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THE ROLE OF PHOTOLUMINESCENCE EXCITATION SPECTROSCOPY IN INVESTIGATION OF QUANTUM CASCADE LASERS PROPERTIES

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ABSTRACT

The properties of quantum cascade laser (QCL) structures have been investigated by optical technique based on spontaneous emission measurements: photoluminescence excitation (PLE) spectroscopy. Three types of test structures used for obtaining final QCL device were examined, i.e., single sequence of coupled quantum wells, which form an active region of the device, 30 sequences of this active region separated by 25 nm thick AlGaAs barriers and finally complete, undoped structure consisting of 30 of sequences repeated active regions and superlattice injectors. The results has been compared with numerical simulations. The role of such measurements has also been discussed.

1. Introduction

The quantum cascade lasers (QCLs) [1], [2] have unique properties, especially power, tuning range and ability to work pulsed at room temperature, which make them very suitable for a broad range of applications. This kind of lasers is based on specific approach, different than in conventional diode lasers (LDs).

Diode lasers relay on transitions between energy bands: electrons and holes injected into the active layer radiatively recombine across the band gap. The efficiency of such laser is not high, because only one photon is generated during each recombination process.

The QCL is an unipolar laser. It involves only one type of carriers and is bases on phenomenas of quantum mechanics: tunneling and quantum confinement. Electrons make intersubband transitions between conduction band states arising from size quantization in ultrathin alternating semiconductor layers, creating structure of quantum well. If the layer size is comparable to the electron's de'Broglie wavelength, the electron motion is restricted to plate perpendicular to the layer. Electrons can only jump from one state to other by discrete steps, every time emitting photon of light. Therefore, the emitted wavelength is not determined by the band gap of used materials (like it occurs in the diode lasers) but by the thickness of the constituent layers. Such mechanism of light generation makes the QCLs much more powerful than the conventional lasers.

2. Experiment

2.1. PLE technique

Intersubband transitions are forbidden for light propagating along *Z* axis (growth direction) in the structure and direct measurement of this is impossible without special sample preparation. Good, but indirect way to obtain further information about QCLs properties is photoluminescence excitation spectroscopy (PLE). This technique is usually widely used for studying the absorption processes in semiconductor quantum wells and superlattice structures grown on non-transparent substrates. Some discussions of the relation between PLE and absorption spectra can be found in several textbooks [3], [4].

Three processes can be distinguished in a typical photoluminescence experiment [5]. First, photons of the light beam incident on the sample are absorbed by the structure. It causes excitation of carriers from the valence into the conduction band and creation of electron-hole pairs. Then, the non-equilibrium electron and hole distributions tend to relax back into the quasi-equilibrium state. The initial intraband relaxation is caused by energy transfer to the crystal lattice. Finally, the electron–hole pairs recombine radiatively under emission of light, which is called photoluminescence.

In our PLE experiment, we tune the pump beam energy in some range above the band gap of quantum well material. When this energy is in resonance with one of the excitation levels, strong absorption proceeds and the carriers are excited into this energy level. These excited carriers thermalize to the first level of QW contributing the PL emission. During experiment, the intensity of PL signal is recorded as a function of the pump beam energy. We can determine the electronic properties of investigated structure by analyzing this dependence.

2.2. Experimental set-up

The PLE experiment requires tunable source of excitation. We used Coherent 890 Ti:Sapphire laser pumped by Ar^+ laser. The exciting beam was focused on the sample by special imaging optics. The excitation wavelength above 700 nm at 100 mW of power allows penetration of the multilayer structure deeply enough to excite the optically active region, but does not cause heating effects. Because the Ti:Sapphire laser power varies together with the wavelength of emission (Fig. 1) we used the

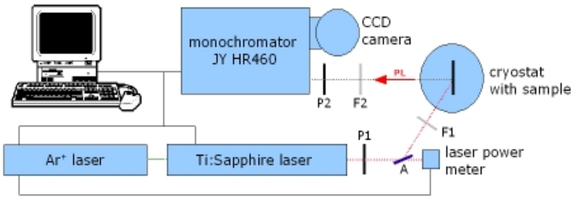

Fig. 1. Scheme of the setup used in PLE experiment.

procedure of stabilization of exciting beam power. Our setup allows to check the power of laser before each measurement and adjust it to suitable level by regulation of pumping Ar^+ laser. Thanks to using such procedure we can be sure that the power density of exciting beam does not change during experiment, therefore the quantity of carriers generated in investigated structure is the same during each measurement. In this way we can compare the intensity of photoluminescence on various energies of excitation.

