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Citation: Applied Physics Letters **82**, 2455 (2003); doi: 10.1063/1.1566467
View online: http://dx.doi.org/10.1063/1.1566467
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/82/15?ver=pdfcov
Published by the AIP Publishing

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(Received 7 October 2002; accepted 12 February 2003)

The evolution on the picosecond scale of the macroscopic (nonequilibrium thermodynamic) state of a highly excited photoinjected plasma in bulk GaN is analyzed. We present the equations of evolution for the quasitemperature (level of excitation) of the hot carriers and of the optical phonons. A hot-phonon temperature overshoot is evidenced, as well as a preferential production of phonons in excess of equilibrium in a reduced off-center region of the Brillouin zone. A comparative analysis of the influence of the length of the exciting laser pulse is also performed. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1566467]

Group-III nitrides (mainly AlN, GaN, InN and their ternary compounds) are receiving considerable attention as a result of the technological advantages they provide when used in diodes and lasers emitting in the blue-green and ultraviolet spectral regions.[1] Despite intensive efforts expended in recent years, the light-emission mechanisms in III-nitride-based devices are not completely understood, either in the case of low-dimensional (wells, wires, and dots) or tridimensional (mainly epilayers) systems. Quantum confinement, interface related states, compositional inhomogeneities, and polarization (spontaneous and piezoelectric) effects are the key competing mechanisms for the understanding of the light emission characteristics of III-nitride-based low-dimensional devices.[2] On the other hand, research for the understanding of light emission from tridimensional III-nitride samples have been focused mainly on GaN, whose stimulated emission at low temperatures is known.[3]

Excitonic and electron–hole plasma recombination, below and above the Mott transition (roughly at 10^{18} cm^{-3}), respectively, are recognized as the key mechanisms of light emission from tridimensional GaN epilayers.[4–11] Band-to-band instead of excitonic transition was shown to be the dominant transition in metalorganic-chemical-vapor-deposition-grown GaN epilayers at room temperature.[4] The stimulated emission threshold in SiC/GaN and sapphire/GaN films was assigned recently to be due to excitonic processes at carrier temperatures smaller than 200 K, and to the existence of an electron–hole plasma when higher.[7] All these results suggest that recombination in the latter is the gain mechanism in GaN-based blue lasers. Consequently, for a deep understanding of the optical gain mechanisms in III-nitride-based optoelectronic devices, it is necessary to focus on the ultrafast processes developing in the coupled nonequilibrium hot-carrier and hot-phonon systems during the ultrafast dissipation of their energy in excess of equilibrium.

We consider this question, performing a theoretical study of the ultrafast phenomena developing in the photoinjected plasma in GaN using a kinetic theory derived from a Gibbs' ensemble formalism for nonequilibrium systems.[12] An extensive detailed description shall be given elsewhere;[13] here, we report and comment on the main physical results. To deal with the irreversible thermodynamic evolution and transport properties of an electron–hole plasma in GaN, we recall that the statistical formalism requires, as a first step, the choice of a basic set of variables to describe the macrostate of the nonequilibrium semiconductor system. For the present case, we select the carriers' Hamiltonian $\hat{H}_o$, and number operators $\hat{N}_{\rm e}$ and $\hat{N}_{\rm h}$ (e stands for electrons, and h for holes), and the number of LO phonons, $\hat{\nu}_{\vec{q}}=b_{\vec{q}}^{\dagger}b_{\vec{q}}$, where $b_{\vec{q}}(b_{\vec{q}}^{\dagger})$ are annihilation (creation) operators of longitudinal optical (LO) phonons in mode $\vec{q}$. The LO phonons are the relevant ones in the highly polar III-nitrides, and those which are specifically warmed up in the process, while the transverse optical (TO) phonons and acoustic (AC) phonons are slightly warmed up, and considered as remaining in equilibrium with the thermal reservoir at temperature T_o.

