Many Particle Theory for Luminescence in Quantum Wells

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A Green's function theory is applied to the description of luminescence and absorption spectra of low dimensional semiconductors. Progress in the numerical solution of the Bethe Salpeter equation for coupled band quantum wells with a T-matrix structure for the polarisation function and carrier-carrier dephasing is given within an approach that satisfy the Kubo-Martin-Schwinger sum rule and eliminates typical artifacts in computed optical spectra.

I Introduction

Photo-luminescence Spectroscopy (PL) is one of the major non-destructive characterisation techniques of novel semiconductor materials. [1] As the growth and optical measurement techniques progress, a parallel development takes place on the theoretical analysis of optical spectra, and it is now a well-established fact that many-body effects are crucial for the understanding of the optical properties of semiconductor materials.[2] Keldysh Green's functions are particularly well- suited to describe both steady state and non- equilibrium situations, and the corresponding diagrams allow for a systematic inclusion of increasingly higher- order effects.[3] If the semiconductor media have low effective dimensionality, like quantum wells dots and wires, quantum confinement of the charged carriers and non- parabolic band structure fundamentally alter the electronic structure. [4] Recent theories have been able to incorporate all these effects consistently and be able to describe their interplay in the resulting optical spectra. [5] However, there is, so far, no approach free of approximations. Different techniques have been used to obtain numerical codes, fast enough for systematic computations allowing direct comparison with experiments and containing enough predictable power, and several discrepancies found in computations in the past are being progressively eliminated. One example is that if the band occupation, or equivalently, the Pauli-blocking factor is written under the quasi-particle approximation, i.e. with the Fermi functions evaluated at the dispersion relations, $f_e(e_e(k))$, $f_h(e_h(k))$, a spurious absorption develops below the gain region. The difficulty can be eliminated when non diagonal dephasing terms are considered in the (coherent) polarisation. [6, 7] Another consequence of the quasi-particle approximation, is the development of negative luminescence on the high energy side of the spectra. None of these artifacts appear in an alternative approach presented recently, [5] since by making sure that the polarisation function satisfies the Kubo-Martin-Schwinger (KMS) condition. [2, 8], high k-values contributions that ultimately give rise to the artifacts are eliminated. Furthermore, the theory automatically guarantees that the switch from absorption to gain occurs exactly at the chemical potential, so that the technique proposed in Ref. [9] to extract the gain spectra from spontaneous emission data can be safely used without a discrepancy between "experimental" and "computed" total chemical potential differences. The theory has, very successfully, predicted several important experimental results and provided explanations for why certain previous approaches actually worked. Specifically for the gain case, the available experimental data for a systematic comparison were obtained for laser structures and materials in which high-order sub-band transitions played a negligible role, and , e.g., strong inhomogeneities masked details of the dephasing mechanism as in Ref. [10]. So, instead of computing those effects in detail, a fit to the GW dephasing expression (corresponding to carrier- carrier spectrum) has been used. [11] The aim of the present paper is not to compare and contrast the different approaches in the literature, but to present a step forward towards the computation of Photo-luminescence (PL) spectroscopy, within the context of a T-matrix based theory. Here, instead of a partly phenomenological dephasing, or frequency- independent results that requires non- diagonal dephasing to remove inconsistencies, an iterated GW carrier self-energy is used giving rise to a k- and frequency-dependent dephasing, extending the approach of Ref. [12] for bulk materials to the MQW case. It further illustrates the relevance of a frequency

dependent dephasing to explain certain features of optical spectra.

II Main equations

The time evolution of the quantum statistical averages describing the interacting carriers (G), photons (D) and plasmons (W), within the Keldysh formalism, given by Dyson equations. [13] Coulomb corrections to the free particle propagators, G_0, D_0, and W_0, are given by the respective self-energies, Σ, P, and p. Functional derivative or diagrammatic techniques allow the consideration of increasingly higher order Coulomb corrections in the self-energies. The carrier and photon self-energies, in a schematic notation, are (for details see Refs. [5, 12]).

