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Physica E 24 (2004) 272–277
PHYSICA E
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Temperature-induced broadening of the emission lines from a quantum-dot nanostructure

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Received 19 February 2004; accepted 6 May 2004

Available online 10 July 2004

Abstract

We report on the effect of temperature fluctuations on the midinfrared electroluminescence from a cascade of coupled AlInAs quantum dots and GaAs quantum wells. The observed line width is significantly broadened with increasing temperature. We then present our theoretical results on homogeneous line broadening due to temperature fluctuations for our experimental system. Our numerical simulations clearly indicate that, temperature fluctuations can account for the observed finite width of the emission lines at high-temperatures.

Keywords: Quantum dots; Intersub-band transitions; Quantum wells

There has been an upsurge of interest in nanostructured systems with reduced dimensionality. A prominent example is the quantum dot (QD) interband laser with its performance now being comparable to that of quantum well (QW) lasers [1]. One other important system that is our main focus here is the QD cascade structures [2–4] with intersublevel transitions. Earlier theoretical work on the effect of temperature fluctuations on the spectral properties of individual semiconductor QDs [5] indicated that homogeneous broadening of the line width can be attributed to temperature fluctuations which reduces the optical gain at higher temperatures. Temperature dependence of the homogeneous line width of the ground state exciton in InGaAs/GaAs self-assembled quantum dots has also been investigated [6] in single dot photoluminiscence spectroscopy. In this paper, we report on our experimental observation of broadening of the emission spectra of a QD–QW cascade structure. We also report on our theoretical work for the temperature dependence of the homogeneous line broadening, following the prescription of Ref. [5], that explains the temperature dependence of the observed spectra.

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1386-9477/$ - see front matter
doi:10.1016/j.physe.2004.05.003

The QD nanostructure consists of coupled AlInAs self-assembled QDs and GaAs QWs with optical transitions between the electronic sublevels of the AlInAs quantum dots and the subbands of the GaAs quantum wells. The layer design and the electronic band structure are described in Ref. [2]. The sample for the present electroluminescence work described below contains a stack of 10 layers of $Al_{0.5}In_{0.5}As$ QDs each grown on top of a 54.4 nm thick superlattice with GaAs QWs and $Al_{0.45}Ga_{0.55}As$ barriers. Each QD layer is capped by a 6.9 nm thick tunneling barrier to inject electrons from the superlattice into the QD sublevels while preventing electron leakage into the two-dimensional wetting layer. The QDs are formed during the deposition of 11.5 monolayers of highly strained $Al_{0.5}In_{0.5}As$ on $Al_{0.45}Ga_{0.55}As$ at a growth rate of 0.4 A/s and a temperature of 490°C, using a Gen II Applied Epi molecular beam epitaxy system. The growth sequence on a n-type semi-insulating GaAs substrate starts with an 800 nm thick n^+-GaAs contact layer and a 200 nm thick n^--GaAs buffer layer grown at 630°C followed by the QD–QW stack grown at 490°C and is terminated with a 500 nm n^--GaAs buffer layer and a 200 nm n^+-GaAs contact layer. The samples are processed into $120 \times 120\ \mu m^2$ mesa with Ti/Pt/Au top and bottom metallization and mounted in a variable-temperature He-flow cryostat.

Electroluminescence measurements were performed using a Fourier transform infrared spectrometer with step scan and lock-in detection techniques. The samples are pumped at a pulse repetition frequency of 93 kHz and a pulse width of 4 μs. The emitted light is detected with a liquid nitrogen-cooled HgCdTe detector with a cutoff at 100 meV. The emission spectra obtained from two adjacent mesas at 15 mA pumping current and at various heat sink temperatures are displayed in Fig. 1. The peaks at 158 meV are attributed to transitions from the discrete electronic states of a resonant subensemble of QDs [2]. The line width is clearly broadened from 10 meV at 70 K to 14 meV at 200 K and 16 meV at 240 K.

In order to understand the observed broadening of the emission peaks in Fig. 1, we have performed a detailed theoretical study as described below.

