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Inter- and intra-subband LO phonon emission rates in GaAs/AlGaAs quantum disks

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Abstract

The inter- and intra-subband relaxation processes of electrons in a GaAs/AlGaAs quantum disk are investigated via femtosecond reflectance change. It is found that the time constants for quantum disks depend on the shape and size of the disk. A possible origin of the modification of the scattering cross-section in a disk of mesoscopic size is discussed. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Inter- and intra-subband relaxation; Quantum disks; Scattering cross-section

1. Introduction

The energy relaxation phenomenon of non-equilibrium carriers in quantum dots/disks is one of the key issues to promote the performance of electro-optical devices. The energy relaxation process in a quantum dot/disk is different from that in a bulk material due to the change of the phonon emission rate in low-dimensional structures [1]. The phonon emission rate may be modified by other scattering processes to randomize the momentum such as carrier–carrier scattering and ionized impurity scattering (Rutherford scattering). If the size of the device is comparable with the mean free path of carries, the two-dimensional electron gas is confined within an arbitrary geometrical structure and the in-plane motion of carriers in the disk is mostly ballistic. Therefore, the carriers in the disk will experience reflection (scattering) on the potential wall at the edge of the disk and the trajectory of the ballistic motion of an electron is modified by the reflection at the boundary of the disk. Since the Coulomb scattering is of long-range nature, it is a strong function of the trajectory and might be modified by the presence of a convex/concave potential wall near the scattering center. That is, the scattering rate might be a function of the geometry of the sample. Recent studies on a double quantum well reveal that the tunneling time is a strong function of the shape and the size of the sample [2,3]. In this paper, we will present that the inter- and intra-subband energy relaxation rates are much reduced in a GaAs/AlGaAs quantum disk, and depend on the geometry. The possible origin is discussed in relation to the modification of the scattering cross-section in the disk.

The reflectance change in photo-excited semiconductors is related to the change of the

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occupation density in the conduction band [4]. Thus, time-resolved photoreflectance spectra can probe directly the energy relaxation process of photo-excited carriers. In this paper, studies of these processes have been carried out with the time-resolved photoreflectance (PR) spectroscopy.

2. Sample structure and experimental method

A 13 nm-wide GaAs single quantum well embedded between $Al_{0.3}Ga_{0.7}As$ cladding layers was grown by MBE on a semi-insulating (0 0 1) GaAs substrate. The top AlGaAs and GaAs cap layers are of n-type doped to supply electrons into the quantum wells. The carrier density in the quantum well (QW) is $6 \times 10^{11}\,cm^{-2}$ and the mean free path is estimated to be $0.3 \sim 0.5\,\mu m$ at 77 K.

Assuming rectangular potential wells, the electron subband separation between the first two confined subbands was estimated to be 61 meV. Thus, the inter-subband relaxation of electrons from the upper subband to the lower subband in the QW is via the emission of LO phonons. Using electron beam lithography and wet chemical etching, array of circular or square disks with a width of 200–400 nm was made to confine carriers within the disks. All the sizes are comparable with the carrier mean free path of 300–500 nm at 77 K.

The reflectance change at 77 K was measured as a function of time and energy using a mode-locked Ti-sapphire laser (the peak wavelength 785 nm, the pulse width 20 fs, the auto correlation width at the sample surface 100 fs). The pulse train was split into an intense pump and a weak probe pulse beam, and they were linearly polarized perpendicular to each other so that we could neglect the coherent artifact. The electrons excited in the quantum wells by the pump pulse have a two-dimensional kinetic energy of 0–120 meV, and the decay processes of the electrons were studied by measuring the reflectance change of the probe pulse as a function of the delay time and the wavelength (energy) [3,4]. The nominal electron density excited was $< 10^{11}\,cm^{-2}$, where the decay time is rather insensitive to the excitation intensity [5,6]. As for the tunneling processes in an asymmetric double quantum well, the results have been published elsewhere [3], so in this report we will focus the discussion on the inter- and intra-subband relaxation process in a single QW.

3. Inter- and intra-subband LO phonon emission time

Fig. 1 shows photoluminescence (PL) and photoreflectance (PR) spectra of the as-grown 2 DEG sample at 77 K. The PL spectrum exhibits two peaks, a peak at 1.53 eV is from the quantum well and that at 1.52 eV is from the bulk GaAs. The PR spectrum shows that the optical absorption/emission between the second subbands in the conduction band and valence band ($(e, hh)_2$) is at about 1.61 eV.

Fig. 2 shows typical traces of the reflectance change, $\Delta R/R$, in various samples as a function of the delay time measured at 1.615 eV. Since the difference energy between the first two confined subbands is larger than the LO phonon energy, the inter-subband relaxation emitting LO phonons predominates the process. Figs. 2(a) and (b) show the reflectance changes for square disks and for the circle disks, respectively. In order to neglect the excitation intensity dependence, the measurements were carried out under the low excitation intensities.

