K_{0123} \), which makes the integration over times more difficult.

\( e^{-iHt}\langle v \rangle = \langle v \rangle \) and \( \langle v | I_2 \rangle \) and \( \langle I_3 | v \rangle \) reduce to one term. As the non-zero contributions of \( I_1 I_0 \) to \( k_{2103} = \langle v | I_2 I_1 I_0 I_3 | v \rangle \) are the ones in \( U^\dagger B_m \) and \( U B^\dagger_m \) only, \( k_{2103} \) splits in two. Its “easy” term, containing \( U^\dagger B_m \), reads \( k_{2103}^{(easy)} = i e^{(3\tau_1 + 2\tau_2 + \tau_3)} a_{2103} j_m(r, t_0) \) with

\begin{equation}
a_{2103} = \langle v | U^{-iH'\tau_2} U^\dagger e^{-iH\tau_1} B_m e^{iH'(\tau_1 + \tau_2 + \tau_3)} U^\dagger | v \rangle,
\end{equation}

where \( H' = H - \omega \), while the \( \tau_i \)'s, defined as \( t_i = \tau_i + t_{i-1} \), run from \(-\infty \) to \( 0 \). The \( a_{2103} \) middle state being the vacuum, the middle \( H \) can be replaced by 0 and the right \( H' \) by \( E'_m \). As \( \langle v | B_m U^\dagger | v \rangle = \mu_m^* \), the formal integration of this “easy” term over the \( \tau_i \)'s readily gives a \( a_{2103} j_m(r, t_0) \) contribution to \( K_{2103} \), with

\begin{equation}
a_{2103} = \frac{\mu_m^*}{(E'_m - 3i\epsilon)(E'_m - i\epsilon)} \langle v | U \frac{1}{H' - E'_m + 2i\epsilon} U^\dagger | v \rangle.
\end{equation}
The $k_{2103}$ “tricky” term reads $k_{2103}^{(\text{tricky})} = i e^{i(3\tau_1 + 2\tau_2 + \tau_3)} b_{2103} j_m^*(r, t_0)$ with

$$b_{2103} = \langle v | U e^{-iH'\tau_2} U e^{-iH'\tau_1} B_m^\dagger e^{iH'(\tau_1 + \tau_2 + \tau_3)} U^\dagger | v \rangle ,$$

and $H'' = H - 2\omega$. The integrations over $\tau_1$ and $\tau_2$ are not straightforward because these times appear in different places. We can put the $\tau_1$'s together by passing $e^{-iH''\tau_1}$ over $B_m^\dagger$ through eq. (9). This splits $b_{2103}$ as $c_{2103} + d_{2103}$: From the first term of eq. (9) we get

$$c_{2103} = e^{-i\mu_m \tau_1} \langle v | U e^{-iH'\tau_2} U B_m^\dagger e^{iH'(\tau_2 + \tau_3)} U^\dagger | v \rangle .$$

$\tau_2$ still is in two places. To go further, we rewrite $UB_m^\dagger$ as $B_m^\dagger U + [U, B_m^\dagger]$, i.e., $B_m^\dagger U + \mu_m - \sum_q \mu_q D_{qm}$, due to eqs. (3,6). In the part with $B_m^\dagger U$, we can replace the left $H'$ by $E_m'$ since the middle state is then the vacuum. As $\langle v | UB_m^\dagger | v \rangle = \mu_m$, the two first terms of $UB_m^\dagger$ give a $\beta_{2103} j_m^*(r, t_0)$ contribution to $K_{2103}$, with

$$\beta_{2103} = \frac{\mu_m}{E_m' + 3i\epsilon} \times \langle v | U \left( \frac{1}{H' - E_m' - 2i\epsilon} - \frac{1}{2i\epsilon} \right) \frac{1}{H' - i\epsilon} U^\dagger | v \rangle .$$

