Forschungszentrum Jülich
in der Helmholtz-Gemeinschaft

Institute of Thin Films and Interfaces
Institute 1: Semiconductor Thin Films and Devices

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Publisher and Distributor: Forschungszentrum Jülich GmbH
Zentralbibliothek
D-52425 Jülich
Telefon (02461) 615368 - Telefax (02461) 61-6103
e-mail: zb-publikation@fz-juelich.de
Internet: http://www.fz-juelich.de/zb

Editorial: Institute of Thin Films and Interfaces
Institute 1: Semiconductor Thin Films and Devices

Published in full on the Internet
Persistent Identifier: urn:nbn:de:0001-00257
http://nbn-resolving.de/urn/resolver.pl?urn=urn:nbn:de:0001-00257

On the semiclassical approach to the optical absorption spectrum: A counter-example

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(February 23, 2005)

Abstract

We investigate an electron-hole two level system coupled to a single photon mode in the presence of an external light field. For a specific choice of the Coulomb matrix element and for a specific class of initial states, we obtain an exact analytical expression for the polarization function $\langle e^+h^+\rangle(t)$ which is valid *for arbitrary electric field amplitudes.* As an interesting application, we demonstrate that neither the formula $A(\omega) = Im(P(\omega)/E(\omega))$ nor the improved formula $A(\omega) = 2 \cdot (\omega/c_0) \cdot n_b \cdot Im\sqrt{1 + \chi(\omega)/\varepsilon_b}$ can be applied to calculate the optical absorption spectrum $A(\omega)$ in the limit of vanishing (phenomenological) damping constants.

I. INTRODUCTION

In solid state physics, the calculation of optical absorption spectra is often based on a *semiclassical approach.* Here, the electron-hole system is coupled to a *classical exciting light field*, and the absorption coefficient $A(\omega)$ is obtained from the well-known equation

$$A(\omega) = 2 \cdot \frac{\omega}{c_0} \cdot n_b \cdot Im\sqrt{1 + \frac{\chi(\omega)}{\varepsilon_b}} \, . \qquad (1)$$

Basic definitions as well as various illustrations of Eq. (1) can be found in many textbooks (see, e.g., Ref[1]). For this reason, we are not going to explain further details here (as these are not decisive for our argumentation). In the limit $\chi(\omega)/\varepsilon_b \ll 1$, $A(\omega)$ is - apart from unimportant factors - given by the formula[1]

$$A(\omega) = Im \frac{P(\omega)}{E(\omega)} \,. \tag{2}$$

Eqs. (1) and (2) are well established in the literature. In many cases, the desired optical absorption spectrum can only be obtained in the framework of a numerical approach. In order to perform a numerical calculation explicitly, the introduction of phenomenological damping constants is indispensable. From a strictly formal point of view, however, there is no doubt that such a procedure has to be regarded mainly as a technical trick: *Any relaxation process - if it exists at all - has its origin in the Hamiltonian.* For this reason, Eqs. (1) and (2) should also be valid in the limit of vanishing (phenomenological) damping constants - provided that $A(\omega)$ is introduced as a *distribution.*

In the present article, we present a counter-example: We analyze an electron-hole two level system coupled to a single photon mode in the presence of an external light field. For a specific choice of the Coulomb matrix element and for a specific class of initial states, the complete infinite hierarchy of coupled equations for the expectation values can be solved analytically in closed form. Our result is valid *for arbitrary electric field amplitudes.* Making use of the *exact formula* for the polarization function $\langle e^+h^+\rangle(t)$, we shall demonstrate that neither Eq. (2) nor Eq. (1) can be applied to calculate the optical absorption spectrum $A(\omega)$ in the limit of vanishing (phenomenological) damping constants.

We emphasize that our statements are valid *even in the case that the particle-photon coupling constant vanishes.* Here, an additional numerical study demonstrates that the formula $Im(P(\omega)/E(\omega))$ leads to an incorrect result *also in a vicinity of the specific Coulomb matrix element mentioned above.*

II. AN ELECTRON-HOLE TWO LEVEL SYSTEM COUPLED TO A SINGLE PHOTON MODE IN THE PRESENCE OF AN EXTERNAL LIGHT FIELD

A. Introduction of the model Hamiltonian

In the following part of our considerations, we are concerned with an electron-hole two level system coupled to a single photon mode. In the presence of an external light field, the corresponding model Hamiltonian H reads as follows:

$$H = H_0 + V(t) \ , \tag{3}$$

$$H_0 = H_1 + H_2 \ , \tag{4}$$

$$H_1 = \varepsilon_0 \cdot e^+ e + \varphi_0 \cdot h^+ h - 2 \cdot W^{eh} \cdot e^+ h^+ h e \ , \tag{5}$$

$$\begin{aligned} H_2 = {} & \hbar\Omega \cdot a^+ a + \ : \left(e^+ + h\right) \left(M \cdot a + M^* \cdot a^+\right) \left(e + h^+\right) \ : = \\ = {} & \hbar\Omega \cdot a^+ a + \left(M \cdot a + M^* \cdot a^+\right) \cdot \left(e^+ h^+ + he + e^+ e - h^+ h\right) \ . \end{aligned} \tag{6}$$

The symbol $: \ldots :$ in Eq. (6) denotes *normal order*. The mathematical structure of the semiclassical light-matter interaction operator $V(t)$ is determined by the particle-photon interaction operator[2] in Eq. (6):

$$V(t) = S(t) \cdot \left(e^+ h^+ + he + e^+ e - h^+ h\right) \ . \tag{7}$$

Here, $S(t)$ contains the amplitude $E(t)$ of the exciting light field as well as the coupling matrix element. We require in advance that $E(t)$ fulfills the following conditions:

$$E(t) = 0 \ , \ if \ t \leq 0 \ ; \ E(t) \rightarrow 0 \ , \ if \ t \rightarrow \infty \ ; \ \int_0^{+\infty} |E(t)| dt < +\infty \ . \tag{8}$$

Direct inspection of Eq. (6) shows that the particle-photon interaction operator is in fact of standard type (and contains, e.g., operator products of *density type*, too). However, if we consider the non-trivial case $M \neq 0$, it is immediately seen that the vacuum state $|0\rangle$ is *not* an eigenstate of H_0. Consequently, if the initial state $|\Psi(t=0)\rangle = |0\rangle$ is chosen, *dynamical vacuum fluctuations do appear even in the absence of an external light field.* Moreover, the ground state of H_0 contains, in general, electrons, holes and photons. It is obvious that the latter statements must lead to a critical analysis of the *standard theory* of electron-hole-photon interaction itself. In a further article (see Ref.[2]), we have discussed the physical consequences to a great extent and in a quite general context. In our present work, however, we still take the standard particle-photon interaction operator in Eq. (6) for granted.