In the PLE experiment the intensity of spontaneous emission from the sample is detected at a fixed photon energy. In our experiment the photoluminescence signal is dispersed by a HR460 JobinYvon monochromator and detected by a CCD3000 JobinYvon camera. We can observe not only the intensity of PL at one energy, but the whole emission spectrum from the structure at once. The results may be illustrated in the form of maps, where on X axis is the wavelength of photoluminescence, on Y axis – the wavelength of exciting beam and Z axis is colored scale of PL intensity.

Due to the fact that the oscillator strength of interband transitions in quantum well is stronger than that of intersubband transitions, we made this investigation in low temperature. The sample was cooled down to 77 K by using a LN cryostat. The temperature in cryostat was monitored by LakeShore 330 temperature controller.

Figure 2 shows the schemes of these structures. In the first case it is a single sequence of coupled quantum wells, which form an active region of the device (Fig. 2a). Next structure consists of 30 sequences of this active region separated by 25 nm wide AlGaAs barriers (Fig. 2b). The last one is complete undoped structure – 30 sequences of repeated active regions and superlattice injectors (Fig. 2c). Each of the investigated structures were

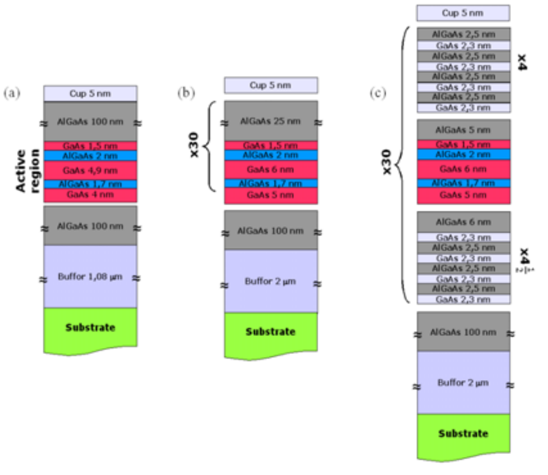

Fig. 2. Schemes of the structures used in our PLE experiments: a) an active region of quantum cascade laser, b) 30 sequences of active region separated AlGaAs barriers, c) 30 sequences of repeated active regions and superlattice injectors.

grown by molecular beam epitaxy (MBE). Next paragraphs contain detailed information about thickness and composition of particular layers are below.

3. Results and discussion

In this paper we present the results of investigation of these three structures. In each case we show a PLE map and the PLE signal at energy corresponding with 1e-1hh transition. The vertical lines on the PLE map represent photoluminescence spectra at particular energy (wavelength) of excitation, presented on the horizontal axis. Colors show the intensity of luminescence – the red one represents the highest PL signal, and the blue one – the lowest signal. On the graphs below maps, the relation between photoluminescence intensity and the energy of excitation is shown. Additionally, the transition energies obtained from numerical simulation are shown. All the measurements were made in the temperature of liquid nitrogen.

3.1. Active region

The first of investigated structures consists of three GaAs quantum wells (which width is properly 1.5 nm, 4.9 nm and 4 nm) separated by AlGaAs barriers (2 nm and 1.7 nm of width). Such structure can create an active region of quantum cascade laser (Fig. 2a).

In Fig. 3 we present the results of measurements of this structure. The analysis of this map allows to notice that the highest photoluminescence signal is at 1.583 eV energy of excitation. This energy corresponds with the 1e-1hh transition in QW.

