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# Microscopic Theory of Optical Nonlinearities and Spontaneous Emission Lifetime in Group-III Nitride Quantum Wells

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## Abstract

Microscopic calculations of the absorption/gain and luminescence spectra are presented for wide bandgap  $\text{Ga}_{1-x}\text{In}_x\text{N}/\text{GaN}$  quantum well systems. Whereas structures with narrow well widths exhibit the usual excitation dependent bleaching of the exciton resonance without shifting spectral position, a significant blue shift of the exciton peak is obtained for wider quantum wells. This blue shift, which is also present in the excitation dependent luminescence spectra, is attributed to the interplay between the screening of a strain induced piezoelectric field and the density dependence of many-body Coulomb effects. The calculations also show an over two orders of magnitude increase in the spontaneous electron-hole-pair lifetime with well width, due to the reduction of the electron-hole wavefunction overlap in the wider wells. The resulting decrease in spontaneous emission loss is predicted to lead to improved threshold properties in wide quantum well lasers.

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The physical properties of wide bandgap group-III quantum-well systems are currently under intense investigation, mostly because of their substantial application potential as light emitters and semiconductor lasers in the ultraviolet to blue-green wavelength region. [1] Additionally, these materials exhibit interesting excitation dependent nonlinear behavior due to the intricate interplay between the strong attractive electron-hole Coulomb interaction, which lead to significant excitonic signatures in the optical spectra, and the quantum-confined Stark effect caused by piezoelectric fields. Such fields are present, for example, in wurtzite  $\text{Ga}_{1-x}\text{In}_x\text{N}/\text{GaN}$  systems because of the strain induced lattice mismatch. [2] The relative magnitude of excitonic and piezoelectric effects depends sensitively on quantum-well width and plasma density, because of many-body interactions leading to effects including screening, dephasing, bandgap renormalization and phase-space filling. [3,4]

In this paper, we present a theoretical investigation of the intricate interplay of the different nonlinear effects and study their influence on the absorption/gain and luminescence spectra, as well as intrinsic spontaneous carrier lifetime. For this purpose, we use a previously developed microscopic theory for the excitation dependent optical response. [4] This theory is based on the semiconductor Bloch equations [3] where the damping and dephasing processes are treated at the level of quantum kinetic theory. The resulting equations have been applied successfully to analyze a wide variety of semiconductors under high excitation conditions. [5] [6] In the present calculations, bandstructure effects are treated at the level of  $k \cdot p$  theory, with the effects of the screened piezoelectric field systematically included by the simultaneous solution of the coupled Poisson equation and the Luttinger Hamiltonian in the envelope approximation. [7]

Numerically solving the semiconductor Bloch equations under small signal conditions [5] and for the material parameters of  $2\text{nm}$  and  $4\text{nm}$   $\text{Ga}_{0.2}\text{In}_{0.8}\text{N}/\text{GaN}$  quantum wells [6] yields the gain/absorption spectra shown in Figs. 1a and 2a, respectively. For the narrow quantum well (Fig. 1a), we see that the low density exciton resonance is gradually bleached with increasing carrier density. For high densities, optical gain develops in the spectral vicinity of the original exciton resonance. As is usual in intrinsic quantum well systems, there is negli-

gible shift of the excitonic peak spectral position during the plasma bleaching process. This excitation independent exciton energy is a consequence of the strong cancellation between the weakening of the exciton binding energy and the reduction of the bandgap energy, i.e. between the non-diagonal (field renormalization) and diagonal (self energy) contributions in the microscopic polarization equations. [3] [4]

