FEMTOSECOND SPECTRAL HOLE-BURNING IN GaAs/AlGaAs MULTIPLE QUANTUM WELLS

J. Oudar, J. Dubard, F. Alexandre, D. Hulin, A. Migus, A. Antonetti

JOURNAL DE PHYSIQUE
Colloque C5, supplément au n°11, Tome 48, novembre 1987

FEMTOSECOND SPECTRAL HOLE-BURNING IN GaAs/AlGaAs MULTIPLE QUANTUM WELLS

J.L. OUDAR, J. DUBARD[(1)], F. ALEXANDRE, D. HULIN*, A. MIGUS* and A. ANTONETTI*

Centre National d'Etudes des Télécommunications, 196, Avenue Henri Ravera, F-92220 Bagneux, France
**Laboratoire d'Optique Appliquée, Ecole Polytechnique, ENSTA, F-91120 Palaiseau, France*

Résumé : La saturation sélective des états couplés optiquement (trou spectral) a été observée en régime femtoseconde dans une structure à puits quantiques multiples de GaAs/AlGaAs. La comparaison avec des résultats antérieurs obtenus sur le GaAs massif dans les mêmes conditions d'excitation montre que les collisions entre porteurs sont environ deux fois plus efficaces dans la structure à puits quantiques, en raison de la réduction des effets d'écran à deux dimensions, ainsi que de l'augmentation de la renormalisation de bande interdite par rapport au matériau massif.

Abstract : The dynamical spectral-hole-burning effect, due to selective saturation of the optically coupled states, has been observed in the femtosecond regime in a GaAs/AlGaAs multiple quantum well structure. Comparison with previous results on bulk GaAs under similar excitation conditions shows that carrier-carrier collisions are about twice as efficient in the quantum wells, due to the reduction of screening efficiency in two-dimensional structures, as well as the increase of band-gap renormalization with respect to the bulk material.

Ultrashort light pulses are a very effective means to generate highly-non-equilibrium carriers and the time-resolved optical techniques can be used to obtain various informations on the intraband relaxation mechanisms [1-3]. Recently we have observed [2], for the first time in a direct-gap semiconductor, a femtosecond spectral hole-burning due to nonthermalized photoexcited carriers in a μm-thick GaAs layer. Due to a careful choice of the pump linewidth (5 meV) and central wavelength (20 meV above the band-gap), the electron-hole pair dephasing time could be resolved by studying the spectral dependance of the absorption saturation around the pump wavelength in a pump-probe experiment with 100 fs probing resolution. Since the sample was at low temperature and the above-band-gap pump excess energy was much smaller than the polar optical phonon energy (37 meV), the intercarrier collisions were the dominant scattering process. These experiments have thus provided a unique opportunity to study the carrier-carrier scattering rates within a photogenerated electron-hole (e-h) plasma, and the thermalization dynamics of initially quasi-monoenergetic electron distributions.

This technique is used here to study such collision rates in GaAs/AlGaAs Multiple Quantum Well Structures (MQWS), so as to compare them with our previous results on bulk GaAs. Other experiments on non-thermal distributions in such MQWS [3] have only yielded qualitative results because the pump spectrum was too large to allow the determination of the e-h homogeneous linewidth through a frequency-domain measurement. The present experiment is made under similar excitation conditions (pulse duration, spectral width, intensity, and photon excess energy above the band gap) as in Ref. 2, so as to have direct access to the effect of the quantum well reduced dimensionality.

[(1)] Present address : Department of Physics, North Texas State University, Denton, TX 76203, U.S.A.

The molecular-beam-epitaxy-grown MQWS sample consists of 60 GaAs wells of 114 Å thickness separated by $Al_{0.3}Ga_{0.7}As$ barriers of 79 Å, sandwiched between two $Al_{0.3}Ga_{0.7}As$ layers, each of 1 μm thickness. The GaAs substrate was removed by selective etching on a surface of about 1 mm^2 and antireflection coatings were deposited, thereby allowing to measure MQWS transmission spectra.

Pump-and-probe measurements were performed as described in Ref. 2. The pump pulse was obtained by spectral filtering of a white-light continuum by means of a narrow bandwidth (5 meV) interferential filter. This selected pulse (bandwidth-limited 450 fs duration) was further amplified in a styryl-9 dye solution [4], raising the available pump energy to the μJ level. The probing pulse was a broad-band continuum pulse of 100 fs duration, which determines the time resolution of the experiment. Its intensity was low enough to keep the sample in the linear regime in absence of the pump, as confirmed by comparison with a conventionally measured absorption spectrum. The pump and probe pulses were obtained in two separate continuum cells so as to avoid any possible artifact due to coherence coupling.