B. Eigenstates and energy spectrum of H_0

It turns out that Hamiltonian (4) can be diagonalized analytically in closed form at least for a specific choice of the Coulomb matrix element. In order to demonstrate this, we now introduce the additional condition

$$\varepsilon_0 + \varphi_0 - 2 \cdot W^{eh} = 0 \tag{9}$$

for the Coulomb matrix element W^{eh} and proceed as follows:

First of all, we remark that the electron-hole subspace of the physical system under consideration is defined by the basic states

$$|1\rangle = |0_{eh}\rangle \ , \ |2\rangle = e^+|0_{eh}\rangle \ , \ |3\rangle = h^+|0_{eh}\rangle \ , \ |4\rangle = e^+h^+|0_{eh}\rangle \ . \tag{10}$$

For the following part of our considerations, we need the basic states

$$|e_1\rangle = \frac{1}{\sqrt{2}} \cdot (|1\rangle + |4\rangle) \ , \ |e_2\rangle = |2\rangle \ , \ |e_3\rangle = |3\rangle \ , \ |e_4\rangle = \frac{1}{\sqrt{2}} \cdot (|1\rangle - |4\rangle) \ . \tag{11}$$

From Eqs. (9), (10) and (11), we obtain the relations

$$H_1|e_i\rangle|\Phi_P\rangle = \tilde{E}_i \cdot |e_i\rangle|\Phi_P\rangle \tag{12}$$

and

$$H_2|e_i\rangle|\Phi_P\rangle = \left(\hbar\Omega \cdot a^+a + P_i \cdot \left(M \cdot a + M^* \cdot a^+\right)\right)|e_i\rangle|\Phi_P\rangle \ , \tag{13}$$

where

$$\tilde{E}_1 = 0 \ , \ \tilde{E}_2 = \varepsilon_0 \ , \ \tilde{E}_3 = \varphi_0 \ , \ \tilde{E}_4 = 0 \tag{14}$$

and

$$P_1 = 1 \ , \ P_2 = 1 \ , \ P_3 = -1 \ , \ P_4 = -1 \ . \tag{15}$$

$|\Phi_P\rangle$ denotes an element of the photonic Fock space.

In view of Eqs. (12) and (13), Hamiltonian (4) can immediately be diagonalized. The eigenstates $|\Psi_{in}\rangle$ are given by

$$|\Psi_{in}\rangle = |e_i\rangle|P_{in}\rangle \ , \tag{16}$$

where

$$|P_{in}\rangle = \frac{1}{\sqrt{n!}} \cdot \left(a^+ + \gamma_i\right)^n \cdot \exp\left(-\frac{1}{2} \cdot |\gamma_i|^2\right) \cdot \exp\left(-\gamma_i^* \cdot a^+\right) |0_{Phot.}\rangle \tag{17}$$

and

$$\gamma_i = \frac{M \cdot P_i}{\hbar\Omega} \ . \tag{18}$$

The energy spectrum of H_0 is obtained from the equation

$$E_{in} = \tilde{E}_i + \left(n - |\gamma_i|^2\right) \cdot \hbar\Omega = \tilde{E}_i + n \cdot \hbar\Omega - \frac{|M|^2}{\hbar\Omega} \ . \tag{19}$$

Direct inspection of Eq. (19) shows that Hamiltonian (4) is bounded from below: *No renormalizations are necessary in order to get finite results.* The ground-state energy E_0 is found to be

$$E_0 = E_{10} = E_{40} = -\frac{|M|^2}{\hbar\Omega} \ . \tag{20}$$

From Eq. (20), we conclude that there exist two eigenstates which belong to the ground-state energy of H_0; these are

$$|\Psi_{10}\rangle = |e_1\rangle|P_{10}\rangle = \frac{1}{\sqrt{2}} \cdot (|1\rangle + |4\rangle) \cdot \exp\left(-\frac{1}{2} \cdot |\gamma_1|^2\right) \cdot \exp\left(-\gamma_1^* \cdot a^+\right) |0_{Phot.}\rangle \tag{21}$$

and

$$|\Psi_{40}\rangle = |e_4\rangle|P_{40}\rangle = \frac{1}{\sqrt{2}} \cdot (|1\rangle - |4\rangle) \cdot \exp\left(-\frac{1}{2} \cdot |\gamma_4|^2\right) \cdot \exp\left(-\gamma_4^* \cdot a^+\right) |0_{Phot.}\rangle \ . \tag{22}$$

Consequently, Hamiltonian (4) in combination with the additional condition (9) for the Coulomb matrix element W^{eh} does provide an example for *a degenerate ground-state energy.* Obviously, this property is in marked contrast to the basic features of, e.g., polaronic systems. As has been demonstrated in the literature, the ground-state energy of a large class of generalized *Fröhlich models* is in fact a simple eigenvalue[3–5] .

According to Eqs. (21) and (22), the eigenstates $|\Psi_{10}\rangle$ and $|\Psi_{40}\rangle$ are many-particle states. The same statement is true for the excited states

$$|\Psi_{1n}\rangle = |e_1\rangle|P_{1n}\rangle \ , \ |\Psi_{4n}\rangle = |e_4\rangle|P_{4n}\rangle \ , \ n \geq 1 \ ; \tag{23}$$

these eigenstates belong to the energies

$$E_{1n} = E_{4n} = n \cdot \hbar\Omega + E_0 \ , \ n \geq 1 \ . \tag{24}$$

Apart from the solutions (21), (22) and (23), there exist further eigenstates of Hamiltonian (4) which can be obtained from Eq. (17) by taking $i = 2$ or $i = 3$:

$$|\Psi_{2n}\rangle = |e_2\rangle|P_{2n}\rangle \ , \ |\Psi_{3n}\rangle = |e_3\rangle|P_{3n}\rangle \ , \ n \geq 0 \ . \tag{25}$$

