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Picosecond infrared spectroscopy of hot carriers in a modulation-doped $Ga_{0.47}In_{0.53}As$ multiple-quantum-well structure

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Intersubband scattering and cooling of hot carriers is investigated in an n-type modulation-doped $Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As$ multiple-quantum-well structure. The nonlinear absorption is monitored in several picosecond pump-and-probe experiments after selective excitation of the electrons to the $n=2$ subband. In addition, the relaxation of hot electron-hole pairs created by interband absorption is studied. An upper limit of 3 ps for the lifetime of the electronic $n=2$ subband is estimated. The cooling is dominated by LO-phonon scattering; the energy loss rate is reduced by a factor of up to 130 compared to the value obtained from theoretical calculations.

The investigation of hot carriers in ternary semiconductors is of fundamental physical interest and of importance for the design of semiconductor devices. In recent years, ternary quantum-well (QW) structures of high quality have been grown by molecular-beam epitaxy.[1-4] The cooling of hot carriers in $Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As$ multiple-quantum-well structures (MQW's) has been studied in picosecond photoluminescence experiments.[5] Intrasubband scattering of hot carriers by polar optical and acoustic deformation-potential interaction was identified as the dominant cooling mechanism. A strong reduction of the energy-loss rate by a factor of up to 100 compared to theoretical calculations of LO-phonon scattering was deduced from the experimental data.

In the present paper, we address the question of intersubband scattering and cooling of electrons and of electron-hole plasmas. Scattering of electrons from the $n=2$ to the $n=1$ subband in $GaAs/Ga_{1-x}Al_xAs$ MQW's has been investigated both experimentally and theoretically.[6-9] Scattering times in the range from 10 up to several hundred ps depending on the QW thickness have been reported. Theoretical calculations for infinitely deep potential wells predict intersubband scattering times in the range of 1 ps for a QW thickness between 5 and 10 nm.[7] In this paper, we report the first picosecond experiments on intersubband scattering in n-type modulation-doped $Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As$ MQW's with a QW thickness of 8.2 nm. A scattering time between 0.5 and 3 ps is deduced from our data. In addition, the nonlinear absorption of the sample after picosecond excitation gives interesting information on the cooling of hot carriers.

The n-type modulation doped MQW structure studied in our experiments was grown by molecular-beam epitaxy.[3] The sample consists of 50 $Ga_{0.47}In_{0.53}As$ quantum wells of a width of 8.2 nm separated by n-doped, 23.4-nm-thick $Al_{0.48}In_{0.52}As$ barriers grown on a (100) InP substrate. The electron density in each QW is 4×10^{11} cm^{-2} (5×10^{17} cm^{-3}). The energy offset of the valence and conduction bands of this system amounts to 0.21 and 0.5 eV, respectively.[4] Three subbands of the electrons exist within the QW's.

Tunable pump and probe pulses in the near infrared are generated by two traveling-wave dye lasers,[10] which are pumped by a modelocked Nd: yttrium aluminum garnet (YAG) laser. The pulse durations vary from 7 ps at 1180 nm (1.05 eV) to 14 ps at 1350 nm (0.92 eV). Tunable pulses in the wavelength range from 4 to 9.5 μm with a duration of 8 ps are generated by difference frequency mixing in the nonlinear crystal $AgGaS_2$.[11] The energy of the excitation pulses in the different experiments ranges from 2 to 20 nJ corresponding to excitation densities between 2×10^{11} and 2×10^{12} cm^{-2} per QW. The energy of the probe pulses is less by two orders of magnitude.

The dipole moment of the subband transition is oriented parallel to the stack axis of the MQW sample.[12] For this reason, the polarization of the excitation pulses is adjusted parallel to the plane of incidence. The semiconductor sample is oriented under Brewster angle in the infrared beam. All measurements are performed at a sample temperature of 10 K.

