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Nonlinear absorption and gain in InGaAs/GaAs quantum wells

Giuliano Coli,[a] Adriana Passaseo, Donato Greco, and Roberto Cingolani
Dipartimento di Scienza dei Materiali, Università di Lecce, 73100 Lecce, Italy

Marco Tagliaferri, Paolo di Trapani, and Alessandra Andreoni
Istituto di Scienze Matematiche, Fisiche e Chimiche, Università di Milano 22100 Como (Italy)

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We present a detailed study of the excitonic nonlinearities in InGaAs/GaAs multiple quantum wells based on both stationary and transient pump-and-probe transmission spectroscopy. Bleaching of the excitonic resonance and free carrier gain have been observed. A quantitative analysis of the observed nonlinearity is provided by means of a rigorous solution of the Bethe–Salpeter equation for the investigated heterostructures. © *1997 American Institute of Physics.*
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Excitonic nonlinearities play a fundamental role in the design of optical modulators. The main issues are the quantitative understanding of the density dependence of the exciton oscillator strength, and the cross over from excitonic absorption to optical gain when the Mott threshold is overcome. Among the various III–V material systems available for optoelectronic devices, InGaAs/GaAs heterostructures are good candidates for application in the near infrared region. In this letter, we have investigated the excitonic nonlinearity of high-quality InGaAs/GaAs quantum wells grown by metal organic chemical vapor deposition (MOCVD) on (100) GaAs substrates. Both stationary conditions and time resolved pump-and-probe transmission spectroscopy have been used in order to measure the density dependence of the exciton bleaching and the occurrence of optical gain. The quantitative analysis of the nonlinear optical spectra has been performed by means of an upgraded Green-function theory based on a suitable solution of the Bethe–Salpeter equation, including the k dependence of dipole matrix elements.

We studied three $In_{0.12}Ga_{0.88}As/GaAs$ multiple quantum well (MQW) samples grown by MOCVD on semi-insulating GaAs substrates buffered by 300-nm-thick undoped GaAs. The samples comprised 15 quantum wells of well width $L_W = 3$, 6, and 15 nm, respectively, separated by 35-nm-thick GaAs barriers. All the structures were covered by a topmost GaAs capping layer (5 nm). Details about growth and linear optical characterization of these samples can be found in our previous work.[1] Stationary condition pump-and-probe (P&P) transmission spectroscopy was performed by using the third harmonic of a Nd:YAG laser as the pump and the tunable output of a parametric oscillator (Spectra-Physics MOPO 710) as probe. Stimulated emission spectra were collected simultaneously in order to correlate the onset of lasing with the presence of gain in the P&P spectra. Time resolved P&P spectra were measured with a frequency-doubled, chirped-pulse amplified, feedback-controlled mode-locked Nd:glass laser (Light Conversion Twinkle) having a pulse width of 1.2 ps. An optically delayed broadband probe of similar duration was obtained by continuum generation in a water cell pumped by a fraction of the pump pulse.

In Fig. 1, we show representative stationary P&P spectra of the 9 nm MQW, together with the corresponding stimulated emission spectra (inset). The salient features of the experiment are the following:

(i) At intermediate power densities (up to about 1 kW/cm^2), a small blue shift of the exciton resonance (about 1 meV) is observed. The oscillator strength slightly reduces with respect to the unperturbed one, whereas the continuum is almost unaffected by the pump. This demonstrates that space phase filling is not enough to substantially reduce the electrostatic energy of pair.

(ii) Above 1.6 kW/cm^2, the excitonic resonance and the continuum enhancement are satured. Under this condition, strong screening of the Coulomb interaction occurs due to the high density of free carriers in the bands.

