Analysis of Energy Loss Rates of High-Density Electron-Hole Plasma in GaN and ZnO

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We investigate the energy loss rates of optically generated hot carriers in GaN and ZnO as a function of carrier temperature $T_c$ for various carrier densities by employing the Kogan formula extended to degenerate electron-hole plasma. The energy losses of the carriers by various carrier-photon interactions are evaluated within the random phase approximation. Our results show that the energy losses are dominantly due to hole-phonon couplings and, at high carrier temperatures, the carriers lose their energies via both long-range polar and short-range non-polar optical-phonon scatterings. As $T_c$ is reduced, the energy loss via carrier-optical-phonon coupling diminishes quickly, and the energy relaxation is dominated by the piezoacoustic coupling to the lattice at relatively higher values of $T_c$ compared to that in GaAs.

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I. INTRODUCTION

Recently, energy loss rates (ELRs) in wide-band-gap semiconductors such as ZnO and GaN have gained renewed interest because of their practical importance in application to short-wavelength optoelectronic devices [1–5]. However, not all the energy relaxation mechanisms of these materials are well investigated, nor is there much information about the ELRs of wide-band-gap materials. Here we present a study of the phonon-related ELRs for the hot electron-hole plasma in GaN and ZnO as a function of carrier temperature $T_c$ for various carrier densities $n(\equiv n_e=n_h)$. The ELRs are evaluated by following the approach of Pugnet et al. with the use of the Kogan formula extended to degenerate electron-hole plasma [6, 7]. The hot carriers of temperature $T_c$ are assumed to originate through high-power short-pulse lasers, and we limit our interest to samples of low lattice temperature $T_L(\sim 4 \text{ K})$, so that relaxation processes involving more than one phonon can be neglected.

II. CALCULATIONAL METHODS

We consider the cases where the carrier-phonon interactions are weak enough to treat them by perturbation theory. A carrier in a state $|i\rangle$ can transfer to a state $|f\rangle$ by absorption or emission of a phonon of wave vector $\mathbf{q}$.

The ELR per electron-hole pair is determined by calculating the transition rate $w_{i \rightarrow f}$ multiplied by the energy gained by the phonons from the carriers during the transition $|i\rangle \rightarrow |f\rangle$, and then dividing the energy gain by the plasma density. The electron-phonon energy relaxation rate is given, in terms of density matrix $\rho_i$, by [8]

$$
P = \sum_{i,f} \rho_i w_{i \rightarrow f} \left( E_f^{ph} - E_i^{ph} \right),
$$

where the initial-state density matrix $\rho_i$ is assumed to be the product of canonical distributions of the carriers and the lattice, $\rho_i = \rho_c(T_c)\rho_L(T_L)$, and $w_{i \rightarrow f} = \frac{2\pi}{\hbar} |\langle f|H_{e-ph}|i\rangle|^2 \delta(E_f-E_i)$. Here, $H_{e-ph}$ is the electron-phonon interaction, which depends on the specific form of the carrier-lattice coupling (see equations below) and induces transition $|i\rangle \rightarrow |f\rangle$ with energy $E_i = E_i^e + E_i^{ph}$ and $E_f = E_f^e + E_f^{ph}$. The interaction through specific coupling (denoted by a parameter $j$) is represented by

$$
H^{(j)}_{e-ph} = \sum_{\mathbf{q},\lambda} \left( M_{\mathbf{q},\lambda}^1 \hat{n}_{-\mathbf{q}} \hat{a}_{\mathbf{q}\lambda} + M_{\mathbf{q},\lambda}^{(j)*} \hat{n}_{\mathbf{q}} \hat{a}^\dagger_{-\mathbf{q}\lambda} \right),
$$

where $M_{\mathbf{q},\lambda}^{(j)}$ and $\hat{n}_{\mathbf{q}}(=\sum_n \hat{n}_{n\mathbf{q}})$ are the (bare) electron-phonon scattering matrix element and the Fourier transform of the electron density operator $\hat{n}(\mathbf{r}) = \langle \psi^\dagger(\mathbf{r})\psi(\mathbf{r}) \rangle = \sum_{\mathbf{q}} \hat{n}_{\mathbf{q}} e^{-i\mathbf{q}\cdot\mathbf{r}}$; $\hat{n}_{\mathbf{q}} = \sum_{\mathbf{k}\nu} c^\dagger_{\mathbf{k}\nu} c_{\mathbf{k}\nu}$, respectively. (Here, $c_{\mathbf{k}\nu}$ and $c^\dagger_{\mathbf{k}\nu}$ are annihilation and creation operators for electrons in band $\nu$, and $\mathbf{q}$ and $\lambda$ denote the phonon wave vector and polarization, respec-
tively; \(\vec{q}, \lambda\) and \(\vec{q}^\dagger, \lambda\) are annihilation and creation operators, respectively, for phonons with frequency \(\omega_{\vec{q}, \lambda}\).