As for the third term of $UB_m^\dagger$, in $D_{qm}$, it physically comes from Pauli “interaction”, so that it is zero for boson-excitons. The contribution it induces to $K_{2103}$ reads $\gamma_{2103} j_m^*(r, t_0)$ with

$$\gamma_{2103} = \sum_q \frac{-i\mu_q}{E_m' + 3i\epsilon} \int_{-\infty}^0 \tau_2 e^{2i\tau_2} d\tau_2 \times \langle v | U e^{-iH'\tau_2} D_{qm} e^{iH'\tau_2} \frac{e^{iH'\tau_2}}{H' - i\epsilon} U^\dagger | v \rangle .$$

Since $H'$ acts on one-excitations only, the integration over $\tau_2$ can be performed exactly, using eq. (3). As $\langle v | B_p D_{qm} B_m^\dagger | v \rangle = 2\lambda_{pqmn}$, due to eq. (6), we find [18]

$$\gamma_{2103} = -\frac{2}{E_m' + 3i\epsilon} \sum_{p,q,n} \frac{\lambda_{pqmn} \mu_p \mu_q \mu_n^*}{(E_m' - i\epsilon)(E_m' - E_q' - 2i\epsilon)} .$$

In passing $e^{-iH''\tau_1}$ over $B_m^\dagger$ in eq. (14), we also generate a $d_{2103}$ term which comes from $W_m^\dagger(\tau_1)$ in eq. (9), i.e., Coulomb interactions with the $m$ exciton. This term produces a $\delta_{2103} j_m^*(r, t_0)$ contribution to $K_{2103}$ with

$$\delta_{2103} = i \int_{-\infty}^0 \tau_2 e^{2i\tau_2} \langle v | U e^{-iH'\tau_2} U W_m^\dagger \frac{e^{iH'\tau_2}}{H' - i\epsilon} U^\dagger | v \rangle ,$$

(19)
where $\mathbf{W}_m^\dagger$ is linked to $W_m^\dagger(\tau_1)$ through

$$\mathbf{W}_m^\dagger = -i \int_{-\infty}^0 d\tau_1 W_m^\dagger(\tau_1) e^{(3\epsilon+i(H' + 2\omega))\tau_1}.$$  \hspace{1cm} (20)

Using eq. (10), it reads

$$\mathbf{W}_m^\dagger = \int_{-\infty}^{+\infty} dy \frac{1}{2i\pi} \frac{1}{y - H'' - i\eta} V_m^\dagger \times \frac{1}{(y - H' + i\epsilon')(y - H' - E_m' - i\eta)}.$$  \hspace{1cm} (21)

with $\eta$ chosen such that $\epsilon' = 3\epsilon - \eta > 0$, to insure convergence for $\tau_1 \to -\infty$.

The integration over $\tau_2$ can be performed exactly, using again eq. (3). This leads to

$$\delta_{2103} = \frac{1}{E_m' + 3i\epsilon} \sum_{p,q,n} \frac{w_{pqmn}}{(E_n' - i\epsilon')(E_n' - E_q' - 2i\epsilon')} \cdot \hspace{1cm} (22)

w_{pqmn} = (E_m' + 3i\epsilon) \langle v | B_p B_q \mathbf{W}_m^\dagger B_n^\dagger | v \rangle \text{ corresponds to Coulomb processes between the } m \text{ exciton and the photcreated ones (} p, q \text{) and } n. \text{ When inserted in } w_{pqmn}, \text{ the } \mathbf{W}_m^\dagger \text{, of eq. (21) can have its two right } H' \text{ replaced by } E_n'. \text{ Integration over } y, \text{ done by residues, yields }

$$w_{pqmn} = \langle v | B_p B_q \frac{1}{E_n' - H'' - 3i\epsilon} V_m^\dagger B_n^\dagger | v \rangle.$$  \hspace{1cm} (23)

