Two-color midinfrared saturation spectroscopy of intersubband transitions in multiquantum wells

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A two-color pump–probe study of bound-to-bound and bound-to-quasibound intersubband transitions in InGaAs/AlGaAs and GaAs/AlGaAs multiquantum wells (MQW), with transition energies near 250 meV has been performed. Electrons were optically excited from the lowest to the upper subband using short midinfrared pulses of intensities up to 10^8 W/cm^2 from a dual optical parametric generator. Differential transmission spectra, taken with the weaker probe pulses, evidence homogeneous line broadening behavior. From the width of the coherence peak, centered near the pump frequency, and arising from a dynamic population grating, the upper state lifetime (bound-to-bound transition) was found to be 0.6 ps, in good quantitative agreement with absorption saturation data. © *1996 American Institute of Physics.* [S0003-6951(96)04248-9]

Among the attractive optical properties of quantum well (QW) semiconductor structures are giant second-order and third-order nonlinearities[1] which are significantly enhanced near intersubband resonances in the mid-IR. This resonance enhancement depends on the width of the homogeneous component, Γ_{hom}, of the intersubband absorption line and hence on the electron dephasing time, T_2. A knowledge of T_2 is also vitally important for the accurate modeling of novel devices such as QW subband lasers[2] and infrared detectors.[3]

Whether or not typical QW intersubband absorption lines are homogeneously or inhomogeneously broadened remains an open question[4,5] and the uncertainty means that T_2 cannot be deduced from linear QW intersubband absorption measurements where inhomogeneous broadening arising from QW thickness variations and from conduction band nonparabolicity may be present, neither can Γ_{hom} be obtained directly from intersubband lifetime measurements, because the intersubband lifetime is generally longer than the electron dephasing time.

Recently, cw induced absorption measurements at $\leqslant$100 K have shown[6] intersubband absorption lines in both GaInAs/AlInAs and GaAs/AlGaAs multiquantum well (MQW) samples (with subband splittings of 129 and 112 meV, and linewidths 5.1 and 6.2 meV, respectively) which are homogeneously broadened. CO_2 laser excitation from the $n=1$ to $n=2$ subbands induced absorption at the $n=2-n=3$ transition energy, but the induced absorption spectrum remained unchanged (and closely Lorentzian) as the excitation energy was tuned through the $n=1-n=2$ absorption peak. This implies a dominantly homogeneous broadening mechanism since inhomogeneous broadening would give an induced absorption spectrum which shifted at least as fast as the excitation energy as QW regions with different well-widths were excited.

We present here new results from two-color intersubband pump–probe absorption saturation measurements which give a valuable insight into the nature of the broadening mechanism of the $n=1-n=2$ subband transition. An intense spectrally narrow pump pulse resonantly excites electrons from the ground to the first excited state and a much weaker probe pulse scans through the resulting absorption spectrum and measures the differential transmission change (DTC) with respect to the unexcited sample.

Both samples studied were grown by molecular beam epitaxy on semi-insulating GaAs substrates with a linear graded InGaAs buffer for strain compensation[7] and had similar subband absorption peak energies, but one (sample No. 4009) had a ''bound-to-bound'' (BB) transition and the other (sample No. 3917) was a ''bound to quasibound'' (BQB) transition. The BB sample consisted of 75×5.85 nm thick $In_{0.5}Ga_{0.5}As$ wells, uniformly doped across the well region at sheet charge density $n_s=7.6\times10^{11}$cm^{-2} per QW and separated by 20 nm thick undoped $Al_{0.45}Ga_{0.55}As$ barriers. The BQB sample was made by cladding the main 3.5 nm thick absorbing GaAs QW (uniformly QW doped at $n_s=7\times10^{11}$ cm^{-2}) on each side by two additional 1.75 nm wells, each spaced from each other and from the main QW well by 4.69 nm thick $Al_{0.45}Ga_{0.55}As$ layers to serve as Bragg reflectors for the electron wave function[8] (inset of Fig. 1). Each of the 100 well periods was separated by a 15 nm thick $Al_{0.45}Ga_{0.55}As$ barrier giving an overall MQW period of 44.2

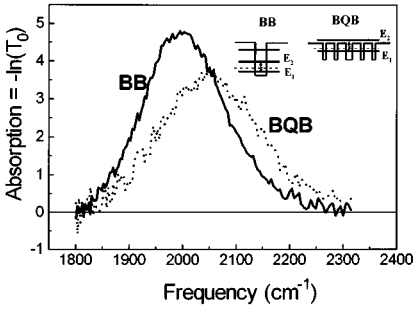

FIG. 1. Small-signal absorption spectra ($T=300$ K) of the MQW samples taken with an optical parametric generator: (a) bound-to-bound transition (BB) peaked at $E_{12}=248$ meV (2000 cm^{-1}) with a FWHM linewidth $\Delta E_{12}=22.1$ meV (178 cm^{-1}) and (b) bound-to-quasibound transition (BQB) peaked at $E_{12}=254$ meV (2049 cm^{-1}) with $\Delta E_{12}=27.6$ meV (223 cm^{-1}. Inset shows energy levels and corresponding transitions for each case (dashed line is the electron Fermi energy).