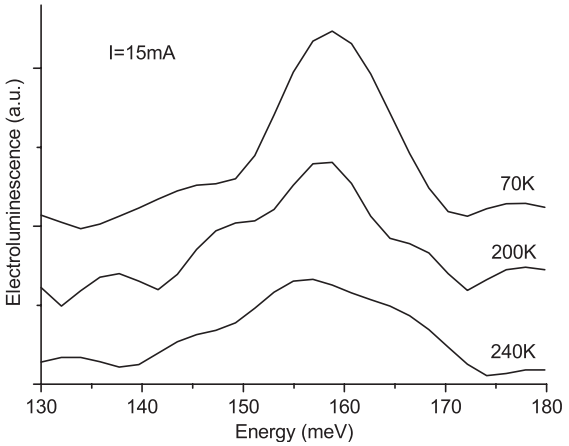

Fig. 1. Electroluminescence spectra at different heat sink temperatures from 10 periods of the coupled QD–QW cascade structure. The QD layers contain AlInAs QDs in the AlGaAs/GaAs material system.

Assuming that the homogeneous line broadening is due to temperature fluctuations, we can follow the procedure outlined in Ref. [5] to investigate the temperature dependence of the broadening. Such temperature fluctuations are standard temperature fluctuations of thermodynamically small systems, such as a QD that is in contact with a thermal reservoir, e.g. phonons. It should be noted however that there are three main differences between our system and the system considered in Ref. [5]: (i) In our case, the optical transitions are *intraband intersubband transitions*, i.e. the electrons are in the conduction band and the optical transition corresponds to electron transition from one subband to another. In Ref. [5], only interband transitions, i.e. transitions between conduction and valence bands are considered. (ii) The QD in Ref. [5] is spherically symmetric, while in our case, the QD has almost cylindrical symmetry. More precisely, our QD system has a large in-plane radius and a small height. This geometry of the QD allows us to consider only in-plane temperature fluctuations and disregard the temperature fluctuations in the direction perpendicular to the plane. (iii) The electron in Ref. [5] is in the QD before and after the transition, i.e. the transition is of the dot-to-dot type. In our case, after the transition the electron is in the QW, i.e. the transition is of the dot-to-well type.

Following the procedure described in Ref. [5], we present the time-dependent electron wave function in the initial state in the form

$$\Psi_{\mathrm{i}}(t,r) = \exp\left[-\frac{\mathrm{i}}{\hbar}\left(E_{\mathrm{i}}t + \int_0^t \Delta E_{\mathrm{i}}(t_1)\,\mathrm{d}t_1\right)\right]\psi_i(r), \quad (1)$$

where ΔE_{i} is the fluctuation of the electron energy due to temperature fluctuations. The temperature fluctuations $\Delta T(t,r)$ result in the fluctuations of the conduction band edge energy E_{c} from which fluctuations of the electron energy can be derived according to the equation

$$\Delta E_{\mathrm{i}}(t) = \frac{\partial E_{\mathrm{c}}}{\partial T}\int \mathrm{d}r \Delta T(t,r)|\psi_i(r)|^2.$$

The time-independent part of the electron wave function $\psi(r)$ can be written as a product of $\chi_2(z)$ and $R(\rho)$, $\rho = (x,y)$

$$\psi_i(r) = \chi_2(z)R(\rho),$$

where $\chi_2(z)$ corresponds to the second subband of the size quantization (due to the confinement potential in the z-direction) and $R(\rho)$ is the electron wave function of the ground state of a QD. In what follows, we assume that the confinement potential of the QD has the parabolic form, i.e.

$$R(\rho) = \frac{1}{\pi l^2}\exp\left(-\frac{\rho^2}{2l^2}\right),$$

where l is the oscillator strength.

Since after the transition, the electron is in the QW, i.e. there is no in-plane confinement potential, the final state of the electron is characterized by the in-plane wavevector, k. Then, similar to Eq. (1), the time-dependent electron wave function in the final state can be written as

$$\Psi_k(t,r) = \exp\left[-\frac{\mathrm{i}}{\hbar}\left(E_k t + \int_0^t \Delta E_k(t_1)\,\mathrm{d}t_1\right)\right]\psi_k(r), \quad (2)$$

where

$$\Delta E_k(t) = \frac{\partial E_{\mathrm{c}}}{\partial T}\int \mathrm{d}r \Delta T(t,r)|\psi_k(r)|^2 = \frac{\partial E_{\mathrm{c}}}{\partial T}\int \mathrm{d}r \Delta T(t,r)|\chi_1(z)|^2, \quad (3)$$

$$\psi_k(r) = \chi_1(z)\frac{\exp(\mathrm{i}k\rho)}{S^{1/2}}.$$

Here, S is the area occupied by the QW and $\chi_1(z)$ corresponds to the first subband of the size quantization. Note that the energy fluctuations $\Delta E_k(t)$ do not depend on k [Eq. (3)].