$$\Sigma(1,2) = \Sigma_{\rm GW}(1,2) + \Sigma_T(1,2), \quad (1)$$

$$P(1,2) = G(1,2)\,G(2,1) + G(1,3)\,G(4,1)\,T(3,4,5,6)\,G(5,2)\,G(2,6). \quad (2)$$

where $\Sigma_{\rm GW}(1,2) = G(1,2)\,W(1,2)$, $\Sigma_T(1,2) = T''(1,3,2,4)\,G(4,3)$. T'' denotes the T-matrix without the first two iterations of the ladder scheme, which gives rise to the Bethe-Salpeter equation

$$T(1,2,3,4) = W(1,2)\,\delta_{13}\,\delta_{24} + W(1,2)\,G(1,5)\,G(6,2)\,T(5,6,3,4). \quad (3)$$

In this paper, T-matrix correlations beyond GW in the spectral density of photons are considered but the carrier self-energies are restricted to GW, i.e., $\Sigma(1,2) = G(1,2)\,W(1,2)$, since the T-matrix contributes significantly to carrier self- energy in quantum wells and superlattices only at low temperatures. [14]

Under stationary conditions, to be describe here, a Fourier-transform with respect to time, yields $\Sigma^r_n(k,\omega) = \Sigma^r_{n,X}(k,\omega) + \Sigma^r_{n,c}(k,\omega)$. Spectral information is included in the retarded Keldysh components, superscript ^{r}. The dephasing, responsible for the gain/absorption broadening, is given by the imaginary part of $\Sigma^r_{n,c}(k,\omega)$, namely

$$\hbar\,\Gamma_n(k,\omega) = \sum_{\mathbf{q}\,\mathbf{q}'\,\lambda} \frac{\hbar\,\Gamma_n(k,\omega)\,F_n(k',p',q')\,W^r(q)^2}{\hbar^2\,(\omega + e_n(k) - e_n(\mathbf{k}') - e_\lambda(\mathbf{p}') + e_\lambda(q'))^2 + \hbar^2\,\Gamma_n^2}, \quad (4)$$

where $f^<$ is a Fermi distribution, $f^> = 1 - f^<$, $\mathbf{p}' = \mathbf{q}' + \mathbf{k} - \mathbf{k}'$, and $F_n(k,p',q') = f^>(e_n(k'))f^>(e_\lambda(p'))f^<(e_\lambda(q')) + f^<(e_n(k'))f^<(e_\lambda(p'))f^>(e_\lambda(q'))$. The iterated dephasing is evaluated at $k = 0$ only and for simplicity, and we further ignore the real part of $\Sigma^r_n(k,\omega)$ without affecting the general conclusions presented here.

The emission $I(\omega)$ and absorption spectra $\alpha(\omega)$ are given respectively by

$$I(\omega) = \frac{\hbar\omega^2/2\pi^2 c\; Im\{P^r(\omega)\}}{1 - exp(\beta(\hbar\omega - \mu))}, \quad \alpha(\omega) = c/(2\omega\sqrt{\epsilon(\infty)})\; ImP^r(\omega), \quad (5)$$

where μ is the total chemical potential, and $I(\omega)$ has been derived from the KMS relation. [5] In the numerical data presented here, the integral Equation 2 is solved through numerical matrix inversion.

III Numerical Results and Discussion

The solid lines of Fig. 1 shows absorption (a,c) and Photoluminescence (b,d) of a 2.5 nm ZnSe/(Zn,Mg)(S,Se) quantum well at 300 K and excited with N = $5 \times 10^{15}\, carriers/cm^3$, as a function of detuning

with respect to the free carrier band gap. The lineshapes computed with the iterated GW dephasing at k=0 (solid lines) are sharper then several experimental findings for 300 K, since higher order diagrams, carrier- phonon scattering and inhomogeneous broadening (present in most samples) have not been considered, as the goal here is to illustrate the importance of the iterated frequency- dependent dephasing. For comparison, the dephasing is computed two expressions that simulate the full Eq. 4,

$$\Gamma = \Gamma_0 \frac{1}{1 + exp[(\hbar e^e(k) + \hbar e^h(k) - \hbar\omega)/\theta]}, \quad (6)$$

