Measurement of subband electronic temperatures and population inversion in THz quantum-cascade lasers

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(Received 8 November 2004; accepted 1 February 2005; published online 10 March 2005)

We compare the electronic temperatures and the population inversion both below and above the lasing threshold in three quantum-cascade lasers (QCLs) operating at 2.8 THz, 3.2 THz, and 3.8 THz using microprobe band-to-band photoluminescence. In the lasing range, while the ground-state temperature remains close to the lattice one (90 K–100 K), the upper radiative state heats up to ~200 K. From the measured thermal resistance and the power dependence of the ground-state electronic temperature, we get a value of the electron-lattice energy relaxation rate comparable with that typical of midinfrared QCLs. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1886266]

Quantum-cascade lasers (QCLs) promise mW-level continuous-wave (cw) power in the range (1–10 THz) for potential applications in spectroscopy, imaging, and sensing. Following the first report, based on chirped superlattices,[1] different schemes have been proposed for the gain medium design, namely, the resonant-phonon,[2–5] the bound to continuum,[6] and the interlaced photon-phonon[7] designs. In this letter, we focus on the resonant-phonon scheme, based on resonant tunneling and fast electron-longitudinal optical (LO) phonon scattering to selectively depopulate the lower radiative state. QCLs based on the above scheme and fabricated with high-confinement low-loss metal waveguides have been demonstrated in the range of 2.1 THz–3.8 THz, and have shown the highest operating temperature in both pulsed (137 K) and cw operation (97 K).[2–5] While early resonant-phonon QCLs displayed relatively high threshold current densities because of parasitic electronic transport channels,[2] significant improvements have been obtained using a design aimed at decreasing the parasitic coupling between the injector states and the initial state of the phonon-assisted transition in the next module.[3]

In this work, we report on the measurement of electronic and lattice temperatures and the relative subband populations in resonant-phonon THz QCLs. Detailed knowledge on the nature of the electronic distribution in THz QCLs is of paramount importance as a guide for the design of improved structures aimed at high-temperature operation. The existence of nonequilibrium electronic distributions in midinfrared (mid-IR) QCLs has been predicted theoretically[8–10] and assessed experimentally.[11–13] In THz QCLs, hot-electron distributions may arise from the detailed balance between the injection and the energy relaxation rates, i.e., inter- and intrasubband electron-electron (*e*-*e*), electron-LO phonon, electron-impurity, and interface roughness scattering. At electronic sheet densities $\geq 10^{11}$ cm^{-2}, the *e*-*e* scattering is fast enough to create Boltzmann-type subband distributions characterized by electronic temperatures T_e^i that may exceed the lattice one (T_L) at injected currents close to the laser threshold.[12,13] Subband thermalization and the related concept of subband temperature is questionable at densities $<10^9$ cm^{-2},[8,10] but it still holds at the densities used in THz QCLs ($3-5\times 10^{10}$ cm^{-2}).[9,14]

In THz QCLs, the photon energy is smaller than the LO phonon energy E_{LO} and the electron-LO phonon scattering between radiative subbands is energetically forbidden at very low electronic temperatures (T_e). However, the strong T_e dependence of the nonradiative relaxation rate $\tau_{5\to 4}^{-1} \propto \exp[(E - E_{\mathrm{LO}})/k_B T_e]$ significantly reduces the gain and increases the threshold current density at high T_e. We show here that optimizing the quantum design may improve the electron-lattice coupling and hence the electrical and optical performance of THz QCLs.[4]

We have compared three QCLs that operate at 2.8 THz (Sample a), 3.2 THz (Sample b), and 3.8 THz (Sample c).[15,16] The conduction-and valence-band structures for a single period of Sample a are shown in Figs. 1(a) and 1(b); the band structures of Samples b and c are similar. Our experimental method is based on the microprobe band-to-band photoluminescence (PL)[17] that proved successful for the investigation of mid-IR QCLs.[12,13,18,19] We kept the laser-induced electron heating below a negligible level (~3 K) by using an incident optical power of ~9 μW. Thus, the electronic distribution remains unperturbed and the laser excita-