As shown in Fig. 2a, the situation is quite different in the  $4\text{nm}$   $\text{Ga}_{0.2}\text{In}_{0.8}\text{N}/\text{GaN}$  quantum well structure. Because of the weaker quantum confinement in this relatively wide quantum well, the piezoelectric field is able to significantly reduce the overlap between the quantum confined electron and hole wavefunctions. Consequently, the interband dipole matrix element or oscillator strength is substantially smaller than is the case for the narrow  $2\text{nm}$  quantum well. This intrinsic quantum confined Stark effect (QCSE) also significantly red shifts the exciton absorption peak relative to the flat band situation. As the plasma density increases, the screening of the QCSE increases the electron-hole wavefunction overlap, and hence, the exciton oscillator strength. Simultaneously, there is a weakening of the piezoelectric field induced red shift, leading to the net blue shift in the exciton resonance and absorption edge with increasing plasma density, as shown in Fig. 2a. These piezoelectric field related nonlinearities occur in addition to the usual many-body nonlinearities. Due to the stronger piezoelectric field effects in the  $4\text{nm}$  quantum well, the compensation between self-energy and field renormalization contributions to the microscopic interband polarization is perturbed, resulting in the excitation dependent blue shifting of the exciton resonance and absorption edge. A similar blue shift, also resulting from a perturbation of the above mentioned compensation effects by a real space charge separation, was observed and microscopically analyzed for type-II quantum wells [8]. Figure 2(a) also shows that the interband absorption, i.e. that part of the spectra well above the absorption edge, increases with increasing excitation density. This again reflects the increasing electron-hole wavefunction overlap resulting from the gradual screening of the intrinsic QCSE.

Using the absorption/gain spectra in Figs. 1a and 2a, and applying a phenomenological relationship between stimulated and spontaneous emission, [9] we obtain the spontaneous

emission spectra shown by the solid curves in Figs. 1b and 2b. These spectra show the increasing spontaneous emission with increasing plasma density. Whereas an excitation independent peak energy of the luminescence is obtained for the narrow quantum well system, the wide quantum well luminescence exhibits the excitation dependent blue shift, whose origin is as discussed for the absorption/gain spectra. Our approach for obtaining the spontaneous emission spectra is of course not rigorous. However, it does circumvent the complexities associated with quantizing the electromagnetic field, which then allows the inclusion of the details of the multiband quantum well bandstructure into the numerical calculations. To show the accuracy of this phenomenological approach, we plotted the results (dashed curves) using the semiconductor quantum luminescence theory developed recently by Kira et al.. [10]. Due to its numerical complexity, this theory can currently only be evaluated for a two-band effective mass model, and using effective dephasing rates extracted from the semiclassical quantum kinetic calculations. The former limits the comparison to low plasma densities, where multiband as well as bandmixing effects are negligible. Comparison of the solid and dashed curves shows that for low densities, where strong excitonic resonances are present in the optical spectra, we obtain deviations between the quantum theory results and the phenomenological conversion. As discussed in [10] these differences are expected. For increasing densities the agreement improves considerably, being almost quantitative for elevated densities where optical gain is present.

The ordinates of Figs 1 and 2 show absorption and spontaneous emission amplitudes that differ considerably between wide and narrow quantum wells. This difference should appear in the radiative carrier lifetime, which is relatively straightforward to measure in experiments. Integrating the luminescence spectra in Figs. 1b and 2b gives the spontaneous emission rate,  $w_{sp} = \int_0^\infty d\omega se(\omega)$ , which is the inverse of the radiative carrier lifetime  $\tau_{sp}$ . The circles in Fig. 3 shows the results from the luminescence spectra obtained via the phenomenological conversion approach. At low carrier densities where the two-band effective mass model is valid, the carrier lifetime can also be obtained from the quantum luminescence theory by directly computing the total radiative decay rate of electron-hole

pairs. The results are shown by the squares, which confirm that both approaches yield close to identical results. These results show that while the narrow  $2\text{nm}$  well has lifetimes typical of conventional III-V materials, the wide  $4\text{nm}$  quantum well shows radiative lifetimes at low plasma densities and room temperature that are two orders of magnitude larger. Similar trends have been observed experimentally for the GaN/AlGaN system at low temperature [11]. This long lifetime is a direct consequence of the electron-hole wavefunction separation which is substantial in the wide quantum wells. The figure also shows the carrier density dependence of the radiative lifetime. Here the lifetime decreases with increasing plasma density, due to increased screening of the quantum confined Stark effect and to increased carrier-carrier collisions.