The absorption spectrum of the MQW structure in the near infrared between 0.85 and 1.15 eV is shown in Fig. 1. This spectrum was measured by a standard infrared spectrometer. Two steps of nearly equal absorbance are observed. They are due to transitions from the $n=1$ and $n=2$ valence bands to the corresponding conduction bands (see arrows 1 and 2 in the inset of Fig. 1). The infrared transition from the $n=1$ to the $n=2$ subband is located at an energy of 0.148 eV (transition 3 in the inset of Fig. 1); the spectral width (full width at half maximum) of the corresponding absorption band amounts to 7 meV.[13] In contrast to $GaAs/Ga_{1-x}Al_xAs$ and $Ga_xIn_{1-x}As/InP$ heterostructures, here the energy of the $n=2$ subband is substantially lower than the confinement energy of the QW's of 0.5 eV.

Four different pump and probe schemes are applied in

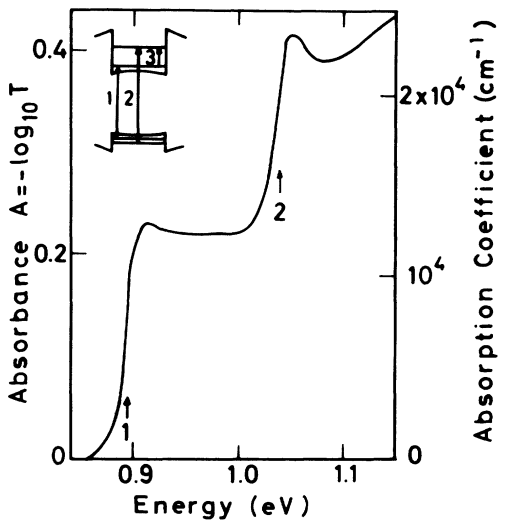

FIG. 1. Interband absorption spectrum of the n-type modulation doped $Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As$ MQW sample. The relevent transitions are marked in the inset.

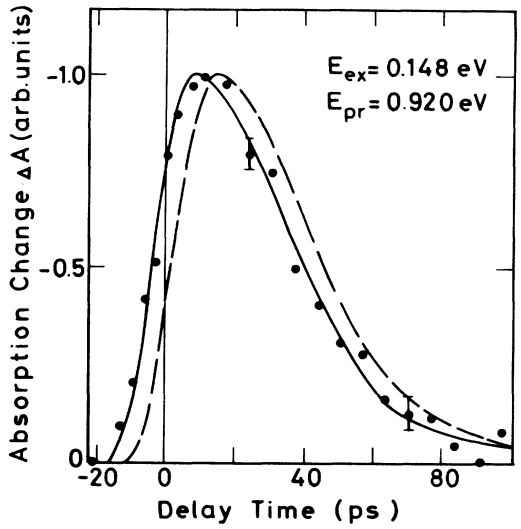

FIG. 2. Picosecond kinetics of the nonlinear absorption change at probe energy $E_{pr}=0.92$ eV after excitation of electrons via the intersubband transition at 0.148 eV (points). The change of absorption is plotted vs the delay time between pump and probe pulses. Solid line: calculated time dependence of the signal for a subband lifetime $\tau_{sub}<3$ ps. Dashed line: calculated signal for $\tau_{sub}=5$ ps.

the picosecond experiments.

(i) Electrons are excited to the $n=2$ subband via the intersubband transition at 0.148 eV (transition 3 in Fig. 1). The energy of the probe pulses has a value of 1.033 eV, i.e., states near the minimum of the $n=2$ subband and high-lying states of the $n=1$ subband are probed (transitions 1 and 2 in Fig. 1). Even for excitation of more than 50% of the electrons by the pulse at 0.148 eV, no absorption change was observed. The expected absorption decrease caused by occupation of the states in the $n=2$ subband could not be detected, i.e., no significant amount of electrons accumulates in the $n=2$ band. This fact indicates a very fast depopulation of the higher subband. Considering the excitation intensity and the sensitivity of our system one estimates a lifetime of the $n=2$ band of less than 3 ps.

(ii) Electrons are again excited to the $n=2$ subband by an infrared pulse at 0.148 eV. The probe pulse at 0.92 eV monitors the change of the absorption between the $n=1$ valence and conduction subbands (transition 1 in Fig. 1). The electronic states monitored by the 0.92-eV pulse are located above the initial Fermi level of the electrons at 0.88 eV, which is defined by the doping concentration.