The corresponding energy eigenvalues are found to be

$$E_{2n} = \varepsilon_0 + n \cdot \hbar\Omega + E_0 \ , \ E_{3n} = \varphi_0 + n \cdot \hbar\Omega + E_0 \ , \ n \geq 0 \ . \tag{26}$$

C. Solution of the time-dependent Schrödinger equation

The solution $|\Psi(t)\rangle$ of the time-dependent Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}|\Psi(t)\rangle = H(t)|\Psi(t)\rangle \tag{27}$$

can be represented by a superpositon of the eigenstates $|\Psi_{in}\rangle$:

$$|\Psi(t)\rangle = \sum_{i=1}^{4} \sum_{n=0}^{\infty} c_{in}(t) \cdot |\Psi_{in}\rangle \ . \tag{28}$$

According to our results from above, the coefficients $c_{in}(t)$ are explicitly given by the equation

$$c_{in}(t) = c_{in}(0) \cdot \exp\left(-\frac{i}{\hbar} \cdot \left(E_{in} \cdot t + P_i \cdot \int_0^t S(\tau) d\tau\right)\right) \ . \tag{29}$$

D. A first approach to the optical absorption spectrum in the limit of vanishing (phenomenological) damping constants

Our results from above exhibit an interesting feature of the model Hamiltonian under consideration: Because of the fact that $S(t)$ is a real number for any t, the relation

$$|c_{in}(t)| = |c_{in}(0)| \tag{30}$$

is immediately derived from Eq. (29). The latter equation demonstrates that

$$\langle \Psi(t)|H_0|\Psi(t)\rangle = \langle \Psi(0)|H_0|\Psi(0)\rangle \tag{31}$$

is valid for any $t \geq 0$: The total energy of the particle-photon system is just a constant. In combination with condition (8), we obtain additionally

$$\langle H(t \to -\infty)\rangle = \langle H(t \to +\infty)\rangle \ , \tag{32}$$

i.e., the optical absorption spectrum $A(\omega)$ of the particle-photon system is equal to zero:

$$A(\omega) = 0 \ . \tag{33}$$

Later on, we shall see whether the formula $A(\omega) = Im[P(\omega)/E(\omega)]$ leads to the same result - or not.

E. Solution of the hierarchy equations

After these preparations, we shall now analyze the dynamical behaviour of the system under consideration in more detail. We already know that there exist two eigenstates which belong to the ground-state energy of H_0; these are $|\Psi_{10}\rangle$ and $|\Psi_{40}\rangle$. In the following part of our considerations, we are going to determine the time evolution of the expectation values $\langle\hat{E}(a^+)^k a^l\rangle(t)$ under the initial condition

$$|\Psi(t=0)\rangle = c_{10}\cdot|\Psi_{10}\rangle + c_{40}\cdot|\Psi_{40}\rangle\ ,\ |c_{10}|^2+|c_{40}|^2 = 1\ , \tag{34}$$

where $c_{10}\equiv c_{10}(0)$ and $c_{40}\equiv c_{40}(0)$. $\hat{E}$ is an electron-hole operator sequence in normal order which is defined by the equation

$$\hat{E}|e_i\rangle = \sum_{l=1}^{4}\lambda_{il}\cdot|e_l\rangle\ . \tag{35}$$

From Eqs. (29) and (35), we obtain:

$$\langle\Psi(t)|\hat{E}(a^+)^k a^l|\Psi(t)\rangle = \gamma^k\cdot(\gamma^*)^l\cdot\left((-1)^{k+l}\cdot\lambda_{11}\cdot|c_{10}|^2+\lambda_{44}\cdot|c_{40}|^2\right)+ \tag{36}$$
$$+\exp\left(-2\cdot|\gamma|^2\right)\cdot\gamma^k\cdot(\gamma^*)^l\cdot\Lambda(t)\ ,$$

where

$$\Lambda(t) := (-1)^l\cdot\lambda_{14}\cdot c_{10}\cdot c_{40}^*\cdot\exp\left(-2\cdot\frac{i}{\hbar}\cdot\int_0^t S(\tau)d\tau\right)+ \tag{37}$$
$$+(-1)^k\cdot\lambda_{41}\cdot c_{10}^*\cdot c_{40}\cdot\exp\left(2\cdot\frac{i}{\hbar}\cdot\int_0^t S(\tau)d\tau\right)$$

and

$$\gamma := \frac{M}{\hbar\Omega}\ . \tag{38}$$

Eq. (36) is remarkable because it contains the solution for the complete infinite system of coupled equations for the expectation values under the initial condition (34). We stress explicitly that we have checked the validity of relation (36) for various equations of motion at the beginning of the infinite hierarchy, too.

By taking $\hat{E}=1$ and $k=l=1$, for example, we obtain the photon number $\langle a^+a\rangle(t)$:

$$\langle a^+a\rangle(t) = |\gamma|^2\ . \tag{39}$$

It turns out that the photon number is just a constant, i.e., it does not depend on the strength of the external light field.

By taking $k = l = 0$, we obtain the occupation numbers of the electron-hole system:

$$\langle e^+ e\rangle(t) = \langle h^+ h\rangle(t) = \frac{1}{2} - \frac{1}{2} \cdot \exp\left(-2 \cdot |\gamma|^2\right) \cdot \Lambda_{occ.}(t) \;, \tag{40}$$

where

$$\begin{aligned} \Lambda_{occ.}(t) &:= c_{10} \cdot c_{40}^* \cdot \exp\left(-2 \cdot \frac{i}{\hbar} \cdot \int_0^t S(\tau) d\tau\right) + \\ &+ c_{10}^* \cdot c_{40} \cdot \exp\left(2 \cdot \frac{i}{\hbar} \cdot \int_0^t S(\tau) d\tau\right) \end{aligned} \tag{41}$$

determines the time evolution of the expectation values under consideration. Because of $|c_{10}|^2 + |c_{40}|^2 = 1$, we conclude from Eq. (41) that

$$-1 \leq \Lambda_{occ.}(t) \leq 1 \tag{42}$$

is true, i.e., we may derive upper and lower bounds for the expectation values $\langle e^+ e\rangle(t)$ and $\langle h^+ h\rangle(t)$:

$$\frac{1}{2} - \frac{1}{2} \cdot \exp\left(-2 \cdot |\gamma|^2\right) \leq \langle e^+ e\rangle(t) = \langle h^+ h\rangle(t) \leq \frac{1}{2} + \frac{1}{2} \cdot \exp\left(-2 \cdot |\gamma|^2\right) \;. \tag{43}$$

Obviously, both expectation values are not negative and bounded from above by 1 - as it must be. In the strong-coupling limit, we obtain $\langle e^+ e\rangle(t) = \langle h^+ h\rangle(t) \approx 1/2$.