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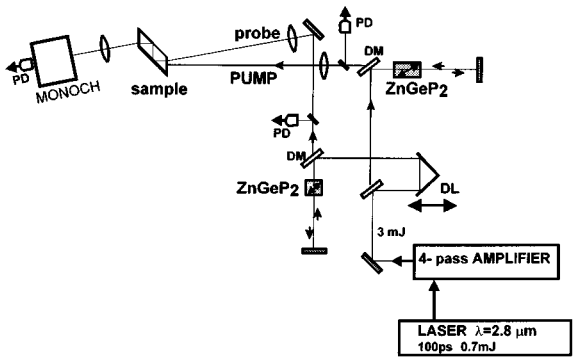

FIG. 2. Schematic of the two-color midinfrared picosecond spectroscopy system: DL–variable optical delay line, DM–dichroic mirrors, PD–infrared photodetectors.

nm. Both samples had similar small signal absorption spectra peaking at ~ 5 μm wavelength (Fig. 1).

All absorption experiments were performed at 300 K in a 45° multipass waveguide geometry with six reflections through the QW layers. Intersubband transitions were isolated from substrate phonon overtone absorption by ratioing the *p*- (absorbing) and the *s*- (nonabsorbing) polarized spectra.

The IR pulses were generated in two traveling-wave optical parametric generators (OPGs), each composed of a $ZnGeP_2$ crystal[9,10] in a double-pass configuration (Fig. 2). They were simultaneously pumped at $\lambda \sim 2.8$ μm by amplified 100-ps pulses from an actively mode-locked, *Q*-switched and cavity dumped Er^{3+}:Cr^{3+}:YSGG laser[9] (repetition rate 3 Hz). Each OPG was independently tunable in the range 3.9–10 μm. The ''pump'' OPG used type-II phase matching giving a 90 ps pulse of ~ 10 cm^{-1} spectral width, i.e., ~5% of the subband absorption linewidth and effectively monochromatic for our purposes. The ''probe'' OPG gave a 75 ps ~ 500 cm^{-1} wide mid-IR quasicontinuum resulting from the type-I phase matching near degeneracy.[10]

All measurements were taken at zero (within 1 ps) pump–probe delay. The pump pulse was focused to a 175 μm diam spot and the probe, focused down to 135 μm, had ~ 100 times weaker intensity. Pump and probe beams were separated (Fig. 2) by an angle of 10.5°, which is a compromise between minimizing the liner scattering of the pump light into the probe channel and maximizing the spatial overlap of the two beams inside the 45° waveguide MQW sample.

Outside the sample the pump beam intensity reached $I_{pu} \sim 100$ MW/cm^2 and the transmitted probe continuum was dispersed with a 1 cm^{-1} resolution grating monochromator. The transmitted and reference optical signals were detected with linear pyroelectric, InSb (77 K) and MCT (77 K) detectors and were ratioed with a double-modulation technique to remove linearly scattered light from the pump beam.

The laser pulses are long compared with typical inter- and intrasubband relaxation times (0.1–1 ps) and to the electron dephasing time ($T_2 \leqslant 100$ fs), i.e., the measurement regime is quasistationary. The pulses are short enough to avoid heating effects though; from total absorbed energy calculations we estimate a maximum lattice temperature rise of <1 K.

DTC spectra (Fig. 3) were taken at pump intensities exceeding MQW intersubband saturation intensities which were separately measured[11] to be $I_{sat} = 9.7$ MW/cm^2 (BB sample) and $I_{sat} = 6.8$ MW/cm^2 (BQB sample). As the pump photon energy with a very narrow spectral width was tuned throughout the absorption peak the DTC spectral *shape* (Fig.