In Fig. 2, the measured change of absorption $\Delta A=-\ln(T/T_0)$ is plotted as a function of the delay time between pump and probe pulses; T_0 and T represent the transmission of the sample prior to and after excitation, respectively. We find a bleaching of the sample, which rises without any detectable delay in relation to the integrated density of absorbed photons and decays within a time interval of 80 ps. Approximately 50% of the electrons in the sample are excited by the pulse in the medium infrared; the absorption change observed at the maximum of the curve in Fig. 1 has a value of $\Delta A=0.1$.[14]

The very rapid rise of the bleaching shows again that the electrons excited to the $n=2$ subband are scattered back very fast to the $n=1$ band. We estimate a scattering time shorter than the temporal resolution of our experiment of 3 ps. Each excited electron reenters the $n=1$ subband with an excess energy of 0.148 eV. The carriers relax on a subpicosecond time scale to a Fermi distribution with a high temperature. The electronic states at 0.92 eV probed in our experiment are located on the high-energy tail of the distribution function. The population of these levels leads to a decrease of absorption, since the final states of the interband transitions are partly blocked. Cooling of the electron distribution results in the depopulation of the probed levels and a concomittant decay of the signal.

The solid and the dashed lines in Fig. 2 are calculated from a theoretical model. The absorption change ΔA is given by $\Delta A=-\alpha_0(E)ndf_e$, where $\alpha_0(E)$ is the absorption coefficient at the energy E in absence of free carriers; $d=8.2$ nm and $n=50$ are the QW thickness and the number of QW's in our sample, respectively; $f_e=1/\{\exp[(E-E_{Fe})/kT_c]+1\}$ is the Fermi distribution of the electrons, which depends on E, the Fermi energy E_{Fe}, and the time dependent carrier temperature $T_c(t)$. The calculated absorption change $\Delta A(E,t)$ is convoluted with a Gaussian-shaped picosecond probe pulse for comparison with the time dependence of the signal.

The transient electron temperature $T_c(t)$ depends on the energy supplied by the excitation pulse, on the density of excited carriers, on the intersubband scattering time τ_{sub}, and on the energy-loss mechanisms of the hot electrons. The theoretical treatment of carrier cooling includes scattering of hot carriers by polar optical and acoustic deformation potential interaction. The energy loss by LO phonon emission is the dominant cooling mechanism for $T\geq 40$ K;[5] in our model, the energy loss is calculated for a two-dimensional carrier system obeying the Fermi statistics.[15] A phonon energy of 31 meV is used. The energy-loss rate $(dE/dt)_{LO}$ by phonon emission is proportional to $1/\tau_e$, where $\tau_e=0.16$ ps is the electron-

LO-phonon scattering time in GaInAs.[5] Good agreement of the theoretical calculation (solid line) and the data points of Fig. 2 is obtained for $\tau_{sub} < 3$ ps and an energy-loss rate $(dE/dt)_{LO}$, which is reduced by a factor of $F = 30$ compared to the result calculated from the formalism in Ref. 15. The slowing down of the cooling process, which is probably caused by phonon excess population and screening of the electron-LO-phonon interaction in the system has been observed in time-resolved photoluminescence experiments with n-doped $Ga_xIn_{1-x}As/Al_xIn_{1-x}As$ MQW's.[5,16]

The calculated time dependence $T_c(t)$ of the electron temperature is plotted in Fig. 4 (dashed line). $T_c(t)$ rises to 370 K after excitation of the sample and decays to 80 K within 100 ps. The dashed line in Fig. 2 represents the result of the theoretical model for a subband lifetime $\tau_{sub} = 5$ ps. Our data do not agree with the calculated signal, e.g., the rise is delayed in relation to the data points. This finding demonstrates again that the subband lifetime in our sample is considerably shorter than 5 ps.

A comment should be made on the precise determination of the zero point of the time scale in the experiment. Interchanging the intensities of pump and probe pulse, the excitation now at 0.92 eV creates additional electrons in the $n = 1$ subband; the infrared pulse at 0.148 eV monitors the increase of the intersubband absorption caused by the excess electrons. The measured signal follows the convolution of the probe pulse with the temporal development of the electron density. The latter is proportional to the time integral over the excitation pulse, which reaches half of its value at time zero.