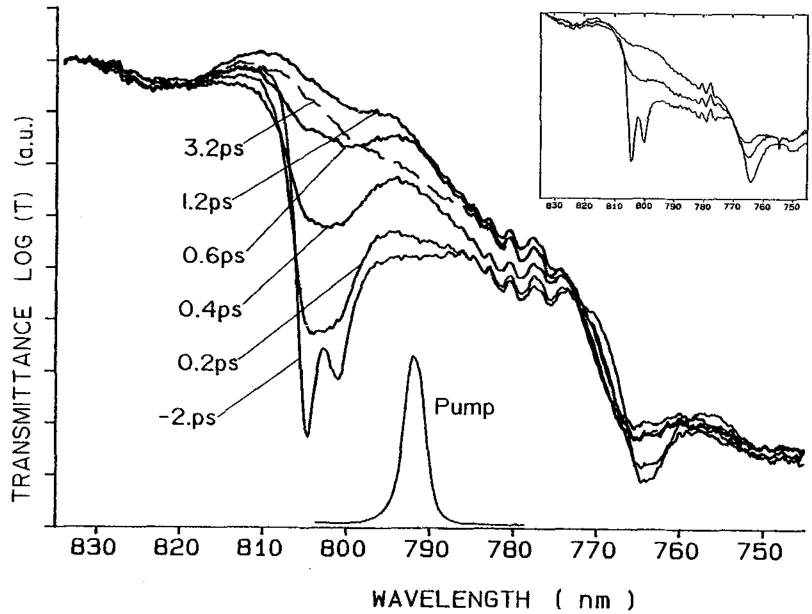

FIG. 1 : Time-resolved transmission spectra at various probe delays after the beginning of the pump pulse, under narrow-band excitation. Insert : transmission spectra under broad-band excitation (the wiggles around 780 nm are instrumental)

A typical set of transmission spectra is shown on Fig. 1, at selected values of the delay between the probe and pump pulses. The pump intensity spectrum (bottom curve in Fig. 1) has a central wavelength of 792 nm. The pump excess energy above the band-edge of the n = 1 heavy-hole to conduction subbands is approximately 20 meV, so that the results are directly comparable to the bulk GaAs results [2]. The curve labeled -2 ps is characteristic of the unexcited sample, while during and after the pump pulse the transmission increases due to the absorption saturation. A very distinct feature in the vicinity of the pump wavelength is especially apparent in the leading edge of the pump pulse (curves labeled 0.2 and 0.4 ps), which progressively disappears after the pump maximum intensity (occuring at 0.5 ps, curve not shown), leaving place to a smooth variation of transmission (curve 3.2 ps). This feature is indicative of a dynamical spectral hole-burning which corresponds to the preferential filling of the subband states directly coupled to the pump pulse. As shown in the insert of Fig. 1 this does not occur under broad-band pumping (715-815 nm), where one observes transmission spectra characteristic of thermalized carrier distributions. Also apparent on Fig. 1 are some interesting data on the n = 2 exciton (centered at 764 nm). From the very beginning of the pump pulse, a significant screening occurs, as well as a red shift of about 4 meV of the onset of the n = 2 transition. Since the n = 2 subbands are essentially empty these are pure many-body effects (exciton screening and band-gap renormalization) induced on the n = 2 subband states by an e-h plasma in the n = 1 subbands.

From the spectral hole-burning lineshape one can deduce a value of the dephasing time T_2 of the e-h transition at the energy of the pump pulse. This provides a direct determination of the carrier-carrier scattering rates within the electron-hole plasma. In Ref. 2 the data were analyzed by describing the interband transitions as an assembly of two-level systems, each valence band state being optically coupled to only one conduction band state, because of momentum conservation. At a given energy separation E, these two-level systems have a finite homogeneous linewidth $\Gamma(E) = 1/T_2$ and a joint density of states $\rho(E)$. By describing the photo-excited carrier distribution at time t as the sum of a thermalized part $f_{th}(t, E)$ and a non-thermal part $f_{nt}(t, E)$, one obtains the following expression [2] for the absorption coefficient α normalized to that of the unexcited sample α_0 :

$$\frac{\alpha}{\alpha_0}(t,h\omega) = \left[1 - f_{th}(t,h\omega)\right] \left[1 - \left\{1 - \frac{1}{(1+I)^{1/2}}\right\} \left(\frac{1}{1+\Delta^2}\right)\right] \qquad (1)$$

where $\Delta = T_2(\omega-\omega_p)/[1+(1+I)^{1/2}]$ is the normalized detuning parameter with respect to the pump frequency ω_p, and I is the light intensity normalized to the saturation intensity of the two-level systems. The last bracket in eq.(1) corresponds to the hole-burning, in the form of a power-broadened Lorentzian, while the first bracket of the right-hand size is the usual term describing the absorption saturation by thermalized distribution of carriers, when neglecting band-gap renormalization and screening of the excitonic effects.