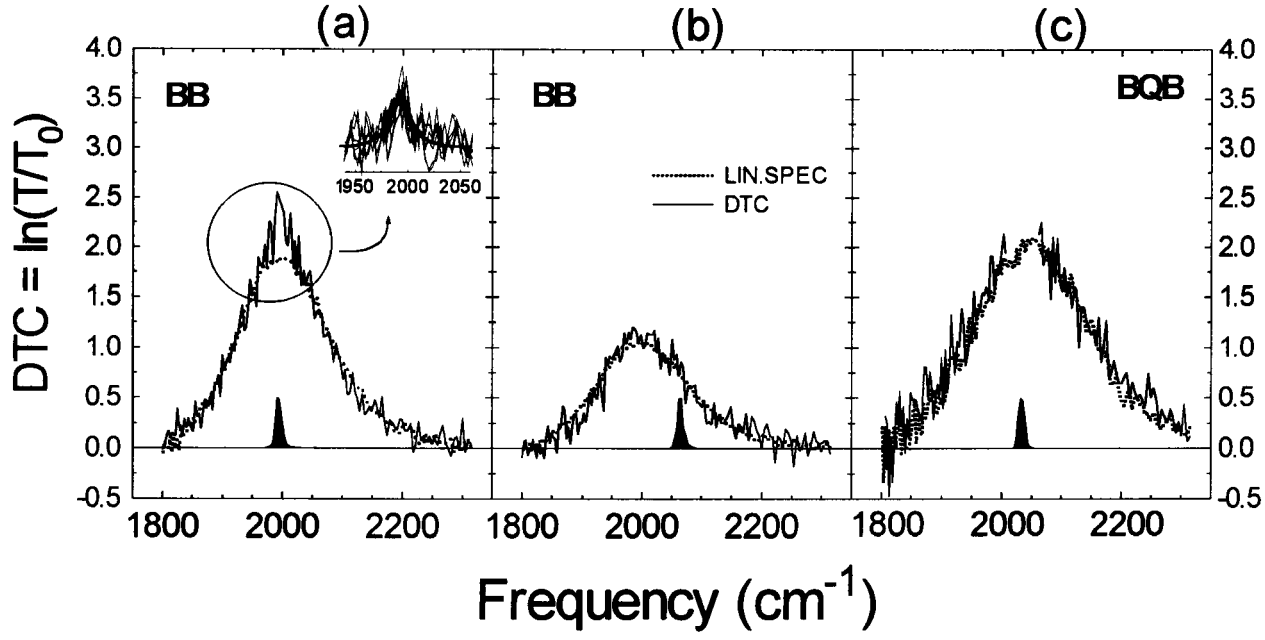

FIG. 3. DTC spectra (solid line) for (a) BB sample, pumped at the intersubband line center; (b) BB sample, pumped in the blue wing of the absorption line; (c) BQB sample, pumped at the intersubband line center. In (c) the DTC spectrum close to the pump frequency is affected by the linear scattering of the pump beam and is omitted. Filled curve show pump beam spectra. Dotted line–linear absorption spectra, scaled to match the DTC amplitude. Inset in (a) shows the ''coherence peak'' occurring near the pump frequency, fitted to a Lorentzian curve.

3) remained independent of the photon energy and was identical to the linear absorption spectrum, whereas the DTC *amplitude* tracked the amplitude of the linear absorption at the pump photon energy. In the presence of a significant inhomogeneous broadening component a ''spectral hole'' of width of order $1/T_2$ would have been seen, ''burnt'' into the absorption line. Its absence unambiguously evidences a dominantly homogeneous 300 K line broadening mechanism allowing the dephasing times to be extracted from the linewidths as $T_2 = h/\pi\Delta E_{12}$, giving 59 fs (BB sample) and 48 fs (BQB sample).

Conduction band nonparabolicity gives different effective masses in the two subbands leading to an inhomogeneous line broadening effect which we here estimate using $m^*(In_{0.5}Ga_{0.5}As) = 0.43m_0$, and the nonparabolicity parameter, $\gamma = 1.13 - 10^{-18}$ m^2 (Ref. 6). In sample No. 4009 with $n_s = 7.6\times 10^{11}$ cm^{-2} we find $\Delta E_{\text{inhom}} = 11.6$ meV (94 cm^{-1}), rather smaller than the measured linewidth (22.1 meV). The inclusion of the depolarization and many body[4,5,12] effects is expected to further reduce the inhomogeneous broadening effect of nonparabolicity and we thus conclude that, along with well-width fluctuations, nonparabolicity broadening plays a negligible role here.

