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Vertical motional narrowing of exciton-polaritons in GaN based multiple quantum wells

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Abstract. Numerical simulation of time-resolved light reflection from GaN/AlGaN single and multi-quantum wells (MQWs) revealed a pronounced vertical motional narrowing effect. This effect consists in reduction of the inhomogeneous broadening of the exciton peaks and increase of the decay-time of the time-resolved reflection due to the averaging of the vertical disorder in the structure by extended exciton–polariton modes. The theory predicts pronounced oscillations in the time-resolved reflection of MQWs resulting from the interference of exciton–polariton modes in the structure.

Random potential fluctuations strongly affect the confined exciton states in GaN/AlGaN and GaN/InGaN quantum wells (QWs) thus reducing the advantages of these structures for opto-electronics applications [1, 2]. The disorder arises mainly due to random polarization fields [3, 4] and is manifested by a huge Stokes-shift and inhomogeneous broadening of the exciton lines. Recently, it has been shown experimentally [5] and theoretically [6] that the radiative coupling between excitons confined in different QWs in multiple quantum well (MQW) structures may reduce effectively the disorder effect on the exciton. This effect known as the vertical motional narrowing (VMN) effect manifests itself in the increase of the decay-time of the time-resolved reflection signal from MQWs as a function of the number of wells. It results from the averaging of the disorder potential in a MQW structure by extended exciton–polariton modes that occupy the entire structure.

Four parameters govern the VMN, namely, the exciton radiative recombination rate, Γ_0, the exciton inhomogeneous broadening, Δ, the spacing between wells, d, and the number of the wells, N. The parameter $\eta\Delta$ is in the range of 10 meV for the best quality MQW structures based on nitrides available now [1, 2]. The exciton oscillator strength in nitrides is an order of magnitude larger than in GaAs, so that $\eta\Gamma_0$ achieves 0.4 meV in GaN/AlGaN QWs [7]. The coincidence of the strong disorder and huge exciton oscillator strength makes us to expect an extremely pronounced VMN effect in nitride-based MQWs.

In this paper we examine theoretically the time-resolved response of GaN/AlGaN MQWs. We show that, except for Bragg-arranged MQWs, the VMN effect governs the excitonic decay rate in these structures.

According to the semi-classical model [8], the exciton inhomogeneous broadening can be accounted for by averaging of the non-local dielectric susceptibility with a continuous distribution function of the exciton resonance frequency (chosen to be a Gaussian function for simplicity):

$$\tilde{\chi}(\omega,\gamma) = \frac{\eta}{\sqrt{\pi}\Delta}\int \chi(\omega,\nu,\gamma)\exp\left[-eta^2\left(\frac{\nu-\omega_0}{\Delta}\right)^2\right]\mathrm{d}\nu \tag{1}$$

where ω_0 is the central frequency of the exciton distribution, $\chi(\omega, \nu, \gamma)$ is the dielectric susceptibility of the single free exciton state with the resonance frequency ν and homogeneous broadening γ.

Solving the Maxwell equations within the non-local dielectric response theory [8], one can find the amplitude reflection coefficient for light incident on a single quantum well (SQW) characterized by the response function (1) in form:

$$r_{QW} = -\frac{\sqrt{\pi}\Gamma_0 w(z)}{\Delta + \sqrt{\pi}\Gamma_0 w(z)} \tag{2}$$

where $w(z) = e^{-z^2} erfc(-iz)$, $z = (\omega - \omega_0 + i\gamma)/\Delta$, and $erfc(z)$ is the complementary error function. The frequency-dependent reflection amplitude of the whole structure $r(\omega)$ can be expressed in terms of r_{QW} using the transfer matrix method. The time-resolved reflection of the MQW system is a Fourier transform of $r(\omega)$ convoluted with the spectral function of the incident pulse [9]:

$$r(t) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} r(\omega) g(\omega) e^{-i\omega t}. \tag{3}$$

In the numerical calculations, for all the QWs under consideration we have taken $\eta\gamma = 0.1$ meV, $d = 70$ nm, $\eta\omega_0 = 3.6$ eV. We will consider either "disordered" QW structures with $\eta\Delta = 5$ meV, for GaN/AlGaN QWs [1] or "homogeneous" QWs with $\Delta = 0$ (for the reference). The value of 5 meV for the inhomogeneous broadening should hopefully be achieved by future optimization of material quality.

Figure 1 shows the time-resolved reflection spectra of a disordered SQW (a), of disordered MQWs containing 10 wells (b), 50 wells (c), 100 wells (d), and of a homogeneous SQW (e). Comparing the curves (a) and (e) in Fig. 1, one can see that the exciton inhomogeneous broadening induces a dramatic decrease of the decay time of the time-resolved reflection. This reflects appearance of additional channels of energy relaxation for excitons as the disorder increases in the structure [8]. Note the strong decrease of the exciton decay time with increase of the number of QWs in the structure. This is a manifestation of the VMN effect, which is visibly stronger than one observed experimentally in the GaAs/AlGaAs MQWs [5]. Note also pronounced oscillations in the spectra of MQWs and a disordered SQW (Fig. 1(a-d)). These oscillations have been widely discussed during recent years [5, 6, 8–10]. Basically, they arise both from the interference of different exciton–polariton modes and the interference within each single exciton–polariton mode due to the inhomogeneous broadening.