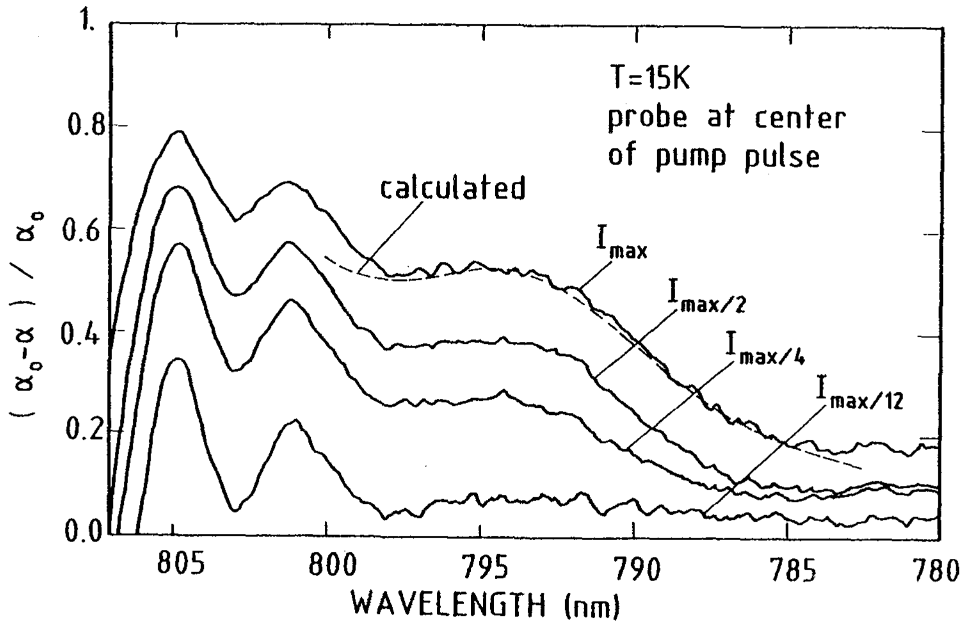

FIG. 2 : Normalized difference spectra at various pump intensities

On Fig. 2 are reproduced some normalized difference spectra $(\alpha_0 - \alpha)/\alpha_0$ together with a curve calculated from eq.(1). The experimental data here were obtained at a fixed delay (corresponding to the maximum of the pump pulse) but different excitation intensities. At the lowest intensity (0.15 I_{max}) the main absorption changes come from the saturation of the heavy-hole and light-hole excitonic absorption at 805 and 801 nm, but some hole-burning also occurs around the pump wavelength. The amplitude of the nonlinear excitonic features then saturates at higher intensities, and is superimposed on a larger smoothly varying background. This is consistent with the current description of excitonic nonlinearities in MQWS [5,6], in terms of phase-space filling and exchange effects. At high intensities the amplitude of the hole-burning feature becomes quite significant, and its lineshape can be compared to the theoretical one with reasonable accuracy. To obtain a good fit with the calculated curve of eq.(1) it was found necessary to allow for a frequency offset (red-shift) of the center of the non thermal distribution with respect to the pump wavelength. Such a red-shift could come from the effects neglected in the above-discussed two-level model, such as the energy renormalization of the nonthermal carriers created at earlier times, or a strong energy dependence of the carrier-carrier scattering rates (being faster at higher energies). The best fit was obtained with the parameters indicated in Table I.

	Calculated curves (best fit)			
Experimental curve	Dephasing time T_2(fs)	Normalized intensity I	Energy offset (see text)	Carrier density (arb. units)
I_{max}	150	2.5	3 meV	1
$I_{max}/2$	170	1.3	3	0.7
$I_{max}/4$	170	0.65	3	0.5
$I_{max}/12$	150	0.15	0	0.15

Table 1

One notes that the dephasing time does not vary strongly with the carrier density, as already noted for bulk GaAs [2,7]. This is due to the fact that for the carrier-carrier scattering rates one can distinguish three regimes of carrier densities :

(1) at low densities N (Boltzmann limit, and negligible screening of Coulomb interaction) the scattering rate per carrier is expected to increase with N, i.e. with the number of scattering centers

(2) intermediate density : the scattering probability saturates as a function of N, because

(a) the increased screening of coulomb interaction

(b) the decreased number of empty states available as final scattering states both compensate the increase of scattering centers

(3) strong degeneracy : the above mechanisms have a maximum efficiency, which results in a decreasing of the scattering probability per carrier.

As a result the carrier-carrier scattering rate goes through a maximum in the intermediate density regime in which the present experimental data were obtained.