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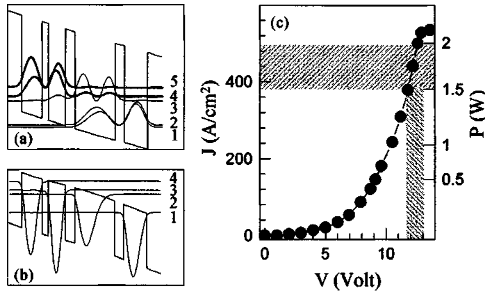

FIG. 1. Conduction (a) and valence (b) band structures of Sample a calculated with a voltage drop of 65 mV per stage using a self-consistent method based on the iterative solution of the Schrödinger and Poisson coupled equations. A 66% conduction-band offset is used. Beginning with the left-most injection barrier, the layer thicknesses measured in Å are 56/81/25/67/39/160/36/93. The underlined layer is doped at $n=1.9\times10^{16}$ cm^{-3} that corresponds to a sheet density of 3×10^{10} cm^{-2}. The energy levels are labeled using increasing integers starting from the ground state either in the conduction or valence bands. (c) Current density vs voltage characteristic of Sample a measured at the heat sink temperature of 50 K. The shaded area shows the lasing region. The right axis shows the electrical power (P).

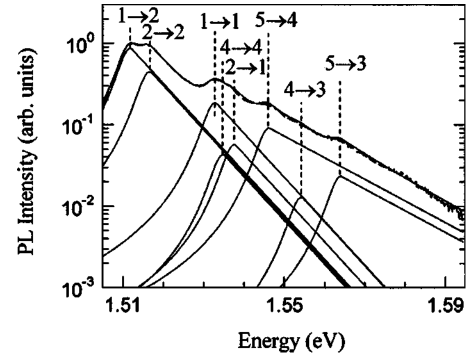

FIG. 3. Dashed line: PL spectrum at $P=1.5$ W. Solid line: Calculated PL components peaked at the theoretical energies of relevant $j\rightarrow k$ transition [see Figs. 1(a) and 1(b)]. The low-energy side of each curve is a Lorentzian with half width at half maximum=3.2 meV. The high-energy side is an exponential decay function $\propto\exp[-(E-E_{j\rightarrow k})/k_BT_e^j]$.

tion only provides holes for band-to-band radiative recombination.[20] The photoexcited holes quickly relax (~200 fs) to one of the valence subbands 1–4 [Fig. 1(b)] and can probe the electron population in the conduction subbands 1–5 [Fig. 1(a)].

Figure 2 shows a set of PL spectra for different values of the electrical power (P). We focus on Sample a, as its P values are sufficiently low to reach the lasing threshold ($P_{th}\sim1.5$ W, $J_{th}\sim400$ A/cm^2) well below the maximum heat dissipation rate of our microcryostat. Similar results are found for Samples b and c. Each spectrum shows a main peak that corresponds to the transition $1\rightarrow2$ between the injector ground state [level 1 of Fig. 1(a)] and the valence subband 2 [see Fig. 1(b)]. The energy E_P of this peak redshifts with P due to the Joule heating (inset, Fig. 2). To ease the comparison, each spectrum is plotted as a function of the energy difference ΔE with respect to the corresponding E_P value. The structure on the high-energy tail of the peak $1\rightarrow2$ is due to the allowed transitions $j\rightarrow k$ between conduction and valence subbands by comparison with the calculated energies E_{jk}.