The ELR per particle, \(P_j\), due to the \(j\)th type of carrier-phonon coupling, is written as

\[
P_j = 2 \sum_{\vec{q} \nu} |M_{\vec{q}, \nu}|^2 \omega_{\vec{q}, \nu} \left[ N(\omega_{\vec{q}, \nu}/T_L) - N(\omega_{\vec{q}, \nu}/T_e) \right] \times \Pi_2(\vec{q}, \omega_{\vec{q}, \nu}; \nu),
\]

where \(N(\omega_{\vec{q}, \nu}/T) = [\exp(h\omega_{\vec{q}, \nu}/k_B T) - 1]^{-1}\) is the occupation number of phonon (of type \(j\)) of wave vector \(\vec{q}\) and \(\Pi_2(\vec{q}, \omega_{\vec{q}, \nu}; \nu)\) is the imaginary part of the retarded density-density correlation function for carriers in band \(\nu\) [9]:

\[
\Pi_2(\vec{q}, \omega_{\vec{q}, \nu}; \nu) = -\frac{1}{2V N(\omega_{\vec{q}, \nu}/T_c)} \int_{-\infty}^{+\infty} dt \, e^{i\omega t} \times \langle \tilde{n}_{\vec{q}, \nu}(0) \tilde{n}_{\vec{q}, \nu}(t) \rangle,
\]

where \(V\) is the volume of the sample. In Eq. (4), \(\langle \tilde{n}_{\vec{q}, \nu}(0) \tilde{n}_{\vec{q}, \nu}(t) \rangle\) means the full statistical average of \(\tilde{n}_{\vec{q}, \nu}(0) \tilde{n}_{\vec{q}, \nu}(t)\) over an equilibrium ensemble of the carriers (in the absence of interaction with phonons).

If one neglects the inter-carrier Coulomb interaction, \(\Pi(\vec{q}, \omega_{\vec{q}, \nu}; \nu)\) can be approximated by the zeroth order expression \(\Pi^{(0)}(\vec{q}, \omega_{\vec{q}, \nu}; \nu) = m^2 / (2\pi\hbar^2) \cdot q^{-4} Q(q, \omega_{\vec{q}, \nu}; \nu)\), where \(Q(q, \omega_{\vec{q}, \nu}; \nu)\) is defined by

\[
Q(q, \omega_{\vec{q}, \nu}; \nu) = \ln \left\{ 1 + \exp \left[ \frac{\mu_\nu}{k_B T_c} - \frac{\hbar^2}{8 m k_B T_c} \left( q - 2 \omega_{\vec{q}, \nu}/\hbar \right)^2 \right] \right\}.
\]

Here, \(\mu_\nu\) is the quasi-chemical potential of the band \(\nu\) (= conduction and valence bands occupied by electrons and holes) and is determined by the particle number density \(n_\nu = 1 / 2\pi \left( \frac{2 m^*}{\hbar^2} \right)^{3/2} \int d^3 r \exp(-\mu_\nu) / k_B T_c |E|^{1/2}\) [10]. Hence \(n_\nu\) is the carrier concentration in the \(\nu\)th band. In this work we use the quasi-Fermi energy evaluated from the relation [10]

\[
\mu_\nu \simeq k_B T_c \ln \left( \frac{n_\nu}{n_0} \right) + 4.8966851 \ln \left( 0.04496457 \frac{n_\nu}{n_0} + 1 \right) + 0.1333760 \frac{n_\nu}{n_0},
\]

where \(n_0 = 1 / 4 \left( \frac{2 m^* k_B T_c}{\hbar^2} \right)^{3/2}\).

