Dynamics of nonlinear optical properties in $In_xGa_{1-x}As/InP$ quantum-well waveguides

C. Cacciatore
CSELT-Centro Studi e Laboratori Telecomunicazioni, via G. Reiss Romoli 274, 10148 Torino, Italy

L. Faustini
University of Brescia, Department of Electornics for Automation, via Branze 38, 25123 Brescia, Italy

G. Leo
Terza University of Rome, Department of Electronic Engineering, via della Vasca Navale 84, 00146 Roma, Italy

C. Coriasso, D. Campi, A. Stano, and C. Rigo
CSELT-Centro Studi e Laboratori Telecomunicazioni, via G. Reiss Romoli 274, 10148 Torino, Italy
(Received 1 October 1996)

We report the dynamics of the optical nonlinearity, measured by a weak probe propagating in single-mode $In_xGa_{1-x}As/InP$ quantum-well waveguides, at wavelengths corresponding to the heavy-hole exciton resonance or longer. The experiments are conducted at moderate pump energies (ranging from 0.1 to a few pJ/pulse). We discovered a contribution from the weak longitudinal component of the waveguided TM mode to the dynamical behavior, and show that this dominates the absorption of TM-polarized light and the relevant optical nonlinearities at wavelengths close to the heavy-hole exciton resonance. Moreover, while at the heavy-hole exciton resonance we observe induced transmission, in the transparent region, we observe an induced-absorption behavior. We evaluate the strong impact on the optical response of waveguides of this behavior, whose origin is purely excitonic, as we show in the framework of a simple model. We further show that the diffusion of carriers out of the waveguiding region is negligible with respect to carrier recombination processes. [S0163-1829(97)50408-5]

Optical nonlinearities in multiple-quantum-well (QW) structures attract much attention due to both potential applications and fundamental reasons. In particular, the QW's based on III-V semiconductors are relevant to the realization of optically controlled waveguiding devices for telecommunication applications. One nonlinear mechanism exploits the variation of the optical properties that can be induced by photogeneration of carriers. It is of utmost technological relevance that a spectral region there exists where the refractive variations induced by photogeneration are sizable, while the absorption coefficient is low enough to permit propagation over optical paths of practical length. Waveguiding devices are generally operated in that spectral region, that is, at wavelengths longer than that corresponding to the exciton resonance lowest in energy. Despite some work already done[1–5] knowledge of the nonlinear behavior and, in general, of the optical properties in that spectral region is far from being complete, particularly in waveguides.

In this contribution we report room-temperature, pump-probe experiments in which the dynamics of waveguides based on $In_xGa_{1-x}As/InP$ QW's is characterized with picosecond time resolution and in a spectral region ranging from the heavy-hole (hh) exciton resonance to a region of almost complete transparency. A variety of dynamical behaviors is observed, including exponential decay of optically induced transparency *or* absorption, and nonexponential decay, with sharp transients in the 100-ps time scale. Some of the behaviors are known, and can be explained in terms of nonlinear displacement of Fabry-Pérot oscillations, but some are newly observed here, as will be discussed in what follows. All these observed behaviors could be explained within a model in which all basic parameters are evaluated independently of the waveguide experiments.

The waveguide used in our experiment, with uncoated facets, acted as a nonlinear Fabry-Pérot resonator, whose absorption coefficient and refractive index could be changed by the carrier pairs excited through the absorption of the pump pulses: this in turn caused changes in the cavity transmission experienced by the probe pulses, coupled into the waveguide at varying delay with respect to the pump pulses. The electric field of the pump beam was polarized parallel to the QW layers, and thus it excited the TE-propagation mode in the waveguide, while the electric field of the probe beam was polarized perpendicularly to the QW layers and thus it excited the TM-propagation mode in the waveguide. In what follows we will refer in short to pump and probe beams as being TE and TM polarized, respectively. Pump and probe pulses had the same wavelength ranging from that corresponding to the exciton resonance lowest in energy (1.523 μm), to about 1.6 μm. A NaCl:OH color-center laser, synchronously pumped at 76 MHz by a Nd:YAG (YAG denotes yttrium aluminum garnet) mode-locked laser was used as the source in the exeriments, whose wavelength could be tuned from 1.48 to 1.60 μm. The pulse width was about 10 ps, slightly in excess of the round-trip time of the waveguides investigated here. The pump-probe delay could be varied from -0.5 to $+9$ ns. Both TE- and TM-polarized beams were end-fire coupled into, and out of, the waveguide. The outcoupled signal was spatially filtered through an iris, in order to ensure that only the guided light was detected. Po-

