Hot-phonon generation in THz quantum cascade lasers

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Abstract. Observation of non-equilibrium optical phonons population associated with electron transport in THz quantum cascade lasers is reported. The phonon occupation number was measured by using a combination of micro-probe photoluminescence and Stokes/Anti-Stokes Raman spectroscopy. Energy balance analysis allows us to estimate the phonon relaxation rate, that superlinearly increases with the electrical power in the range 1.5 W – 1.95 W, above laser threshold. This observation suggests the occurrence of stimulated emission of optical phonons.

1. Introduction

In semiconductor structures the emission of optical phonon is the dominant process that controls the energy relaxation of carriers and the typical value for the electron-optical phonon scattering rate is $(2\mathrm{ps})^{-1}$. The anharmonic decay into acoustical modes controls the lifetime of optical phonons, which is in the range 3-9 ps [1]. Thus, for large excited carrier densities, the process of electron cooling may generates a significant non-equilibrium (hot) optical-phonon population. Electrical methods to create a hot population of selected phonons have been proposed [2] and stimulated emission of acoustic phonons in a GaAs/AlAs superlattice has been recently observed [3]. In a previous investigation we have demonstrated the generation of a hot-optical phonon population associated with electron transport in InGaAs/AlInAs-based mid-IR quantum cascade lasers (QCLs) [4].

In QCLs, electrons experience a sequence of transitions down the ladder of electronic states of a periodic structure [5]. In THz QCLs energy relaxation of electrons injected by resonant tunnel into excited subbands proceeds via intra- and inter-subband transitions, the dominant non-radiative channel generally being e-e-scattering. When energetically permitted, as in the case of a resonant-phonon THz QCL, scattering due to electron-optical phonon interaction has a major role as inter-subband relaxation mechanism and may lead to the creation of a nonequilibrium optical phonon population. Considering the potential energy drop per stage necessary for band structure alignment and the energies of relevant optical phonons, in a resonant-phonon THz QCL each electron generates at least one optical phonon per stage. Since THz QCLs may include more than one hundred of periods, quite large optical phonon generation rates can be achieved. Inclusion of the phonon ensemble in the band structure engineering will give a more realistic description and control not only on the electronic processes and lifetimes, but also on the phonon processes as well. However, the large hot-optical phonon generation achievable in QC structures could be exploited to use this kind of laser as a source of optical phonons. Such a device, would have potential applications for the generation of high-frequency phonon beams used in phonon optics, phonon spectroscopy and nanostructures imaging [6].

In this work we report the observation of a non-equilibrium optical phonon population generated by electron transport in a resonant–phonon THZ QCL emitting at 2.8 THz operated in dc mode below and above laser threshold, using a combination of micro-probe Stokes/anti-Stokes (S/AS) Raman and photoluminescence (PL) spectroscopy.

2. Results and discussion

The investigated structure includes a 10-μm thick active region composed of 176 periods of a GaAs/$Al_{0.15}Ga_{0.85}$ heterostructure grown by MBE on semi-insulating GaAs (001). The conduction and valence band structure and the optical and electrical characteristics have been reported elsewhere [7]. The microprobe apparatus used for Raman and PL measurements is described in Ref. 4. The PL and Raman signals were obtained by focusing the 647.1 nm line of a Kr^+ laser onto the QC laser front facet to a spot of ~ 1 μm diameter with an 80x microscope objective. Raman measurements were performed using an incident power density of ~3 $\times 10^4$ W cm^{-2}. To suppress the Rayleigh scattering and simultaneously record both S and AS spectra we used an interference notch filter. To determine the local lattice temperature, we exploited an experimental approach based on the measurement of band-to-band PL spectra in operating QCLs, using the shift of the PL spectra as a thermometric property [8] and to avoid additional heating of the sample we used a low incident power density (~10^3 W/cm^2), also thanks to the much higher PL signal with respect to the Raman one.