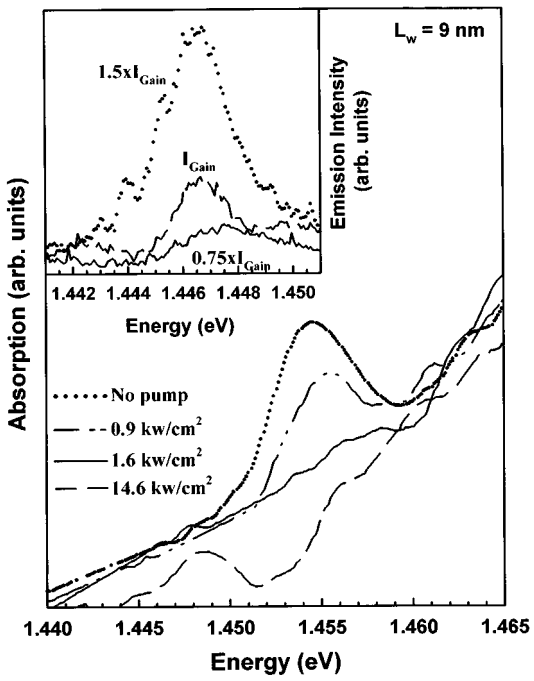

FIG. 1. Stationary condition P&P absorption spectra collected at different power densities from the $L_w = 9$ nm MQW sample. The temperature is 10 K. Inset: stimulated emission spectra collected under the same excitation conditions of the P&P spectra.

[a]Electronic mail: coli@axpmat.unile.it

(iii) Optical gain occurs at $I_{gain} \approx 14.6$ kW/cm^2 and is overwhelmed by Fabry–Perot oscillations.[2] Simultaneously, stimulated emission appears in the photoluminescence (PL) spectra (see inset of Fig. 1), with a characteristic Stokes shift of about 8 meV at threshold. The integrated area of the stimulated emission at $1.5\times I_{gain}$ is three times larger than at I_{gain}, thereby showing the expected superlinear increase. An overall red shift of about 15 meV with respect to the spontaneous emission band is seen at this photogeneration rate, consistent with the simultaneous observation of negative absorption (gain) in the P&P spectra.

A quantitative analysis of the above data is performed by solving the Bethe–Salpeter equation[3–5] (BSE), providing the dressed electron-hole pair propagator for the interacting Fermi sea. By integrating the solution, including the k dispersion of matrix elements in the bands $\mu(\mathbf{k})$, we obtain the dielectric susceptibility

$$\chi(\omega)=\chi_{\rm fc}(\omega)+\chi_{\rm exc}(\omega)+\chi_{\rm enh}(\omega). \tag{1}$$

In the reciprocal space ($\mathbf{k}$,$\mathbf{p}$)

$$\chi_{\rm fc}(\omega)=-\frac{1}{\pi}\cdot\sum_{\mathbf{k},\mathbf{p}}\mu(\mathbf{k})G_0(\mathbf{k},\mathbf{p})\mu^{\rm cc}(\mathbf{p}) \tag{2}$$

is the free carrier (fc) contribution, whereas

$$\chi_{\rm exc}(\omega)=-\frac{1}{\pi}\cdot D_0(\omega)\,\frac{L(\omega)^2}{1+L(\omega)/V}\sum_{\mathbf{k},\mathbf{p}}\mu(\mathbf{k})\mu^{\rm cc}(\mathbf{p}) \tag{3}$$

is the exciton contribution. The enhancement due to the electrostatic correlation between carriers is given by

$$\chi_{\rm enh}(\omega)=+\frac{1}{\pi}\cdot L(\omega)\sum_{\mathbf{k},\mathbf{p}}\sum_{\mathbf{q}}\mu(\mathbf{k})G_0(\mathbf{k},\mathbf{q})\mu^{\rm cc}(\mathbf{p}). \tag{4}$$

