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ZnO-based terahertz quantum cascade lasers

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ABSTRACT

High-power terahertz sources operating at room-temperature are promising for many applications such as explosive materials detection, non-invasive medical imaging, and high speed telecommunication. Here we report the results of a simulation study, which shows the significantly improved performance of room-temperature terahertz quantum cascade lasers (THz QCLs) based on a ZnMgO/ZnO material system employing a 2-well design scheme with variable barrier heights and a delta-doped injector well. We found that by varying and optimizing constituent layer widths and doping level of the injector well, high power performance of THz QCLs can be achieved at room temperature: optical gain and radiation frequency is varied from 108 cm^{-1} @ 2.18 THz to 300 cm^{-1} @ 4.96 THz. These results show that among II–VI compounds the ZnMgO/ZnO material system is optimally suited for high-performance room-temperature THz QCLs.

1. Introduction

The Terahertz (THz) spectral region is defined as the electromagnetic wave with a frequency range between 0.1 THz and 10 THz. In the last two decades the problem of fabrication of terahertz devices like THz sources and THz detectors has attracted many research groups because these devices could be used in different kinds of applications such as detection of explosive materials, non-invasive THz imaging in medicine, security applications, high speed telecommunications and short-distance communications in air as required for pure in-house applications [1]. All these applications require relatively high power terahertz sources with milliwatt-level output power, which could operate at room temperature. In recent years, different concepts and approaches of terahertz source were suggested, such as resonant-tunneling diodes (RTDs), large-area photoconductive antennas, photomixers, quantum cascade lasers (QCLs), and difference frequency generation QCLs (DFG-QCLs). Resonant-tunneling diodes (RTDs) are usually based on InGaAs/GaAs and have a microwatt-level output power at room-temperature and their maximum operating frequency is less than 2 THz [2]. Terahertz sources such as optical photomixers and DFG-QCLs can be tunable over a wide range of terahertz spectra from 1.2 THz to 5.9 THz, but they also have low output power, which limits their use in practical applications. THz QCLs can produce milliwatt-level output power, but they require cryogenic cooling down to less than 200 K [1]. The first demonstration of THz QCLs (4.4 THz at 50 K) has been reported by Köhler *et al.* in 2002 [3]. The most reported THz QCL devices are based on the AlGaAs/GaAs material system. The maximum operation temperature of 200 K was reported by Fathololoumi *et al.* in 2012 [4] for 3.22 THz $Al_{0.15}Ga_{0.85}As$/GaAs THz QCLs in pulse mode operation based on a 4 well diagonal design scheme with longitudinal-optical phonon (LO-phonon) depopulation of the lower laser state. To overcome the problem of low-temperature operation of the THz QCLs several approaches have been suggested such as QCLs with a tall quantum barrier, QCLs with variable barrier heights and others. Also other material systems have been considered and studied such as InGaAs/GaAs, AlGaN/GaN, InGaN/GaN, ZnMgCdSe/ZnCdSe, ZnMgSe/ZnSe, ZnMgO/ZnO, SiGe/Si [1,4–6]. For design and fabrication of high-performance room-temperature THz QCLs, a II—VI wide bandgap material system like ZnMgO/ZnO has several advantages in comparison to the AlGaAs/GaAs such as higher value of LO-phonon energy (72 meV for ZnO vs. 36 meV for GaAs) and higher conduction band offset (CBO) values ($\tilde{2}$.38 eV for ZnO/MgO vs. 0.72 eV for GaAs/AlAs) [5]. It is well known that ZnO-based materials have spontaneous and piezoelectric polarizations, which play an important role regarding optical properties and resulting built-in electric field in c-plane ZnO-based heterostructures. To overcome this problem it was suggested to use non-polar m-plane grown heterostructures. Recently, Bajo *et al.* [7] reported on successful fabrication m-plane ZnMgO/ZnO multi-quantum wells with low