The well width and excitation dependent oscillator strength due to the quantum confined Stark effect have important implications to group-III nitride lasers. On the one hand, the QCSE reduces the gain in the wide quantum wells. On the other hand, it also reduces their spontaneous emission losses. Figure 4 shows the peak gain versus spontaneous emission contribution to the current density, for the  $2\text{nm}$  and  $4\text{nm}$  quantum wells. The spontaneous emission current is given by  $J_{sp} = edw_{sp}$ , where  $e$  is the electron charge, and  $d$  is the quantum well width. These curves give the theoretical limit to the threshold current density for a given threshold gain,  $G_{th} = G_{pk}$ . From the figure, we see that the reduction in spontaneous emission loss more than compensates the gain reduction in the wide quantum well, so that for typical threshold gains of  $G_{th} = 10^2$  to  $10^3 \text{cm}^{-1}$ ,  $J_{sp}$  for the wide well is far lower than that for the narrower well system. Hence, the reduced electron-hole dipole matrix element in the wide  $\text{Ga}_{0.2}\text{In}_{0.8}\text{N}/\text{GaN}$  quantum well systems actually benefits laser operation. Comparison of the two curves close to transparency illustrates the very different physical mechanisms leading to the onset of gain. In the  $2\text{nm}$  quantum well, the onset of gain is due to band filling. In contrast, a significant population inversion already exists in the  $4\text{nm}$  quantum well at the onset of gain. Here, the appearance of gain is due to the switch on of the interband dipole matrix element by the screening of QCSE. We wish to emphasize that in an experiment, gain in the  $4\text{nm}$  quantum well will not occur as close to the origin as shown

in Fig. 4, because of the presence of nonradiative losses.

In summary, the microscopic calculations of the optical absorption/gain and luminescence properties of wide bandgap  $\text{Ga}_{0.2}\text{In}_{0.8}\text{N}/\text{GaN}$  quantum well systems predict interesting well-width dependent nonlinearities. A blue shift with increasing plasma density in absorption and luminescence in relatively wide wells occurs as a consequence of the screening of the piezoelectric field induced quantum confined Stark effect. The quantum confined Stark effect also results in carrier lifetimes in the wide wells that are significantly longer than in typical III-V semiconductors. This effect reduces spontaneous emission loss in wide quantum well structures, and can benefit laser threshold properties.

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### Figure Captions

Figure 1. Calculated (a) TE absorption/gain for  $2\text{nm}$   $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}/\text{GaN}$  quantum well at  $T = 300K$ , and carrier densities  $N = 10^{12} \text{ cm}^{-2}$  to  $6 \times 10^{12} \text{ cm}^{-2}$  in  $10^{12} \text{ cm}^{-2}$  increments. (b) shows the spontaneous emission spectra for the densities  $1, 2$  and  $3 \times 10^{12} \text{ cm}^{-2}$ . The solid lines are obtained from the absorption/gain spectra, and the dashed lines are obtained from the semiconductor luminescence equations [10], see text. The material parameters are taken from [6].

Figure 2. Same as Fig. 1, but for a  $4\text{nm}$  structure. The densities in (b) are  $1, 3$  and  $5 \times 10^{12} \text{ cm}^{-2}$ .

Figure 3. Radiative lifetime versus carrier density for  $2\text{nm}$  and  $4\text{nm}$   $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}/\text{GaN}$  quantum well at  $T = 300K$ . The circles are computed from the absorption/gain spectra of Figs. 1 and 2, and the squares are obtained directly from the semiconductor luminescence equations.

Figure 4. Peak gain vs. spontaneous emission current density for  $2\text{nm}$  and  $4\text{nm}$   $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}/\text{GaN}$  quantum well at  $T = 300K$ .