From the analysis of the Raman spectra, measured in backscattering from the laser facet ($1\bar{1}0$ direction), we determine the phonon population. In fact, the phonon occupation number N can be determined from the ratio ρ of the S/AS intensities for a given phonon mode:

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

where $\sigma_S(\omega_L)$ and $\sigma_{AS}(\omega_L)$ are the S and AS Raman cross sections at the laser frequency ω_L and ω is the phonon frequency. These cross sections have a strong ω_L dependence close to fundamental electronic resonances, thus relation (1) cannot be used under these conditions. If N is higher than its equilibrium value at the lattice temperature (N_0), the phonon population is out of thermal equilibrium and $N' = N - N_0$ are the phonons generated in the non-equilibrium process.

In mid-IR QC lasers, the strongest electron–phonon interaction is associated with in-plane propagating interface phonons [9]. However, as demonstrated in GaAs-based superlattices [10], larger well widths, as those typically used in the case of THz QCLs, lead to a reduction of the electron-IF scattering and to prevailing electron-LO and TO phonon interactions. It is necessary to keep in mind that:(*i*) the observation of LO phonons in backscattering from the device facet ($1\bar{1}0$) is forbidden by the Raman selection rules, while the observation of transverse interface modes (IF_{TO}) is allowed when using a crossed or parallel configuration of polarization [11]; (*ii*) the symmetry selection rules are broken close the resonances. However near resonance conditions equation (1) is not applicable. Hence, we have focused our attention on the study of scattering processes by GaAs-like IF_{TO} and the Raman signals were detected using an exciting laser energy of $\hbar\omega_L$=1.92 eV (647.1 nm), for which the GaAs-like TO phonons Raman signals are expected to be not affected by electronic resonances [12].

Note that, due to Raman scattering momentum conservation rules, in our experiments we can probe only phonons having a wave vector of ~0.75×10^6 cm^{-1} and in the device can be generated also phonons with larger wave vectors. Therefore, N' represents the nonequilibrium population of the zone-center IF_{TO} phonon modes.

Figure 1 shows three typical S and AS Raman spectra taken from the active region in crossed polarization condition, while no current flows in the device and with injected currents I = 89 mA and I = 148 mA, respectively below and above laser threshold. The corresponding dissipated electrical power are P = 1 W and P = 1.85 W. The Raman spectra, as predicted by the selection rules, reveal the presence of GaAs-like IF_{TO}.

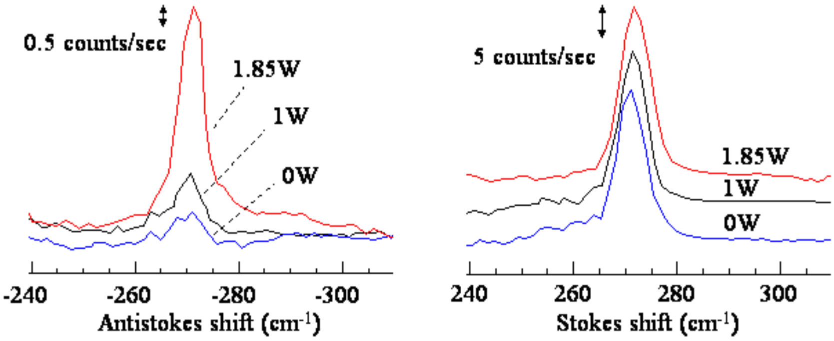

Figure 1. Stokes and anti-Stokes Raman spectra measured with device off, P = 1 W and P = 1.85 W. The heat sink temperature was kept at 80K.

In order to check that no significant resonance effects exists under the chosen excitation condition, we studied the ratio ρ varying the energy of the electronic transition, by changing the lattice temperature. Figure 2a shows the intensity ratio between AS and S lines, measured for the GaAs-like IF_{TO}, by probing the device with zero current, while varying the heat sink temperature (T_H). The experimental data were fitted by means of the equation $y=k\cdot exp[\hbar\omega/k_B\cdot(T_H+B)]$. The parameter B represents the heating effect induced by the Kr^+ pump laser. An excellent reproduction of the experimental data was obtained using k = 1.21 ± 0.015 and B = 23.5 ±1.7 K. In figure 2b is reported the additional device heating induced by the incident Kr^+ exciting laser power, extracted from PL measurements with device-off. The obtained B value agrees very well with the temperature increase (~24.3 K) measured at the power densities of 3×10^4 W cm^{-2}, employed in the Raman measurements.