The average values over the nonequilibrium ensemble of these dynamical quantities, that is, the basic macrovariables which characterize the nonequilibrium thermodynamic state of the system, are the carriers' energy $E_c(t)$ and density $n(t)$ (the same for electrons and holes since they are produced in pairs),[14] and the LO-phonon population in each mode $\vec{q}$, $\nu_{\vec{q}}(t)$. Let us consider an intrinsic material, or, since doping is unavoidable when growing these materials, we consider that its concentration is much smaller (orders of magnitude) than the photoinjected one, and can be neglected. Associated with the basic variables of Eqs. (1)–(3), is a set of intensive nonequilibrium thermodynamic variables that the formalism introduces, which are the so-called carriers' quasitemperature $T_c^*(t)$, which is a measurement in Kelvin degrees of the kinetic energy of the photoexcited carriers; the so-called qua-

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sichemical potentials $\mu_e(t)$ and $\mu_h(t)$; and the quasitemperature associated to each LO-phonon mode, $T^*_{\vec{q}}(t)$.[15,16] These intensive nonequilibrium thermodynamic variables (so-called Lagrange multipliers in the variational statistical formalism)[12] are present in the collision operators on the right-hand side of Eqs. (1)–(3). Hence, in order to close these equations, it is required to relate them to the basic variables, relations which constitute a kind of nonequilibrium equations of state.[12] The solution of the problem requires, then, to solve the coupled system of kinetic and state equations.

The equations describing the irreversible evolution of the macrostate of the system are[13–15,17]

$$\frac{d}{dt}E_c(t) = -\sum_{\alpha,\eta,i}\dot{E}^i_{\alpha,\eta}(t), \tag{1}$$

$$\frac{d}{dt}n(t) = \dot{n}_R(t) + \dot{n}_D(t), \tag{2}$$

$$\frac{d}{dt}\nu_{\vec{q}}(t) = \dot{\nu}_{\vec{q},C}(t) + \dot{\nu}_{\vec{q},AN}(t). \tag{3}$$

In Eq. (1), the right-hand side stands for the rate of energy in excess of equilibrium, which is transferred to the phonons; η=LO,TO,AC, and the upper index i refers to the different types of interactions, PD, PZ, and FR for deformation potential, piezoelectric, and Fröhlich interactions, respectively; in Eq. (2), $\dot{n}_R$ refers to the rate of change of n due to radiative recombination, and $\dot{n}_D$ to the rate of change due to ambipolar diffusion. The first one is unimportant in the picosecond range we are analyzing, and the latter is written as $\dot{n}_D = -n(t)/\tau_D(t)$, where $\tau_D(t)$ is a characteristic time for diffusion.[18,19] Finally, in Eq. (3), the first term on the right accounts for the change in time of the LO-phonon population in each mode $\vec{q}$ as a result of scattering with carriers, and the second due to anharmonic interactions with the AC phonons. The change in the thermodynamic state of TO and AC phonons can be disregarded, as compared with the one of LO phonons in this highly polar semiconductor, as already noticed.

Let us consider the case of an exciting laser pulse with a photon-energy-generating carriers with an excess kinetic energy of 810 meV, which produces a photoinjected carrier system with an initial quasitemperature $T^*_c(0) \simeq 3140$ K. The intensity of the pulse is taken as to produce a photoinjected concentration of 4×10^{18} cm^{-3}, with, first, a pulse duration of 0.1 ps and second, of 6 ps. In the given conditions, the carriers (electron and hole pairs) are on the metallic side of Mott transition.[8] The coupled set of nonlinear equations of evolution are solved using the parameters for zinc-blende GaN (effective masses, Fröhlich coupling strength, energy gap, etc.) available in the literature (details in Ref. 13).