$$\Gamma = \Gamma_0 \frac{exp(-\beta\mu_h) + exp[(\hbar e^e(k) + \hbar e^h(k) - \hbar\omega)/\lambda]}{exp(-\beta\mu_h) + 1}, \quad (7)$$

where $e^e(k)$, $e^h(k)$ are renormalised energies, μ_h is the holes chemical potential, $\beta = 1/K_B T$, with the temperature T in Kelvins, and μ_h is the holes chemical potential. The fit- formulas depends on two phenomenological parameters that must be adjusted in comparison, e.g. with PLE or absorption spectra: θ, and λ which is are tail factors, and Γ_0. They can be alternatively (in some cases) be adjusted using the dephasing calculated here and the curves generated with it, as suggested by the numerical results presented.

This type of expression has helped to, very successfully, reproduce a wealth of experimental absorption data, e.g. as in Refs. [11, 10] (and references therein). For high performance device simulators, these formula may be quite useful by adjusting the parameters either to experiments or calculated data, allowing for easy and fast programming. Many simulators include many other effects corresponding to all characteristics of the devices and approximations for the full microscopic expressions are frequently necessary with the present numerical capabilities available.

Figure 1 displays calculated normalised Photoluminescence (a,b,c) and corresponding absorption spectra (d,e,f) of a 3.0 nm ZnSe/ (Zn,Mg)(S,Se) quantum well at 300 K and excited with N = $1 \times 10^{11}\, carriers/cm^2$ as a function of detuning with respect to the free carrier band gap. In all plots, the solid lines are computed with the iterated GW dephasing of Eq. 4 at k=0. In (a,d) the dotted, dot- dashed, and dashed curves are respectively for (see Eq. 6) Γ_0 = 0.2, 0.1, 0.025. $\times$ e_0, and θ=108 $\times$ e_0, where the 2D binding energy is e_0 = 79.6 meV. In (b,e) the dot-dashed and long- dashed curves are, respectively, for $\Gamma_0 = 0.025\ e_0$, and θ = 108, 0.65 $\times$ e_0. In (c,f) The dotted and dot- dashed curves are for Γ_0 = 0.025, and $\theta = 0.65 \times e_0$, and $\lambda = 0.3 \times e_0$.

The numerical study presented here shows that the fit formulas are consistent with the GW calculation for a certain choice of parameters, and fails for others. As a matter of fact, in (a,d) the θ factor is so large in the spectra as to make them indistinguishable of those computed with a constant dephasing $\Gamma = \Gamma_0$, or in other words, with a Lorentzian lineshape. Note that, as long as the polarisation function satisfies the KMS condition, no spurious absorption develops below the gain region, [15] and all plots in (d) could well be used as representative absorption curves. However, as shown in (a) the PL rises unrealistically in the low- energy side of the broader spectra. In other words, a theory that yields reasonable absorption and gain spectra, not necessarily gives realistic PL or Photo luminescence Excitation Spectroscopy spectra (PLE). Note that this approach can be extended to computed both single and two beam PLE. [5, 10, 16] So, as long as the carrier occupation functions can be described by quasi- equilibrium conditions, not only the KMS relation must hold, but the dephasing must be sharply frequency dependent for PL (and PLE) spectra which are consistent with quantum statistical mechanics and still realistic. The computations including the GW dephasing thus increase the predictability of the theoretical approach. At this point an important comment is in order, i.e. weather or not an excited semiconductor at steady- state should be described by quasi- equilibrium is a whole other matter, and further theoretical and experimental work is at this point necessary to determine if this approximation, so successful so far, really is correct. In other words, if the system, even at steady state is far from equilibrium, the KMS condition does not have necessarily to hold anymore. In summary, a T-matrix based many-body theory for the optical spectra of semiconductor superlattices has been presented and its relevance for the realistic computation of photoluminescence spectra has been outlined. In the present approach there is no need for phenomenological parameters to obtain spectral line broadening, thus increasing the predictability of the approach, which can be used as a starting point for simulations of a wealth of different optical spectra and the simulation optoelectronic device characteristics.