In the rest of this paper we report an analytical theory aimed to express the decay time of the coherent optical response of MQWs and the period of oscillations via the structure parameters and excitonic characteristics. We adopt the effective dielectric media (EDM) approximation [11] which describes the MQW structure as a homogeneous layer containing a single inhomogeneously broadened exciton resonance. This approximation is valid if excited states and A, B, C, bands are ignored, if $\Gamma_0 \ll \gamma$ and if one can find an integer n such as:

$$kd - n\pi \ll 1 \tag{4}$$

where k is the wave vector of light in the media at the exciton resonance frequency. The formalism allowing to calculate the time-resolved response of a semiconductor film containing an inhomogeneously broadened exciton resonance has been developed in Ref. [10].

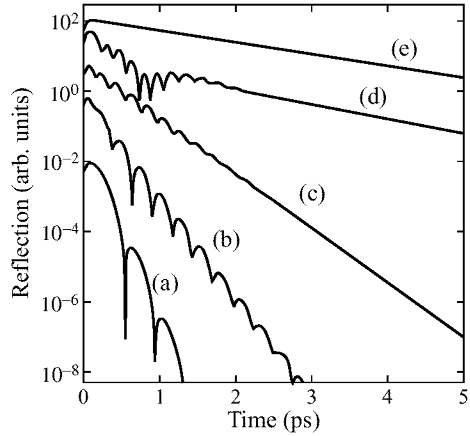

Fig. 1. Time-resolved reflection of GaN/AlGaN quantum well structures: (a) SQW with $\eta\Delta = 5$ meV; (b) MQW, $N = 10$, $\eta\Delta = 5$ meV; (c) MQW, $N = 50$, $\eta\Delta = 5$ meV; (d) MQW, $N = 100$, $\eta\Delta = 5$ meV; (e) SQW with $\eta\Delta = 0$ meV.

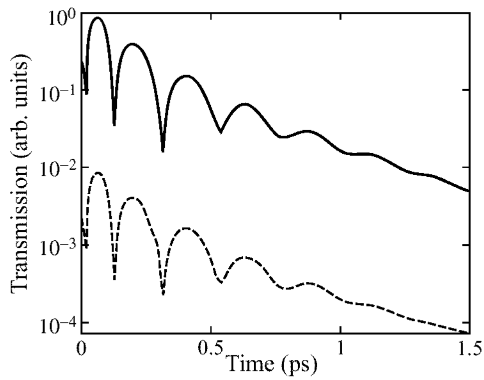

Fig. 2. Time-resolved transmission of light through the GaN/AlGaN MQW structure containing 50 QWs (solid) and through the thin film of a bulk semiconductor characterized by an effective dielectric susceptibility according to the EDM approximation (dashed).

To describe the MQWs one should substitute in that formalism the thickness of the film by Nd and the exciton longitudinal-transverse splitting ω_{LT} by $2\Gamma_0/kd$ [11].

Figure 2 shows the time-resolved transmission of the structure (c) calculated within the EDM approximation (dashed line) in comparison with the exact calculation (solid line). Note, that the criterion (4) is satisfied for this structure, with $n = 1$. An excellent agreement between the exact and approximate calculations allows us to apply the steepest descent method presented in Ref. [10] to find the decay time τ and the period of oscillations T in the time-resolved reflection of the MQWs. Using Eq. (27) from Ref. [10] one can obtain

$$T(t) = \frac{\pi\sqrt{t}}{\sqrt{N\Gamma_0}[1 + (3\Delta^2 t/4N\Gamma_0)]}. \tag{5}$$

In order to find a compact analytical formula for the decay time τ one has to extend substantially the formalism of Ref. [10] and the EDM approximation. Let us remind that the spectrum of exciton–polariton eigen-modes in a MQW structure consists of a few super-radiant modes having a huge oscillator strength and of many dark modes having a very small oscillator strength. If the condition (4) is satisfied, i.e. either for short-period MQWs or for quasi-Bragg-arranged MQWs, there is only one super-radiant mode with an extremely short decay-time. At the pico-second time-scale the exciton radiative decay is governed by the dark modes, which form two bands similar to the bulk exciton–polariton bands. That is why the EDM approximation works so well for this kind of MQWs.

In the limit $\Delta \ll N\Gamma_0$ one can assume the sum of the oscillator strengths of the exciton–polariton modes to be independent of Δ. Thus, the oscillator strength of the dark modes can be estimated as a total oscillator strength (proportional to $N\Gamma_0$) minus the oscillator strength of the super-radiant mode, which decreases with increase of Δ. Within this hypothesis and with use of the Eq. (A6) from Ref. [10] one can find the decay rate of the time-resolved optical response of a MQW structure as

$$\frac{1}{2\tau} = \gamma + \frac{\Delta^2}{2[N\Gamma_0 - (\Delta^2/2N\Gamma_0)]}. \tag{6}$$

In conclusion, we have shown that the vertical motional narrowing effect is of great importance for the coherent optical spectra of the GaN/AlGaN MQWs. The approximate analytical expressions for the decay rate and period of oscillations of the time-resolved reflection and transmission spectra of MQWs have been derived.

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