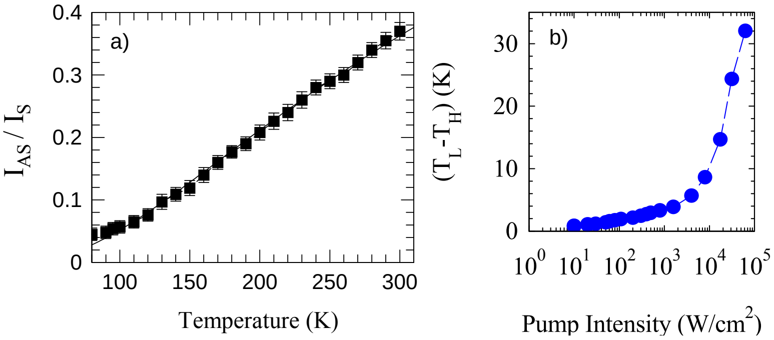

Figure 2. (a) Ratio I_{AS}/I_S, plotted as a function of T_H obtained with the device off. The solid line represents the fit function $y=k\cdot exp[\hbar\omega/k_B\cdot(T_H+B)]$. b): device lattice heating induced by the exciting Kr^+ laser plotted as a function of the laser incident power density.

The k value extracted from the fit corresponds to a constant ratio $\sigma_{AS}/\sigma_S = 1.04 \pm 0.015$ in the investigated temperature range 80-350 K, thus demonstrating the absence of resonant effect on the GaAs-like IF_{TO}. Moreover, the intensity ratio between two-phonon and one-phonon Raman bands in our spectra is about 6%, which is typical under off-resonance conditions. Thus, we can safely use the relation (1) to extract the phonon occupation number N.

The thermal equilibrium values N_0 can be readily obtained from the expression $N_0=1/\exp(\hbar\omega/k_B\cdot T_L)$. The local lattice temperature $T_L= T_H+\Delta T_1+\Delta T_2$ were determined by considering the two different factors that make T_L different from the heat-sink one:(*i*) the heating effect ΔT_1 induced by the Kr^+ laser high pumping power density (~3×10^4 W/cm^2); (*ii*) the Joule heating effect ΔT_2 induced by the dissipate electrical power. The first contribute can be extracted by fig. 2b (ΔT_1=24.3 K); the second one was extracted from the PL measurements. In this case, we have used a low incident power (~10^3 W/cm^2), so keeping the Kr$^+$ induced heating less than 3 K (see fig. 2b).

The N and N_0 values for GaAs-like IF$_{TO}$ phonons are plotted in figure 3 as a function of P, together with the excess phonon population $N'=N-N_0$ values. The N values remain nearly equal to the calculated N_0 values at low dissipated power (< 1.5 W), while significantly departs in the lasing range.

Energy balance analysis allows to estimate the phonon relaxation rate, i.e., the scattering rate of GaAs-like IF$_{TO}$ phonons with respect to the total phonon scattering rates in our system. The relation between the electron (G_e) and phonon (G_p) generation rates per unit volume is $G_p=NG_e=(\Delta E/\hbar\omega)\cdot G_e$, where N is the number of phonons generated by a single electron, ΔE is the total electronic energy variation in the cascade process. The phonon generation rate must be equal to the phonon decay rate. Under the assumption that the optical phonons decay before they equilibrate within the optical branch and deformation potential-type interaction, one readily obtains:

$$T_r = \tau_p \cdot r = \frac{N'}{I\Delta V}\cdot\frac{\hbar\omega}{6\pi^2}Ad\cdot\left(k_{\max}-k_{\min}\right) \tag{2}$$

where I is the injected current, ΔV the applied voltage, A the device area, d the thickness of the active region, k_{min} and k_{max} are the electron wavevector limits, τ_p is the GaAs-like IF$_{TO}$ lifetime and r is the relative phonon relaxation rate.