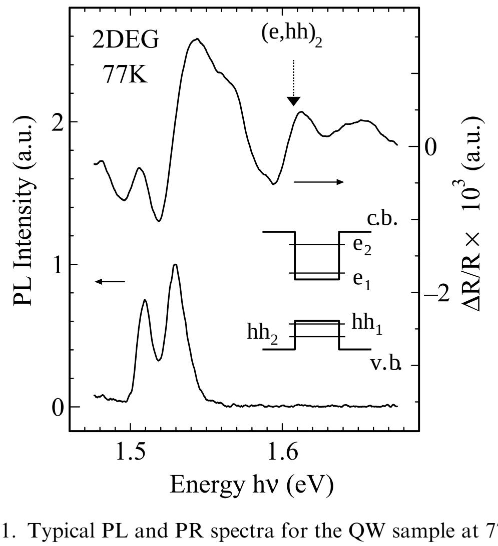

Fig. 1. Typical PL and PR spectra for the QW sample at 77 K. The inset shows the band diagram of the QW.

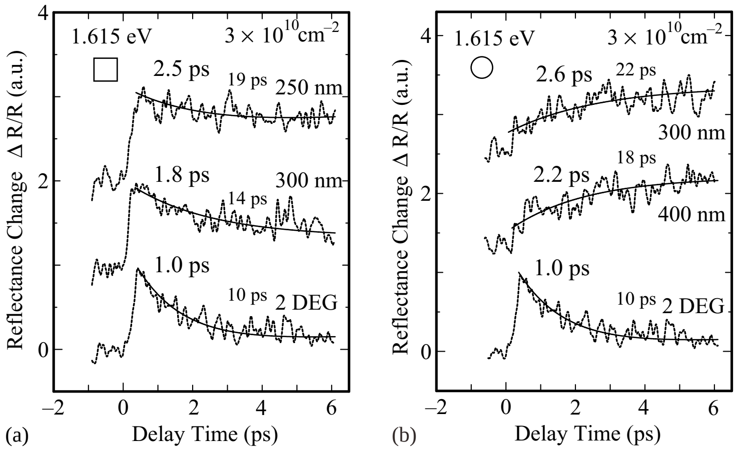

Fig. 2. Typical reflectance change $\Delta R/R$ in a square disk (a) and in a circular disk (b) as a function of the delay time measured at $h\nu = 1.615\,\mathrm{eV}$.

The decay process of carrier density at a level can be described by two time constants, τ_1: a decay (escape out) time into lower levels, and τ_2: an accumulation (in-coming) time from upper levels. The observed decay curves can be fitted with the two-time-constant response function. In an as-grown 2 DEG sample, we found a fast decay time (τ_1) on the order of 1.0 ps and the second time constant (τ_2) was on the order of 10 ps. The decay time constant of 1.0 ps obtained for the 2 DEG is on the same order of the inter-subband LO phonon emission time [6,7] and the second time constant represents the energy relaxation time from higher energy states within the well. Similar measurements/fitting procedures were performed for various disk samples. The time constant τ_1 (τ_2) was 1.8 ps (14 ps) for a square disk 300 nm wide and 2.5 ps (19 ps) for a sample 250 nm wide. In a circular disk, it was 2.2 ps (18 ps) for a sample 400 nm wide and 2.6 ps (22 ps) for 300 nm wide. It is notable that the time constants for the circular disk are longer than those for the square disks and the 2 DEG sample. Moreover, the smaller the size of the disk is, the longer the decay time constant is. These results show that the emission of LO phonon is suppressed in disks and depends on the shape and size.

In Fig. 3, the reflectance changes in the circular disks at 1.580 eV are displayed. The first subbands in the conduction band and valence band $((\mathrm{e},\mathrm{hh})_1)$ are at about 1.530 eV from the PL spectrum. Therefore, the characteristic time obtained at 1.580 eV should represent the intra-subband LO phonon emission time in the quantum well. Figs. 3 (a) and (b) show the reflectance changes for different excitation intensities. When the excitation intensity is $6.0 \times 10^{10}\,\mathrm{cm}^{-2}$ (Fig. 3(a)), the fast decay time τ_1 of 0.3 ps obtained for the as-grown 2 DEG sample was on the same order of the intra-subband LO phonon emission time in a bulk GaAs [8], and the second time constant τ_2 was 7 ps. It was found that the τ_2 at 1.580 eV was shorter than that at 1.615 eV. This reflects the fact that near the bottom of the subband the relaxation process emitting acoustic phonons should take part to enhance the relaxation time. In circular disks, the time constant τ_1 (τ_2) is 0.5 ps (10 ps) for a sample 400 nm wide, while it is 0.8 ps (15 ps) for 300 nm wide and 1.0 ps (18 ps) for 200 nm. For $3.0 \times 10^{10}\,\mathrm{cm}^{-2}$ (Fig. 3(b)), on the other hand, τ_1 (τ_2) was 0.8 ps (6 ps) for a sample 400 nm wide, 1.0 ps (8 ps) for 300 nm wide and 1.2 ps (11 ps) for 200 nm. From Figs. 3(a) and (b) the decay time constants for the circular disk are longer than those for the 2 DEG, and the smaller the size of the disk, the longer the decay time constant is. It means that the emission of LO phonon is suppressed. Therefore, we conclude that the intra-subband LO phonon emission time depends on the size of the quantum disk.