$D_0(\omega)$ is the unperturbed density of states (DOS) whereas

$$L(\omega)=\frac{1}{D_0(\omega)}\,\frac{1}{2}\sum_{\mathbf{k},\mathbf{p}}\sum_{\mathbf{k}_1,\mathbf{k}_2}G_0(\mathbf{k},\mathbf{k}_1,\omega)\times V_S(|\mathbf{k}_1-\mathbf{k}_2|)G_0(\mathbf{k}_2,\mathbf{p},\omega) \tag{5}$$

is the mean self-energy correction weighed by the renormalized, naked pair propagator[3]

$$G_0(\mathbf{k},\mathbf{p},\omega)=\frac{[1-f_e(k)-f_h(p)]\,\delta_{\mathbf{k},\mathbf{p}}}{\hbar\omega-\left(E_{\rm gap}+\Sigma+\dfrac{\hbar^2k^2}{2m^*}\right)+i\eta}.$$

m^* is the reduced mass for pairs, η is a damping factor, and $\mu^{\rm cc}$ is the complex conjugate of the matrix element μ. Further details of our model will be presented in a forthcoming article. For the present letter, it is important to know that Eqs. (1)–(5) are quite general, as they hold for bulk as well as confined heterostructures and for any form of the dipole matrix element, screened Coulomb potential (V_s), and self-energies corrections (Σ). Solving the BSE, we have assumed that the mean self-energy correction, weighted by the dressed propagator, does not substantially differ from that of the diluted Fermi gas, coherently with the ladder approximation. In our calculations, we have included a form factor[6] in order to take into account the effects of finite thickness on Coulomb potential. The screened Coulomb potential and self-energy corrections have been calculated in the static limit, within the single plasmon pole approximation, according to Refs. 7 and 8, respectively. The single-particle energy correction causes band gap shrinkage and chemical potential renormalization in each band,[4] whereas the nonlinear effects associated with the inhibition of transmission probability (band-filling effects) are taken into account by the filling factor[3,4] in the numerator of $G_0(\mathbf{k},\mathbf{p},\omega)$. In this way, Fermi distributions $[f_{e,h}(k)]$ and inverse screening lengths also include the effective temperature of particles.[8] Within our two-band model, we used an approximate single-particle dipole moment for the heavy hole exciton, in transverse electrical (TE) polarization,[9] which includes renormalization effects.[10] In Fig. 2, we show our theoretical calculations. The calculated density dependence of the absorption spectrum reproduces the overall behavior of the P&P spectra quite well. The exciton oscillator strength reduces by one half at carrier density of the order of 1.5×10^{11} cm^{-2}, which is less than classical Mott saturation threshold (2.0×10^{11} cm^{-2}). Furthermore, at a low density, the dotted and dashed dotted curve reproduce the blue shift of the excitonic resonance (1 meV) observed in the P&P spectra.

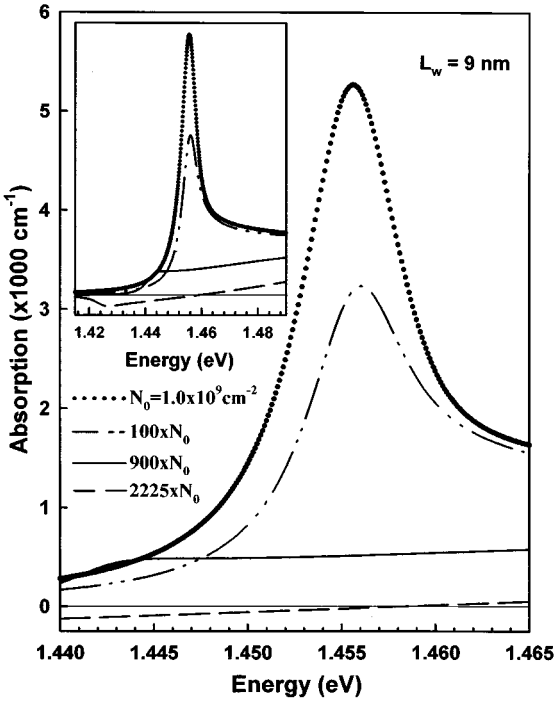

FIG. 2. Pump-and-probe spectra calculated according to Eqs. (1)–(5). The main features of the experimental spectra reported in Fig. 1 are well reproduced.

The dashed line shows that a gain of about 200 cm^{-1} occurs at a carrier density of the order of 2.2×10^{12} cm^{-2}. At this concentration, the cross over from gain to absorption occurs at the same energy of the experimental one. Under this condition, a band gap renormalization of about 35 meV is obtained from the calculated spectra. The above data indicate the efficient many-body nonlinearity of excitons in the InGaAs/GaAs material system. Further insight in this problem can be obtained by the time resolved P&P spectra.