Figure 4 shows the estimated T_r as a function of the dissipated electrical power, at T_H = 80 K. At low dissipated power (P < 1.5W) we found a nearly constant value ($T_r \sim 0.5$ ps), correspondingly the lattice temperature increases up to ~ 144 K [7]. Experimental investigations of the GaAs-like TO phonon lifetime give a value $\tau_p \sim 5$ ps at temperature T = 80 K [13], thus the observed T_r value indicates that ~ 10 % of the total electron-phonon energy relaxation processes involves GaAs-like IF$_{TO}$ phonons.

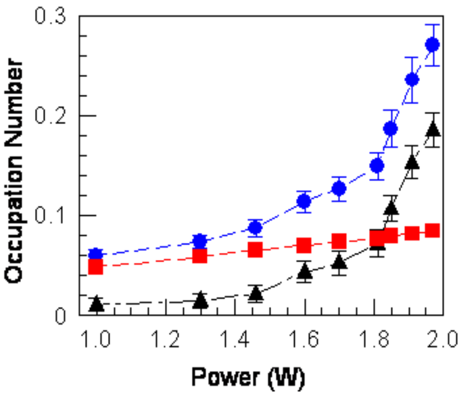

Figure 3. (color online) GaAs-like IF$_{TO}$ phonon occupation numbers N(●) determined by Raman S and AS measurements as a function of the electrical power, displayed together with the equilibrium phonon population N_0 (■) and the excess phonon population N'(▲).The dashed curves are guides for the eye.

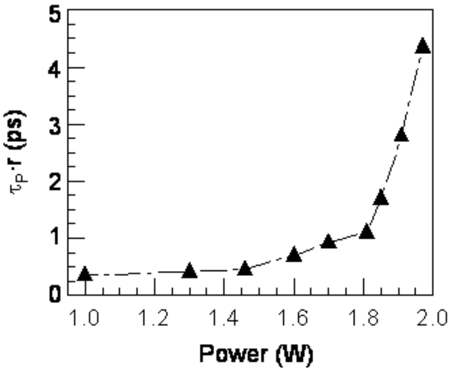

Figure 4. Estimated T_r values plotted as a function of the dissipated electrical power, at an heat sink temperature of 80 K. The dashed line is a guide for the eye.

However, above the lasing power threshold (~ 1.4 W) we observed a *super linear* increases of the phonon occupation with the electrical power (see Fig. 3) and consequently an increase of T_r, that reaches a value of ~ 5 ps at P = 1.95 W, correspondingly the lattice temperature rises up to ~153 K. A phonon lifetime decreases should be expected at increasing temperature, consequently our observation implies an increases of the GaAs-like IF_{TO} phonon relaxation rate *r*. This behavior is characteristics of phonons stimulated emission, which seems to be triggered by the active region subbands population inversion. Generation of coherent phonons has been theoretically predicted, as a result of population inversion in the electron subsystem, in the case of an electrically biased doped semiconductor superlattice in the hopping electron transport regime [2], i.e. phonon-assisted transitions of carriers between bound-electron states in adjacent quantum wells, a condition very similar to the case of QCL structures. It is worth noting that we do not observed any spectral linewidth narrowing, but the full width half maximum of the GaAs-like IF_{TO} peak remains nearly constant (~ 6 cm^{-1}). This unchanged phonon signal lineshape can be explained considering that all the different GaAs wells composing the active region heterostructure contributes to the GaAs-like IF_{TO} Raman signal and the fluctuations of the GaAs layers width causes a phonon peak broadening which may hidden the observation of linewidth narrowing.

3. Conclusion

In summary, we have observed non-equilibrium optical phonon generation in an operating GaAs/AlGaAs THz QCLs device. Above laser threshold, we observed a super linear increase of the GaAs-like IF_{TO} relaxation rate with the electrical power, which suggest the occurrence of stimulated emission of phonons.

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