A further important example is the polarization function $\langle e^+ h^+\rangle(t)$. By taking $\hat{E} = e^+ h^+$ and $k = l = 0$, we obtain from Eq. (36):

$$\langle e^+ h^+\rangle(t) = \frac{1}{2} \cdot \left(|c_{10}|^2 - |c_{40}|^2\right) - \frac{1}{2} \cdot \exp\left(-2 \cdot |\gamma|^2\right) \cdot \Lambda_{pol.}(t) \;; \tag{44}$$

here, we have introduced the abbreviation

$$\begin{aligned} \Lambda_{pol.}(t) &:= c_{10} \cdot c_{40}^* \cdot \exp\left(-2 \cdot \frac{i}{\hbar} \cdot \int_0^t S(\tau) d\tau\right) + \\ &- c_{10}^* \cdot c_{40} \cdot \exp\left(2 \cdot \frac{i}{\hbar} \cdot \int_0^t S(\tau) d\tau\right) \;. \end{aligned} \tag{45}$$

According to Eqs. (7), (40), (44) and (45), the semiclassical interaction energy $\langle V(t)\rangle$ is thus given by

$$\langle V(t)\rangle = S(t) \cdot \left(|c_{10}|^2 - |c_{40}|^2\right) \;. \tag{46}$$

Remark: In the following part of our considerations, the coefficients c_{10} *and* c_{40} *are chosen to be real numbers.* From Eqs. (44) and (45), we conclude that

$$P(t) := \langle e^+ h^+ \rangle(t) = \frac{1}{2} \cdot \left(|c_{10}|^2 - |c_{40}|^2 \right) + \tag{47}$$
$$+ i \cdot \exp\left(-2 \cdot |\gamma|^2\right) \cdot c_{10} \cdot c_{40} \cdot \sin\left(\frac{2}{\hbar} \cdot \int_0^t S(\tau) d\tau\right)$$

is true under these circumstances.

Last but not least, we mention the expectation values $\langle e^+ h^+ a \rangle(t)$ and $\langle hea \rangle(t)$. Under the additional condition from above, we obtain from Eqs. (36) and (37):

$$\langle e^+ h^+ a \rangle(t) = -\frac{\gamma^*}{2} \cdot \left[1 - 2 \cdot c_{10} \cdot c_{40} \cdot \exp\left(-2 \cdot |\gamma|^2\right) \cdot \cos\left(\frac{2}{\hbar} \cdot \int_0^t S\left(\tau\right) d\tau\right)\right] \ , \tag{48}$$

$$\langle hea \rangle(t) = -\frac{\gamma^*}{2} \cdot \left[1 + 2 \cdot c_{10} \cdot c_{40} \cdot \exp\left(-2 \cdot |\gamma|^2\right) \cdot \cos\left(\frac{2}{\hbar} \cdot \int_0^t S\left(\tau\right) d\tau\right)\right] \ . \tag{49}$$

Direct inspection of these equations shows that the expectation value $\langle hea \rangle(t)$ is - in general - not comparatively small. As an illustration, we examine some specific cases in more detail:

First case:

$$c_{10} \cdot c_{40} \cdot \cos\left(\frac{2}{\hbar} \cdot \int_0^t S\left(\tau\right) d\tau\right) \geq 0 \ . \tag{50}$$

Here, we obtain

$$0 \leq 1 - 2 \cdot c_{10} \cdot c_{40} \cdot \exp\left(-2 \cdot |\gamma|^2\right) \cdot \cos\left(\frac{2}{\hbar} \cdot \int_0^t S\left(\tau\right) d\tau\right) \leq 1 \ , \tag{51}$$

$$1 + 2 \cdot c_{10} \cdot c_{40} \cdot \exp\left(-2 \cdot |\gamma|^2\right) \cdot \cos\left(\frac{2}{\hbar} \cdot \int_0^t S\left(\tau\right) d\tau\right) \geq 1 \tag{52}$$

and therefore

$$|\langle e^+ h^+ a \rangle(t)| \leq |\langle hea \rangle(t)| \ . \tag{53}$$

If we take, for example, $c_{10} \cdot c_{40} \geq 0$, condition (50) is fulfilled for sufficiently small t. A further example is to choose the time t such that $(2/\hbar) \cdot \int_0^t S(\tau) d\tau = (2n+1) \cdot (\pi/2)$ is true; if this condition is fulfilled, one obtains immediately $\langle e^+ h^+ a \rangle(t) = \langle hea \rangle(t) = -\gamma^*/2$.

Second case:

$$|\gamma| \gg 1 \ . \tag{54}$$

Here, we conclude from Eqs. (48) and (49) that

$$\langle e^+ h^+ a\rangle(t) \cong \langle hea\rangle(t) \cong -\frac{\gamma^*}{2} \tag{55}$$

is true under all circumstances.

Our results from above confirm our previous conclusions in Ref.[2]. The analysis of quantum-field theoretical hierarchy equations is often based on the approximation $\langle hea\rangle(t) = 0$ which seems to be justified by the physical interpretation that an electron-hole pair and a photon are removed from the system under consideration[6]. In the framework of the standard model, however, Eqs. (48) and (49) demonstrate once more that such an approximation is - in general - *not* correct.

III. SEMICLASSICAL APPROACH TO THE OPTICAL ABSORPTION SPECTRUM

We have already seen in section II, Part D that the optical absorption spectrum $A(\omega)$ of the particle-photon system under consideration is equal to zero: $A(\omega) = 0$. This statement is true because of the fact that (i) the total energy $\langle H_0\rangle$ of the particle-photon system is just a constant and (ii) the semiclassical interaction energy $\langle V(t)\rangle$ vanishes in the limits $t \to -\infty$ and $t \to +\infty$ under the conditions from above (see Eq. (8)). Now, the following question arises: What do we obtain from the well-known formula $A(\omega) = Im[P(\omega)/E(\omega)]$?