(iii) Hot electron-hole pairs in the $n = 1$ valence and conduction subbands are generated by strong interband excitation at 0.968 eV (transition 1 in Fig. 1). In Fig. 3(a), the absorption change measured at 0.954 eV is plotted versus the delay time between the pump pulse and the probe pulse (points); approximately 6×10^{11} electron-hole pairs per cm^2 are created in each QW by the excitation pulse with an excess energy of 0.1 eV. The strong bleaching of the sample decreases within 60 ps to a residual absorption change, which decays via carrier recombination with a time constant of approximately 250 ps [not shown in Fig. 3(a)]. The absolute value of ΔA at the maximum of the curve is approximately 0.3. The dashed line in Fig. 3(a) represents the cross-correlation trace between pump and probe pulses.

The bleaching of the sample is due to band filling by hot carriers. The absorption change ΔA is given by

$$\Delta A(E) = -\alpha_0(E)nd\{f_e[\bar{m}_e(E-E_G),E_{Fe}] + f_h[\bar{m}_h(E-E_G),E_{Fh}]\}$$

in the parabolic band approximation, where α_0 is the absorption coefficient in absence of free carriers and nd is the thickness of the 50 QW's of the sample; $f_{e,h}$ and $E_{Fe,h}$ are the Fermi distributions and quasi-Fermi levels of electrons and holes, respectively (E_G: band gap). Values of $m_e = 0.041m_0$ and $m_h = 0.38m_0$ (m_0, free-electron mass) are taken from the literature[3] to determine the mass factors $\bar{m}_e = m_h/(m_e+m_h)$ and $\bar{m}_h = m_e/(m_e+m_h)$.[17] The time-dependent $\Delta A(t)$ is calculated from the temporal development of the carrier temperature $T_c(t)$, which is assumed to be the same for electrons and holes. The theoretical model for $T_c(t)$ includes carrier cooling by LO phonon and acoustic deformation potential scattering as well as plasma heating by recombination[18] and Auger processes.[19] The detailed calculation will be published elsewhere. The numerical evaluation shows, that for $T \geq 40$ K the energy loss of the plasma by LO phonon emission strongly dominates the relaxation kinetics. The theoretical energy-loss rate $(dE/dt)_{LO}$ is characterized by the electron and hole LO-phonon scattering times $\tau_{e,h}$, which have values of $\tau_e = 0.16$ ps and $\tau_h = 0.09$ ps, respectively.[5]

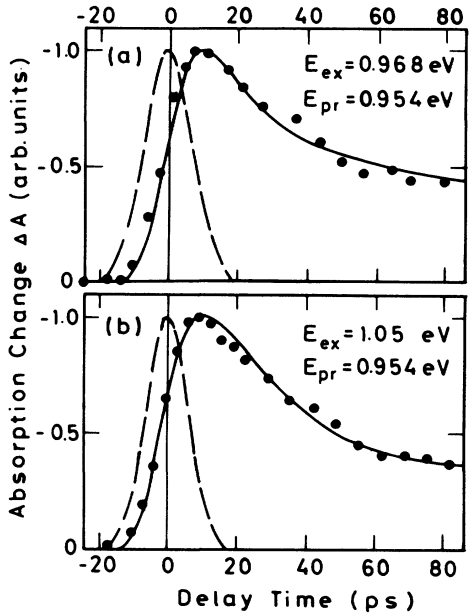

FIG. 3. Measured time dependence of the nonlinear absorption (points) for various excitation (E_{ex}) and probe (E_{pr}) energies. The solid lines are calculated from a theoretical model of carrier cooling. Dashed lines: cross-correlation traces of pumped and probe pulses.

The solid line in Fig. 3(a) represents the calculated absorption change, which matches well the data points; the corresponding time dependence of the carrier temperature is plotted in Fig. 4 (solid line). The latter curve is obtained with a reduction of the theoretical energy-loss rate $(dE/dt)_{LO}$ by a factor of $F = 130$. A similar value of F is estimated from time resolved photoluminesence measurements with the same excitation density of 6×10^{11} cm^{-2}.[16]

(iv) Finally, intersubband scattering of hot electron-hole pairs is investigated with pump pulses at 1.05 eV. The absorption coefficients of the $n = 1$ and $n = 2$ interband transitions 1 and 2 are nearly identical at this energy, i.e., half of the total excited electrons are promoted to the $n = 1$ and to the $n = 2$ subband. The pump pulse generates 6×10^{11} electron-hole pairs per cm^2 in each QW with an excess energy of 0.185 eV. The resulting absorption change monitored at 0.954 eV in the $n = 1$ bands (transition 1 in Fig. 1) is plotted in Fig. 3(b); the maximum signal has a value of $\Delta A = 0.46$.