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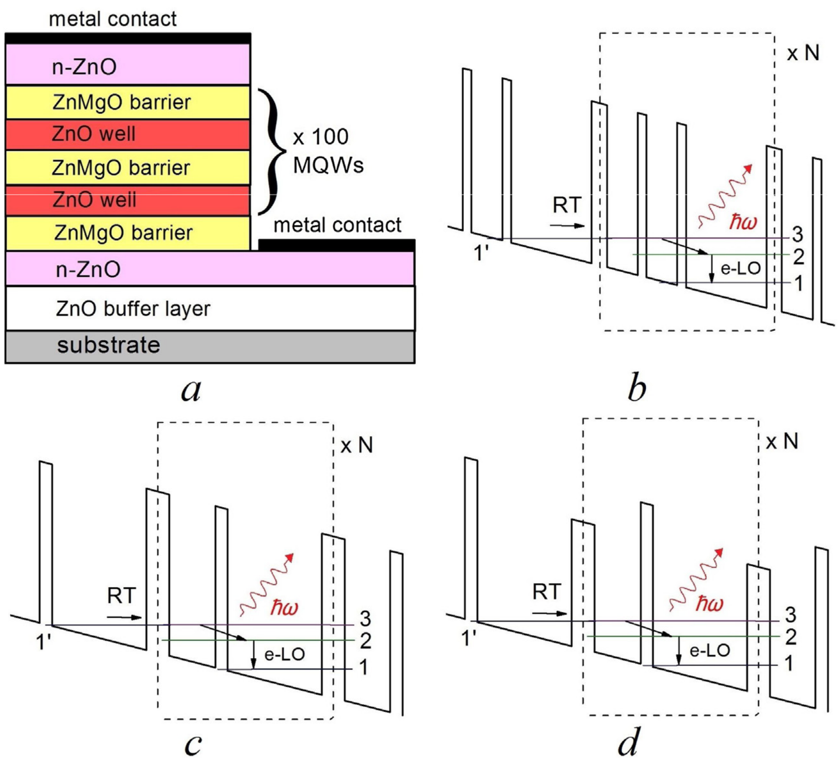

Fig. 1. (a) Schematics structure of THz QCL device based on ZnMgO/ZnO multi-quantum well heterostructues. Conduction band profiles of THz QCL devices based on different designs: (b) 3-well design scheme with fixed barrier heights, (c) 2-well design scheme with fixed barrier heights, (d) 2-well design scheme with variable barrier heights.

defects density and investigated intersubband absorption in such structures.

In this paper we report on the design and simulation study of high power room-temperature THz QCLs based on a non-polar m-plane ZnMgO/ZnO material system and employing a 2-well design scheme with variable barrier heights and delta-doped injector well. We show that by varying and optimizing the constituent layer widths and doping level of the injector well, high power performance of THz QCLs can be achieved at room temperature: optical gain and radiation frequency is varied from 108 cm^{-1} @ 2.18 THz to 300 cm^{-1} @ 4.96 THz.

2. Device design, methods and model

In this paper we report on the results of a numerical study of ZnMgO/ZnO-based THz QCLs with different layer structures and designs as shown in Fig. 1a–d, which include a 3-well design scheme with fixed barriers, a 2-well design scheme with fixed barrier, and a 2-well design scheme with variable barrier heights. The electronic quantum transport and optical gain of ZnMgO/ZnO-based THz QCLs were investigated numerically within single band effective mass approximation. The time-independent Schrödinger equation within the single-band effective mass approximation can be written as:

$$\left[-\frac{\hbar^2}{2}\frac{\partial}{\partial z}\frac{1}{m(z)}\frac{\partial}{\partial z}+(eV(z)\Delta E_C(z))\right]\Psi(z)=E\Psi(z), \quad (1)$$

where $m(z)$ is the position-dependent electron effective mass in the z direction, $V(z)$ is the electrostatic potential, $\Delta E_C(z)$ is the stepwise function due to the conduction band discontinuity, $\Psi(z)$ is the eigen wave function, and E is the energy eigenvalue.

