Investigation of possibility of VLWIR lasing in HgCdTe based heterostructures

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Abstract. The optical properties of a number of Hg₁₋ₓCdₓTe bulk epilayers (x = 0.152 – 0.23) and heterostructures with quantum wells (QW) based on narrow gap HgCdTe are examined aiming to reveal the prospects of such structures for laser development in long wave infrared and very long wave infrared ranges. Experimental evidence of long wavelength superluminescence, i.e. amplification of spontaneous emission, at 8.4 µm in narrow gap HgCdTe bulk epitaxial film at 100 K is reported. Employing heterostructures with QW is demonstrated to be promissory for furthering the radiation wavelength to 10 - 30 µm range.

HgCdTe material has been intensively studied for more than half a century due to its paramount importance for infrared optoelectronics. Development of IR sensors has driven the HgCdTe growth techniques, in particular molecular beam epitaxy (MBE), which nowadays allows producing high quality epitaxial structures with in situ control of layers composition and thickness. Besides bulk epilayers with high uniformity and low residual carrier density (~10¹⁴ cm⁻³) MBE can also provide HgCdTe based heterostructures with quantum wells (QW) energy spectrum of which can be tuned by changing QW width and Cd content in it. A number of peculiar fundamental properties [1, 2] were demonstrated in such systems as well as pronounced long-wavelength photoconductivity (PC) up to THz range [3, 4]. However, processes of light emission from narrow gap HgCdTe were less investigated; in particular, spectra of interband photoluminescence (PL) from HgCdTe based structures in very long wavelength infrared range (VLWIR) are scarce in literature. Stimulated emission was investigated only in wavelength region up to 5.3 µm [5, 6]. In this work we have examined the optical properties of a number of Hg₁₋ₓCdₓTe bulk epilayers and heterostructures with QW based on narrow gap HgCdTe aiming to reveal the prospects of such structures for laser development in LWIR and VLWIR ranges.
All structures under study were grown on semi-insulating GaAs(013) substrates with ellipsometric control of the layer content and thickness [7]. The “active” parts of the structures were grown on a relaxed CdTe buffer layer about 5 µm thick. Typical residual carrier density in active layer is about \( n = 5 \times 10^{14} \text{ cm}^{-3} \). PL spectra studies were carried out in a closed-cycle cryostat optically connected to a Fourier transform infrared spectrometer Bruker Vertex 80v, operating in step-scan mode. Details on experiment can be found in [8].