In Figure 1(a) is shown the evolution of the carriers' quasitemperature and the quasitemperature of several LO-phonon modes in the case of the short pulse (0.1 ps). It can be noticed a cooling down of the carriers in the picosecond scale, with their quasitemperature displaying a rapid decrease in the, say, first 6 to 7 ps, followed by a smooth change (a near plateau) in the tens of picoseconds region, and attaining final thermal equilibrium with the lattice, at 300 K, after roughly 100 ps. Concerning the phonon modes, they warm up but in a differentiated way, namely, a strong one for those in an off-center region of Brillouin zone, roughly for wave numbers in the interval 1×10^5 to 1×10^8 cm^{-1}. This is also noticed in the case of GaAs,[20–22] and it is a result of the interplay of the conservation of energy and momentum in the scattering events, together with the dependence on wave number of the Fröhlich potential and the density of states (this is discussed in Ref. 23). Moreover, similarly to the case of GaAs,[20–22] it is possible to notice the existence of the phenomenon called *hot-phonon (HP) temperature overshoot*; that is, at certain time intervals, a few phonon modes (contained in the off-center region of the Brillouin zone just mentioned) acquire a quasitemperature that is larger than that of the carriers. The cooling down of the carriers is slow (in a hundred of picoseconds time scale) as a result of this "*phonon bottleneck*," and also because of the effect of ambipolar diffusion. In Fig. 1(b) is shown the case when the excitation is performed using a longer pulse (6 ps) than in the previous case. It can be noticed that the phonons in the modes of the privileged off-center region of the Brillouin zone are warmed up in a stronger way than in the case of Fig. 1(a). As a

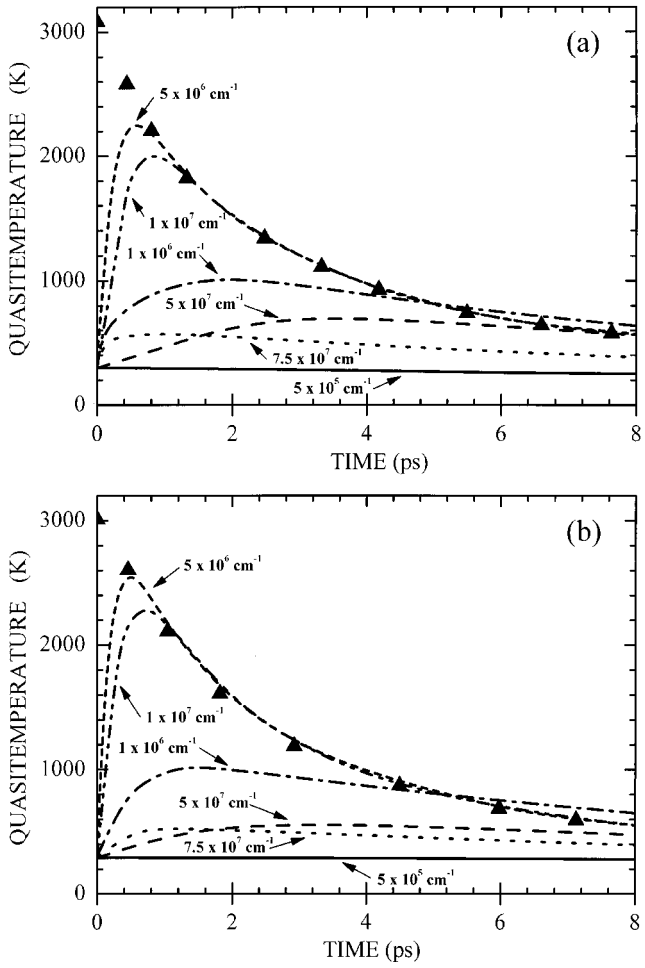

FIG. 1. Time evolution of the carriers' quasitemperature (triangles) and of the quasitemperatures of the LO phonons in modes $q=5\times10^5$ cm^{-1} (solid line), 1×10^6 cm^{-1} (point long-dashed line), 5×10^6 cm^{-1} (dashed line), 1×10^7 cm^{-1} (double point-dashed line), 5×10^7 cm^{-1} (long-dashed line), and 7.5×10^7 cm^{-1} (points). The upper figure (a) corresponds to the case of excitation with a laser pulse of 0.1 ps, and the lower one (b) to a laser pulse of 6.0 ps.