The electrostatic potential $V(z)$ is determined by solving the Poisson equation, which is given in the absence of polarization by [8]:

$$\frac{\partial}{\partial z}\left[-\varepsilon(z)\frac{\partial}{\partial z}V(z)\right]=q\left(N_D(z)-n(z)\right), \quad (2)$$

where $\varepsilon(z)$ is the position-dependent dielectric constant, $V(z)$ is the electrostatic potential, $N_D(z)$ is the ionized donor doping concentration, and $n(z)$ is the free electron concentration. The system of coupled Eqs. (1) and (2) was solved numerically using nextnano.MSB solver software [9] which is based on the modified non-equilibrium Green's function (NEGF) method. The used nextnano.MSB software includes the following scattering mechanisms: electron-longitudinal optical (LO) phonon scattering, electron-electron scattering, electron-longitudinal acoustic (LA) phonon scattering. It is a memory efficient method and it has successfully predicted the experimental results of AlInAs/InGaAs- and GaAs/AlGaAs-based mid-infrared and THz QCL lasers [10]. The details of a model used in this paper can be found in Ref. 10. For $Zn_{1-x}Mg_xO$ quantum barriers the material parameters were linearly approximated between the respective values of ZnO and MgO. At a temperature of T = 0 K the CBO of $Zn_{1-x}Mg_xO$ used in the calculations is equal to 2.38x eV, and the bandgap energy is equal to 3.437 + 3.40x for $x < 0.4$. The other material parameters used were taken from [5,11,12] and summarized in Table 1. In order to enhance quantum transport in the considered quantum structures, the first well of QCLs were n-doped with a concentration varied from 1×10^{16} cm^{-3} to 1×10^{19} cm^{-3}. The temperature was varied from 100 K to 300 K. The cross section area of all the investigated QCL devices is of 0.15×1.80 mm^2.

Table 1
Summary of material parameters.

Material	ZnO	MgO
Bandgap energy at 0 K (eV)	3.437	6.8
Varshni parameters		
α (meV/K)	0.2	0.3
β (K)	325	295
Electron effective mass (m_0)	0.24	0.35
Material density (kg/m^3)	5606	3580
LO-phonon energy (meV)	72	92
Deformation potential (eV)	−3.80	−1.95
Sound velocity (m/s)	6080	9080
Low frequency dielectric constant ε_0	8.10	9.83
High frequency dielectric constant ε_∞	4.11	2.95

3. Results and discussion

The scheme and structures of numerically investigated ZnMgO/ZnO THz QCL devices with different designs are shown in Fig. 1b–d. These THz QCLs are based on designs with diagonal laser transitions, employing resonant-tunneling and intra-well depopulation of lower laser state mechanisms. As reference, we have chosen the design of ZnMgO/ZnO THz QCL suggested by Bellotti *et al.* [5]. This THz QCL device consists of three ZnO quantum wells and $Zn_{0.85}Mg_{0.15}O$ quantum barriers (Fig. 1b). The layer thicknesses of one cascade of such a THz QCL starting from the injector barrier in nm is: **3.0**/3.1/**2.5**/2.4/**3.4**/5.5, where the underlined quantum well is homogeneously n-type doped with a concentration of 3×10^{16} cm^{-3}. Our approach for high-power room-temperature ZnMgO/ZnO THz QCLs are based on a 2-well design scheme employing the fixed and variable barrier heights (Fig. 1c and 1d) and a delta-doped injector well of 1×10^{18} cm^{-3}. The layer thicknesses of one cascade of such structures starting from injector well in nm are equal to **2.7**/6.0/**2.6**/4.0 and **2.7**/6.0/**1.5**/4.0, respectively. The THz QCL with fixed barriers consist of $Zn_{0.85}Mg_{0.15}O$ quantum barriers, while devices based on a design with variable barrier heights consist of $Zn_{0.80}Mg_{0.20}O$ and $Zn_{0.70}Mg_{0.30}O$ quantum barriers. For comparison, we have also simulated the best experimental $Al_{0.15}Ga_{0.85}As$/GaAs THz QCL based on a 3-well design scheme with fixed barriers [4]. The layer thicknesses of one cascade of this THz QCL starting from the injector barrier in nm is: **4.3**/8.9/**2.46**/8.15/**4.1**/16, where the last quantum well is homogeneously n-type doped with a concentration of 6×10^{16} cm^{-3}. In Fig. 1b–d states 1, 2, and 3 represent the ground, lower laser and upper laser states respectively. States 1′ and 3″ represent the ground state of the preceding period and the upper laser state of the following period, respectively. The electrons are injected into upper laser level 3 from the injector level 1′ by resonant-tunneling (RT). The THz laser emission is due to the 3 → 2 photon-assisted diagonal tunneling transition. The depopulation of the lower laser level 2 occurs directly by fast intra-well electron-longitudinal-optical (e-LO) phonon scattering back into the injector level 1, where $E_{21} \approx \hbar\omega_{LO} \approx 72$ meV. THz QCL structures use diagonal laser transitions (3 → 2) that suppresses nonradiative transitions.