In order to calculate $P(\omega)$, we need the polarization function $P(t)$. In section II.E, we have already applied the standard formula $P(t) = \langle e^+ h^+\rangle(t)$ which is well established in the literature (see, e.g., Ref.[7]). If the coefficients c_{10} and c_{40} are chosen to be real numbers, $P(t)$ is given by Eq. (47).

As far as $P(\omega)$ is concerned, we first remark that

$$P(\omega) = \int_{-\infty}^{+\infty} P(t) \cdot e^{i\omega t} dt = \tag{56}$$
$$= \int_{-\infty}^{0} P(0) \cdot e^{i\omega t} dt + \int_{0}^{+\infty} (P(t) - P(\infty)) \cdot e^{i\omega t} dt + \int_{0}^{+\infty} P(\infty) \cdot e^{i\omega t} dt$$

is true under these circumstances. Introducing the distributions

$$\int_{-\infty}^{+\infty} e^{i\omega t} dt = 2\pi \cdot \delta(\omega) \ ; \ \int_{0}^{+\infty} e^{i\omega t} dt = \pi \cdot \delta(\omega) + i \cdot P\left(\frac{1}{\omega}\right) \ , \tag{57}$$

we obtain from Eq. (56):

$$P(\omega) = \pi \cdot Q \cdot \delta(\omega) + \exp\left(-2 \cdot |\gamma|^2\right) \cdot c_{10} \cdot c_{40} \cdot R(\omega) \ , \tag{58}$$

where

$$Q := |c_{10}|^2 - |c_{40}|^2 + i \cdot \exp\left(-2 \cdot |\gamma|^2\right) \cdot c_{10} \cdot c_{40} \cdot \sin\left(\frac{2}{\hbar} \cdot \int_{0}^{+\infty} S(\tau) d\tau\right) \tag{59}$$

and

$$R(\omega) := -\sin\left(\frac{2}{\hbar} \cdot \int_{0}^{+\infty} S(\tau) d\tau\right) \cdot \left(P\left(\frac{1}{\omega}\right) - \frac{1}{\omega}\right) + \tag{60}$$
$$-\frac{2}{\hbar\omega} \cdot \int_{0}^{+\infty} \cos\left(\frac{2}{\hbar} \cdot \int_{0}^{t} S(\tau) d\tau\right) \cdot S(t) \cdot \cos\omega t \ dt +$$
$$-\frac{2i}{\hbar\omega} \cdot \int_{0}^{+\infty} \cos\left(\frac{2}{\hbar} \cdot \int_{0}^{t} S(\tau) d\tau\right) \cdot S(t) \cdot \sin\omega t \ dt \ .$$

In a next step, we split the Fourier transforms of $P(t)$ and $E(t)$ into

$$E(\omega) = E_R(\omega) + i \cdot E_I(\omega) \tag{61}$$

and

$$P(\omega) = P_R(\omega) + i \cdot P_I(\omega) \ ; \tag{62}$$

consequently, the imaginary part of $P(\omega)/E(\omega)$ is given by

$$Im \frac{P(\omega)}{E(\omega)} = \frac{P_I(\omega) \cdot E_R(\omega) - P_R(\omega) \cdot E_I(\omega)}{E_R^2(\omega) + E_I^2(\omega)} \ . \tag{63}$$

Remark: In order to avoid confusions, we omit the expression $\sim (P(1/\omega) - 1/\omega)$ *(see Eq. (60)) in the following part of our considerations. It is needless to emphasize that our further conclusions are not affected.*

From Eq. (58), we thus obtain

$$P_I(\omega)\cdot E_R(\omega) - P_R(\omega)\cdot E_I(\omega) = D(\omega)\cdot\pi\cdot\delta(\omega) + \tag{64}$$
$$-\frac{2}{\hbar\omega}\cdot\exp\left(-2\cdot|\gamma|^2\right)\cdot c_{10}\cdot c_{40}\cdot\left(\Sigma_I(\omega)\cdot E_R(\omega) - \Sigma_R(\omega)\cdot E_I(\omega)\right)$$

under these circumstances. Here, we have introduced the abbreviation

$$D(\omega) := \left(|c_{40}|^2 - |c_{10}|^2\right)\cdot E_I(\omega) + \tag{65}$$
$$+\exp\left(-2\cdot|\gamma|^2\right)\cdot c_{10}\cdot c_{40}\cdot\sin\left(\frac{2}{\hbar}\cdot\int_0^{+\infty} S(\tau)d\tau\right)\cdot E_R(\omega)$$

and the *non-linear integral transforms*

$$\Sigma_R(\omega) := \int_0^{+\infty}\cos\left(\frac{2}{\hbar}\cdot\int_0^t S(\tau)d\tau\right)\cdot S(t)\cdot\cos\omega t\ dt \tag{66}$$

and

$$\Sigma_I(\omega) := \int_0^{+\infty}\cos\left(\frac{2}{\hbar}\cdot\int_0^t S(\tau)d\tau\right)\cdot S(t)\cdot\sin\omega t\ dt\ . \tag{67}$$

Now, it is obvious that the second term on the right-hand side of Eq. (64) is - in general - different from zero. The proof proceeds by *reductio ad absurdum:* We assume that the equation $\Sigma_R(\omega)/\Sigma_I(\omega) = E_R(\omega)/E_I(\omega)$ is valid for arbitrary amplitudes $E(t)$ and replace $E(t)\rightarrow\lambda\cdot E(t)$; under these circumstances, we obtain the relation

$$\frac{\int_0^{+\infty}\cos\left(\frac{2}{\hbar}\cdot\lambda\cdot\int_0^t S(\tau)d\tau\right)\cdot S(t)\cdot\cos\omega t\ dt}{\int_0^{+\infty}\cos\left(\frac{2}{\hbar}\cdot\lambda\cdot\int_0^t S(\tau)d\tau\right)\cdot S(t)\cdot\sin\omega t\ dt} = \frac{E_R(\omega)}{E_I(\omega)}\ . \tag{68}$$

