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Miriam S. Vitiello,1,a) Rita C. Iotti,2 Fausto Rossi,2 Lukas Mahler,3 Alessandro Tredicucci,3 Harvey E. Beere,4 David A. Ritchie,4 Qing Hu,5 and Gaetano Scamarcio1,b)
1CNR-Istituto di Fotonica e Nanotecnologie UOS Bari and Dipartimento Interateneo di Fisica “M. Merlin,” Università degli Studi di Bari, Via Amendola 173, 70126 Bari, Italy
2Dipartimento di Scienze Applicata e Tecnologia, Politecnico di Torino, Corso Duca degli Abruzzi 24, 10129 Torino, Italy
3NEST, CNR-Istituto Nanoscienze and Scuola Normale Superiore, Piazza san Silvestro 12, 56127 Pisa, Italy
4Cavendish Laboratory, University of Cambridge, Madingley Road, Cambridge CB3 0HE, United Kingdom
5Department of Electrical and Computer Science and Research Laboratory of Electronics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA

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We report on the experimental observation of non-equilibrium longitudinal (LO) and interface transverse-like (IF-TO) optical phonons populations associated with electron transport in resonant-phonon THz quantum-cascade lasers. The measured LO-phonon occupation numbers are in excellent agreement with the result of Monte Carlo simulations at a phonon in-plane wave number \( q = 4.2 \times 10^{-14} \text{ cm}^{-1} \), and they remain about 40% larger than the IF-TO ones. © 2012 American Institute of Physics. [doi:10.1063/1.3687913]

In semiconductor heterostructures and related devices, the energy relaxation of carriers is mostly controlled by the electron-phonon interaction.1 If the excess energy supplied to the electronic ensemble is sufficiently high, the electron cooling processes may generate non-equilibrium optical-phonon populations which, in turn, may cause substantial changes in the carrier relaxation kinetics.

The latter effects are expected to be very pronounced in quantum cascade lasers (QCLs) in which the light-emission mechanism involving intersubband transitions between quantized conduction band energy states exploits the electron-phonon interaction to achieve population inversion.

In a terahertz QCL, the energy relaxation of carriers proceeds via a combination of intra- and inter-subband transitions, while the dominant non-radiative e-e-scattering modifies the electron distribution among the laser subbands. However, when energetically permitted, as in the case of resonant-phonon active region designs,2 the electron-optical phonon interaction may have a major role as inter-subband relaxation mechanism and may lead to the creation of a non-equilibrium optical phonon population.3 Considering the potential energy drop per stage necessary for band structure alignment and the energies of the relevant optical phonons, in a resonant-phonon THz QCL,9 each electron generates at least one optical phonon per stage. Since resonant-phonon THz QCL structures employ more than one hundred periods, a large rate of optical phonon generation can be achieved. This means that the inclusion of the phonon ensemble in the band structure engineering is necessary to give a more realistic description and control not only on the electronic processes and lifetimes but also on the phonon processes as well.

In addition, the large hot-optical phonon generation achievable in principle in THz QC structures could be exploited to use similar structures as sources of quasi-monochromatic optical phonon distributions. Such a device would have potential applications for the generation of high-frequency phonon beams used in phonon optics, phonon spectroscopy, and nanostructures imaging.4

In the present letter, we report on the experimental observation of non-equilibrium optical phonon populations generated by electron transport in THz QCLs emitting at 3.2 THz.5

The investigated devices are based on GaAs/Al\(_{0.15}\)Ga\(_{0.85}\)As quantum cascade heterostructures employing a resonant phonon gain medium design.5,6 The active region, grown by molecular beam epitaxy on an undoped GaAs substrate, includes \( N = 200 \) periods for a total thickness of \( \sim 10.8 \mu \text{m} \) and is embedded between two contact GaAs layers. Their thickness/doping values are \( 80 \text{ nm}/5 \times 10^{18} \text{ cm}^{-3} \) and \( 700 \text{ nm}/3 \times 10^{18} \text{ cm}^{-3} \). A schematic diagram of the active region design is sketched in Fig. 1. At variance from chirped superlattice and bound-to-continuum designs relying on intraminband scattering and transport for depopulation of the lower radiative state, the investigated design uses resonant tunneling and subpicosecond longitudinal-optical (LO)-phonon scattering assisted depopulation. Moreover, the photon energy is smaller than the longitudinal-optical phonon one (\( E_{\text{LO}} = 36 \text{ meV} \) in GaAs), thus thermally activated LO-phonon scattering leads to a rapid decrease of the upper state lifetime at an increasing temperature, which reduces the laser gain.

The wafer (sample a) was processed in a double-metal waveguide using gold-gold wafer bonding followed by substrate removal and processed into 82-μm wide ridges by wet etching. Devices were metalized before wafer bonding with Ti/Au (5 nm/700 nm) and the ridges were contacted with Ti/Au (5 nm/200 nm). 5-μm wide, 10-μm long top apertures have then been defined by optical lithography on top of each metalized ridge, and the 80-nm heavily doped upper cladding layer was removed by wet etching using the top metal
The Raman spectrum (Refs. 6–8) and micro-Raman spectroscopy.3 PL and Raman identification of microprobe band-to-band photoluminescence (PL) waveguides inhibit in this case the laser action.