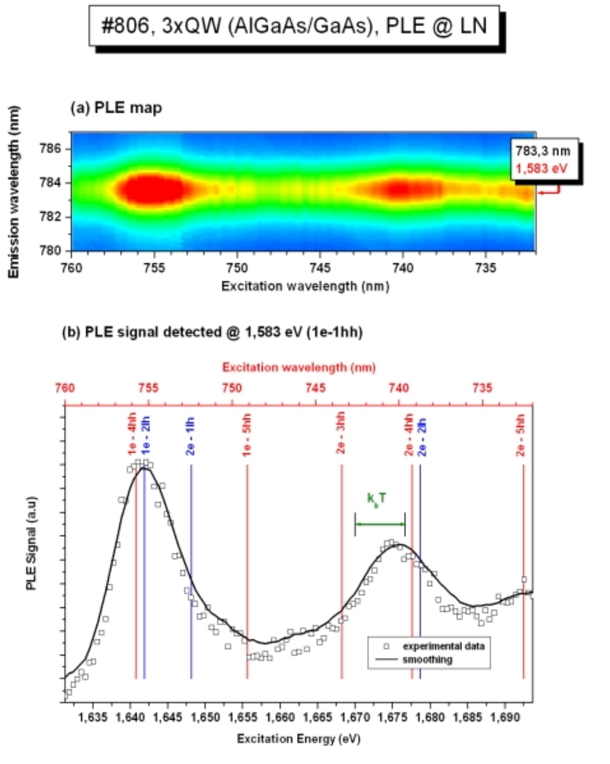

Fig. 3. The results of PLE investigation of active region of QCL; a) map of changes in PL signal at particular wavelengths (energies) of excitation. Red color represents the highest intensity of PL and blue color – the lowest one; b) PLE signal at energy corresponding with 1e-1hh transition. The squares represent experimental data, the black line is smoothing of this data, vertical lines shows the transition energies according to the results of simulation.

We carried out the numerical simulation of energy levels using software package *nextnano*^{3} [6]. In this case the nominal value of Al content in QW barriers is 30%, but taking into account the specificity of MBE growing process we may expect differences in Al content on various areas of the sample. On the basis of simulations carried out at various values of Al content we determined this as 24% in the point of measurement.

In next stage of results analysis we can plot the intensity of PL signal corresponding with the 1e-1hh transition, recorded as a function of the pump beam energy. Figure 3b shows this PLE signal and the results of numerical simulation. Taking into account the k_BT factor, which at this temperature is 7 meV, we can notice that the two picks visible in the PLE signal correspond with the transitions: 1e-4hh (1.641 eV) and 2e-4hh (1.677 eV) respectively. We have very good conformity between experimental data and numerical simulation results.

From the point of view of quantum cascade laser design, we are interesting in positions of energy levels in conduction band of the structure. On the basis of our results we can determine the difference between energies of the first and the second level, which equals to 34 meV according to experimental data and 36.8 meV according to simulation results.

3.2. 30 sequences of active region

In the next part of experiment, we investigated structure consisting of 30 repetitions of QCL active region (3xQW), similar like in previous case, but of different QWs width (properly 1.5 nm, 6 nm and 5 nm). The geometrical parameters and Al content of barriers are the same. To prevent coupling of the quantum wells areas, the width of AlGaAs layers between particular repetition is 25 nm (Fig. 2b).

Figure 4 presents the results of measurement of this structure. In this case, the energy of 1e-1hh transition is 1.574 eV. In it very similar value that we obtained from numerical simulation. Figure 4b shows the PLE signal at this energy. We can see a lot of features corresponding with particular optical transitions. The differences between experimental data