It has been predicted[13] that the electron redistribution between the subbands at optical saturation may produce a many-body shift in the absorption energy which is most pronounced in stepped well profiles with large differences in spatial extents of the subband wave functions. We did not see this here: even though the upper subband was substantially populated the absorption line shifted by less than 1 meV in both samples.

A sharp (24.4 cm^{-1} FWHM) feature centered at the pump frequency was observed in the DTC spectrum of BB sample [Fig. 3(a), inset]. This coherence peak (CP), also called a ''coherence artefact,'' has the same origin as that first observed in time-resolved pump–probe studies of germanium.[14] Interference of the pump and probe beams forms a spatially modulated carrier density distribution which in turn forms a standing-wave index grating, diffracting some of the strong pump beam into the probe direction. In these single-color experiments in germanium, where the population lifetime is much longer than the laser pulse coherence time, the CP has a temporal width determined solely by the laser coherence time.[14] In our experiment though, the pump pulse has a narrow spectral width and its coherence time is larger than the expected MQW population lifetime.

As it was shown theoretically[15] and experimentally,[16] if the pump–probe experiment is carried out in the spectral domain, the CP spectral width in the DTC spectrum can give a valuable insight into the carrier dynamics. If the pump–probe beat frequency is nonzero, an interference between the probe and pump beams produces a traveling-wave grating which diffracts and Doppler shifts some of the pump beam so as to be indistinguishable from the probe. This can only happen as long as the subband population lifetime, τ, is short enough for the subband populations to follow the moving (with the time period, corresponding to the pump–probe beat frequency) interference pattern and gives a Lorentzian CP[15] of spectral FWHM $\Delta\nu_{cp} = (1 + I/I_{\text{sat}})/\pi\tau$, where I is the pump light intensity and I_{sat} is the intersubband saturation intensity. Deconvolving the pump linewidth (10 cm^{-1}) from the experimental CP width [Fig. 3(a)] gives $\Delta\nu_{cp} = 22.3$ cm^{-1} and we find a CP amplitude corresponding to an effective intensity of $\sim 0.3\, I_{\text{sat}}$. On the basis, analysis of the CP gives an estimate of $\tau = 0.6$ ps, in good agreement with the $\tau = 0.4$ ps value we obtained from absorption saturation measurements on the same sample.[11]

In conclusion our experiments unequivocally show that the intersubband absorption line behaves as homogeneously broadened for both bound-to-bound and bound-to-quasibound transitions. At 300 K, the spectral width in our samples, appears to be determined solely by the electron dephasing time. We have also shown that two-color spectroscopy can be successfully used to study femtosecond-scale MQW carrier population dynamics.

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[1] G. Almogy and A. Yariv, J. Nonlinear Opt. Phys. Mater. **4**, 401 (1995).
[2] J. Faist, F. Capasso, D. L. Sivco, C. Sirtori, A. L. Hutchinson, and A. Y. Cho, Science **264**, 553 (1994).
[3] B. F. Levine, J. Appl. Phys. **74**, R1 (1993).
[4] M. Zaluzny, Phys. Rev. B **43**, 4511 (1991).
[5] P. von Allmen, Phys. Rev. B **46**, 13351 (1992).
[6] J. Faist, F. Capasso, C. Sirtori, D. L. Sivco, A. L. Hutchinson, S. N. G. Chu, and A. Y. Cho, Appl. Phys. Lett. **63**, 1354 (1993).
[7] H. C. Chiu and J. S. Harris, Jr., J. Vac. Sci. Technol. B **12**, 1019 (1994).
[8] F. Capasso, C. Sirtori, J. Faist, D. L. Sivco, S. N. G. Chu, and A. Y. Cho, Nature (London) **358**, 565 (1992).
[9] K. L. Vodopyanov, JOSA B **10**, 1723 (1993).
[10] K. L. Vodopyanov and V. G. Voevodin, Opt. Commun. **117**, 277 (1995).
[11] K. L. Vodopyanov, V. Chazapis, C. C. Phillips, B. Sung, and J. S. Harris, Jr., Semicond. Sci. Technol. (to be published).
[12] R. J. Warburton, C. Gauer, A. Wixforth, and J. P. Kotthaus, Phys. Rev. B **53**, 7903 (1996).
[13] D. J. Newson and A. Karobe, Appl. Phys. Lett. **51**, 1670 (1987).
[14] C. V. Shank and D. V. Auston, Phys. Rev. Lett. **34**, 479 (1974).
[15] M. Sargent, P. E. Toschek, and H.-G. Danielmeyer, Appl. Phys. **11**, 55 (1976).
[16] F. Keilman, Appl. Phys. **14**, 29 (1977).