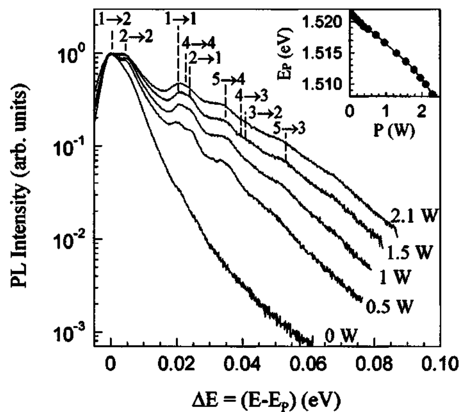

FIG. 2. Representative PL spectra of sample (a) at different electrical powers, each plotted as a function of the energy difference ΔE with respect to the corresponding main peak energy E_P. The heat sink temperature is 50 K. The dashed vertical lines labeled $j\rightarrow k$ mark the energies of the transitions between levels in the conduction (j) and valence (k) bands [see Figs. 1(a) and 1(b)]. Inset: Main peak ($1\rightarrow2$) energy E_P as a function of the electrical power. The line is a guide for the eyes.

The analysis of the PL line shape is based on the following expression:

$$I_{\mathrm{PL}}(E)\propto\sum_{j=1}^{5}\sum_{k=1}^{4}A_{jk}E_{jk}^4|\langle\psi_j|\psi_k\rangle|^2\mathcal{L}(E), \qquad (1)$$

where $A_{jk}=n_j\cdot p_k$, n_j, and p_k are the populations of the conduction and valence subbands. The term $\langle\psi_j|\psi_k\rangle$ is the overlap integral of the envelope functions. The line shape function $\mathcal{L}(E)$ is obtained joining a Lorentzian with a phenomenological broadening $\Gamma/2=3.2$ meV on the low-energy side, and an exponential decay $\propto\exp[-E/k_BT_e^j]$ on the high-energy side. T_e^j is the electronic temperature of the conduction jth subband. For $P\geqslant1$ W, an excellent reproduction of the PL is obtained considering the $j\rightarrow k$ transitions that have an overlap integral >0.2 and leaving T_e^j,A_{jk} as fitting parameters. Figure 3 illustrates the application of this method for the PL spectrum of Sample a measured at $P=1.5$ W.[21] However, for $P<1$ W, the results becomes unclear, since the number of allowed transitions considerably increases due to the lower localization of the wave function $j=5$ and the occurrence of resonances between subbands originating in adjacent periods. Therefore, in the range $P<1$ W we have restricted our analysis to the main PL band and estimated only the ground-state electronic temperature.

The fitting parameters T_e^j are plotted in Fig. 4(a) as a function of P together with T_L, extracted by comparing E_P against a calibration curve obtained by probing the device with zero injected-current while varying the heat sink temperature.[19] We found that the electronic temperatures of the subbands $j=1-4$ are nearly equal and increase linearly with P with a slope $R_e=28.0$ K/W, slightly larger than the thermal resistance $R=dT_L/dP=25.3$ K/W.[22] On the other hand, the temperature of the upper laser level T_e^5 reaches ~200 K in the range of $P=1$ W–2.2 W, i.e., it is higher by ~100 K than T_L. The existence of differences as high as 25–40% in the subband electronic temperatures is predicted by Monte Carlo simulations both in mid-IR[10] and THz[23] QCLs. In our case, we tentatively ascribe the large difference between T_e^5 and $T_e^{1,2}$ to the reduced efficiency of intersubband e-e scattering channels coupling electrons in the $j=5$ and $j=1,2$ levels, with respect to intrasubband e-e processes, as calculated for prototype THz QCLs structures.[14] One impor-

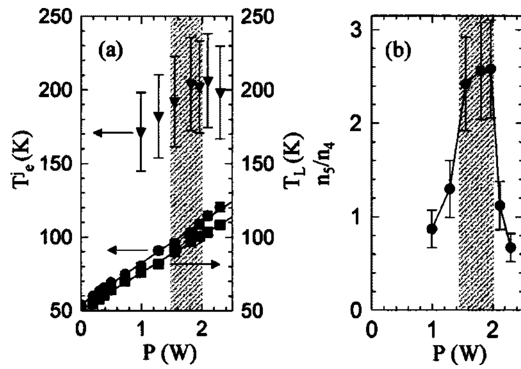

FIG. 4. (a): Mean lattice temperature (■) and electronic temperatures T_e^j; $j=1,2$ (●); $j=5$ (▼) in the active region of Sample a measured as a function of the electrical power at a heat sink temperature of 50 K. The lines are linear fits to the data. The small difference (~3 K) between T_e and T_L at zero-electrical power is a well known effect due to the heating of the electronic ensemble induced by the probe laser. (b) Relative populations n_5/n_4 between the upper ($j=5$) and lower ($j=4$) laser level as a function of the dissipated electrical power calculated for Sample a. The shaded areas mark the lasing region.

