Invited Paper

Recent progress in terahertz quantum cascade lasers

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Abstract: Terahertz quantum cascade lasers are semiconductor devices based on electron transitions between subbands. They have the advantages of small size, adjustable frequency, and fast response speed. Their working frequencies are between microwave and infrared light, covering the fingerprint spectrum of many gas molecules, compounds and condensed matters. They have potential applications in astronomical observation, public safety, biomedicine and other fields. In recent years, the performance of terahertz quantum cascade lasers has been significantly improved, and their applications in high-resolution spectroscopy, terahertz imaging, wireless broadband communications and other fields have also attracted attention. This article reviews the development of terahertz quantum cascade lasers, briefly describes their working principles and device structures, introduces the latest progress in device performance, frequency comb, and some new technologies. On this basis, their future development is prospected.

Keywords: Terahertz technology; Quantum cascade lasers

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1. Introduction

Terahertz (THz) is a frequency region between microwaves and infrared light, usually defined as electromagnetic waves with frequencies between 100 *GHz* and 30 *THz* (wavelength 3 *mm*–10 *μm*). THz waves have wide bandwidth and low photon energy. Their energy covers the vibrational-rotational energy of many molecules and can penetrate many non-polar media. These properties make THz waves have broad potential applications in spectroscopy, imaging, and communications [1-3]. In particular, the rapid development of THz science and technology in recent years has made THz waves useful in many valuable fields, such as airport security inspection, astronomical and atmospheric composition monitoring, pharmaceutical and coating inspection, and semiconductor integrated circuit diagnosis [4]. Currently, wireless communication technology is entering the 6G era, and higher communication frequencies are required to meet the growing demand for bandwidth. With the development of subsequent versions of wireless communication technology, the communication frequency is expected to move from the microwave to the THz frequency. It is foreseeable that the commercialization will be one of the main driving forces for the development of the THz technology.

Terahertz quantum cascade lasers (THzQCLs) are based on the electronic intersubband transitions in quantum structures. They are the only compact laser systems at this frequency so far and have a huge impact on the THz field. The first THzQCL was successfully developed in 2002 [5], and the device performance has been greatly improved during more than two decades. At the moment, the emission frequency covers the range from 1.2 *THz* [6] to 5.6 *THz* [7]. The maximum output power is 2.4 *W* [8], the single-mode continuously turning range reaches 650 *GHz* [9], the radiation bandwidth exceeds 2.6 *THz* [10], the maximum operating temperature is 261 *K* [11], and the far-field beam pattern is significantly improved [12]. THzQCLs have short inter-subband transition lifetime, which has been used to achieve active mode locking [13], fast modulation [14], and ultrafast detection [15]. THzQCLs also have strong nonlinear effects, and therefore, optical frequency combs are realized through four-wave mixing [16], room temperature THz lasers are developed through mid-infrared laser intracavity frequency difference [17], and high-order wave mixing [18] and inter-subband polaritons [19] are investigated. The development of phase-locking technology has greatly improved their frequency stability and the linewidth close to the quantum limit [20] is observed. These technologies have been successfully used in high-resolution spectroscopy [21], metrology [22] and THz imaging [23].

In this perspective article, we focus on the principles and the latest development of THzQCLs, including active regions and waveguide structures, device performance, frequency combs, and some new technologies. We also discuss the problems and provide an outlook on future developments.