Introducing the power series expansion of the cosine-function, Eq. (68) reads as follows:

$$\frac{\sum_{k=0}^{\infty} a_k(\omega)\cdot\lambda^{2k}}{\sum_{k=0}^{\infty} b_k(\omega)\cdot\lambda^{2k}} = \frac{E_R(\omega)}{E_I(\omega)}\ , \tag{69}$$

where

$$a_k(\omega) := \frac{(-1)^k}{(2k)!}\cdot\left(\frac{2}{\hbar}\right)^{2k}\cdot\int_0^{+\infty}\left[\int_0^t S(\tau)d\tau\right]^{2k}\cdot S(t)\cdot\cos\omega t\ dt \tag{70}$$

and

$$b_k(\omega) := \frac{(-1)^k}{(2k)!}\cdot\left(\frac{2}{\hbar}\right)^{2k}\cdot\int_0^{+\infty}\left[\int_0^t S(\tau)d\tau\right]^{2k}\cdot S(t)\cdot\sin\omega t\ dt\ . \tag{71}$$

According to our assumptions from above (see Eq. (8)), we obtain the inequalities

$$|a_k(\omega)| \leq G \cdot \frac{1}{(2k)!} \cdot \left(\frac{2 \cdot G}{\hbar}\right)^{2k} \;,\; |b_k(\omega)| \leq G \cdot \frac{1}{(2k)!} \cdot \left(\frac{2 \cdot G}{\hbar}\right)^{2k} \;, \tag{72}$$

where

$$G := \int_0^{+\infty} |S(\tau)| d\tau < +\infty \;. \tag{73}$$

In combination with a well-known *Theorem of Weierstraß*[8], we conclude from Eqs. (72) and (73) that the power series on the left-hand side of Eq. (69) are in fact *absolutely convergent and even uniformly convergent with respect to* ω, i.e., we obtain well-defined expressions for arbitrary values of λ and ω. Now, direct inspection of Eqs. (70) and (71) shows that the coefficients $a_k(\omega)$ and $b_k(\omega)$ are - in general - different from each other; consequently, Eq. (69) exhibits a contradiction, because the left-hand side does - in general - explicitly depend on λ. This finishes our proof.

In the last part of our considerations, we choose $c_{10} = \pm 1/\sqrt{2}$ and $c_{40} = \pm 1/\sqrt{2}$; here, *we obtain* $P(t \leq 0) = 0 \;,\; \langle V(t) \rangle = 0$ *under all circumstances and*

$$\begin{aligned} P(\omega) = \pi \cdot i \cdot \exp\left(-2 \cdot |\gamma|^2\right) \cdot \left(\pm\frac{1}{2}\right) \cdot \sin\left(\frac{2}{\hbar} \cdot \int_0^{+\infty} S(\tau) d\tau\right) \cdot \delta(\omega) + \\ + \exp\left(-2 \cdot |\gamma|^2\right) \cdot \left(\pm\frac{1}{2}\right) \cdot R(\omega) \;, \end{aligned} \tag{74}$$

$$\begin{aligned} & P_I(\omega) \cdot E_R(\omega) - P_R(\omega) \cdot E_I(\omega) = \\ & = \exp\left(-2 \cdot |\gamma|^2\right) \cdot \left(\pm\frac{1}{2}\right) \cdot \sin\left(\frac{2}{\hbar} \cdot \int_0^{+\infty} S(\tau) d\tau\right) \cdot E_R(\omega) \cdot \pi \cdot \delta(\omega) + \\ & -\frac{2}{\hbar\omega} \cdot \exp\left(-2 \cdot |\gamma|^2\right) \cdot \left(\pm\frac{1}{2}\right) \cdot \left(\Sigma_I(\omega) \cdot E_R(\omega) - \Sigma_R(\omega) \cdot E_I(\omega)\right) \;. \end{aligned} \tag{75}$$

We thus conclude that *the optical absorption spectrum is - in general - different from zero, too; moreover,* $A(\omega)$ *may become negative.* We emphasize that these results are valid *even in the case that the particle-photon coupling constant vanishes*, i.e., the coupling of the electron-hole system to the quantized electromagnetic field is not decisive in this context. It is also important to realize that our conclusions are only based on the mathematical structure of Eq. (47) - and not on specific examples for the exciting light field.

We summarize our basic results:

Theorem:

Consider a particle-photon system, as defined by Eqs. (4), (5) and (6). Assume (i) that the Coulomb matrix element W^{eh} *fulfills the condition* $\varepsilon_0 + \varphi_0 - 2 \cdot W^{eh} = 0$ *and (ii) that the initial state is given by*

$$|\Psi(t=0)\rangle = c_{10} \cdot |\Psi_{10}\rangle + c_{40} \cdot |\Psi_{40}\rangle \ , \ |c_{10}|^2 + |c_{40}|^2 = 1 \ , \tag{76}$$

where $|\Psi_{10}\rangle$ *and* $|\Psi_{40}\rangle$ *are defined by Eqs. (21) and (22), respectively. Then, the following relations are valid for any* $t \geq 0$*:*

$$\begin{aligned} \langle\Psi(t)|\hat{E}(a^+)^k a^l|\Psi(t)\rangle = \gamma^k \cdot (\gamma^*)^l \cdot \left((-1)^{k+l} \cdot \lambda_{11} \cdot |c_{10}|^2 + \lambda_{44} \cdot |c_{40}|^2\right) + \\ + \exp\left(-2 \cdot |\gamma|^2\right) \cdot \gamma^k \cdot (\gamma^*)^l \cdot \Lambda(t) \ , \end{aligned} \tag{77}$$

$$\begin{aligned} \Lambda(t) := (-1)^l \cdot \lambda_{14} \cdot c_{10} \cdot c_{40}^* \cdot \exp\left(-2 \cdot \frac{i}{\hbar} \cdot \int_0^t S(\tau)d\tau\right) + \\ +(-1)^k \cdot \lambda_{41} \cdot c_{10}^* \cdot c_{40} \cdot \exp\left(2 \cdot \frac{i}{\hbar} \cdot \int_0^t S(\tau)d\tau\right) \ ; \end{aligned} \tag{78}$$

$$\langle e^+h^+\rangle(t) = \frac{1}{2} \cdot \left(|c_{10}|^2 - |c_{40}|^2\right) - \frac{1}{2} \cdot \exp\left(-2 \cdot |\gamma|^2\right) \cdot \Lambda_{pol.}(t) \ , \tag{79}$$

$$\begin{aligned} \Lambda_{pol.}(t) := c_{10} \cdot c_{40}^* \cdot \exp\left(-2 \cdot \frac{i}{\hbar} \cdot \int_0^t S(\tau)d\tau\right) + \\ -c_{10}^* \cdot c_{40} \cdot \exp\left(2 \cdot \frac{i}{\hbar} \cdot \int_0^t S(\tau)d\tau\right) \ . \end{aligned} \tag{80}$$