tant implication of our findings is that the high T_e^5 values lead to relatively fast nonradiative relaxation times $\tau_{5\rightarrow 4,3} \approx 1.3$ ps, and thus it is a key limiting factor for the operation at high temperatures of the investigated THz QCLs.

The expression $(R_e-R)^{-1}$ gives the strength of the electron-lattice energy relaxation rate $(\tau_E)^{-1}$. In resonant-phonon THz QCLs, the latter rate is controlled by the LO-phonon-assisted transitions $3,4\rightarrow 1,2$. Simple rate equation arguments give $\tau_E^{-1}=P/N_eNk_B(T_e-T_L)$, where N_e is the number of electrons per stage and N is the number of stages. Since we have shown in our system that the vast majority of electrons share the same temperature, we can rewrite $\tau_E^{-1}=[N_eNk_B(R_e-R)]^{-1}$. From the measured R_e and R values, we obtained $\tau_E^{-1}=4.9$ ps^{-1}, $\tau_E^{-1}=1.67$ ps^{-1}, and $\tau_E^{-1}=1.05$ ps^{-1} for Samples a, b, and c, respectively. The largest electron-lattice coupling for Sample a confirms the efficacy of its design that has been aimed at increasing the matrix elements associated with the $3,4\rightarrow 1,2$ transitions. Also, the energy separation between the above levels is slightly higher than E_{LO} at all values of the electric field above the alignment. The more efficient carrier thermalization in Sample a reduces thermal backfilling of the subband $j=4$, helps in keeping the optical gain closer to the designed value, and thus improves optical performance. In fact, the larger τ_E values in Samples b and c, respectively, are reflected in the higher measured laser thresholds: $J_{\mathrm{th}}=450$ A/cm^2 and $J_{\mathrm{th}}=630$ A/cm^2.

Figure 4(b) shows the ratio n_5/n_4 as a function of P for Sample a, determined from the ratio A_{54}/A_{44}. The population inversion ($n_5/n_4>1$) occurs when P is slightly >1 W, in agreement with the measured laser threshold of ~1.5 W. It then starts to decrease beyond 2.0 W where the injector subbands become misaligned with the $j=5$ level, the device enters a region of negative differential resistance, and lasing ceases. The fact that n_5/n_4 remains approximately constant during laser operation likely reflects the clamping of the gain (and thus the population inversion n_5-n_4) that occurs at lasing threshold.

In conclusion, we note that optimizing the phonon-mediated transitions ($3\rightarrow 2$, $3\rightarrow 1$) in THz QCLs leads to electron-lattice energy relaxation rates comparable with those of mid-IR QCLs ($\tau_E^{-1}=4$ ps^{-1}).[13] However, the performance is limited by the hot electron distribution in the upper radiative state.

This work was partly supported by MIUR (FIRB-RBAU01E8SS), ESA-ESTEC(16863/02/NL/PA), and by AFOSR, NASA, and NSF at MIT. One of the authors (B.W.) would like to thank H. Callebaut for helpful discussions. Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed–Martin Company, for the U. S. Dept. of Energy (Contract No. DE-AC04-94AL85000).

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[21] Note that the transition $3\rightarrow 2$ does not contribute to the luminescence, despite the large value of the overlap integral $(\langle\psi_3|\psi_2\rangle=0.4-0.5)$ in the whole range of investigated powers and thus demonstrates that the electron-LO phonon interaction efficiently depletes the subband $j=3$.
[22] The thermal resistance of Devices b and c are $R=18.6$ K/W and $R=18.0$ K/W. These values are 2.5–3.5 times larger than in mid-IR QCLs, due to the thicker active layers used for THz QCLs.
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