2. Active region and waveguide structures

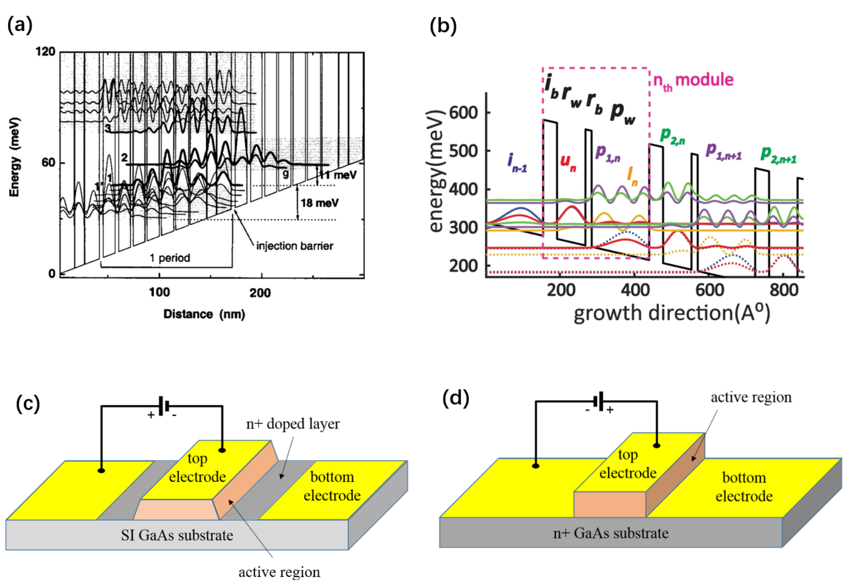

Fig. 1 Band structure of the active regions: (a) bound-to-continuum structure and (b) resonant phonon structure. Waveguide structure of (c) surface-plasmon waveguide and (d) metal-metal waveguide.

The active region of THzQCL provides gain for light amplification. According to the mechanisms of electron injection and extraction, the active region is mainly divided into four types. As shown in Figure 1(a), the bound-to-continuum (B-to-C) design belongs to the first type [24]. The electron injection to the upper laser level (level 2) and the electron extraction from the lower laser level (level 1) are both mainly relied on the resonant tunneling in the miniband. The devices using this structure have low threshold current density and low power consumption, which are beneficial for the continuous-wave (CW) operation. As shown in Figure 1(b), one of the second types is the resonant-phonon design [11]. As the main feature of this type, strong longitudinal optical (LO) phonon scattering is used for the electron extraction form the lower laser level. This active region type has larger gain and better temperature performance. Currently, the highest operating temperature of THzQCL is achieved by devices using this type of active region design. The third type is the result of the combination of the first type and the second type. The electron injection adopts the resonance tunneling, and the electron extraction adopts the miniband transport plus LO phonon scattering [25]. High-power devices in CW mode mostly use this type of active region design. In the fourth type, the electron injection is assisted by LO phonon scattering [26]. Devices using this structure have strong electron injection capabilities and can maintain effective electron injection under low frequency and high temperature conditions. Although the operating temperature of this type is still lower than theoretical calculation, this active region design has shown advantages in low-frequency devices [27].

The waveguide is used to confine the light into the gain medium. For the THzQCLs, the heavily doped dielectric waveguide, commonly used in infrared lasers, is no longer applicable due to its strong free carrier absorption. The new waveguide structure is one of the key breakthroughs to the first demonstration of THzQCL. Currently, there are two types of THz waveguides: semi-insulating substrate-surface plasmon (SISP) waveguide and metal-metal (MM) waveguide [5, 28], as shown in Figure 1 (c) and (d). In the SISP waveguide, the light is confined between the top metal layer and the bottom plasmonic layer, and the heavily doped GaAs substrate is replaced by a semi-insulating GaAs substrate and a thin heavily doped layer (600 *nm*). Overlap between the optical field and the heavily doped substrate is avoided, thereby reducing free carrier absorption and waveguide losses. In the MM waveguide, the active region is sandwiched between two layers of metal, which greatly improves the light confinement factor. The SISP waveguide have advantages of high output coupling efficiency and good beam quality, while the MM waveguide results in low threshold current and high operating temperature. In addition, the waveguide metal material will also affect the waveguide loss. Gold is the most used waveguide material and has good stability and good process compatibility. Copper and silver have also been reported as waveguide metals with lower loss [29, 30].

3. Device performance

The frequency performance of THzQCLs is important for the applications in spectroscopy and communication. The high-frequency emission, single-mode tuning range, and spectral bandwidth of the devices have been improved in recent years. Although the frequency of THzQCL can be flexibly adjusted by changing the size of quantum wells, the lasing frequency is mostly between 2-5 *THz*. It becomes more difficult to lase at both lower and higher frequency sides. At the low-frequency side, C. Walther et al. optimized the selectivity of electron injection and extraction, and observed 1.2 *THz* laser radiation [6]. Later, A. Wade et al. further reduced the scattering process between laser energy levels by applying an external magnetic field, reducing the lasing frequency to 0.68 *THz* [31]. Recently, several teams made a breakthrough at the high-frequency device. As shown in Figure 2(a), the highest lasing frequency was 5.6 *THz* in pulsed mode [7], and 5.1 *THz* in CW mode [32].