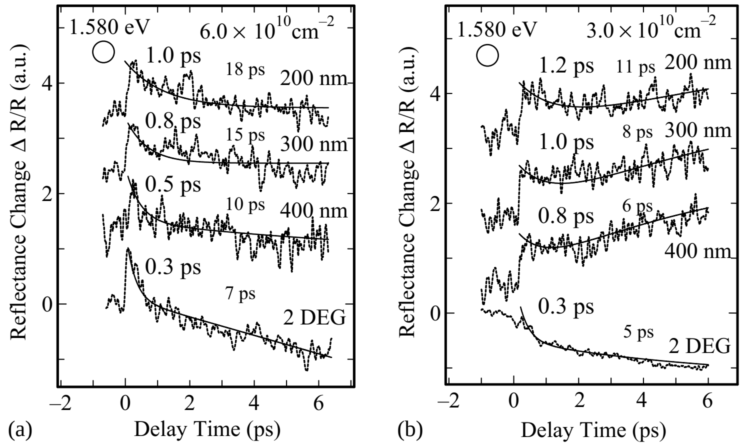

Fig. 3. Reflectance change $\Delta R/R$ in the circular disks as a function of the delay time measured at $h\nu = 1.580\,\text{eV}$ ((a) $6.0 \times 10^{10}\,\text{cm}^{-2}$ (b) $3.0 \times 10^{10}\,\text{cm}^{-2}$).

In Fig. 2(a) and (b), the sign of the reflectance change is inverted. As already shown in Fig. 1, the PR signal intensity is sensitive to the photon energy (wave length) near the special energy. The sign of the reflectance change is thus also the function of the wave length as well. Moreover, in a quantum well sample, the sign depends on the thickness of the top AlGaAs layer. Nevertheless, the sign of the bleaching signal due to the photo-excited carrier is same for each sample. Therefore, the sign of the reflectance signal was inverted in accordance with the PR spectrum. Actually, in the present 2 DEG sample we have observed similar inversion by reducing the thickness of the cap layer by wet chemical etching. We found that in spite of the change, the decay time constants are same at low excitation intensities. Therefore, we might neglect the inversion of sign when the decay process is the issue, and we could determine the time constants assuming exponential decay curves.

As for the origin of the suppression of LO phonon emission rate in semiconductors, various mechanisms have been studied. In the present case, the planar size is so large that we might neglect the quantum size effect in the $x-y$ plane. The blue shift of the PL peak was not so significant. Thus, the suppression of LO phonon emission is not attributed to the formation of discrete quantum levels. The screening and the hot phonons have often been found to predominate the similar phenomena [9]. In both cases, the suppression is a strong function of the carrier density. We carried out the measurements under the low excitation intensity and the dependence of the decay process on the excitation intensity was rather weak. We should think of another mechanism. One possibility is the strong modification of the carrier–carrier scattering and Coulomb scattering in a disk. Since the carrier–carrier scattering and Coulomb scattering (Rutherford scattering) randomize the momentum of electrons/holes, the phonon emission rate might be modified by these scattering processes. In Figs. 2 and 3, modification depended on the geometry and size of the samples, and we might take into account the shape of the potential wall at the boundary of the disk.

On the basis of semi-classical model for the Rutherford scattering, the scattering cross-section was investigated with Monte Carlo method. We simulated the trajectory of an electron in the square and circular disk using the billiard model and estimated the differential scattering cross-section σ for each collision after the reflection at the potential wall. We investigated how the scattering cross-section is modified by the geometry as well as the size.

We found that the average value of σ for more than 500 collisions is a strong function of the geometry and size. It was shown that the average value of σ in a circular disk, of which diameter is

200 nm, is about a half of that in a square disk of 200 nm long side, and the average value of σ in a square disk with 400 nm long side is twice of that in a sample with 200 nm long side. The average value of σ is reduced in a circular than those in square disks. The results are in agreement with the experimental observation.

4. Summary

The inter- and intra-subband LO phonon emission processes in GaAs/AlGaAs quantum disks have been studied using the time-resolved PR spectroscopy. It was found that the LO phonon emission times for the disks are longer than those for the as-grown 2D sample and depend on the planar shape and size. We conclude that the origin of the suppression of LO phonon emission rate is due to the variation of the scattering cross-section near the potential wall with mesoscopic curvature. The Rutherford scattering cross-section in the disk was investigated numerically and the result was in agreement with the experiment result.

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