The normalized curves of Figs. 3(a) and 3(b) agree within the experimental accuracy. This finding gives direct evidence of a very fast intersubband scattering

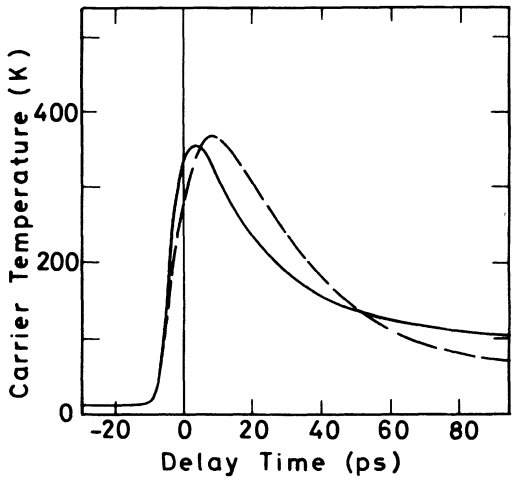

FIG. 4. Temporal development of the carrier temperature as obtained from the theoretical model of carrier cooling. Dashed line: cooling kinetics of the electrons as determined from a fit of the data in Fig. 2. Solid line: kinetics of carrier temperature of a hot electron-hole plasma as obtained from the analysis of the measurement in Fig. 3(a).

time: The electron-hole pairs excited to the $n=2$ subband contribute to the absorption change in the $n=1$ bands without any detectable time delay. A delayed contribution to the signal would occur for a subband lifetime longer than the temporal resolution of our experiment. The kinetics of Fig. 3(b) is reproduced well by our theoretical model of carrier cooling in the $n=1$ bands without taking into account the intersubband scattering process (solid line). The energy-loss rate $(dE/dt)_{LO}$ is reduced by a factor of $F=130$ in the calculation, in agreement with the result of Fig. 3(a). The larger absolute value of the measured absorption change compared to experiment [(iii)] is caused by the higher excess energy of the carriers in (iv), which leads to higher plasma temperatures.

We now return to the discussion of the lifetime τ_{sub} of the electrons in the $n=2$ subband. Our various data suggest an upper limit of $\tau_{sub} < 3$ ps. A lower limit of approximately 500 fs is calculated from the spectral width of the infrared transition between the $n=1$ and $n=2$ subband of the electrons. However, the latter estimate is incomplete since further contributions to the linewidth have to be considered, e.g., the nonparabolicity of the bands and fluctuations of the QW thickness. Theoretical calculations of the intersubband scattering time τ_{sub} based on a model for infinitely deep potential wells have been reported in Ref. 7; τ_{sub} is given by $\tau_{sub}=\tau_e\Gamma$, where τ_e is the electron-phonon scattering time and $\Gamma \gtrsim 1$ is a reduction factor, which depends on the material parameters. We estimate $\tau_{sub}=1.8$ ps for our MQW system, in good agreement with the experimental result.

Recently, intersubband relaxation times in the range of 10 ps have been reported for $GaAs/Ga_{0.65}Al_{0.35}As$ MQW's[9] of a QW thickness of approximately 5 nm. These time constants are considerably longer than the intersubband scattering times calculated in Ref. 7 (with the GaAs parameters) and exceed the values found in our work. This discrepancy may be due to an incomplete confinement of the carriers to the QW's of the $GaAs/Ga_{1-x}Al_xAs$ system; the $n=2$ subband energy is close to the confinement energy for a QW of a thickness of less than 6 nm. The repopulation of the $n=1$ subband should be delayed when the excited carriers are scattered out of the QW's and have to be recaptured for relaxation.

It should be noted that the $Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As$ system fulfills the approximation of infinitely deep QW's considerably better than the $GaAs/Ga_{0.65}Al_{0.35}As$ system because of the larger conduction-band offset. Our extensive data are consistent with the picture of rapid intersubband relaxation and considerably slower cooling of hot carriers.

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