Intersubband Relaxation in GaAs-$Al_xGa_{1-x}As$ Quantum Well Structures Observed Directly by an Infrared Bleaching Technique

A. Seilmeier and H.-J. Hübner
Physik-Department, Technische Universität München, D-8000 München, Federal Republic of Germany

G. Abstreiter
Physik-Department, Technische Universität München, D-8046 Garching, Federal Republic of Germany

and

G. Weimann and W. Schlapp
Forschungsinstitut der Deutschen Bundespost, D-6100 Darmstadt, Federal Republic of Germany
(Received 22 June 1987)

Direct intersubband excitations in narrow, modulation doped GaAs-$Al_xGa_{1-x}As$ quantum well structures are studied by picosecond infrared spectroscopy. The bleaching of the intersubband absorption induced by a high-intensity picosecond pump pulse is studied by a delayed probe pulse. Typical intersubband relaxation times are of the order of 10 ps at 300 K for a subband splitting of about 150 meV. Polar LO-phonon scattering is discussed as the most relevant relaxation mechanism.

PACS numbers: 73.40.Lq, 42.65.Re, 73.60.Br, 78.30.Fs

Semiconductor heterostructures and multi-quantum-well structures are of great interest because of their novel electronic and optical properties. In the GaAs-$Al_xGa_{1-x}As$ system, the lower conduction-band edge of GaAs leads to the formation of two-dimensional subbands in the binary semiconductor. The carrier confinement produces significant changes in the density of states, the electron-phonon interaction, and excitonic effects. The subband structure has been studied mainly by resonant inelastic light scattering techniques.[1] Recently, however, direct infrared intersubband absorption experiments have also been performed,[2-4] where special emphasis was put on fast-infrared-detector applications. Intersubband absorption had been studied earlier by F. Koch and co-workers in various semiconductor surface space-charge layers.[5] On the other hand, little work has been done to investigate the carrier dynamics in systems of reduced dimensionality. Kash, Tsang, and Hvam[6] performed the first time-resolved Raman-scattering experiments to study the optical-phonon emission rates of photoexcited carriers in bulk GaAs. The cooling rates of carriers in quantum well structures have been studied[7-9] with the use of time-resolved photoluminescence. Very recently, Oberli *et al.*[10] used picosecond Raman spectroscopy to get direct information on intersubband relaxation times in wide quantum well structures with energy separations smaller than the energy of an optical phonon. They find cooling rates of the order of several hundred picoseconds. Intersubband relaxation times have also been estimated from transport properties in GaAs-$Al_xGa_{1-x}As$ single heterostructures with high 2D carrier concentrations.[11] Theoretical estimates have also been published.[12-14]

In the present publication we discuss the first direct observations of intersubband relaxation times using an infrared bleaching technique with picosecond time resolution. We use a mode-locked Nd-doped glass laser system (repetition rate of 4 pulses/min) which pumps a traveling-wave infrared dye laser.[15] A second part of the glass laser pulse and the dye laser pulse are mixed in a $AgGaS_2$ crystal. In this way pulses at the difference frequency tunable between 5 and 10 μm are generated.[16] The pulses have an energy of several microjoules, a spectral width of about 10 cm^{-1}, and a pulse duration of 2 ps as determined by a background-free autocorrelation technique. The tunable infrared pulse is divided into a pump and a probe beam which are focused into the sample. A noncollinear geometry with an angle of $\simeq 30°$ between pump and probe beams is used. The change of transmission of the probe beam is studied as a function of delay time.

The samples are grown by molecular-beam epitaxy on (100) semi-insulating GaAs substrates of 350-μm thickness and consist of fifty thin, undoped GaAs layers with individual thicknesses of the order of 50 Å. They are embedded in 400-Å-thick $Al_{0.35}Ga_{0.65}As$ layers, in which the central 100 Å are doped with Si. This leads to a two-dimensional carrier concentration of about $(4 \text{ to } 5) \times 10^{11}$ cm^{-2} per GaAs quantum well as determined from Hall-effect and Shubnikov–de Haas experiments. The total multi-quantum-well structures are cladded in between 0.2-μm-thick $Al_{0.35}Ga_{0.65}As$ layers to avoid surface depletion and substrate effects. The level splitting in the wells depends critically on the well width. Complete confinement of the first-excited subband is only expected for $d_{GaAs} \gtrsim 50$ Å.