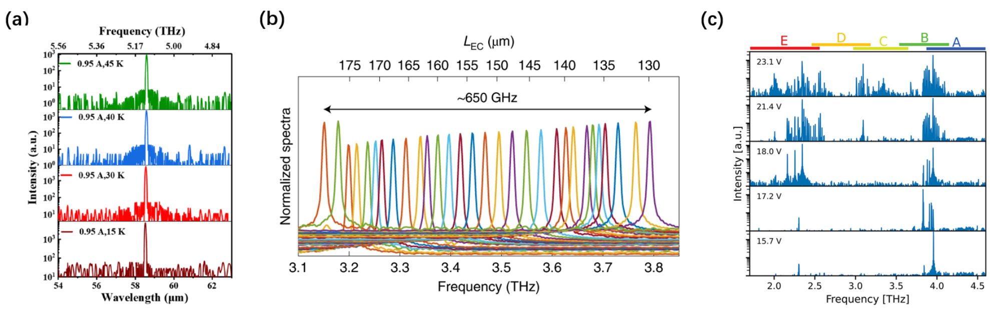

Fig. 2 Emission spectra of the THzQCLs. (a) The high frequency device [32], (b) the tunable single mode device[9], (c) the broad band device[10].

The laser frequency is normally tuned by changing parameters related to the effective laser cavity length. For example, evanescent edge field coupling [33] and microcavity coupling structure [34] were used to achieve 330 *GHz* and 162 *GHz* tuning, respectively. These methods suffered from mode hopping. In 2019, a vertical cavity surface emitting THzQCL structure with metasurface materials was demonstrated, using a short external cavity to increase the free spectral range and avoid mode hopping. As shown in Figure 2(b), centered on 3.47 *THz*, the single mode laser was continuously tuned up to 650 *GHz*, with the maximum tuning range of 880 *GHz* [9] and a large fractional bandwidth of 25%. Recently, G. Y. Xu et al. achieved single-mode continuous tuning of 80 *GHz* in non-Hermitian devices simply by changing the bias voltage.

In terms of spectral bandwidth, three active regions with different emission frequency range were stacked to expand the spectral bandwidth, and the bandwidth of 1 *THz* were experimentally demonstrated [35, 36]. By improving the device process, M. Rösch et al. further increased the bandwidth to 1.9 *THz* [37]. As shown in figure 2(b); the spectral bandwidth exceeding 2.6 *THz* was reported recently by stacking more active region structures [10].

The output power is an important parameter for most applications of THzQCL. Initially, the power of THzQCL was at the range of hundred milliwatt. M. Brandstetter et al. made a breakthrough to the watt-level output by wafer-bonding a symmetrical structure [38]. Short after that, several groups also reported the output power exceeding 1 *W* by improving the active region and device process [39]. The up-to-date maximum power is 2.4 *W* [8]. For single mode devices, the output power of 1.35 *W* was obtained in a vertical cavity surface emitting QCL [40]. In 2020, Y. Jin et al. introduced a short cavity longitudinal coupling scheme, improving the radiation efficiency effectively. As shown in figure 3(a), the output power was increased to 2.03 *W*, with a differential quantum efficiency of 115 photons per electron [41].

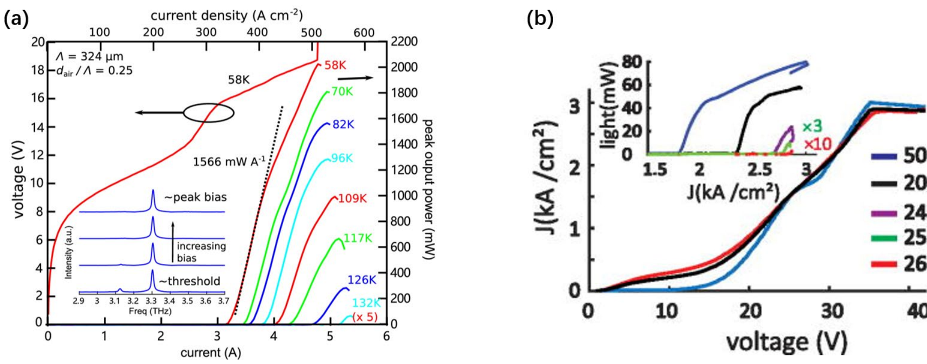

Fig. 3 Temperature dependent L-I-V curves for (a) the high-power single-mode device [41], (b) the high-temperature device [11].