$w_{pqmn}$ cannot be calculated exactly because $H''$ acts on two excitons. It can however be expanded in Coulomb interactions using eq. (8). At lowest order, this leads to replace $H''$ by $E_p' + E_q'$. From eq. (7) and the scalar product of two-exciton states, \( \langle v | B_p B_q B_r^\dagger B_s^\dagger | v \rangle = \delta_{pr} \delta_{qs} + \delta_{ps} \delta_{qr} - 2\lambda_{pqrs}, \) which follows from eq. (6), we end with

$$w_{pqmn} \simeq 2 \frac{\xi_{pqmn}^\text{dir} - \xi_{pqmn}^\text{in}}{E_n' - E_p' - E_q' - 3i\epsilon} + \cdots.$$  \hspace{1cm} (24)

$\xi_{pqmn}^\text{in}$ is an exchange Coulomb scattering with its Coulomb interactions between the “in” excitons ($m, n$) only. Once again [8,9,10], it differs from the “out” exchange Coulomb scattering $\xi_{pqmn}^\text{out} = \sum_{r,s} \xi_{pqrs}^\text{dir} \lambda_{rsmn}$ appearing in the effective bosonic Hamiltonian for excitons [19], widely used up to now, in spite of the fact that it is unphysical because not hermitian.

The “commutation technique” being new, and the project to extract from $\mathbf{J}$ its non-interacting terms ambitious, it seemed to us useful to detail the calculation of a particular $K_{ijkl}$, even if this calculation may appear somewhat technical. The expressions of $\alpha$, $\beta$, $\gamma$, $\delta$’s it provides, are actually quite interesting.
(i) The $\alpha$’s and $\beta$’s, independent from Pauli and Coulomb interactions, contain three exciton-photon couplings $\mu_p \mu_q \mu_n^*$. In the case of Wannier excitons, they generate three factors $L^{D/2}$ and force $Q_p = Q_q = Q_n = Q$ so that no sample volume comes from the sums over $(p, q, n)$. By including the $L^{-D/2}$ factor of $j_m(r)$, these $\alpha$’s and $\beta$’s thus generate volume-linear contributions $L^D$ to $J_m$, in agreement with model Hamiltonians. They have to and do cancel exactly for any kind of excitons.

(ii) The $\gamma$’s and $\delta$’s have the same number of exciton-photon couplings. They however have one additional Pauli “scattering” for $\gamma$’s, and Coulomb scattering for $\delta$’s. As, for Wannier excitons [8,9,12], these scatterings both behave as the exciton volume divided by $L^D$, the $\gamma$ and $\delta$ contributions to the current density are indeed sample volume free.

(iii) Finally, at large detuning, the induced current density is dominated by pure Pauli processes: As obvious from eqs. (6,7), the $\lambda_{pqmn}$’s are dimensionless, while the $\xi_{pqmn}$’s have the dimension of an energy, so that the $\delta$’s do have one more energy denominator than the $\gamma$’s, which makes them smaller in the large detuning limit.

Conclusion

Using a new many-body theory for interacting close-to-bosons, we have succeeded in extracting the various parts of the induced current density linked to uncorrelated excitations and proved their cancellation, without knowing $H$. This formalism actually provides the first clean way to calculate nonlinear susceptibilities. After combining all its contributions, the induced current density appears in terms of the exchange and Coulomb “scatterings” of the “commutation technique” (see eq. (5)). Explicit calculations for Wannier and Frenkel excitons, as well as polarization effects linked to spin degrees of freedom, will be presented elsewhere. We can however say, just from dimensional arguments, that, at large detuning, nonlinear susceptibilities are entirely controlled by Pauli interactions between excitations, without the help of any Coulomb process — result beyond the reach of the effective bosonic Hamiltonians for excitons.

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(14) See e.g., eq. (4) of ref. (1) or eq. (2.109) of ref. (2)
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(16) This grouping was already noted in ref. (4) and ascribed to the classification of the various terms, according to their frequency dependences, which guarantees a cancellation independent from frequencies.
(17) See for instance, M.C., J. Phys. A 34, 6087 (2001)
(18) $\gamma_{2103}$ and $\delta_{2103}$ seem to have singular terms for $q = n$. They do in fact combine with similar ones appearing in $\gamma_{3102}$ and $\delta_{3102}$ to ultimately give eq. (5).
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