In the presence of the electron-electron interactions, the electron-phonon couplings are screened. If we include the screening effect within the random phase approximation (RPA), \(\Pi^{(rpa)}_2 = \frac{\Pi^{(0)}_2}{\tau^2}\), where \(\tau = 1 - \frac{\nu}{\nu(\vec{q}, \omega_{\vec{q}, \nu})}\), \(\nu(q)\) is the bare Coulomb interaction. In this work, we use the static expression for \(\epsilon_{RPA}\) given, in the long wave length limit, by \(\epsilon_{RPA}(q, \theta) = 1 + \frac{q^2}{\pi^2} \frac{2\nu(q)}{\nu(\vec{q}, \omega_{\vec{q}, \nu})}\) [11], where \(q_{DH}\) is the Debye-Hückel screening length \(q_{DH}^2 = \frac{4 \pi^2 e^2}{\epsilon \epsilon_0} \sum \frac{n\\nu}{\hbar \\
\\varepsilon \\
\\nu}.

In polar crystals, the longitudinal optical (LO) mode induces an electrical polarization field which is long-ranged. For the coupling of carriers to polar optical phonons, we have \(\left| M^{(j)}_q(q) \right|^2 = \frac{\nu(q)}{\nu(\vec{q}, \omega_{\vec{q}, \nu})}\), where \(\epsilon_\infty, \epsilon_0\) and \(\omega_{\nu, \nu, \nu, \nu}\) are, respectively, the optical and static dielectric constants and the LO phonon frequency. Then, the ELR per particle through the carrier-LO phonon scattering becomes

\[
P_{\nu,LO} = \sum_{\nu=cond.,val.} \frac{\epsilon^2 m^2 k_B T}{\hbar^2 \nu^2} \left( 1 + \frac{1}{\epsilon_\infty} \right) \omega_{\nu,LO}^2 \times (N(\omega_{\nu,LO}/T_c) - N(\omega_{\nu,LO}/T_e)) \times \int_0^{\infty} dq q^3 \frac{d(q^2 + q_{DH}^2)}{Q(q, \omega_{\nu,LO}; \nu)}.
\]

The sum over \(\nu\) means that the total ELR of the plasma is the sum of the contributions from the electrons in the conduction band and from the holes in the valence bands.

For crystals with no inversion symmetry, such as GaN and ZnO, acoustical phonon modes can also induce an electrical polarization field, which is the origin of the piezoelectricity. The matrix element \(M^{PA}_q(q)\) for piezoacoustic (PA) coupling is given by

\[
\left| M^{PA}_q(q) \right|^2 = \frac{2 \nu(q)}{\rho \nu(\vec{q}, \omega_{\vec{q}, \nu})} \sum_{i \neq j} q_k e_k e_j \xi(\lambda, q) \right| q_j \right|^2
\]

where \(\rho, \xi(\lambda, \vec{q}),\) and \(e_k e_j\) denote, respectively, the mean mass density of the material, the unit vector of the acoustic lattice polarization \(\lambda\) (TA or LA), and the piezoelectric coupling constants of a third rank tensor [12]. Both the transverse and the longitudinal modes contribute to the piezoelectric interaction, and the ELR per particle via piezoacoustical coupling is written, within the Debye model, as

\[
P_{PA} = \sum_{\nu} \epsilon^2 m^2 k_B T \frac{\mathcal{F}}{4 \pi^2 \hbar^3} \int_0^{\infty} dq q^5 \frac{d(q^2 + q_{DH}^2)}{Q(q, \omega_{\nu,LO}; \nu)} 
\]

Here, \(s\) and \(\mathcal{F}\) are, respectively, the mean sound speed and a structure dependent factor given by [12]

\[
\mathcal{F} = \frac{8 \pi}{35 \rho e_0} \left[ 2 \left( e_{31} - e_{33} + e_{15}^2 \right) + \frac{112}{9} e_{15}^2 + \frac{4}{3} \left( e_{31} + 3 e_{33} + 2 e_{15} \right) + \frac{7}{4} e_{33}^2 \right]
\]

