Cooling of an electron-hole plasma in a $Ga_{0.47}In_{0.53}As$ multiple-quantum-well structure

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Time-resolved photoluminescence experiments on a $Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As$ multiplequantum-well structure grown by molecular-beam epitaxy yield a strong dependence of plasma cooling on excitation density. Theoretical fits of the cooling behavior necessitate an enhancement of the polar-optical-phonon scattering time by a factor of 100 when the carrier density is increased from 10^{16} to 10^{18} cm⁻³.

INTRODUCTION

The study of carrier-phonon interactions is of principal interest since they determine basic electrical properties of semiconductors, such as carrier mobility. These interactions govern the cooling process of hot carriers. Therefore, the scattering of carriers by phonons, especially by longitudinal optical (LO) phonons, has been extensively investigated for three-dimensional (3D) and more recently for two-dimensional (2D) semiconductor systems.^{1–7} Excite-and-probe techniques¹ and time-resolved² as well as steady-state^{3,4} photoluminescence methods have been used mostly in the GaAs/Ga_{1-x}Al_xAs system to study the energy relaxation dynamics of the photoexcited carriers. The investigations agree on the following points:

(i) Photoexcited carriers relax within ≤ 10 ps toward an equilibrium distribution, which is described by a carrier temperature T_c and the chemical potentials μ_e and μ_h for electrons and holes, respectively.²⁻⁴

(ii) For $T_c \ge 40$ K, the dominant energy-loss mechanism is the emission of LO phonons via the Fröhlich interaction.³⁻⁵

(iii) At low excitation densities $(n_c \leq 10^{17} \text{ cm}^{-3})$ and for bulk material, the experimentally determined data^{1,5} are in reasonable agreement with the theoretical value for the carrier-LO-phonons scattering rate.

(iv) For high excitation density, a slowing of the cooling rate is observed. 6,7

The mechanism of this slowing down is still questionable. In particular, the influence of dimensionality is not clear. Modulation-doped quantum-well structures exhibit a slowing down even for low-level excitation.⁸

In contrast, only a few experiments have been reported for $Ga_{0.47}In_{0.53}As$, although ternary materials and quantum-well (QW) structures composed of $Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As$ or $Ga_{0.47}In_{0.53}As/InP$ have energy gaps in the spectral range between 1.1 and 1.7 μ m and are therefore interesting for application in optical communication systems.

Recent cw photoluminescence experiments on bulk $Ga_{0.47}In_{0.53}As$ as well as on $Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As$ QW structures revealed carrier phonon scattering times, which are comparable to the theoretical

value.⁹ However, time-resolved experiments on bulk Ga_{0.47}In_{0.53}As could only be explained by assuming a scattering time, which exceeds the theoretical value by more than a factor of $10.^2$

In this paper we present the first measurement to our knowledge of time-resolved luminescence on a $Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As$ multiple-quantum-well (MQW) structure. The cooling of the carriers is studied for excitation densities between 10^{16} and 10^{18} cm⁻³.

Our theoretical analysis of the experimental data takes into account the degeneracy of the electron and hole systems. The results of our study clearly show that the effective carrier-phonon scattering time depends on the excitation density.

EXPERIMENTAL PROCEDURE

The MQW structure for the present study consists of 150 unintentionally doped $Ga_{0.47}In_{0.53}As$ QW's of 3.4 nm width alternating with unintentionally doped 11.4 nm thick $Al_{0.48}In_{0.52}As$ barriers grown by molecular-beam epitaxy (MBE) lattice-matched on a InP(100) substrate. At a temperature of 5 K the band gap of the QW's and of the barriers amounts to 1.00 and 1.54 eV, respectively. Since the QW's are very thin, only transitions between the n = 1 subbands are observed in absorption and photoluminescence, i.e., in our experiment interference by higher subband transitions cannot occur.