It is well known that transitions between subbands exhibit large oscillator strengths. Strong absorption is,

however, only observed for polarization of the infrared light normal to the layers. A special prism geometry is used which leads to a large component of the electric-field vector normal to the layers. Samples with trapezoidal shape of approximately 1-mm length and 10-mm width are prepared. The two long cleavage planes are polished under angles of 20° and 35° as shown in the inset of Fig. 1(a). The multi-quantum-well structure is located at the larger surface of the sample. Infrared absorption and time-resolved experiments are performed with incident light normal to the 20° polished plane. After total internal reflection at the multilayer surface, the beam exits at the back side, oriented under an angle of 35°. The direction of the emerging beam is nearly parallel to the entrance beam. The angle of incidence at the back side is close to the Brewster condition. This reduces the reflection of the excitation pulse back into the sample. Such reflections would result in multiple excitations that especially affect the time-resolved experiments.

First we discuss briefly the infrared absorption of the quantum wells. Spectra of two samples with well thicknesses of 47 and 51 Å are shown in Fig. 1. The well widths are estimated from luminescence measurements. The infrared spectra are obtained with use of a double-beam grating spectrometer. As reference, a GaAs substrate with exactly the same sample geometry as shown in Fig. 1 is used. Consequently, only the absorption due to the heterostructure is shown in Fig. 1. The strong peaks are related to direct intersubband transitions as indicated in the inset.

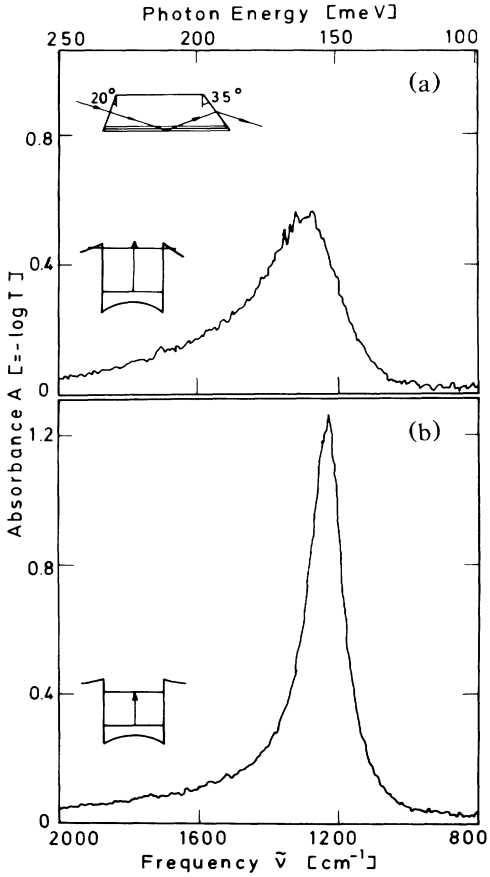

FIG. 1. Infrared absorption of multi-quantum-well samples with well widths of about (a) 47 Å and (b) 51 Å. The strong absorption band is due to intersubband transitions. The sample geometry is shown in the inset of (a).

The absorption maximum of the 51-Å sample is at about 1225 cm^{-1}. The absorbance due to intersubband transitions amounts to $A=1.2$. The 47-Å sample [Fig. 1(a)] exhibits a smaller peak absorbance ($A=0.55$) at 1300 cm^{-1}. The widths of the absorption bands amount to 120 and 270 cm^{-1}, respectively. The width of the 51-Å sample is believed to be partially due to inhomogeneities in the layer thicknesses. The enhanced asymmetry and the further increased linewidth of the 47-Å sample indicate, however, that the energy of the excited subbands is already close to the top of the potential well. So, transitions into continuum states of the conduction band may occur on the high-energy side, leading to a broad tail of the absorption band.

We now estimate the absorption cross section of subband electrons at the absorption maximum. The fifty quantum wells are traversed twice under an angle of 20°. Each well contains 5×10^{11} cm^{-2} electrons. With $A=1.2$ in the 51-Å sample, we calculate an absorption cross section of 10^{-14} cm^2. In other words, approximately 10^{14} photons/cm^2 are necessary to bleach the absorption band. This corresponds to an intensity of $I_{sat}=2$ MW/cm^2 when pulses of 2-ps duration are used.