The higher cavity losses of the single plasmon guides. The latter procedure has the purpose of controlling with a calibrated Si-diode mounted close to the front facet, down to a spot of about 2-μm diameter by using an 80× long-working-distance plano-achromatic microscope objective lens. Raman experiments were performed in a backscattering configuration by probing the pumping laser on the laser facet and on a small aperture on the top surface in order to have access to both interface-transverse optical (IF-TO) or LO phonons, respectively. Indeed, wavevector conservation selection rules allow for interface phonon observation in backscattering from the sample edge. In contrast, LO phonons can only be observed in backscattering from the (001) surface.

The excitation wavelength has been chosen to avoid any resonant effect in the Raman spectra. At ℏωL = 1.92 eV only TO or transverse interface modes are not affected by electronic resonances, while to detect non-resonant LO phonons, a frequency ℏωL = 2.60 eV has been used. The absence of any resonant effect is confirmed by the measured value of the ratio between second order and first order intensities of the Raman spectrum (≈6%), being approximately equal to the expected thermal value.3

The devices were mounted into a helium-flow microcryostat and kept at a fixed heat sink temperature T_s = 56 K controlled with a calibrated Si-diode mounted close to the device. The sample position was varied using a two-dimensional translation stage with a 0.1-μm spatial resolution, along either the z or the x axis. The PL and Raman signals were dispersed using a 0.64-m monochromator and detected by a Si charge coupled device (CCD) cooled to 140 K. We used an interference notch filter to suppress the Rayleigh scattering and simultaneously record both Raman Stokes (S) and anti-Stokes (AS) spectra.

The measured Raman S/AS spectrum gives an estimate of the phonon population. In fact, the ratio ρ between the S/AS intensities for a given phonon mode is related with the phonon population via the relation,

\[ N = \left[ \frac{\rho \sigma_{AS}(\omega_L)}{\sigma_S(\omega_L)} \left( \frac{\omega_L + \omega_0}{\omega_L - \omega_0} \right)^4 - 1 \right]^{-1}, \]

where N is the phonon occupation number, ω0 is the optical phonon frequency, \( \sigma_{AS}(\omega_L) \) and \( \sigma_S(\omega_L) \) are the S and AS Raman cross sections at the laser frequency \( \omega_L \).

After processing, the samples were cleaved in 0.64-mm long cavities and indium soldered on copper mounts. In addition, non-lasing devices of comparable areas (sample b) and long cavities and indium soldered on copper mounts. In addition, non-lasing devices of comparable areas (sample b) and identical active region design were fabricated by optical lithography and wet etching in single plasmon-optical waveguides. The higher cavity losses of the single plasmon waveguides inhibit in this case the laser action.

The phonon population has been measured by a combination of microprobe band-to-band photoluminescence (PL) (Refs. 6–8) and micro-Raman spectroscopy.3 PL and Raman spectra were recorded by focusing the 647-nm line of a Kr laser, working in continuous mode, directly onto the QCL facet, down to a spot of about 2-μm diameter by using a 80× long-working-distance plano-achromatic microscope objective lens. Raman experiments were performed in a backscattering configuration by probing the pumping laser on the laser facet and on a small aperture on the top surface in order to have access to both interface-transverse optical (IF-TO) or LO phonons, respectively. Indeed, wavevector conservation selection rules allow for interface phonon observation in backscattering from the sample edge. In contrast, LO phonons can only be observed in backscattering from the (001) surface.

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charge carriers and lattice degrees of freedom in resonant-phonon device designs, we have, therefore, considered the full electron subsystem, i.e., the complete set of active-region and injector subbands – interacting with out-of-equilibrium bulk (dispersionless) LO-phonon modes. The latter are assumed to decay anharmonically into thermalized acoustic modes (lattice thermal bath) by means of a phenomenological lifetime $\tau$.\textsuperscript{10} In particular, within the Fermi’s golden rule approximation, the LO-phonon interaction translates into the following term in particular, within the Fermi’s golden rule approximation, the electron subsystem, i.e., the complete set of active-region and phonon device designs, we have, therefore, considered the full

generators of the model, $S$ and $T_L$, are concerned, the former is reasonably set to 6 ps,\textsuperscript{13} while the latter is given by the value, which varies with $P$, measured by PL experiments we performed on the operating device. Simulated results, shown in Fig. 2, obtained for our prototypical resonant-phonon terahertz emitting device at a phonon in-plane wavenumber $q = 4.2 \times 10^{14}$ cm$^{-1}$, show a very good agreement with measured data, indicating how the out-of-equilibrium LO phonon population affects the electro-optical device performances.\textsuperscript{13} Conversely, our theoretical approach also allows us to directly investigate the features of the LO phonon distribution over the wavevector $q$, due to the non-trivial coupling with the “hot” electron subsystem.