We excite the sample with a synchronously pumped mode-locked dye laser with a repetition rate of 80 MHz and a pulse duration of 4 ps. The laser pulses with a photon energy of 1.48 eV are absorbed only in the Ga_{0.47}In_{0.53}As QW's. The luminescence is spectrally dispersed by a 0.25 monochromator and is temporally resolved by a Hamamatsu synchroscan streak camera with an S1 photocathode. The time resolution is limited to approximately 30 ps due to the temporal broadening by the spectrometer and the trigger jitter of the streak camera system. The measurements are performed at a sample temperature of 5 K. The maximum excitation intensity is 20 mW corresponding to an excitation density of 1×10^{18} cm⁻³. The excitation density was attenuated down to 1×10^{16} cm⁻³ with neutral-density filters.

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FIG. 1. Time-integrated photoluminescence spectrum of a Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As MQW structure using $\lambda_{exc} = 840$ nm (1.48 eV) and $n_{exc} = 1 \times 10^{18}$ cm⁻³.

RESULTS AND DISCUSSION

Figure 1 shows the *time-integrated* luminescence spectrum of the Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As MQW structure at the highest excitation density of 1×10^{18} cm⁻³ measured with a cooled high-purity Ge detector. The spectrum peaks at 1.00 eV. This value agrees with the energy gap of the MQW as determined by the measurement of the absorption spectrum. The luminescence is detectable up to 1.2 eV on the high-energy side. In Fig. 2 we show the temporal behavior of the luminescence at different photon energies. The luminescence decay strongly depends on photon energy. At 1.12 eV, the decay is faster than the experimental resolution of our equipment. The decay time increases from 100 to 700 ps if the photon energy is changed from 1.06 to 1.00 eV, respectively.

The *transient* photoluminescence spectra of Fig. 3 are calculated from the time-integrated spectrum and the streaks at the different wavelengths. The spectra are nor-



FIG. 2. Three typical streaks of the photoluminescence at wavelengths of 1.24, 1.17, and 1.10 μ m ($h\nu$ =1.00, 1.06, and 1.12 eV).



FIG. 3. Transient photoluminescence spectra at three different delay times (40, 130, and 500 ps) depicted for the highest excitation density.

malized with respect to the maximum of luminescence.

The delay times with respect to the maximum of the excitation pulse are 40, 130, and 500 ps, respectively. A well-established exponential high-energy tail indicates that the carriers are thermalized and can be characterized by a carrier temperature T_c already 40 ps after the excitation. Neglecting reabsorption of luminescence and effects due to inhomogeneous excitation, we can deduce the carrier temperature T_c from the high-energy tail of the transient spectra. This high-energy tail is proportional to $E^2 e^{-E/kT_c}$ for energies high above the quasi-Fermienergy.

The temporal evolution of carrier temperature T_c is plotted in Fig. 4 for four different excitation densities. Substantial differences in the cooling behavior are observed. The curve obtained at the highest excitation density shows a fast cooling from nearly 500 down to 100 K within 200 ps. Above 200 ps the temperature decreases slightly and even after 1 ns the carrier system has a temperature of nearly 50 K and is obviously not in equilibrium with the lattice.¹⁰ For lower excitation density the in-



FIG. 4. Cooling data at four different excitation densities. The solid lines *a*, *b*, *c*, and *d* are theoretical fits using the parameters: $E_{ac} = 5$ eV, $C_{Auger} = 4 \times 10^{-29}$ cm⁶s⁻¹, and F = 350, 140, 40, and 3 for the curves *a*, *b*, *c*, and *d*, respectively. *F* is a phenomenological elongation factor for both the electron- and hole-optical-phonon scattering time.

itial temperature is also lower and a faster temperature decay is observed.

We now discuss the processes which lead to a cooling of the hot carriers, using an evaluation developed for bulk material i.e., 3D calculations.¹¹ For temperatures above 40 K, carrier–polar-optical-phonon (PO) scattering dominates the energy loss. The average energy-loss rate per electron for a degenerate distribution is given by¹¹

$$\left[\frac{dE}{dt}\right]_{\rm PO} = P_0 x_c^{1/2} \frac{2}{\sqrt{\pi}} \frac{1}{F_{1/2}(\eta)} \left[N_q \int_0^\infty f(\varepsilon) [1 - f(\varepsilon + x_c)] \sinh^{-1} \left[\frac{\varepsilon}{x_c}\right]^{1/2} d\varepsilon - (N_q + 1) \int_0^\infty f(\varepsilon + x_c) [1 - f(\varepsilon)] \sinh^{-1} \left[\frac{\varepsilon}{x_c}\right]^{1/2} d\varepsilon \right]$$
(1)