Here, the expressions λ_{ij} *are defined by Eq. (35);* γ *is related to the particle-photon coupling constant* M*:* $\gamma := M/\hbar\Omega$*. As far as the optical absorption spectrum is concerned, the following statements are true: (i) The total energy of the particle-photon system is a constant. (ii) The expression* $Im[P(\omega)/E(\omega)]$ *is - in general - different from zero and may become negative.*

We add four comments:

(C1) It is important to realize once more that the specific choice $c_{10} = \pm 1/\sqrt{2}$ and $c_{40} = \pm 1/\sqrt{2}$ leads to the result $\langle V(t)\rangle = 0$, i.e., *the semiclassical interaction energy vanishes for any* t.

(C2) Turning to the initial state $|\Psi(t=0)\rangle$, we obtain the explicit equations

$$|\Psi(t=0)\rangle = \frac{1}{2}\cdot|1\rangle\cdot(|P_{10}\rangle+|P_{40}\rangle)+\frac{1}{2}\cdot|4\rangle\cdot(|P_{10}\rangle-|P_{40}\rangle)\ , \quad if \quad c_{10}=\frac{1}{\sqrt{2}}\ ,\ c_{40}=\frac{1}{\sqrt{2}} \tag{81}$$

and

$$|\Psi(t=0)\rangle = \frac{1}{2}\cdot|1\rangle\cdot(|P_{10}\rangle-|P_{40}\rangle)+\frac{1}{2}\cdot|4\rangle\cdot(|P_{10}\rangle+|P_{40}\rangle)\ , \quad if \quad c_{10}=\frac{1}{\sqrt{2}}\ ,\ c_{40}=-\frac{1}{\sqrt{2}}\ . \tag{82}$$

At this point, we should emphasize again that - for an interacting particle-photon system - the vacuum state $|0\rangle$ is actually *not* an eigenstate of H_0, the reason being that the particle-photon interaction operator in Eq. (6) is of *standard type.*

If the particle-photon coupling constant is equal to zero, Eqs. (81) and (82) read as follows:

$$|\Psi(t=0)\rangle = |0\rangle\ , \quad if \quad c_{10}=\frac{1}{\sqrt{2}}\ ,\ c_{40}=\frac{1}{\sqrt{2}}\ ,\ M=0\ , \tag{83}$$

$$|\Psi(t=0)\rangle = e^{+}h^{+}|0\rangle\ , \quad if \quad c_{10}=\frac{1}{\sqrt{2}}\ ,\ c_{40}=-\frac{1}{\sqrt{2}}\ ,\ M=0\ . \tag{84}$$

(C3) Making use of the power series expansion of the cosine function again, we obtain the relation

$$\Sigma_I(\omega)\cdot E_R(\omega)-\Sigma_R(\omega)\cdot E_I(\omega) = \sum_{k=1}^{\infty}\frac{(-1)^{k+1}}{(2k)!}\cdot\left(\frac{2}{\hbar}\right)^{2k}\cdot\int_0^{+\infty}\left(\int_0^t S(\tau)d\tau\right)^{2k}\cdot \cdot S(t)\cdot(E_I(\omega)\cdot\cos\omega t - E_R(\omega)\cdot\sin\omega t)\,dt\ . \tag{85}$$

Consequently, if we split $E(t)$ into $E(t)=E_0\cdot f(t)$, we conclude from Eq. (85) that

$$\frac{\Sigma_I(\omega)\cdot E_R(\omega)-\Sigma_R(\omega)\cdot E_I(\omega)}{E_R^2(\omega)+E_I^2(\omega)} \sim E_0^2 \tag{86}$$

is true, i.e., $Im\,(P(\omega)/E(\omega))$ depends explicitly on E_0 (see Eqs. (63) and (64)).

(C4) According to our results from above, it is necessary to demonstrate for any specific model that the expression $Im[P(\omega)/E(\omega)]$ is in fact equal to the optical absorption spectrum. If such a proof is missing, a numerical calculation of $A(\omega)$ (which is based on the formula $A(\omega) = Im[P(\omega)/E(\omega)]$ in combination with phenomenological damping constants) is at least highly questionable. If we use the improved formula[1] $A(\omega) = 2 \cdot (\omega/c_0) \cdot n_b \cdot Im\sqrt{1+\chi(\omega)/\varepsilon_b}$, Eq. (58) leads to a serious mathematical problem, the reason being that the square root contains a distribution. Here, the optical absorption spectrum $A(\omega)$ does in fact not exist.

IV. DOES AN ALTERNATIVE APPROACH TO THE POLARIZATION FUNCTION EXIST? FURTHER RESULTS.

We have already emphasized that the standard formula $P(t) = \langle e^+h^+\rangle$ for the polarization function $P(t)$ is well established in the literature (as an example, we refer again to Ref.[7]). The specific choice for $P(t)$ is further supported by the detailed analysis in Ref.[2]: *Although dynamical vacuum fluctuations do appear in the system under consideration, the expectation value $\langle e^+h^+\rangle$ is in fact equal to zero for any t:*

$$P(t) \equiv \langle e^+h^+\rangle(t) = 0 \quad , \quad if \quad |\Psi(t=0)\rangle = |0\rangle \tag{87}$$

(see Ref.[2] for further details and generalizations). On the other hand, however, the analysis in our present article has demonstrated that the formula $A(\omega) = Im[P(\omega)/E(\omega)]$ becomes questionable, if the standard expression for $P(t)$ is used. At this point, the following question arises: Is it possible to maintain the basic relation $A(\omega) = Im[P(\omega)/E(\omega)]$ for the optical absorption spectrum and to replace the *polarization function* by another one? As an example, we may take the real valued function $P(t) = \langle e^+h^+\rangle + \langle he\rangle$ to describe the response of the electron-hole system to external perturbations. It is obvious that Eq. (87) remains valid under these circumstances. However, a numerical analysis [1] demonstrates immediately that the latter formula leads in fact to wrong results, too.