Using the time-dependent perturbation theory, we calculate the transition probability for the transition from initial state $\Psi_{\mathrm{i}}(t,r)$ to all final states $\Psi_k(t,r)$

$$P(t) = C\int_0^t \mathrm{d}t_1 \int_0^t \mathrm{d}t_2 \times \int \mathrm{d}k \tilde{R}(k)\mathrm{e}^{\mathrm{i}(\omega-\omega_k)(t_1-t_2)}\langle \mathrm{e}^{\mathrm{i}[\phi(t_1)-\phi(t_2)]}\rangle, \quad (4)$$

where C is a numerical constant independent of the frequency ω of the emitted light, $\tilde{R}(k) = \int \mathrm{d}\rho R(\rho)\exp(\mathrm{i}k\rho) = 2\pi l^2\exp(-k^2l^2/2)$, $\omega_k = E_{\mathrm{i}} - E_k$, and

$$\phi(t) = \frac{1}{\hbar}\int_0^t \mathrm{d}t_1[\Delta E_{\mathrm{i}}(t_1) - \Delta E_k(t_1)].$$

The averaging (angular brackets) over the temperature fluctuations in Eq. (4) can be rewritten following Ref. [5] as

$$\langle \mathrm{e}^{\mathrm{i}[\phi(t_1)-\phi(t_2)]}\rangle = \langle \mathrm{e}^{\mathrm{i}\Delta\phi(t_1-t_2)}\rangle = \mathrm{e}^{-\langle\Delta\phi^2(t_1-t_2)\rangle/2}. \quad (5)$$

Substituting Eq. (5) in Eq. (4), the optical transition rate $W = P/t$ is given by

$$W(\omega) = C\int \mathrm{d}k \tilde{R}(k) f(\omega-\omega_k), \quad (6)$$

where

$$f(\Delta\omega) = \int \mathrm{d}t_1 \mathrm{e}^{-\langle\Delta\phi^2(t_1)\rangle/2}\cos(\Delta\omega t_1). \quad (7)$$

In order to determine $F(\Delta\omega)$, we need to calculate the average $\langle\Delta\phi^2(t)\rangle$, which has the form

$$\langle\Delta\phi^2(t)\rangle = \frac{1}{\hbar^2}\left(\frac{\partial E_{\mathrm{c}}}{\partial T}\right)^2\int_0^t \mathrm{d}t_1\int_0^t \mathrm{d}t_2\int \mathrm{d}z \times \int\int \mathrm{d}\rho_1\mathrm{d}\rho_2[R^2(\rho_1)\chi_2^2(z) - S^{-1}\chi_1^2(z)] \times [R^2(\rho_2)\chi_2^2(z) - S^{-1}\chi_1^2(z)] \times \langle\Delta T(t_1,\rho_1)\Delta T(t_2,\rho_2)\rangle. \quad (8)$$

In what follows, we shall disregard the second terms in both square brackets because they are much smaller than the corresponding first terms, which is due to the additional small factor S^{-1}.

The correlation function of the temperature fluctuations can be found from the solution of the Langevin heat diffusion equation

$$\frac{\partial \Delta T}{\partial t} = D_T \nabla^2 \Delta T + F, \tag{9}$$

where D_T is the thermal diffusion constant related to the thermal conductivity ξ and the heat capacity c_v as $D_T = \xi/c_v$. The correlation function of the Langevin force is given by

$$\langle F(t,r)F(t_1,r_1)\rangle = \beta \nabla_r \nabla_{r_1} \delta^3(r-r_1)\delta(t-t_1), \tag{10}$$

where $\beta = 2D_T k_B T^2/c_v$. As we mentioned above, the dependence of the fluctuating functions (ΔT and F) on the z-coordinate is disregarded due to the small size of the system in the z direction. Then, by taking two-dimensional Fourier components of both parts of Eq. (9), the solution of Eq. (9) is obtained in the k-space as

$$\Delta \tilde{T}(t,k) = \int_{-\infty}^{t} \mathrm{d}t_1 \tilde{F}(t_1,k) \mathrm{e}^{-D_T k^2 (t-t_1)}. \tag{11}$$