Figure 3 shows the comparison between the LO and IF-TO like phonon occupation numbers in the investigated THz QC laser (sample a) as a function of $P$. Four regions, clearly correlated with features in the transport measurements (Fig. 3(b)) can be identified. At $P < 2.5$ W, (region I) band structure calculations show that the energy of the injector doublet remains more than 6 meV lower than that of the upper state and transport proceeds via injection into level 3. Under these conditions, the phonon population is approximately thermal. As carriers start being injected into the upper state (region II) $(2.5 < P < 3.3$ W), a non-equilibrium phonon population arises in our structure that increases as the injector level 1 approaches the resonance with subband 5 $(P = 3.4$ W). The large negative discontinuity in the differential resistance at the onset of region III $(2.5 < P < 3.3$ W) indicates that the laser threshold is reached, while the population inversion becomes clamped at the onset for stimulated emission.\textsuperscript{14} Correspondingly, the slope of $N$ vs power decreases implying that the photon emission extracts part of the input power, efficiently cooling both the electron\textsuperscript{15} and optical phonon subsystems. This behavior is very similar to that observed for hot electrons in a terahertz QCL.\textsuperscript{15} A further proof of the optical origin of the latter effect is the fact that this change disappears when the same measurements are performed on a non-lasing device (sample b) as shown in Fig. 2. Finally, in region IV $(P > 4.3$ W) lasing ceases, as revealed by the sudden jump in the differential resistance (Fig. 3(b)).

In this rollover regime, the voltage drop in each stage and the associated subband structure become ambiguous.

\begin{equation}
\frac{d}{dt} f_{\nu k} = \sum_{q} \left( N_q + \frac{1}{2} \right) \sum_{\nu'k'} \left( 1 - f_{\nu' k'} \right) \frac{P_{\nu' k' \rightarrow \nu k} q^2}{2\pi h} - \left( 1 - f_{\nu k} \right) \frac{P_{\nu k \rightarrow \nu' k'} q^2}{2\pi h},
\end{equation}

where $N_q$ is the phonon population of the LO-modes with wave vector $q$, $P_{\nu k \rightarrow \nu' k'} q^2 = C_2 (2\pi h) |g_{\nu k \rightarrow \nu' k'}|^2 \delta(\epsilon_{\nu k} - \epsilon_{\nu' k'} + \epsilon_q)$, where $g_{\nu k \rightarrow \nu' k'}$ is the Frohlich carrier-phonon coupling coefficient, and the sign $\pm$ refers to emission (+) or absorption (−) processes. To properly model the carrier-phonon interacting systems, the Boltzmann transport equation for the electron subsystem, with the contribution in Eq. (1), goes together with the following phonon counterpart:

\begin{equation}
\frac{d}{dt} N_q = \sum_{\nu k} \left( N_q + \frac{1}{2} \right) \sum_{\nu'k'} \left( 1 - f_{\nu' k'} \right) \frac{P_{\nu' k' \rightarrow \nu k} q^2}{2\pi h} - \frac{N_q - N_q^{th}}{\tau},
\end{equation}

with $N_q^{th}$ is Bose-Einstein distribution at the lattice temperature $T_L$. The kinetic equations (2) and (3) are sampled by means of a Monte Carlo simulation scheme able to deal with a variable number of phonons on one side and a “charge conserving scheme” for the electrons on the other. The latter is obtained by imposing periodic boundary conditions. To properly account for relevant intra- as well as inter-subband scattering mechanisms, besides carrier-LO phonon scattering, we have also included carrier-carrier interaction in the electron dynamical equation. For the electron-electron interaction, here we employ the well-established time-dependent static-screening approach commonly used in Monte Carlo simulations of energy relaxation and transport phenomena in two-dimensional systems. Indeed, the impact of more refined screening models on the non-equilibrium many-body dynamics in these devices is still debated.\textsuperscript{11} To limit the number of free-parameters, extrinsic, and strongly material/device dependent, scattering mechanisms, such as carrier-impurity and interface roughness,\textsuperscript{11,12} have not been included in the present simulations. As far as the two phenomenological parameters of the model, $\tau$ and $T_L$, are concerned, the former is reasonably set to 6 ps,\textsuperscript{13} while the latter is given by the value, which varies with $P$, measured by PL experiments we performed on the operating device. Simulated results, shown in Fig. 2, obtained for our prototypical resonant-phonon terahertz emitting device at a phonon in-plane wavenumber

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig3}
\caption{(Color online) (a) LO (●) and IF-TO (▲) phonon occupation numbers in a terahertz quantum cascade laser plotted as a function of $P$, together with the corresponding equilibrium phonon populations (●). (b) Voltage-power and differential resistance vs power characteristics collected at $T_L = 57$ K, while driving the QCL in continuous wave mode. The inset shows a schematic diagram of the employed experimental configuration. The arrows show the direction of the incident and scattered photons in the Raman processes. The grey area show the lasing regime.}
\end{figure}