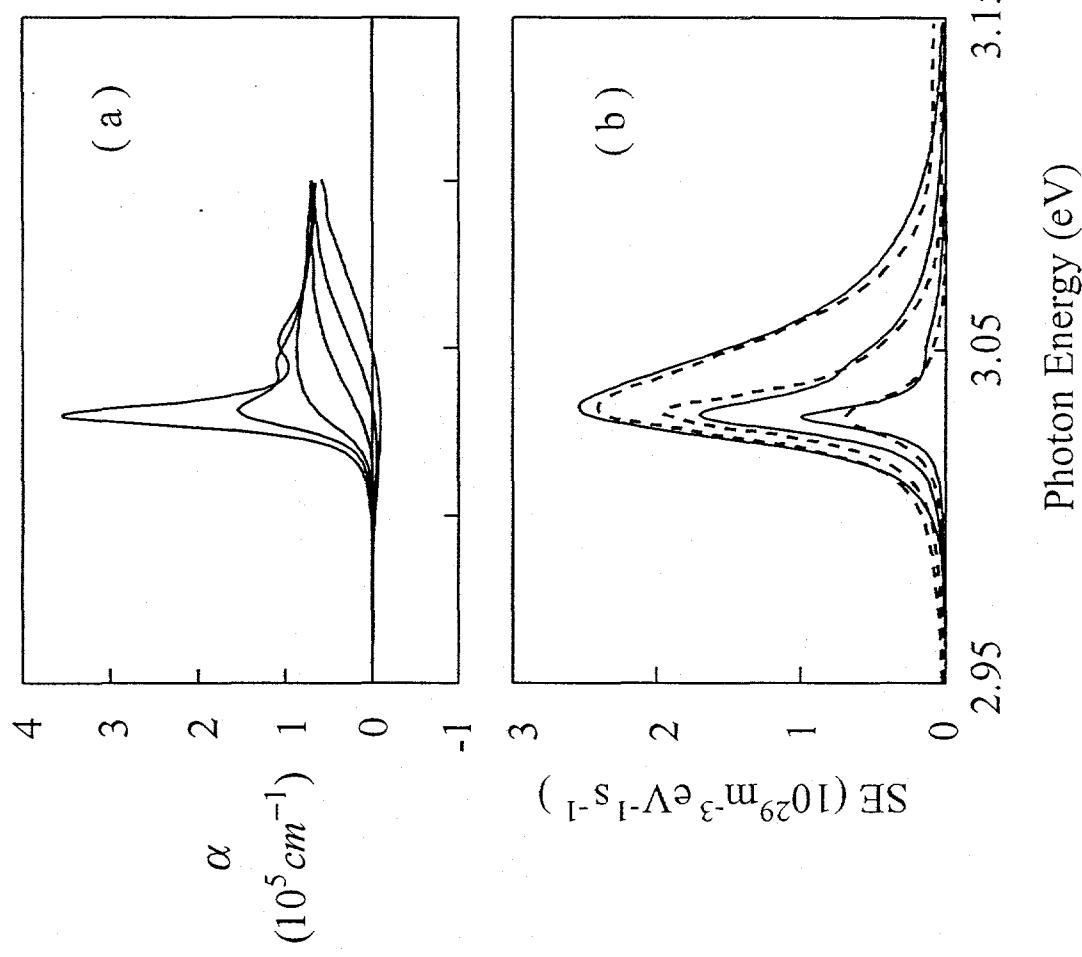


Fig. 1, PRB, Chow