The correlation function of the Fourier component of the force F can be found from Eq. (10) in the form

$$\langle \tilde{F}(t,k)\tilde{F}(t_1,k_1)\rangle = 4\pi^2 \beta k^2 \delta^2(k-k_1)\delta(t-t_1).$$

Taking into account the relation between $\Delta T(t,k)$ and $F(t,k)$ [Eq. (11)], the correlation function of the temperature fluctuations is then

$$\langle \Delta\tilde{T}(t,k)\Delta\tilde{T}(t_1,k_1)\rangle = \frac{4\pi^2\beta}{D_T} \mathrm{e}^{-D_T k^2 |t-t_1|} \delta^2(k-k_1). \tag{12}$$

Substituting Eq. (12) into Eq. (8), we obtain

$$\langle \Delta\phi^2(t)\rangle = \frac{1}{2\pi\hbar^2}\left(\frac{\partial E_c}{\partial T}\right)^2 \frac{\beta}{z_0 D_T} \int_0^t \mathrm{d}t_1 \int_0^t \mathrm{d}t_2 \times \int_0^\infty k\ \mathrm{d}k [\tilde{R}^2(k)]^2 \mathrm{e}^{-D_T k^2 |t_1 - t_2|},$$

where $z_0^{-1} = \int \mathrm{d}z\, \chi_2^4(z)$ and $\tilde{R}^2(k) = \exp(-k^2 l^2/4)$. After the integration over k, t_1, and t_2, the expression for $\langle \Delta\phi^2(t)\rangle$ takes the form

$$\begin{aligned}\langle \Delta\phi^2(t)\rangle &= \frac{1}{2\pi\hbar^2}\left(\frac{\partial E_c}{\partial T}\right)^2 \frac{\beta l^2}{z_0 D_T^3}[(\tau+1)\ln(\tau+1)-\tau] \\ &= \frac{k_B c_v T^2 l^2}{2\pi\hbar^2 \xi^2 z_0}\left(\frac{\partial E_c}{\partial T}\right)^2 [(\tau+1)\ln(\tau+1)-\tau] \\ &= 2\Phi(T)\mathscr{F}(t/t_0),\end{aligned} \tag{13}$$

where $\tau = t/t_0, t_0 = l^2/2D_T$ and $\mathscr{F}(x) = (1+x)\ln(1+x) - x$. The asymptotics of the function $\mathscr{F}$ are: $\mathscr{F}(x) \approx x^2/2$ for $x \ll 1$ and $\mathscr{F}(x) \approx x \ln x$ for $x \gg 1$. Substituting Eq. (13) in Eq. (7), we obtain

$$f(\Delta\omega) = \int \mathrm{d}t_1\, \mathrm{e}^{-\Phi(T)\mathscr{F}(t_1/t_0)} \cos(\Delta\omega t_1).$$

The final expression for the transition rate (shape of the emission line) is then

$$W(\omega) \propto \int \mathrm{d}\omega' W_0(\omega' - \omega_1) f(\omega - \omega'), \tag{14}$$

where $W_0(\omega - \omega_1)$ is the transition rate at low (zero) temperatures, centered at frequency ω_1. In addition to the factor $\tilde{R}(k)$ in Eq. (6), which results in a finite width of the line at low temperature, there are other contributions to $W_0(\omega)$, e.g. inhomogeneous broadening. One of the origins of this inhomogeneous broadening is the fluctuations of the dot geometry. The effect of these fluctuations on line broadening has been extensively studied theoretically in Ref. [3]. We approximate all these contributions by a Lorentzian function, $W_0(\omega) = \Gamma/[2\pi(\omega^2 + (\Gamma/2)^2)]$, with a finite width Γ. In our calculations below we set $\Gamma = 15$ meV.