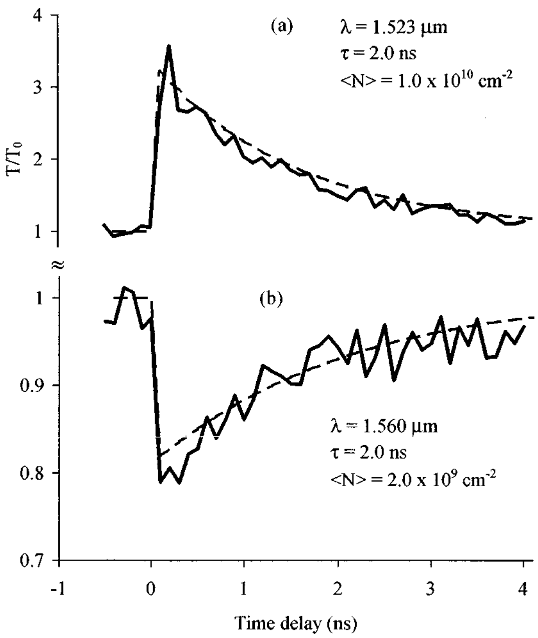

FIG. 1. Transient transmittance changes T/T_0 measured at (a) the hh resonance, 1.523 μm, (b) 1.560 μm as a function of probe time delay (solid lines). Pump pulse energies were about 0.1 pJ. The dashed lines are the calculated transmittance changes for computational conditions consistent with those addressed experimentally; in particular, $\langle N \rangle$ is the carrier density averaged over the waveguide length.

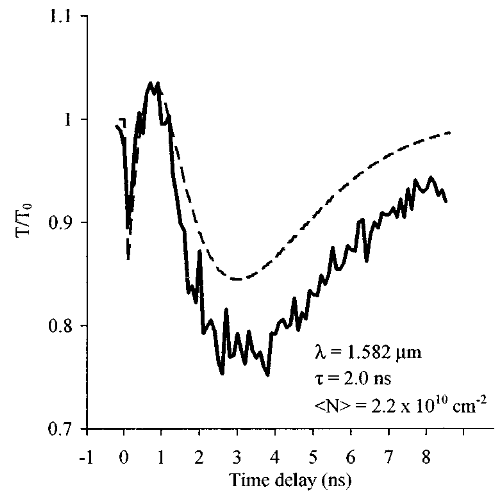

FIG. 2. Transient transmittance changes T/T_0 measured in the highly transparent region, at 1.582 μm, and for a pump pulse energy of about 0.1 pJ. Note the sharp initial transient, occurring over a time scale much smaller than the radiative recombination time constant. $\langle N \rangle$ is the carrier density averaged in the total waveguide length.

larization purity was checked to be better than 1:10^3 in the whole spectral range investigated here. To vary the pump power onto the waveguide the TE-polarized beam was passed through a variable attenuator. In general, the intensity of the probe pulse was smaller, by about a factor 100, as compared to that of the pump pulse, and the pump pulse peak intensity was always kept at least three orders of magnitude smaller than the intensity required to induce sizable virtual-carrier nonlinearities,[6] like the optical (or ac) Stark effect. Thus, all the nonlinear effects observed here are induced by the photogeneration of real carriers.

The semiconductor heterostructure used in this study was grown (on an n-doped substrate) by chemical beam epitaxy and consisted of a stack of thirty 7.0-nm, $In_xGa_{1-x}As$ QW's separated by 11.0-nm, InP barriers. Below this region there was a 0.45-μm InP buffer layer and above it a 0.3-μm InP capping layer. The material in the structure was both undoped and unstrained. Heavy- and light-hole (lh) resonances were clearly visible in the linear absorption spectrum at 1.523 and 1.430 μm, respectively. Ridge waveguides were fabricated by reactive-ion etching into the capping layer, leaving stripes 2 μm wide and 0.1 μm thick, which were cut to a length of 300 μm. The waveguide geometry allowed propagation of one TE and one TM mode.