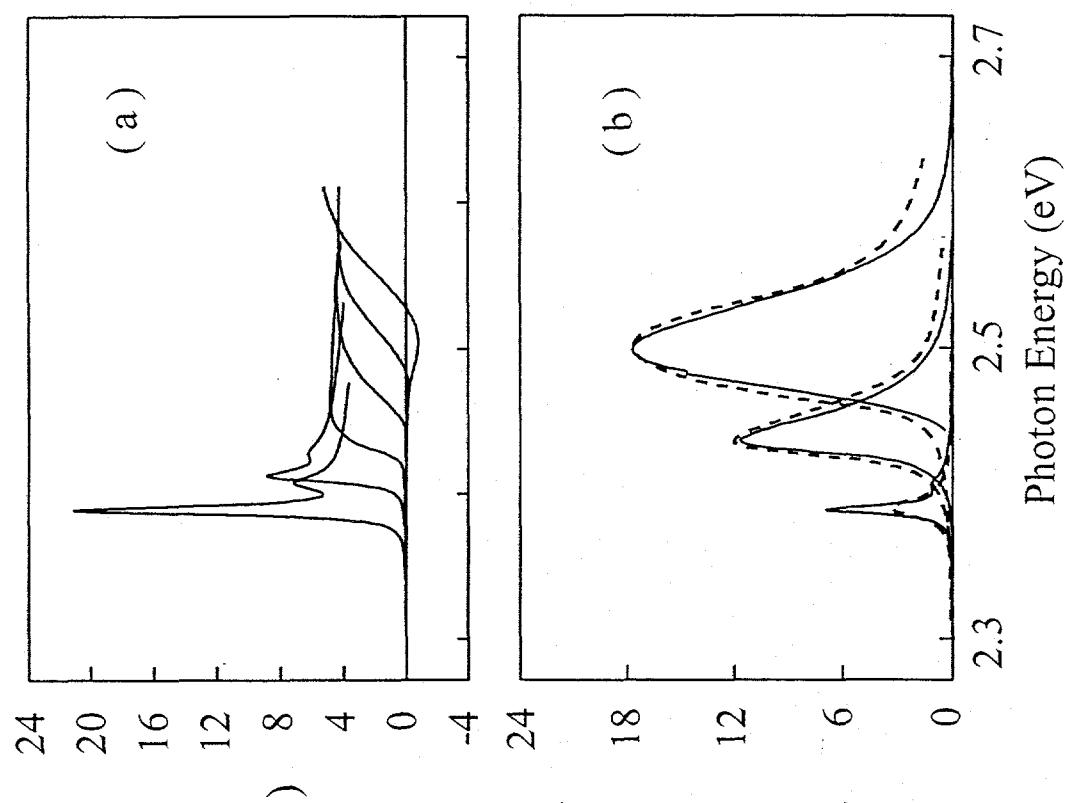


Fig. 2, PRB, Chow

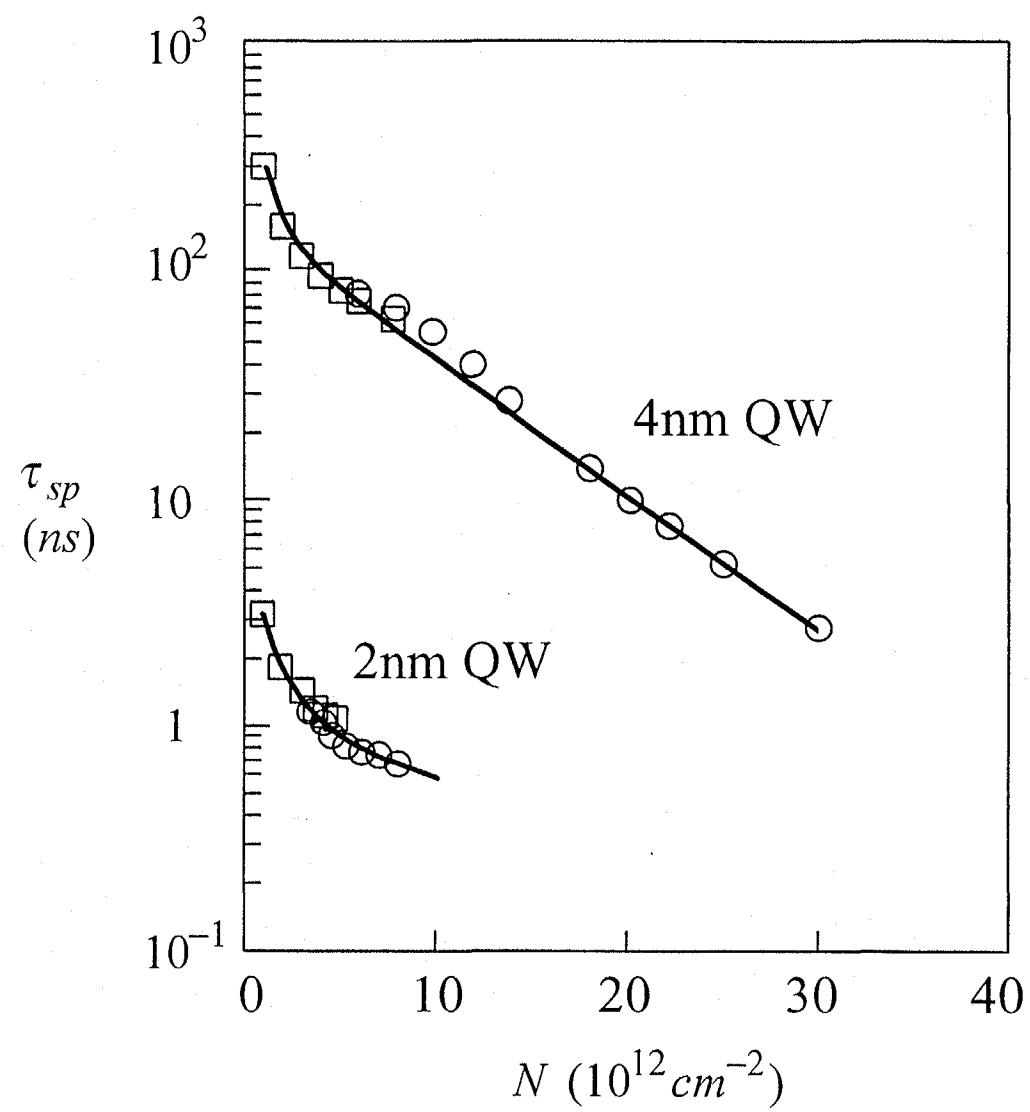