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[1]Our calculations have been performed by a *2.4.20-4GB-athlon* machine (model name: *AMD Athlon(TM) XP1800+*).

We have analyzed an electron-hole two level system coupled to an unlimited number of phonon modes and interacting with the classical exciting light field (coupled to the polarization). As before, the Coulomb interaction between electrons and holes is included and determined by a single Coulomb matrix element W^{eh} (see Eq. (5)). The initial state is the vacuum state $|0\rangle$ which is an *eigenstate* of the underlying Hamiltonian [2], if the exciting light field vanishes. The dispersion of the corresponding phonon branch is taken to be constant (Einstein approximation). If the latter condition is fulfilled, a powerful technique introduced in Ref.[9] is applicable. This technique enables one to take the *exact* equations of motion for the phonon-assisted expectation values up to an arbitrary order with respect to the particle-phonon interaction. The numerical calculation of the optical absorption spectrum is based on the formula $A(\omega) = Im[P(\omega)/E(\omega)]$ in combination with phenomenological damping constants [3].

Now, we choose the Coulomb matrix element W^{eh} such that the ground-state energy of the model Hamiltonian under consideration is *negative*. For a suitable choice of the underlying system parameters, it is then possible to find a counter-example for the alternative expression $P(t) = \langle e^+h^+\rangle + \langle he\rangle$: Our example shows that $A(\omega)$ *changes its sign* in a certain domain of ω. If the standard expression $P(t) = \langle e^+h^+\rangle$ is applied, however, the explicit result for $A(\omega)$ is positive for *arbitrary* values of ω. Clearly, these findings demonstrate that the alternative expression from above has to be excluded.

For sufficiently small values of the Coulomb matrix element W^{eh}, the standard formula $P(t) = \langle e^+h^+\rangle$ leads to the following statements: **(i)** $A(\omega)$ is not negative. **(ii)** The excitonic peak is observed at the expected position. **(iii)** On the high-energy side of the excitonic peak, we find equidistant phonon peaks separated by the LO-phonon energy. **(iv)** On the low-energy side of the excitonic peak, no phonon peaks are observed.

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[2] As usual, it is understood that the energy eigenvalue of $|0\rangle$ is equal to zero.

[3] We choose $E(t) = A \cdot \exp\left(-t^2/4\gamma^2\right) \cdot \cos\omega_0 t$.

Although these results are quite convincing, we have to realize once more that a counter-example for the standard formalism does in fact exist (see again Hamiltonian (3) in combination with the additional condition (9)). We do not believe that the incorrect result (75) for $A(\omega)$ is caused by the degenerate ground-state energy of H_0. Indeed, a further numerical study for $M = 0$ demonstrates that the formula $Im(P(\omega)/E(\omega))$ leads to an incorrect result *also in a vicinity of the specific Coulomb matrix element* $W^{eh} = (\varepsilon_0 + \varphi_0)/2$. We are not going to explain further details here, but only add the following comments: **(v)** In the limit of vanishing (phenomenological) damping constants, a numerical calculation of the *polarization function* is still possible and confirms our analytical result (47) for $M = 0$ and for the initial state $|\Psi(t = 0)\rangle = |0\rangle$ (see also Eq. (83)). **(vi)** If the parameter $\varepsilon_0 + \varphi_0 - 2 \cdot W^{eh}$ is enlarged, we observe that $|P(t)|^2$ is a continuous functional form of W^{eh}. The polarization function $P(t)$ itself has an additional factor $e^{i\omega_P t}$ with the property $\omega_P \to 0$ if $\varepsilon_0 + \varphi_0 - 2 \cdot W^{eh} \to 0$.

These findings clearly demonstrate that the transition from the specific parameter $\varepsilon_0 + \varphi_0 - 2 \cdot W^{eh} = 0$ to the enlarged values $\varepsilon_0 + \varphi_0 - 2 \cdot W^{eh} > 0$ is not accompanied by a dramatic change of $P(t)$. This is further supported by a numerical calculation of the *formal expression* $Im(P(\omega)/E(\omega))$ (here, the introduction of only one additional damping constant is sufficient). The explicit result shows exactly one peak, and only a continuous shift of the peak position - and nothing else - is observed, if the parameter $\varepsilon_0 + \varphi_0 - 2 \cdot W^{eh}$ is enlarged.

We thus conclude that the standard formalism leads to wrong results at least for sufficiently large values of the Coulomb matrix element W^{eh}. If W^{eh} is small enough, the implications of the standard formalism are in excellent agreement with the *spectral properties* of the underlying Hamiltonian (see again comments **(ii)** - **(iv)**). However, the question whether $A(\omega)$ shows the correct peak *heights* or not remains open.

Last but not least, we should emphasize once more that we have analyzed the dynamical behaviour of an *electron-hole system*. From Ref.[2], we already know that *the description of pair creation and annihilation in the framework of standard quantum field theory* is definitely a source of fundamental and serious difficulties.

ACKNOWLEDGMENT

I am grateful to Michael Indlekofer for valuable help. The numerical work was done in part in collaboration with W. Schäfer.

REFERENCES

[1] W. Schäfer, M. Wegener, Semiconductor Optics and Transport Phenomena, Springer-Verlag (Berlin Heidelberg 2002)

[2] F. Kalina, Serious problems of the present theory of electron-hole-photon interaction, Jülich (2004), urn:nbn:de:0001-00217

[3] J. Fröhlich, Fort. Phys. **22**, 159 (1974)

[4] H. Spohn, J. Phys. **A 21**, 1199 (1988)

[5] B. Gerlach, H. Löwen, Rev. Mod. Phys. **63**, 63 (1991)

[6] H. Haken, Quantenfeldtheorie des Festkörpers, Teubner-Verlag (Stuttgart 1993)

[7] D. Steinbach, G. Kocherscheidt, M. U. Wehner, H. Kalt, M. Wegener, K. Ohkawa, D. Hommel, V. M. Axt, Phys. Rev. **B 60**, 12079 (1999)

[8] I. S. Gradshteyn, I. M. Ryzhik, Table of Integrals, Series and Products, Academic Press (1980)

[9] T. Stauber, R. Zimmermann, H. Castella, Phys. Rev. **B 62**, 7336 (2000)