When pump and probe wavelength was tuned at the exciton resonance, and the coupled pump energy was moderate (of the order of 0.1 pJ/pulse or less), the observed nonlinear behavior was *induced transmittance*, showing exponential recovery towards thermodynamical equilibrium, with a time constant $\tau = 2.0 \pm 0.5$ ns [see Fig. 1(a)]. The nonlinear variations that could be induced were found to be progressively smaller as the wavelength was raised, becoming practically undetectable around 1.55 μm. Beyond that wavelength, and at moderate pump intensity, the observed transmittance variations were opposite in sign with respect to those observed at shorter wavelengths, and the nonlinear behavior became *induced absorption*, whose recovery was again exponential, with the same time-constant value as in the above, Fig. 1(b). This recovery time was checked to be determined solely by the carrier lifetime, by performing a pump-probe experiment directly on the heterostructure (with light propagating perpendicular to the layers, at the exciton resonance and prior to processing), which indeed yielded the same value for the time constant within the experimental accuracy: this shows that carrier diffusion out of the waveguiding region can be discounted in the recovery dynamics. In general, as the pump intensity was increased (up to a few pJ/pulse), the recovery behavior was found experimentally to deviate from the exponential behavior, although remaining monotonic. We note that for wavelengths shorter than 1.570 μm no Fabry-Pérot oscillations were visible in the probe transmittance, while these became clearly visible at longer wavelengths, in the spectral region where the absorption was particularly small. In that region the dynamical behavior became more complicated, both in the moderate and in the strong pumping regime, showing generally a nonexponential and, eventually, nonmonotonic recovery, often with sharp transients in the 100-ps time scale. A typical example is shown in Fig. 2. Several experimental conditions were explored, whose results will be reported elsewhere. We further mention that, in general, no photoluminescence emission was observed in the waveguide experiments, irrespective of the pump power, at least down to nW detection sensitivity.

The observed results are noteworthy in two respects: First, the sign of the nonlinear absorptive variations depend on the probe wavelength, and, in particular, we observe an increasing absorption behavior at wavelengths corresponding to the absorption tail of the hh exciton resonance. This induced-absorption behavior is well known and experimentally verified in the literature on QW heterostructures.[7] In this work we emphasize its impact in waveguiding configurations. Second: The absorption variations are observed using a TM-polarized probe light, in a region in which the absorption (and thus the absorption variations) of light with electric field polarized perpendicularly to the QW plane should be vanishing, according to selection rules.

All the experiments could be interpreted within the framework of a model, based on our previous treatment of the excitonic nonlinear optical properties[8] in QW's. Our model describes the optical properties of the QW material and their nonlinear variations induced by a quasiequilibrium carrier density: it was checked to accurately reproduce the results of a stationary, spectral, pump-probe transmittance experiment conducted on the heterostructure. A few parameters could not be obtained within the model and had to be measured or estimated independently. In particular, (i) the spectral width of the hh exciton resonance was measured to be 15 meV from the linear absorption spectrum of the heterostructure. Further, (ii), in order to describe the waveguide experiment, we calculated the complex propagation constants of both TE and TM modes with the effective index method,[9] using the material properties resulting from the model and assuming that the photogenerated carriers decayed exponentially according to the time constant measured on the heterostructure. The waveguide transmittance changes T/T_0, addressed by the experiment, were obtained by inserting the optical properties calculated for the (probe) TM mode into the Fabry-Pérot (Airy) equation. The two main approximations in the model are the effective index method and the assumption of a uniform distribution of carriers along the waveguide, the latter being justified only at wavelengths much longer than that of the hh exciton resonance. However, we obtained very similar results in further work,[10] where we relaxed the uniform-density approximation by using a finite-difference computational scheme.

We note that in the wavelegth range explored here, both pump and probe beams resonantly generate excitons, which decay into electron-hole pairs with a subpicosecond time constant due to collision with LO phonons.[11] The pairs modify both the probability of exciton photogeneration by the probe photons and the refractive index, and these factors determine the propagation properties of the probe beam. From our model and from the experimental outcomes, we obtain the following picture.

(1) Based on symmetry considerations, the oscillator strengths of the hh and lh band-to-band transitions are predicted to be 0 and 1, respectively, for an electric field vector E perpendicular to the QW plane. Now, the solution of the Helmholtz equation for a TM mode in a planar waveguide predicts that the field E has *two* vector components, one perpendicular ($E_\perp$) and one parallel ($E_\parallel$) to the QW plane. The latter is usually neglected because it is very small as compared to the former. From this it is usually concluded that TM-polarized light can only induce lh-related band-to-band or excitonic transitions. We argue that this conclusion is not safe and this explains (i) our observation of linear and nonlinear absorption of TM-polarized light at wavelength strongly detuned from the lh exciton resonance (at least 90 nm); (ii) the marked hh character of the observed nonlinearity. In fact, while $E_\perp$ can only excite lh-related transitions. $E_\parallel$ can excite both hh and lh transitions with strengths $\frac{3}{4}$ and $\frac{1}{4}$, respectvely. In passing, we note that the problem becomes more complicated in ridge waveguides, where the Helmholtz equations predicts *three* components, of which *two* lie in the QW plane. Nevertheless, the second transversal component is vanishingly small compared to the longitudinal one[12] due to the fact that the optical confinement in the lateral direction is very weak compared to that in the direction perpendicular to the QW's. This justifies the slab approximation in this context: an exact evaluation of the three components in this case would require a finite-element calculation which is outside the scope of this paper.