The low operating temperature of THzQCLs is a main barrier to their commercialization. Due to the small photon energy of THzQCL (10 *meV*), the development of high-temperature or even room-temperature devices is extremely challenging. In the first ten years, the device could only work in the temperature range of liquid helium or liquid nitrogen. The operating temperature reached the thermoelectric cooling range till 2019, as a two-well THzQCL lased at 210 *K* (-63°*C*) [42]. A. Khalatpour et al. significantly increased the operating temperature to 250 *K* (-23°*C*) [43]., This laser provided a peak power of tens of milliwatts with a thermoelectric cooler at 230 K. As shown in Figure 3(b), the temperature was raised again to 261 *K* (-12 ℃) in 2023 [11]. The increase in operating temperature is mainly due to new active region designs.

4. THz frequency combs based on QCLs

A miniaturized terahertz frequency comb based on quantum cascade laser was successfully developed in 2014 [16]. The comb performance was largely affected by group velocity dispersion. In this work, a chirped waveguide structure was used for the dispersion compensation. The other methods such as integrating a Gires–Tournois interferometer (GTI) on the back of the laser and external cavity modulation were proposed. In 2021, A. D. Gaspare et al. used a single-layer graphene grating gated modulator for the dispersion compensation, and the frequency comb with 98 mode fingers was demonstrated, broadening the comb spectral bandwidth to 1.2 *THz* [44].

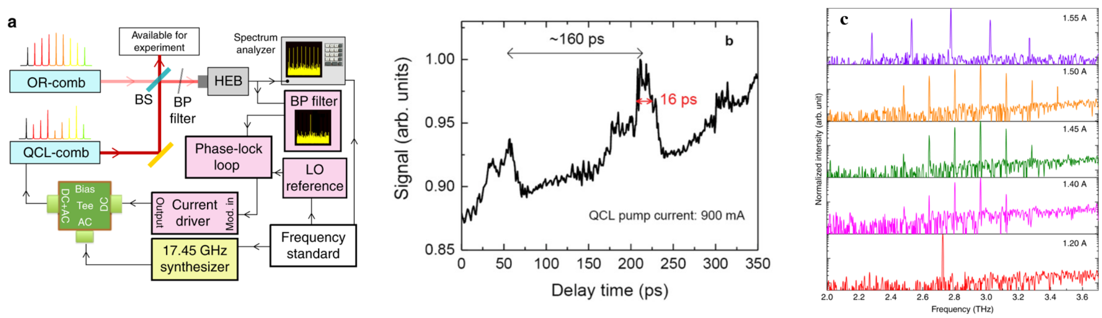

Fig. 4 (a) Schematics of the experimental setup employed for full stabilization of the THzQCL-comb based on dual-comb multi-heterodyne detection [46]. (b) Beam intensity of passively mode locked THzQCL-comb versus delay time [47]. (c) Spectra of THz frequency comb from mid-IR QCLs at room temperature continuous wave operation [48].

The most important feature of frequency comb is the phase relation among comb modes. Precise phase measurement and control methods are required for the correlation analysis. Shifted wave interference Fourier transform spectroscopy (SWIFT) was applied to measure the phase relation, but its real-time performance was limited by the scanning speed of mechanical components. For real-time measurement, a scheme combining dual-comb multi-heterodyne detection with Fourier transform analysis was demonstrated. The comb modes relation was monitored and the time profile of the comb emission was retrieved [45]. As shown in figure 4(a), the two degrees of freedom, the mode spacing and frequency offset, were independently controlled by using a similar scheme [46], so that the comb modes exhibited sub-hertz relative frequency stability and high coherence. This technology lays the foundation for metrology-grade applications. Pulsed THzQCL comb operation is quite challenging and requires active or passive mode locking. H. Li et al. utilized the saturation absorption of multi-layer graphene to generate terahertz pulses [47]. A GTI reflector formed by graphene and silicon lenses was used to compensate the dispersion of the laser gain medium. As shown in figure 4(b), this method resulted in a THz pulse of 16 *ps* and increase in the comb mode.

The cryogenic operation of THzQCL combs also limits their applications. For room temperature operation, a method based on frequency difference was demonstrated [48]. A mid-infrared QCL comb was integrated with a mid-infrared single mode QCL, and due to the nonlinearity of the laser materials, a room temperature THz comb was generated via down-converting their emission. As shown in figure 4(c), difference frequency generated five comb lines spaced 245 *GHz* between 2.2 and 3.3 *THz*.

5. New technologies

Terahertz quantum cascade lasers are mainly based on the GaAs material system. GaN and ZnO materials have larger LO phonon energy than GaAs based materials, which is expected to significantly increase the operating temperature of the THzQCL. The electroluminescence was observed in a GaN-based terahertz quantum cascade laser structure, but the development of lasers has not been successful. Recently, research on ZnO materials made progress, and THz electroluminescence was also observed in a ZnO/ZnMgO based quantum cascade structure for the first time [49]. As shown in figure 5(a), the device adopted a four-well quantum cascade structure and its luminescence centered at 8.5 *THz*. This result is an important step towards the ZnO-based THz lasers.