All structures were firstly characterized by PC spectra and kinetics measurements [3, 4]. Figure 1(a) summarizes the PC decay time data, i.e. information on carrier lifetimes in structures under study. One can see that PC decay time grows with \( x \) up to \( \sim 10 \mu s \) at \( x = 0.22 \) and then slightly decreases. The radiative lifetime is known to decrease with the bandgap [9]; hence, such behavior, i.e. carrier lifetimes increase with the bandgap indicates that recombination is non-radiative. Review of literature data suggests that the dominating mechanism of carrier recombination is Shockley-Read-Hall (SRH) process [9-11]. However, according to recent calculation of carrier recombination rates [11] lifetime of 9.2 µs measured in our bulk epilayer (\( x = 0.22 \)) practically coincides with the calculated radiative lifetime for n-doped (\( 5 \times 10^{14} \text{ cm}^{-3} \)) \( \text{Hg}_{1-x}\text{Cd}_{x}\text{Te} \) with \( x = 0.224 \) at low temperatures (20 K). Thus, one can suggest that in epilayers under study carrier recombination is mostly controlled by radiative processes when \( x \geq 0.22 \) at least at low temperatures. This is confirmed by further decrease in carrier lifetime when \( x \) increases to 0.23 (see Figure 1(a)). Moreover, domination of radiative process over SRH recombination can be enhanced when excitation intensity is increased. At high enough excitation level (in the case of PL measurements, for example) recombination via trap centers tends to saturate while rates of radiative recombination increase and thus radiative recombination readily dominates in solid solutions with \( x = 0.21 \) – 0.23 at low temperature [4, 8]. As a result, this allowed observation of interband PL on record wavelength of 26 µm [8] in bulk \( \text{Hg}_{1-x}\text{Cd}_{x}\text{Te} \) with \( x = 0.189 \). PL measurements in such structures could be the first step in the development of light emitting devices for the spectral range 20 – 30 µm, which is practically unacceptable for \( \text{A}_3\text{B}_5 \) based devices such as quantum cascade lasers, due to strong two-phonon absorption.
However, the next question to be answered is whether the amplification of long-wavelength radiation can be achieved in such structures. In order to reveal the possibility of light amplification in samples under study the PL spectra transformation under intense optical excitation was examined. For this experiment the bulk epilayer with $x = 0.22$ was chosen since it was the narrowest gap structure that according to calculation contained the dielectric waveguide for radiation wavelength corresponding to interband transitions. PL radiation was collected from the edge of the sample since in-plane direction of the epilayer is thought to be optimal for amplification of radiation. Indeed, at high enough excitation level spectra of PL radiation propagating along the film plane is considerably narrower as compared to radiation propagating in $45^\circ$ angle relative to growth direction (see the insert in figure 1(b)). Figure 1(b) demonstrates the genesis of a narrow line of amplified PL on broad band of spontaneous emission as the intensity of pumping grows. Graph $P_1$ represents PL spectra under pulsed excitation with $\sim 10^{12}$ photons in 130 fs pulse, which proved to be almost identical to PL spectra measured under continuous wave excitation with $P = 300$ mW. As the pumping power grows PL line broadens at first due to filling of conduction and valence band by non-equilibrium carriers (compare graphs $P_1$ - $P_2$). Then a narrow line, which is superimposed on the broad background band of spontaneous emission, appears at 1170 cm$^{-1}$ and rapidly grows with pumping intensity (compare graphs $P_2$ - $P_3$) finally slightly shifting to 1196 cm$^{-1}$ (8.4 $\mu$m) (graph $P_4$). At excitation power $P_4$ the full width at half maximum (FWHM) of PL line is almost 3 times smaller as compared to power $P_2$. At this point it should be noted that such experiments were not performed earlier apparently because Auger recombination was believed to be absolutely detrimental for light emission processes in such narrow gap material. However, it is worth mentioning some recent papers that give an optimistic view concerning the negative influence of Auger recombination in narrow gap MBE epilayers. Firstly, calculations of recombination rates that exploit the full band structure obtained in hybrid pseudopotential tight-binding Hamiltonian model predict longer Auger time than those calculated with widely used analytical expressions [11]. Secondly, at high carrier densities Auger recombination rates are affected by Coulomb screening that should also increase the Auger recombination time [9]. Though at rather high temperature as 100K the Auger recombination lifetime is of the same order of magnitude as spontaneous radiative recombination time according to [11] somewhat of Auger mitigation is indirectly confirmed by our experiments as well.
Nevertheless, the FWHM and intensity of superluminescent line demonstrate saturation at higher excitation. This is due to plasmonic contribution to dielectric function in the active layer of the structure. High carrier density formed by intense excitation increases plasma frequency bringing it closer to the PL radiation frequency. This leads to considerable decrease in dielectric function in the active layer, which has dramatic effect on the light confinement. Estimations show that the waveguide in the structure under study almost disappears at $n = 2 \times 10^{18} \text{ cm}^{-3}$. Thus, at high density of non-equilibrium carriers, the amplification of radiation is weak and electron-hole recombination takes place mostly via fast non-radiative processes. However, further amplification can be achieved in structures with separate light and carrier confinement. Indeed, the negative influence of excess carriers on the light confinement can be reduced by placing QWs inside the core layer of the dielectric waveguide. QWs will accumulate the non-equilibrium carriers providing the same gain but the change of dielectric function in thin QW layers will have small impact on waveguide properties of the structure.

QW are also expected to be beneficial for additional suppression of non-radiative processes [12]. Unlike bulk material the energy spectrum of QW structures is characterized by close effective masses of electrons and holes that should result in mitigation of the Auger recombination. This is indirectly confirmed by low temperature quenching of PL in QW structures as compared to bulk samples [13]. PL signal at room temperature is easily detected not only in wide QW but also in rather narrow wells (figure 2(a)). Moreover, SRH recombination can also be suppressed by saturating the trap centers with intense pumping. Time resolved PL measurements in QWs show that the carrier relaxation time grows with excitation intensity and can be as high as 5 $\mu$s under high excitation (figure 2(b)). This infers that carrier density of several $10^{11} \text{ cm}^{-2}$ required for amplification [14] can be achieved under reasonable level of excitation. We calculated mode localization and modal gain coefficients for several optimized designs of epitaxial HgCdTe structures aimed at obtaining stimulated emission in VLWIR range. These structures are supposed to contain 5 HgTe QW in core layer of waveguide. Calculations show [15] that these QW would provide amplification coefficient of several $10 \text{ cm}^{-1}$ at non-equilibrium carrier concentration of $1 - 4 \times 10^{11} \text{ cm}^{-2}$ (for $\lambda \sim 15 - 30 \mu \text{m}$).

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