The dependences of the optical gain of THz laser emission as function of temperature for AlGaAs/GaAs and ZnMgO/ZnO THz QCLs with different design schemes are shown in Fig. 2. It can be seen that optical gain of all devices decreased with increasing temperature. The calculated maximum operating temperature of AlGaAs/GaAs THz QCL is about 200 K, which is in good agreement with experiment [4]. The calculated radiation frequency of this device at 200 K is 3.50 THz, slightly higher than the experimental value of 3.22 THz. Thus, these results confirm the validity of the used model and software. As can be seen from Fig. 2, among all devices, at room temperature the best value of optical gain of 300 cm^{-1} @ 4.96 THz is given by the ZnMgO/ZnO THz QCL with a 2-well design scheme and variable barrier heights. This value is significantly higher than the performance of the other devices: 60 cm^{-1} @ 3.13 THz for ZnMgO/ZnO THz QCL with a 2-well design scheme and fixed barrier heights, and 1.4 @ 7.13 THz for ZnMgO/ZnO THz QCL with a 3-well design scheme and fixed barrier heights [5]. The higher laser performance of ZnMgO/ZnO THz QCLs compared with AlGaAs/GaAs is attributed to the higher LO-phonon energy in ZnO (72 meV for ZnO vs. 36 meV for GaAs). The approach of THz QCL devices with a 2-well design scheme employing alternating variable barrier heights leads to further enhancement of optical gain: the barriers with lower heights enhance electron injection in the active region, while the barriers with higher heights limit escaping of injected carriers from the quantum well and also reduce thermally activated carrier leakages via higher-energy parasitic levels [6]. Moreover, by varying and optimizing constituent layer widths and barrier heights, the optical gain and radiation frequency of ZnMgO/ZnO THz devices at room temperature can be tailored from 108 cm^{-1} @ 2.18 THz (for a device with $Zn_{0.90}Mg_{0.10}O$ and $Zn_{0.80}Mg_{0.20}O$ quantum barriers and layer thicknesses of one cascade in nm of **2.7**/6.2/**1.5**/11.9) to 300 cm^{-1} @ 4.96 THz (for a device with $Zn_{0.80}Mg_{0.20}O$ and $Zn_{0.70}Mg_{0.30}O$ quantum barriers described above and shown in Fig. 2).