In our experimental situation (that is a situation which is commonplace in optoelectronic and photonic devices based on unstrained QW's) the $E_\perp$ component cannot practically induce any transition, because the probe wavelength is strongly detuned from the lh resonance, while the $E_\parallel$ can excite the medium, due to closeness of the hh resonance wavelength. As a result, we calculated (within the effective index appoximation[9]) that the power fraction related to the longitudinal component of the TM mode was $\varepsilon = 2.8\times 10^{-2}$. We found that all the experimental results, otherwise unexplained, could be accurately reproduced by taking this component into account. We stress that the longitudinal component has dramatic impact on the linear and nonlinear properties despite its weakness, due to the relatively high hh absorption coefficient (with respect to the lh absorption coefficient) in the spectral region explored here, and due to a long optical pact, inherent in waveguided configuration. As a result of the discussed mechanism, a TM-polarized probe has a small but *nonvanishing* probability of exciting the hh resonance, as quantitatively described by our model. The strengths of TM-excited hh and lh resonances are ε and $1-\varepsilon$, respectively, however, at wavelength of the hh resonance or longer, the (small) hh excitonic absorption is the only sizable absorption mechanism. We note that a further mechanism exists, which could induce an inherent violation of the transition selection rules in QW's due to valence-band mixing.[13] We did not include this mechanism in our model, and nevertheless obtained a good agreement between modeling and experimental results: we may thus argue that the effects of band mixing are small compared to the mechanism considered here, at least in our case.

(2) The hh resonance is quenched *and* broadened under pumping.[14,15] With pump and probe tuned at the hh exciton peak, the photogenerated carriers inhibit formation of further hh excitons by the probe and the material becomes more transparent as in Fig. 1(a). On the contrary, at sufficiently longer wavelength, the collisional broadening of the absorption peak is the dominant phenomenon and we observe induced absorption of the probe beam[15] as in Fig. 1(b). According to our model, the crossover between induced transparency and induced absorption occurs between 1.535 and 1.545 μm, depending on the photogenerated carrier density (see Fig. 3). We stress that the induced absorption provides a feedback mechanism, which may influence crucially the behavior of waveguiding devices.[5,16] We further emphasize that the broadening mechanism at the origin of the induced absorption in a QW waveguide differs from the renormalization mechanism at the basis of the induced absorption observed in bulk semiconductors.[17] In fact, the exciton reso-

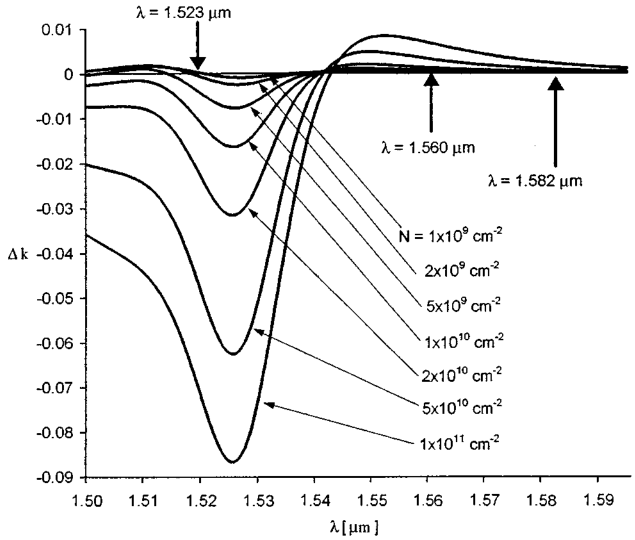

FIG. 3. Calculated spectral variations of the imaginary part of the QW refractive index at various photogenerated carrier densities.

nance spectral position is practically unaffected by the renormalization because the unbinding of the exciton under pumping compensates the gap renormalization[7,8] leaving the exciton resonance position ''locked'' to its position in the linear absorption spectrum, as we could verify in our nonlinear transmission experiment on the heterostructure prior to processing. We further stress that the change of the sign of the nonlinearity cannot be explained without considering the weak longitudinal component of the TM mode.

(3) In a wavelength region where Fabry-Pérot effect can be neglected, an exponential time decay is expected for (a) small probe absorbance in the spatial region modified by the pump and, (b), linear dependence of the absorptive variations on the photogenerated carrier density.[18] In general, large pump intensities induce deviations from the linear regime and the decay becomes nonexponential.