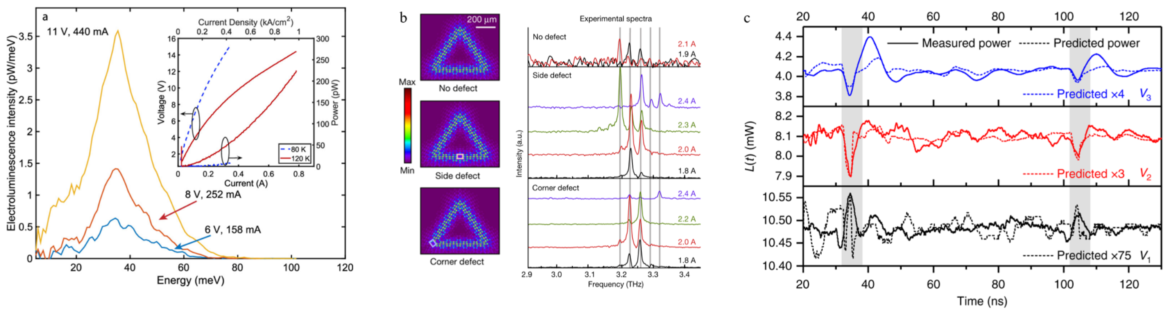

Fig. 5 (a) Electroluminescence spectra of ZnO/ZnMgO cascade structure under different biases. The inset shows the LIV (power-current-voltage) curves [49]. (b) Eigenmode electric field and the corresponding emission spectra of topological THzQCLs with no, side, and corner defect [50]. (c) Temporal output power responses of the acoustic modulated QCL at three different biases [14].

Quantum cascade lasers are sensitive to defects produced during device fabrication, which can lead to frequency variations among devices. In 2020, Y. Q. Zeng et al. reported a new terahertz topological laser with unprecedented stability and manufacturing repeatability [50]. The topological photonic structure was based on the valley Hall effect, and laser emission from topological edge states was observed in the device with triangular cavity. As shown in figure 5 (b), the device worked in multimode and its emission frequency was not affected by the introduction of defects. These results proved the robustness of topological protection, and opened the routes to new type of devices such as robust laser arrays and vortex lasers.

Terahertz quantum cascade lasers have picosecond carrier lifetimes, indicating the potential for high-speed modulation. Traditional electric modulation methods are fundamentally limited by the parasitic impedance. A new acoustic wave modulation technology was demonstrated [14], in which acoustic wave pulses excited by femtosecond laser were used to perturb the energy band structure of the THQCL, thereby achieving high-speed modulation of the laser radiation intensity. As shown in figure 5(c), the modulation rise time was 800 ps and the maximum modulation depth was 6%.

6. Discussions and perspectives

The main challenges include developing room temperature devices and expanding spectral coverage. For GaAs materials, new active region designs could reduce thermal leakage current and improve quantum efficiency. The low-loss waveguide materials, metasurface structures and new photonic crystals could also improve the device performance. In addition, quantum simulation and artificial intelligence could also help the design of new devices. ZnO and GaN based devices are expected to have higher operating temperature and broader spectral range. The improvement of the materials quality could result in the laser operation. The electron-phonon interaction in Si-based nonpolar materials is weak, which can suppress the thermal leak current. The inter-subband electroluminescence has been observed in GeSi materials [51]. Although the emission wavelength is in the mid-infrared range, it provides a reference for THzQCLs. For nanomaterials such as quantum dots, cascade structures are difficult to fabricate, but it is also another option for developing high-temperature devices.

Ultrashort pulsed THzQCL combs, high-power and low-noise optical frequency combs and dual-comb technology are worthy of attention. Soliton combs and harmonic combs also provide an interesting perspective for exploring complex nonlinear systems. THz topological optoelectronic devices are attracting more interests, such as robust laser arrays, unidirectional waveguides, high-order topological devices, wavefront engineered devices, and non-Hermitian devices.

The development of THzQCLs will further promote the applications, especially in high-resolution spectroscopy, hyperspectral imaging, and THz communications. Combined with near-field microscopy, THzQCL based systems can achieve nanometer resolution imaging, providing powerful methods for the research in fields such as material science and biomedicine.

Acknowledgments

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