Picosecond experiments are performed to investigate the carrier depopulation of the excited subband at room temperature. These are the first experiments of this type in the infrared spectral region between 5 and 10 μm. An intense first picosecond pulse, tuned to the frequency of the intersubband absorption maximum, promotes electrons to the upper subband. The pump intensity is high enough ($\simeq5$ MW/cm^2) to assure absorption saturation, i.e., that nearly half the electrons are excited. A second pulse of much lower intensity monitors the recovery of the absorption as a function of delay time. The intensity of the probe pulse has to be well below the saturation intensity I_{sat}, in order to keep bleaching by the probe pulse itself small. Intensities of approximately 200 kW/cm^2 are used.

Time-resolved data are presented in Figs. 2(a) and 2(b) for the 47- and 51-Å sample, respectively. $\ln(T/T_0)$, which is proportional to the population of the upper subband, is plotted as a function of delay time. A rapid rise of the transmission is observed which follows the integral over the autocorrelation of the infrared pulses. The observed increase of the transmission corresponds approximately to complete bleaching of the absorption band. The depopulation of the upper subband proceeds

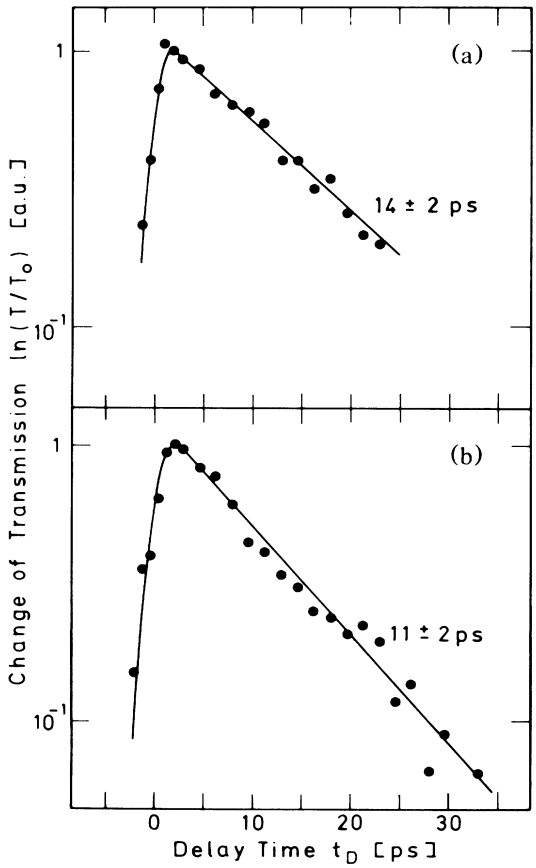

FIG. 2. Bleaching of the probe-pulse intersubband absorption as a function of the delay time for 47- and 51-Å well widths.

with a time constant of 14 ± 2 and 11 ± 2 ps for the 47- and 51-Å quantum wells, respectively. The observed intersubband relaxation time appears to be slightly longer in the thinner quantum wells.

There is considerable interest in elucidating the mechanisms of intersubband relaxation. If one neglects radiative recombination, the excited carriers have to be scattered first with finite $k_{\parallel}$ to the ground subband (τ_{inter}). Then cooling within the lower subband can occur (τ_{intra}). This is shown schematically in Fig. 3. Polar optical-phonon interaction is believed to be the most efficient intraband relaxation mechanism both in bulk GaAs[17] as well as in quantum well structures.[7-9,18] Intraband cooling proceeds on a time scale of 10^{-12} to 10^{-13} s, depending on excitation conditions. The relaxation times of the order of 10 ps, as obtained in the present experiments, therefore probably correspond to intersubband transitions. For subband separations smaller than the LO-phonon energy much longer times have been observed in Raman scattering.[10] From these experiments it was concluded that acoustic-phonon interactions are responsible for the long intersubband time constants.

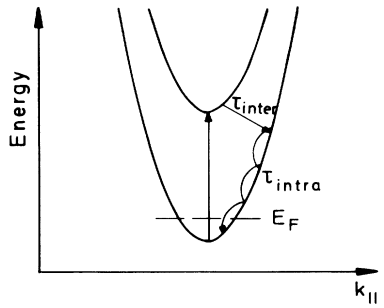

FIG. 3. Absorption and recombination processes between two subbands in $k_{\parallel}$ space. The Fermi energy is indicated by the broken line. The pump pulse excites carriers into the upper subband. Relaxation occurs via intersubband LO-phonon scattering (τ_{inter}) and subsequent intrasubband scattering (τ_{intra}).