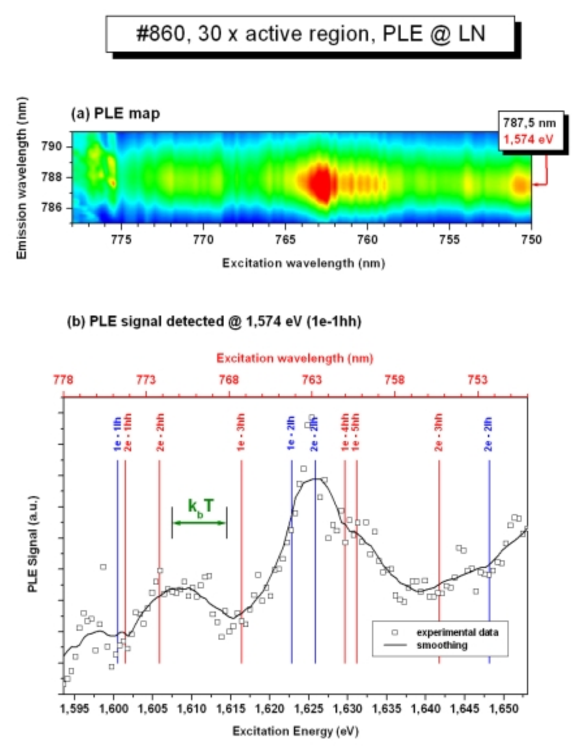

Fig. 4. The PLE results of 30 repetitions of QCL active region; a) the PLE map; like previously, red color represents the highest intensity of PL signal and blue color – the lowest one. b) PLE signal at 1.574 eV (1e-1hh). The squares represent experimental data, the black line is smoothing of this data, vertical lines shows the numerically simulated optical transitions.

and the results of numerical simulations are more significant than previously. However, the conformity is sufficient, if we take into account the $k_B T$.

Analysis of experimental data allows us to determine the energy of 2e-1hh transition, which is about 1.598 eV. The difference between first and second energy level in conduction band of this structure is 24.8 meV. This value, calculated on the basis of numerical simulations results, is 25.3 meV.

3.3. Complete structure of QCL

The last part of experiment was investigation of full, but undoped QCL structure. Detailed information about width and packing of particular layers are shown on Fig. 2c. In comparison with previous two samples, there is new element of structure-superlattice, which consists of 4.5 repetitions of alternately ordered GaAs (2.3 nm of width) and AlGaAs (2.5 nm of width) layers. Such area performs the role of injector. It transfers the electrons from one active region to next one.

Figure 5 shows the results of PLE investigation of third structure. In this case, we can see that the energy of 1e-1hh transition in QW is 1.572 eV. On the basis of numerical results we can conclude that the first peat in PLE spectrum corresponds with 2e-1hh transition (1.603 eV). This allows us to determine the energetic difference between first and second level in conduction band, which equals to 31 meV (28.6 meV from numerical analysis).

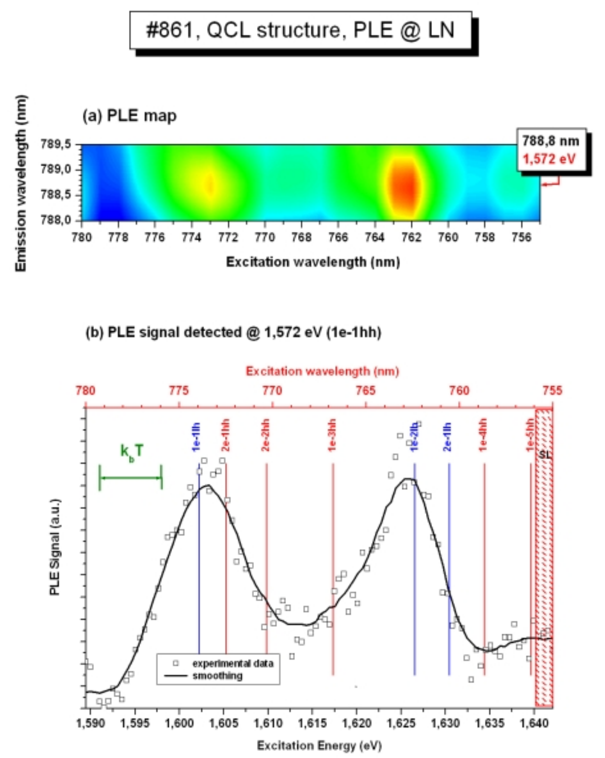

Fig. 5. The results of PLE investigation of full undoped QCL structure; a) PLE map; colored scale of PL intensity, b) PLE signal at e1.572 eV (1e-1hh). The squares represent experimental data, the black line is smoothing of this data, vertical lines shows the numerically simulated optical transitions.

4. Summary

In conclusions, the main point of this paper is that PLE used to study of structures of QCL devices, is very good alternative to other optical characterization techniques. In this work, according the results of measurements and numerical simulations, we occurred the values of optical transitions energies and the differences between first and second energy level in active region of QCL laser. Application an excitation source, which works in wider spectral range would permit to observe other transitions and the energetic distance between second and third energy level, which in very useful in the point of view QCL designing. The use PLE technique is particularly relevant at low temperature when the broadening factor $k_B T$ is lesser than at 300 K. At 77 K we obtained very good conformity between experimental and numerical results.

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