where $P_0 = \hbar \omega_{\rm LO}/\tau_0$, $\hbar \omega_{\rm LO}$ is the energy of the longitudinal optical phonon, $\tau_0 = 0.16$ ps is the electron-LOphonon scattering time, $x_c = \hbar \omega_{\rm LO}/kT_c$, $N_q = 1/(e^{x_0} - 1)$ is the occupation probability for LO phonons with $x_0 = \hbar \omega_{\rm LO}/kT_L$ where T_L is the lattice temperature, $f(\varepsilon) = 1/[1 + \exp(\varepsilon - \eta)]$ is the occupation probability for electrons, with $\varepsilon = E/kT_c$ and $\eta = E_F/kT_c$,

$$F_j(\eta) = \frac{1}{\Gamma(j+1)} \int_0^\infty \frac{\varepsilon^j d\varepsilon}{1 + \exp(\varepsilon - \eta)}$$

is the Fermi integral of index j.¹¹ Deformation potential scattering is the dominant energy loss mechanism in our specimen for temperatures below 40 K. Piezoelectric scattering must be only considered for $T_c \leq 20$ K and can therefore be neglected for our calculations, because we are interested in the temperature range beginning from 20 K. The energy loss by deformation potential (DP) scattering is given by¹¹

$$\left[\frac{dE}{dt}\right]_{\rm DP} = \frac{-8\sqrt{2}}{\pi^{3/2}} \frac{(m^*)^{5/2} E_1^2}{\rho \hbar^4} (kT_c)^{3/2} \\ \times \left[1 - \frac{T_L}{T_c}\right] \frac{F_1(\eta)}{F_{1/2}(\eta)} , \qquad (2)$$

where $E_1 = 5.9$ eV is the acoustic deformation potential for electrons, $m^* = 0.041m_0$ is the effective mass of electrons, and $\rho = 5.59$ g/cm³ is the density of mass of Ga_{0.47}In_{0.53}As.¹²

The LO-phonon energy-loss rate for holes is obtained using the effective mass of heavy holes in Ga_{0.47}In_{0.53}As $m_{\rm hh}=0.5m_0$. The rate must additionally be reduced by a factor of K=2 since the theory of LO-phonon scattering is calculated with *s*-like wave functions, whereas holes are described by wave functions with *p* symmetry having a smaller overlap, which must be compensated.¹³ The deformation potential scattering rate is obtained using the effective mass of heavy holes and a mean acoustic deformation potential $E_{\rm ac}$ for holes.¹³ The deformation potential scattering of holes far exceeds that by electrons because of the large difference in effective masses.

We include two additional mechanisms which lead to carrier heating, namely band-band Auger effect¹⁴ and recombination heating.¹⁵ Recombination heating is important since the carrier lifetime amounts to 800 ps (nearly independent of excitation density) and is comparable to the cooling times.

The unknown parameters $E_{\rm ac}$, τ_0 , and the Auger coefficient in QW's are determined as follows: For low excitation density and temperatures below 40 K, deformation potential scattering of holes is the dominant cooling process. Therefore the cooling curve obtained at very low excitation density yields the deformation potential E_{ac} . At high excitation densities our data are best fitted with a value for the Auger coefficient of 4×10^{-29} cm⁶ s⁻¹, which is close to the value for comparable ternary and quaternary materials.^{16,17} A good overall fit to the cooling curves at higher excitation densities is only obtained by a variation of the electron-phonon scattering time τ_e and hole-phonon scattering time τ_h . For τ_e and τ_h , we use $\tau_e = \tau_{e0}F$ and $\tau_h = \tau_{h0}F$, where τ_{e0} and τ_{h0} are the theoretical values of the electron-phonon respective holephonon scattering time, which amounts to 0.16 and 0.09 ps (with $\varepsilon_0 = 13.73$ and $\varepsilon_{\infty} = 11.37$).¹² The factor F phenomenologically describes the observed slowing of carrier cooling at higher excitation densities. The theoretical fits for the four different excitation densities are included in Fig. 4. From curve d we obtain $E_{ac} = 5$ eV and $F = 3 \pm 2$. The large uncertainty of F at this low excitation density of 8.75×10^{15} cm⁻³ is due to the insensitivity to the parameter F. Higher carrier density, however, cause a strong increase of F: to 40, 140, and 350 for carrier densities of 7×10^{16} cm⁻³, 2.6×10^{17} cm⁻³, and 1×10^{18} cm⁻³, respectively.