In Fig. 3, we show the temporal evolution of the exciton

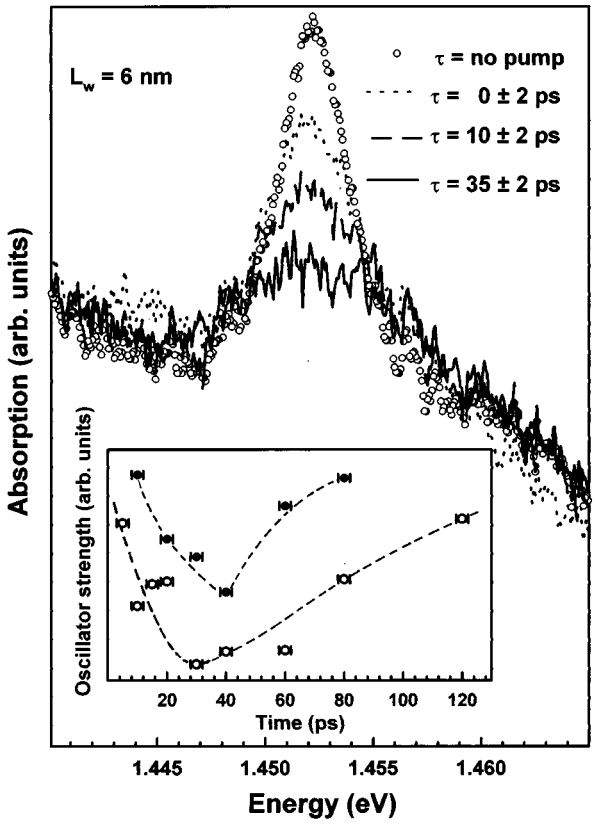

FIG. 3. Time resolved pump-and-probe spectra collected at different delays from the $L_w = 6$ nm sample. The temperature is 77 K. Inset: temporal evolution of the $n = 1$ exciton oscillator strength at two different pump fluences: 509 (empty dots) and 306 μJ/cm^2 (full dots) of the pump intensity.

bleaching of the $L_w = 6$ nm sample, at carrier densities ranging between 1.0×10^9 and 2.0×10^{11} cm^{-2}. The reduction of the oscillator strength occurs on a time scale of about 40 ps, i.e., on a time scale longer than the pulse width. The recovery of the oscillator strength is found to occur on a time scale exceeding 100 ps, which is unexpectedly long.

A clear description of the bleaching dynamics is given in the inset of Fig. 3, where we plot the time dependence of the integrated oscillator strength of the $n = 1$ exciton recorded at two different power densities. Both the bleaching and the recovery are found to be faster at low density, though they do not follow the pump pulse. At high density, the recovery is very slow, and falls in the nanosecond range. This regime will be explored in detail in a forthcoming work. The unexpectedly slow dynamics of the nonlinearity (at least as compared to other III–V heterostructures) is presumably due to the off-resonant pump (about 0.8 eV of initial excess energy for the time resolved P&P measurement). This originates a hot-carrier population which produces, in turn, hot phonon reabsorption which slows down the carrier cooling rate. The carrier temperature estimated from the high energy tail of the spontaneous luminescence, was of the order of 80–100 K. Furthermore, hole localization at compositional disorder in the ternary alloy might cause a partial suppression of the absorption recovery due to exciton localization. Resonant P&P experiments presently in progress, will help to clarify this issue.

In conclusion, we have investigated the nonlinear excitonic absorption of InGaAs/GaAs quantum wells by means of stationary and transient pump-and-probe spectroscopy. The quantitative analysis of the optical spectra, based on the integration of BSE for the dielectric susceptibility, reproduces our P&P experiments nicely thus providing the carrier density dependence of the exciton bleaching and optical gain. The unexpectedly slow dynamics of the nonlinearity has to be considered as a potential problem for the design of fast modulators.

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