Electron-electron scattering is believed to be important only for thermalization within each subband. In addition, because of the presence of band-gap radiations, holes also are generated in the valence bands which complicates the interpretation of light-scattering experiments. The much shorter intersubband relaxation times observed in our infrared experiments are believed to be due to LO-phonon scattering with sufficiently large wave vector. The higher subband separation allows such processes. The measured time constants of about 10 ps at room temperature, with increasing tendency for smaller well width, correspond to LO-phonon intersubband scattering, which requires larger wave vectors with increasing subband splitting. It should be noted that an upper limit for the scattering time of 10 ps was estimated from Raman experiments on narrower quantum wells,[10] in agreement with our results.

Intersubband relaxation has been studied theoretically in the approximation of infinitely deep potential wells.[12] Such theories find intrasubband relaxation to be much faster than intersubband relaxation. This is probably even more pronounced for finite well heights and narrow widths for which the excited subbands are already close to the top of the wells. This leads to a finite probability for the excited carriers to be in the $Al_xGa_{1-x}As$ barriers. Consequently, the overlap integral with the strongly confined wave function in the lower subband is reduced. Therefore, time constants of several picoseconds seem to be quite reasonable.

In conclusion, we have used time-resolved infrared subband spectroscopy to get direct information on intersubband relaxation times. Such investigations are important for an improved understanding of hot-electron effects in quantum well structures and two-dimensional systems.

We thank W. Kaiser and F. Koch for helpful and stimulating discussions.

[1]G. Abstreiter, R. Merlin, and A. Pinczuk, IEEE J. Quan-

tum Electron. **22**, 1771 (1986).

[2]L. C. West and S. J. Eglash, Appl. Phys. Lett. **46**, 1156 (1985).

[3]B. F. Levine, R. J. Malik, J. Walker, K. K. Choi, C. G. Bethea, D. A. Kleinmann, and J. M. Vandenberg, Appl. Phys. Lett. **50**, 273 (1987).

[4]B. F. Levine, K. K. Choi, C. G. Bethea, J. Walker, and R. J. Malik, Appl. Phys. Lett. **50**, 1092 (1987).

[5]For references until 1982, see, for example, T. Ando, A. B. Fowler, and F. Stern, Rev. Mod. Phys. **54**, 437 (1982).

[6]J. A. Kash, J. C. Tsang, and J. M. Hvam, Phys. Rev. Lett. **54**, 2151 (1985).

[7]J. Shah, IEEE J. Quantum Electron. **22**, 1728 (1986), and references therein.

[8]E. Göbel, R. Hoger, and J. Kuhl, in *Proceedings of the MRS-Europe Meeting,* edited by K. Ploog and N. T. Linh (Materials Research Society, Pittsburg, PA, 1986), p. 53.

[9]J. F. Ryan, R. A. Taylor, A. J. Turberfield, A. Maciel, J. M. Worlock, A. C. Gossard, and W. Wiegmann, Phys. Rev. Lett. **53**, 1841 (1984).

[10]D. Y. Oberli, D. R. Wake, M. V. Klein, J. Klem, T. Henderson, and H. Morkoc, Phys. Rev. Lett. **59**, 696 (1987), and Solid State Electron. (to be published).

[11]H. L. Störmer, A. C. Gossard, and W. Wiegmann, Solid State Commun. **41**, 707 (1982).

[12]B. K. Ridley, J. Phys. C **15**, 5899 (1982); F. A. Riddoch and B. K. Ridley, J. Phys. C **16**, 6971 (1983).

[13]P. J. Price, Ann. Phys. (N.Y.) **133**, 217 (1981).

[14]J. P. Leburton, J. Appl. Phys. **56**, 2850 (1984).

[15]T. Elsaesser, H. J. Polland, A. Seilmeier, and W. Kaiser, IEEE J. Quantum Electron. **20**, 191 (1984).

[16]T. Elsaesser, H. Lobentanzer, and A. Seilmeier, Opt. Commun. **52**, 355 (1985).

[17]D. von der Linde and R. Lambrich, Phys. Rev. Lett. **42**, 1090 (1979).

[18]W. H. Knox, R. L. Fork, M. C. Downer, D. A. B. Miller, D. S. Chemla, and C. V. Shank, Phys. Rev. Lett. **45**, 1306 (1985).