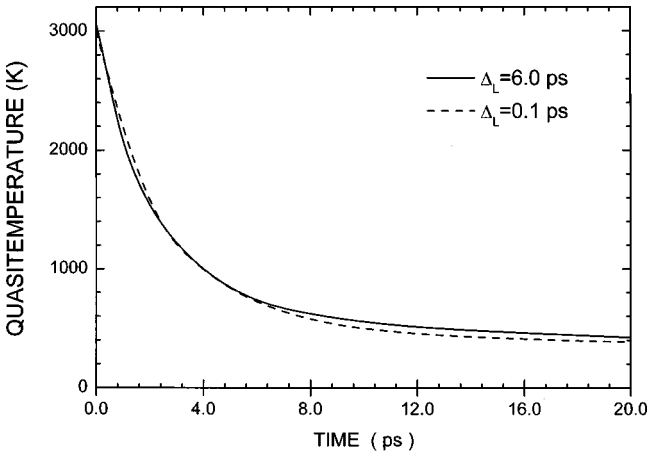

FIG. 2. Time evolution of the carriers' quasitemperature for a short laser pulse of 0.1 ps (dashed line) and a long laser pulse of 6.0 ps (solid line).

consequence, the carriers' quasitemperature becomes, in the first picoseconds, smaller than in the other case, however, approaching the equilibrium value at a slower pace, as is shown in Fig. 2.

The necessity of a hot carriers' picture to describe the evolution in the picosecond scale of the macroscopic state of a highly excited plasma in bulk GaAs has been already pointed out: a HP temperature overshoot was observed by Kim and Yu[20] in photoexcited GaAs using Raman spectroscopy. Rego and Algarte[21] were able to explain this behavior in GaAs by considering hot-phonon effects. By explicitly solving the coupled Boltzmann equations numerically for the injected intraband electrons and the subsequently generated hot phonons in an incoherent relaxation regime, Nahm *et al.*[22] obtained for GaAs that the HP population of some modes increases rapidly in the initial stage, and that the calculated electron quasitemperature displays a slower decay when the HP effect is considered.

Recently, the search for a deep understanding of the lasing mechanism in GaN beyond the Mott transition is evidencing the role of hot phonons and nonthermal carrier states.[11] Femtosecond pump-probe spectroscopy measurements have evidenced the ultrafast carrier dynamics in highly excited GaN epilayers[10] which has been assigned to HP effects slowing down the energy relaxation of the hot carriers at densities ranging from 4×10^{17} to 10^{19} cm^{-3}. Measurements of the hot-hole relaxation dynamics in Mg-doped *p*-type GaN wurtzite films by Ye *et al.*[24] have suggested that LO-optical phonon emissions modified by hot-phonon effects is the dominant energy relaxation process. The results presented here strongly support the HP picture used to explain existing experiments on the relaxation dynamics of photoexcited GaN. Preliminary results also point out the fact that the hot phonons can also play an important role in the high-field transport in III-nitrides.[25]

Summarizing, *phonon bottleneck* (due to hot-phonon temperature overshoot), associated with the fact of a selective warming up of the LO phonons and ambipolar diffusion, slow the cooling down of the carriers. It follows a near thermalization of carriers and LO phonons in the roughly tens of picoseconds time scale. It can also be noticed that, as time elapses, the recombination of the carriers proceeds in a nanosecond time scale, which also comes to influence the relaxation process, however, outside the picosecond range.

We acknowledge financial support provided by the National Research Council (CNPq) through the grant NanoSemiMat Project (No. 550.015/01-9) (V.N.F., A.R.V., and R.L.) and the São Paulo State Research Foundation under grant 2000/04301-5 (A.R.V. and R.L.). Some of the authors (A.R.V., V.N.F., and R.L.) are CNPq Research Fellows.

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