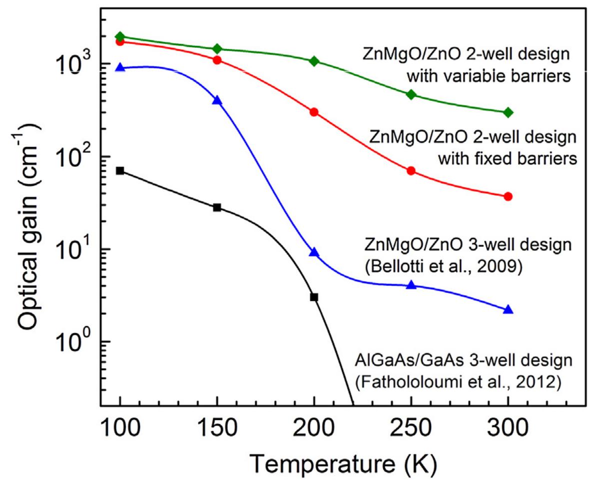

Fig. 2. Calculated optical gain of terahertz laser emission for four THz QCL devices with different designs as function of temperature.

Figures 3(a) and 3(b) shows the current density-voltage characteristics at different temperatures for two ZnMgO/ZnO THz QCL devices with different design schemes: with fixed barrier height and with variable barrier heights, respectively. As could be seen from Fig. 3(a) and 3(b) for both QCL structures a negative differential resistance (NDR) is observed at voltages lower than 60 mV per period. For both QCL structures the increase of temperature increases the current density due to the thermal activation of the free carriers, and also reduces the peak-to-valley (PVR) ratio of the current density. The increase of the temperature from 100 K to 300 K leads to the slightly shifted peak voltage from 29 mV to 39 mV for a QCL structure with fixed barrier heights [Fig. 3(a)], and from 36 mV to 50 mV for a QCL structure with variable barrier heights [Fig. 3(b)]. The shifting of threshold voltage with increase of temperature of both THz QCL lasers is caused by the temperature dependence of the bandgap energy of ZnO and MgO materials, which leads to changing of the shape of the conduction band diagram, as well as positions of the respective ground, upper laser and lower laser levels. The presence of NDR feature in the current density-voltage characteristics of the THz QCL laser were observed experimentally by Kumar *et al.* [13] and Jiang *et al.* [14] for GaAs/AlGaAs THz QCL lasers with different designs, as well as for a ZnSe-based THz QCL in our previous works [6,8]. The nature of the

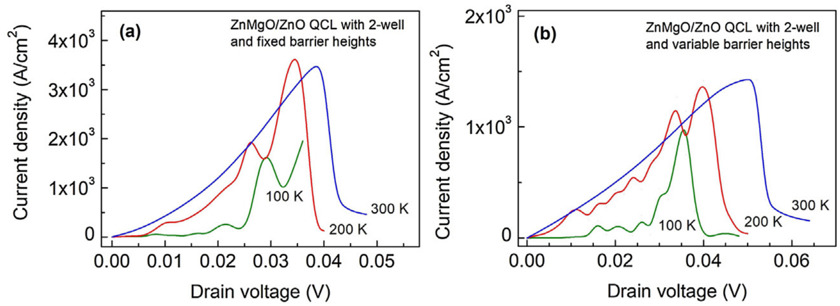

Fig. 3. Calculated optical gain of terahertz laser emission for two ZnMgO/ZnO THz QCL devices with different designs as function of temperature: (a) device with 2-well design scheme with fixed barrier heights, (b) device with 2-well design scheme with variable barrier heights.

NDR is caused by resonant-tunneling transport of free carriers in the quantum cascade structures. As could be seen from Fig. 3(a) and 3(b) the NDR feature in the current density-voltage curve is present up to room-temperature for both QCL structures. This NDR feature could be used for the design and fabrication of electronic devices for generation and amplification of high frequency emission.

4. Conclusions

In summary, for the first time room-temperature non-polar m-plane ZnMgO/ZnO THz QCLs based on a 2-well design scheme with variable barrier heights is suggested and examined numerically here. We show that m-plane ZnMgO/ZnO THz QCLs with a novel design scheme employing variable barrier heights and delta-doped injector well provide improved performance at room temperature (optical gain of 108 cm^{-1} @ 2.18 THz and 300 cm^{-1} @ 4.96 THz) compared to conventional AlGaAs/GaAs and c-plane ZnMgO/ZnO THz QCL devices based on a 3-well design scheme with fixed barrier heights.