Two effects may modify the carrier-LO-phonon scattering rate and therefore the cooling curve for the higher carrier densities. First, screening might weaken the strength of the carrier-phonon coupling. Second, a nonequilibrium LO-phonon population due to the high generation rate of LO phonons and their slow decay (i.e., a LO-phonon bottleneck) might reduce the cooling, i.e., introduce an apparent reduction of the polar-opticalphonon scattering rate. Further investigations are necessary to distinguish between the two effects.

CONCLUSIONS

We have measured the cooling of an electron-hole plasma in a $Ga_{0.47}In_{0.53}As/Al_{0.48}In_{0.52}As$ MQW as a function of excitation density. The cooling is found to be extreme-

ly sensitive to the excitation density. A theoretical model has been used to describe the cooling, taking into account polar-optical-phonon scattering, deformation potential scattering, recombination heating, and auger recombination. We derive effective carrier-phonon scattering times from the good agreement between our theoretical description and measured data. At low excitation densities the observed carrier-phonon scattering time is small and close to the theoretical expectation. For the highest excitation density an apparent reduction of the carrier-LO-phonon

¹D. von der Linde and R. Lambrich, Phys. Rev. Lett. **42**, 1090 (1979).

scattering time by a factor of 350 is found. Finally, we

emphasize the importance of using Fermi-Dirac statistics

for interpretation of the data. The cooling is strongly

- ²K. Kash, J. Shah, D. Block, A. C. Gossard, and W. Wiegmann, Physica 134B, 189 (1985).
- ³J. Shah, Solid-State Electron. 21, 43 (1978).
- ⁴S. A. Lyon, J. Lumin. 35, 121 (1986).
- ⁵C. H. Yang, J. M. Carlson-Swindle, S. A. Lyon, and J. M. Worlock, Phys. Rev. Lett. **55**, 2359 (1985).
- ⁶R. J. Seymour, M. R. Junnarker, and R. R. Alfano, Solid State Commun. **41**, 657 (1982).
- ⁷J. Shah, A. Pinczuk, A. C. Gossard, and W. Wiegmann, Phys. Rev. Lett. **54**, 2045 (1985).
- ⁸J. F. Ryan, R. A. Taylor, A. J. Turberfield, A. Maciel, J. M. Worlock, A. C. Gossard, and W. Wiegmann, Phys. Rev. Lett. 53, 1841 (1985).
- ⁹H. Lobentanzer, W. W. Rühle, W. Stolz, and K. Ploog, Solid State Commun. (to be published).

dependent on degeneracy, and recombination heating cannot be neglected, because recombination lifetimes are comparable to cooling times.

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- ¹⁰For later delay times of 1.6 ns we find carrier temperature ≤ 20 K. This observation gives evidence that lattice heating is not important in our experiment.
- ¹¹J. Shah and R. F. Leheny, Semiconductor Probed by Ultrafast Laser Spectroscopy, edited by R. R. Alfano (Academic, Orlando, 1984), Vol. I, p. 45.
- ¹²T. P. Pearsall, *GaInAsP Alloy Semiconductors* (Wiley, New York, 1982).
- ¹³E. O. Göbel and O. Hildebrand, Phys. Status Solidi 88, 645 (1978).
- ¹⁴A. Haug, Appl. Phys. Lett. 42, 512 (1983).
- ¹⁵D. Bimberg and J. Mycielski, Phys. Rev. B 31, 5490 (1985).
- ¹⁶M. E. Prise, M. R. Taghizadeh, B. S. Wherrett, and S. D. Smith, J. Lumin. **30**, 369 (1985).
- ¹⁷B. Sermage, H. J. Eichler, J. D. Heritage, R. J. Nelson, and N. K. Dutta, Appl. Phys. Lett. **42**, 259 (1983).