(4) At wavelengths where the material becomes sufficiently transparent (Fig. 2) the Fabry-Pérot fringes, and their displacement under pumping, dominate the transmittance of the probe beam, although sizable contributions of absorptive variations are still present. In this region the recovery of the equilibrium transmittance is always nonexponential, and strongly dependent on the wavelength as in Refs. 1 and 18. A very good matching between the modeling and the experiment can be obtained by tuning the wavelength addressed in the model within 1 nm. From our experimental results we evaluate a nonlinear phase shift of about 2 radians at 0.1-pJ pump power.

In conclusion, the results of our experiments can be grouped into three categories (induced transparency, induced absorption, and nonlinear Fabry-Pérot resonator), whose typical dynamics is illustrated in Figs. 1 and 2 and that correspond to three spectral regions. We demonstrate that the absorption of TM light can be related to hh transitions by taking proper account of the electromagnetic theory of waveguides. Our findings are believed to be general for multiple QW waveguides, while the relative extensions of the three regions and the relative weight of the effects can be engineered to a large degree, particularly using strained material,[19] or changing the geometrical parameters of the heterostructure.

[1] I. Gontijo, D. T. Neilson, J. Ehrlich, A. C. Walker, G. T. Kennedy, and W. Sibbet, Appl. Phys. Lett. 66, 1871 (1995).

[2] C. Cacciatore, D. Campi, C. Coriasso, G. Meneghini, and C. Rigo, Electron. Lett. **28**, 1624 (1992).

[3] B. Hubner, R. Zengerle, and A. Forchel, Appl. Phys. Lett. **66**, 3090 (1995).

[4] C. Coriasso, C. Cacciatore, D. Campi, A. Stano, C. Rigo, L. Faustini, and G. Leo (unpublished).

[5] C. Coriasso, D. Campi, C. Cacciatore, L. Faustini, G. Leo, F. Buscaglia, C. Rigo, and A. Stano, Appl. Phys. Lett. **67**, 585 (1995).

[6] W. H. Knox, D. S. Chemla, D. A. B. Miller, J. B. Stark, and S. Schmitt-Rink, Phys. Rev. Lett. **62**, 1189 (1989); A. Von Lehmen, D. S. Chemla, J. E. Zucker, and J. P. Heritage, Opt. Lett. **11**, 609 (1986).

[7] D. S. Chemla, D. A. B. Miller, P. W. Smith, A. C. Gossard, and W. Weigmann, IEEE J. Quantum Electron. **QE-20**, 265 (1984); D. S. Chemla and D. A. B. Miller, J. Opt. Soc. Am. B **2**, 1155 (1985); S. Schmitt-Rink, D. S. Chemla, and D. A. B. Miller, Adv. Phys. **38**, 89 (1989).

[8] D. Campi and C. Coriasso, Phys. Rev. B **51**, 10 719 (1995).

[9] M. J. Adams, *An Introduction to Optical Waveguides* (Wiley, New York, 1981).

[10] G. Osella, P. Debernardi, G. P. Coriasso, and D. Campi (unpublished).

[11] M. Nisoli, V. Magni, S. De Silvestri, O. Svelto, D. Campi, and C. Coriasso, Appl. Phys. Lett. **66**, 227 (1995).

[12] C. Vassallo and Ye-Heng Wang, J. Lightwave Technol. **8**, 56 (1990).

[13] J. S. Weiner, D. S. Chemla, D. A. B. Miller, H. A. Haus, A. C. Gossard, W. Weigmann, and C. A. Burrus, Appl. Phys. Lett. **47**, 664 (1985).

[14] D. R. Wake, H. W. Yoon, J. P. Wolfe, and H. Markoç, Phys. Rev. B **46**, 13 452 (1992).

[15] D. Campi, C. Coriasso, M. Nisoli, and S. De Silvestri, Appl. Phys. Lett. **67**, 953 (1995).

[16] J. E. Ehrlich, G. Assanto, G. I. Stegeman, and T. H. Heng Chiu, IEEE J. Quantum Electron. **QE-27**, 809 (1991).

[17] S. W. Koch, in *Optical Nonlinearities and Instabilities in Semiconductors*, edited by H. Haug (Academic, Boston, 1988), and references therein.

[18] T. Kanetake, H. Inoue, S. Tanaka, and K. Ishida, Electron. Lett. **29**, 1682 (1993).

[19] R. Jin, K. Okada, G. Khitrova, H. M. Gibbs, M. Pereira, S. W. Kock, and N. Peyghambarian, Appl. Phys. Lett. **61**, 1745 (1992).