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References

[1] M.A. Belkin, F. Capasso, New frontiers in quantum cascade lasers: high performance room temperature terahertz sources, Phys. Scr. 19 (2015), 118002.
[2] T. Maekawa, H. Kanaya, S. Suzuki, M. Asada, Oscillation up to 1.92 THz in resonant tunneling diode by reduced conduction loss, Appl. Phys. Express 9 (2016), 024101.
[3] R. Köhler, A. Tredicucci, F. Beltram, H.E. Beere, E.H. Linfield, A.G. Davies, D.A. Ritchie, R.C. Iotti, F. Rossi, Terahertz semiconductor-heterostructure laser, Nature 417 (2002) 156.
[4] S. Fathololoumi, E. Dupont, C.W.I. Chan, Z.R. Wasilewski, S.R. Laframboise, D. Ban, A. Matyas, C. Jirauschek, Q. Hu, H.C. Liu, Terahertz quantum cascade lasers operating up to ∼200 K with optimized oscillator strength and improved injection tunneling, Opt. Express 20 (2012) 3866.
[5] E. Bellotti, K. Driscoll, T.D. Moustakas, R. Paiella, Monte Carlo simulation of terahertz quantum cascade laser structures based on wide-bandgap semiconductors, J. Appl. Phys. 105 (2009), 113103.
[6] V.P. Sirkeli, O. Yilmazoglu, F. Küppers, H.L. Hartnagel, Room-temperature terahertz emission from ZnSe-based quantum cascade structures: A simulation study, Phys. Status Solidi – Rapid Res. Lett. 11 (2017), 1600423.
[7] M.M. Bajo, J. Tamayo-Arriola, A. Jollivet, M. Tchernycheva, F.H. Julien, R. Peretti, J. Faist, M. Hugues, J.-M. Chauveau, A. Hierro, Intersubband absorption in m-plane ZnO/ZnMgO MQWs, Proc. SPIE 10105, Oxide-Based Materials and Devices VIII 1010500 (2017), http://dx.doi.org/10.1117/12.2252056.
[8] V.P. Sirkeli, O. Yilmazoglu, D.S. Ong, S. Preu, F. Küppers, H.L. Hartnagel, Resonant Tunneling and Quantum Cascading for Optimum Room-Temperature Generation of THz Signals, IEEE Trans. Electron Devices 64 (2017) 3482.
[9] Web Site of Nextnano GmbH Company, 2018 (Accessed 16 December 2018) http://www.nextnano.de.
[10] P. Greck, S. Birner, B. Huber, P. Vogl, Efficient method for the calculation of dissipative quantum transport in quantum cascade lasers, Opt. Express 23 (2015) 6587.
[11] V.P. Sirkeli, O. Yilmazoglu, F. Küppers, H.L. Hartnagel, Effect of p-NiO and n-ZnSe interlayers on the efficiency of p-GaN/n-ZnO light-emitting diode structures, Semicond. Sci. Technol. 30 (2015), 065005.
[12] S. Adachi, Properties of Semiconductor Alloys: Group – IV, III-V and II-VI Semiconductors, Wiley, Berlin, 2009.
[13] S. Kumar, C.W.I. Chan, Q. Hu, J.L. Reno, A 1.8-THz quantum cascade laser operating significantly above the temperature of $\hbar\omega/kB$, Nat. Phys. 7 (2011) 166.
[14] A. Jiang, A. Matyas, K. Vijayraghavan, C. Jirauschek, Z.R. Wasilewski, M.A. Belkin, Experimental investigation of terahertz quantum cascade laser with variable barrier heights, J. Appl. Phys. 115 (2014) 163103.