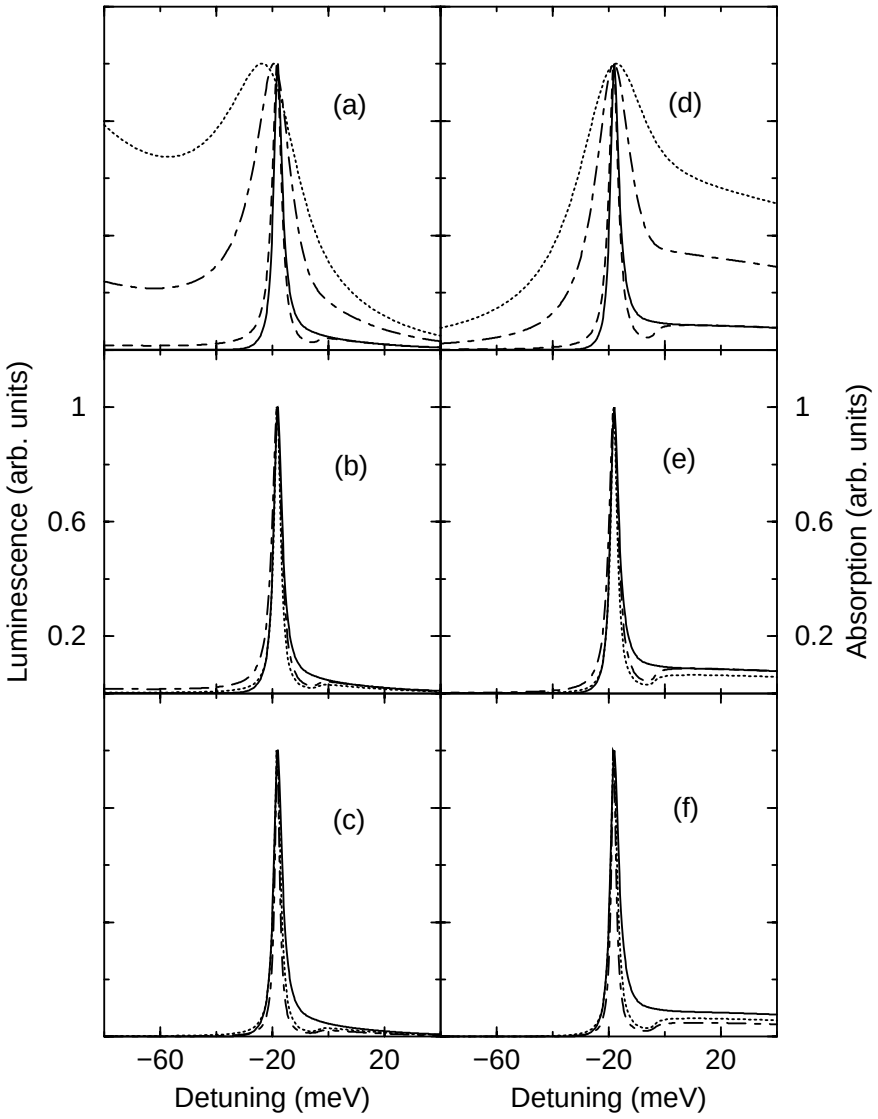

Figure 1. Normalised Photoluminescence (a,b,c) and corresponding absorption spectra (d,e,f) of a 3.0 nm ZnSe/ (Zn,Mg)(S,Se) quantum well at 300 K and excited with N = 1×10^{11} $carriers/cm^2$ as a function of detuning with respect to the free carrier band gap. In all plots, the solid lines are computed with the iterated GW dephasing of Eq. 4 at k=0. In (a,d) the dotted, dot- dashed, and dashed curves are respectively for (see Eq. 6) Γ_0 = 0.2, 0.1, 0.025. $\times$ e_0, and θ=108 $\times$ e_0, where the 2D binding energy is e_0 = 79.6 meV. In (b,e) the dot-dashed and long- dashed curves are, respectively, for Γ_0 = 0.025 e_0, and θ = 108, 0.65 $\times$ e_0. In (c,f) The dotted and dot- dashed curves are for Γ_0 = 0.025, and θ = 0.65 $\times$ e_0, and λ = 0.3 $\times$ e_0.

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