The temperature dependence of the function $\Phi(T)$ [Eq. (13)] comes from the following factors: (a) the explicit T^2 term, (b) through $\xi(T)$, for which we take the following approximation [7] $\xi(T) = 0.36 - 0.0017T + 2.47 \times 10^{-6} T^2$, and (c) through $E_c(T) = E_{c,0} - \alpha T^2/(T+\beta)$ dependence, which gives $\partial E_c(T)/\partial T = \alpha T(T+2\beta)/(T+\beta)^2$, where α and β are material constants. Substituting these dependences into the expression for function $\Phi(T)$ [Eq. (13)], we obtain

$$\Phi(T) = \frac{k_B c_v \alpha^2}{4\pi\hbar^2}\left(\frac{l^2}{z_0}\right)\frac{T^4(T+2\beta)^2}{\xi^2(T)(T+\beta)^4}.$$

There is also temperature dependence of the time t_0

$$t_0 = \frac{l^2}{2D_T} = \frac{l^2 c_v}{2\xi(T)}.$$

It should be noted that the experimentally observed additional peak in the emission spectra (see Fig. 1) at high temperature and low frequency cannot be explained just from temperature broadening of a single line. The explanation of this additional structure requires introduction of optical transitions from an excited state of the system with excitation energy E_{ex}. Such a state will be occupied at high temperatures and will modify expression (14) as

$$W(\omega) \propto \int d\omega' \left[W_0(\omega' - \omega_1) f(\omega - \omega') + \exp\left(-\frac{E_{ex}}{T}\right) \times W_0(\omega' - \omega_2) f(\omega - \omega') \right], \quad (15)$$

where ω_2 is the transition frequency for the excited state.

In our calculations we have used the following parameters for the QD–QW system: $\alpha \approx 2.8 \times 10^{-4}$ eV/K, $\beta \approx 80$ K, $c_v \approx 1.4$ J/cm^3 K, and $l = 15$ nm. The results of calculations are shown in Fig. 2 at different temperatures. There is an additional peak at low frequency and high temperature which is due to the optical transition from an excited state of the system. Appearance of this peak indicates that the corresponding optical transition is the transition from the excited state of the QD to the excited state of the QW, compared to the main optical transition which we can call the transition from the ground state of the QD to the ground state of QW. The low-frequency transitions can be due to the inhomogeneity of the system and come from the region where the QW has a smaller width. Decreasing of the width of the QW compared to the normal region, which is responsible for the main peak, increases the size quantization energy both for the QD and QW. This means that the transition in this region is the transition from the excited state of QD to that of the QW. Because the sensitivity of the size quantization energy to the width of the QW is higher for QW than for QD, the corresponding optical transition results in a low-frequency line. Theoretical results presented in Fig. 2 clearly indicate that temperature fluctuations can be the reason for the observed finite width of the emission line at high temperatures.

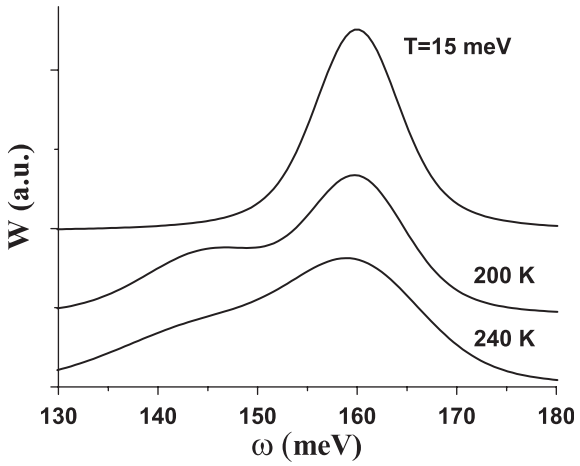

Fig. 2. The shape of the emission line for the QD–QW structure is shown at different temperatures for a parabolic QD with $l = 15$ nm. Broadening of the emission line can be attributed to temperature fluctuations.

In conclusion, generally the main origin of temperature broadening of emission lines is the temperature repopulation of energy levels of the system. The specific feature of our system is that it consists of the QDs. The excitation energy in the QD (energy difference between the levels) is higher than the characteristic temperature range in the present experiments. In such a case, the mechanism of temperature broadening due to temperature fluctuations becomes dominant.

The work of one of the authors (T.C.) is supported by the Canada Research Chair Program.

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