One also notes a significant effect of the reduced dimensionality in the carrier-carrier scattering rates, since the dephasing time was found to be 300 fs for bulk GaAs under quite similar experimental conditions. This is attributed mainly to the reduced screening efficiency of the electron-hole plasma, which is characteristic of two-dimensional (2D), structures [8]. Another possibility is the fact that the band-gap renormalization is larger in 2D than in 3D structures [9], which increases the effective photon excess energy above the band-gap. Since the scattering probabilities increase at higher energies, the band-gap renormalization due to the carriers generated during the leading edge of the pump pulse is thus an indirect cause for larger scattering rates.

Finally Fig.3 shows the time-evolution of the hole burning amplitude (indicative of the carrier thermalization) after the excitation pulse. Although the major part of the non-thermal distribution vanishes very rapidly on the subpicosecond time-scale, a residual hole-burning persists at times longer than 2 ps after the excitation pulse maximum. This fact, also observed on bulk GaAs [2], shows that the inelastic collision processes responsible for the carrier thermalization are much less effective than the quasi elastic ones which contribute to the dephasing time T_2.

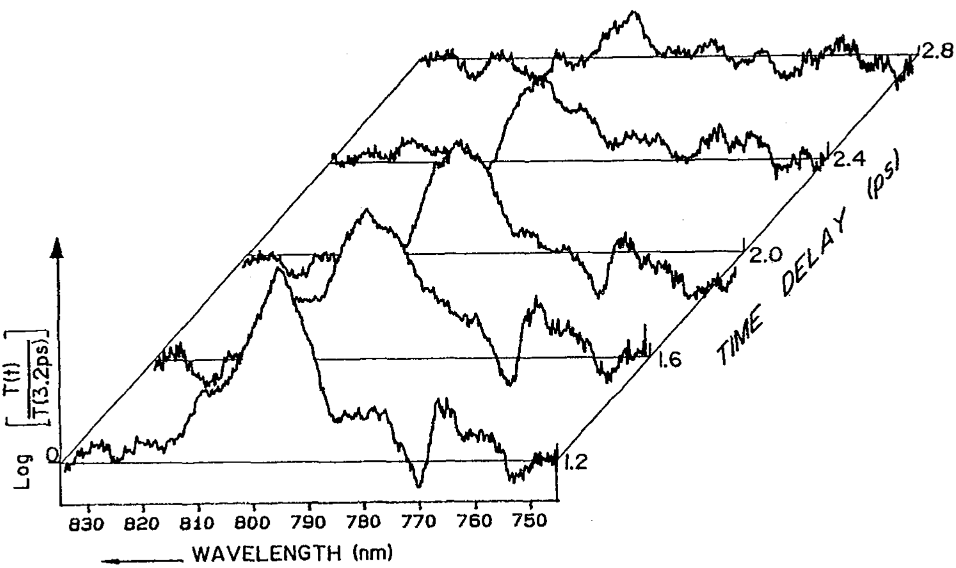

FIG.3 : Hole-burning decay after the end of the pump pulse. The spectrum at +3.2 ps is taken as a reference for the thermalized regime.

In conclusion we have observed the effect of dynamical spectral hole-burning in a GaAs quantum-well structure. Through direct comparison with a previous measurement on bulk GaAs the results allow to study the effect of dimensionality on the carrier-carrier scattering rates. We have found that the near band-edge homogeneous linewidth is about twice broader in the MQWS than in the bulk material, and discussed this result in terms of the screening of coulomb interaction and of the band-gap renormalization in quasi two-dimensional structures.

REFERENCES

1. J. Shah, J. Phys. (Paris), Colloq. C7 42, 445 (1981)
2. J.L. Oudar, D. Hulin, A. Migus, A. Antonetti and F. Alexandre, Phys. Rev. Lett. 55, 2074 (1985)
3. W.H. Knox, C. Hirlimann, D.A.B. Miller, J. Shah, D.S. Chemla and C.V. Shank, Phys. Rev. Lett. 56, 1191 (1986)
4. A. Migus, A. Antonetti, J. Etchepare, D. Hulin, and A. Orszag, J. Opt. Soc. Am. B2, 584 (1985)
5. D.S. Chemla and D.A.B. Miller J. Opt. Soc. Am. B2, 1155 (1985)
6. S. Schmitt-Rink, D.S. Chemla and D.A.B. Miller, Phys. Rev. B32, 6601 (1985)
7. J. Collet, J.L. Oudar and T. Amand, Phys. Rev. B34, 5443 (1986)
8. T. Ando, A.B. Fowler and F. Stern, Rev. Mod. Phys. 54, 437 (1982)
9. G. Tränkle, H. Leier, A. Forchel, H. Haug, C. Ell and G. Weimann, Phys. Rev. Lett. 58, 419 (1987)