Fig. 3, PRB, Chow

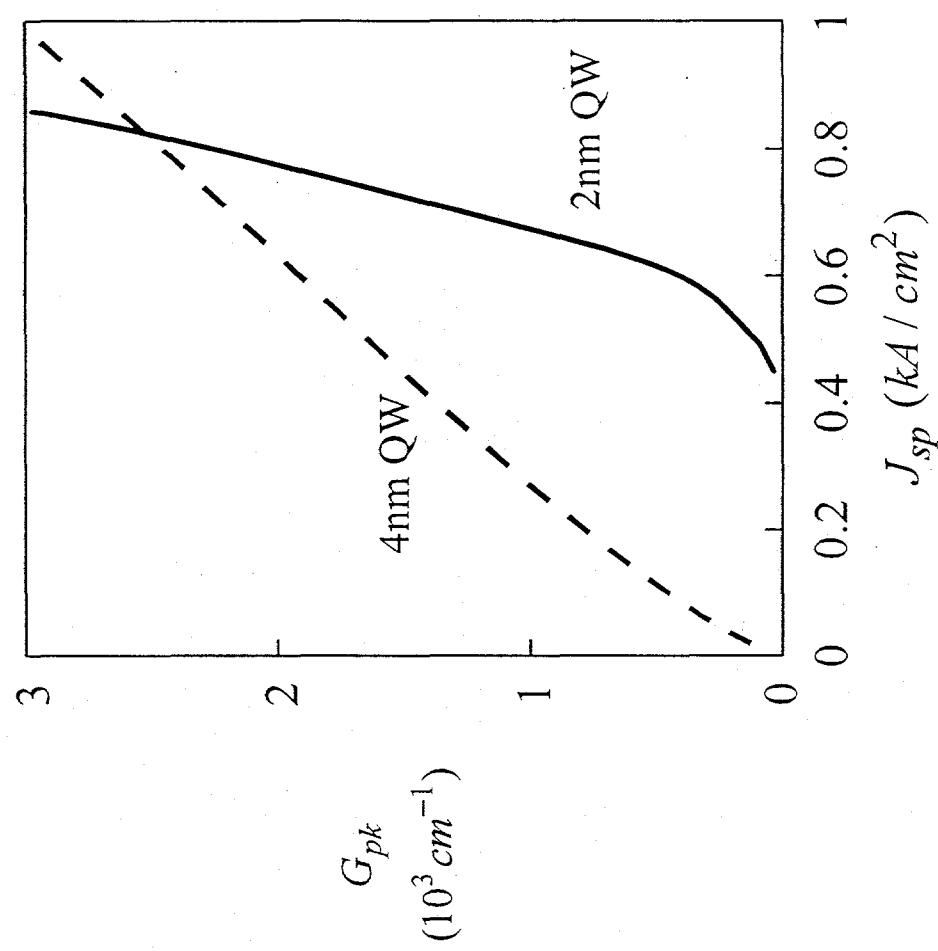


Fig. 4, PRB, Chow