for material of the wurtzite structure, and \(\mathcal{F} = \frac{176 \pi e_{15}^2}{45 \rho \rho_{eo}}\) in the case of the zincblende structure. Here, \(e_{ij}\) is the component of the strain tensor [2,12,13].
Table 1. Physical parameters of GaN and ZnO.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>GaN</th>
<th>ZnO</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\epsilon_\infty$</td>
<td>5.35</td>
<td>3.7</td>
</tr>
<tr>
<td>$\epsilon_0$</td>
<td>10</td>
<td>8.8</td>
</tr>
<tr>
<td>$\hbar \omega_{LO}$</td>
<td>92</td>
<td>72.8</td>
</tr>
<tr>
<td>$m_c/m_0$</td>
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<td>0.28</td>
</tr>
<tr>
<td>$m_{hh}/m_0$</td>
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<td>0.59</td>
</tr>
<tr>
<td>$m_{lh}/m_0$</td>
<td>1.03</td>
<td>0.59</td>
</tr>
<tr>
<td>$\rho$</td>
<td>6.095</td>
<td>5.675</td>
</tr>
<tr>
<td>$\mathcal{E}_c$</td>
<td>5</td>
<td>5</td>
</tr>
<tr>
<td>$\mathcal{E}_v$</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>$D$</td>
<td>$10^9$</td>
<td>$10^9$</td>
</tr>
<tr>
<td>$s$</td>
<td>$3.03 \times 10^5$</td>
<td>$2.73 \times 10^5$</td>
</tr>
</tbody>
</table>

† Mean value of the data in three papers

Fig. 1. Total ELRs per electron-hole pair as a function of carrier temperature for (a) GaN and (b) ZnO single crystals in wurtzite and zincblende structures. Solid and open symbols denote the wurtzite and zincblende structures of the corresponding material.

The matrix element $M_q^{TO}(q)$ for nonpolar-optical-phonon (NPO) coupling is given by $|M_q^{TO}(q)|^2 = \frac{\hbar D^2}{2\rho \omega_{TO}}$ [14,15]. The ELR per particle via NPO phonons is given, by assuming dispersionless nonpolar optical modes of frequency $\omega_{TO}$, by

$$P_{NPO} = \sum_{\nu} \frac{m^2 k_B T_c D^2}{4\pi^3 \hbar^5 \rho} \left\{ N_{TO}(T_c) - N_{TO}(T_L) \right\} \times \int_0^\infty \frac{dq q^5}{(q^2 + q_{BH}^2)^2} \mathcal{Q}(q, \omega_{TO}; \nu), \quad (8)$$
where $D$ is the optical deformation potential constant. On the other hand, $M_{ie}^{D}(q)$ for acoustical deformation potential (ADP) scattering is given by $|M_{ie}^{D}(q)|^{2} = \frac{\hbar^{2}q^{2}^{2}}{2\mu_{ie}^{D}V}$ [14–16], where $\epsilon_{ie}$ denotes the deformation potential constants of the LA waves for the carriers in the conduction and valence bands. By assuming $q = \gamma q$ for acoustic modes, the ELR per particle via ADP scattering is written as

$$P_{ADP} = \sum_{\nu} \frac{m^{2}k_{B}T_{c}c_{\nu}^{2}}{4\pi^{4}k^{2}T_{L}^{2}} \int_{0}^{\infty} dq q^{7} \left( \frac{q^{2} + \gamma^{2}d_{\nu}^{2}}{q^{2} + \gamma^{2}d_{\nu}^{2}} \right) \times \{N_{\nu}(T_{L}) - N_{\nu}(T_{L})\} \mathcal{Q}(q, \gamma q; \nu).$$ (9)

### III. RESULTS AND DISCUSSION

Table 1 shows the physical parameters used in obtaining the ELR for GaN and ZnO. The total ELR via electron-phonon coupling in a given electron-hole plasma is the sum of the individual ELRs via all possible lattice modes by electrons and holes through various coupling mechanisms described in Sec. II.

Figure 1 shows the total ELR per electron-hole pair of (a) GaN, and (b) ZnO, both in the wurtzite and in the zincblende structure at $T_{L} = 4$ K for carrier densities of $n = 10^{14}$, $10^{15}$, $10^{16}$, and $2 \times 10^{19}$ cm$^{-3}$. Solid and open symbols denote, respectively, the wurtzite and zincblende structures of the corresponding single crystals of (a) GaN and (b) ZnO. Of the two types of carriers (electrons and holes) in the plasma, the energy loss arises dominantly by the holes, which have much heavier effective masses than electrons. As the carrier concentration increases, the ELR per electron-hole pair is reduced significantly. The main source of this reduction is conjectured to be the carrier screening of the electron-phonon scattering mechanism.

Figure 2 shows the ELR per electron-hole pair of GaN in wurtzite structure as a function of carrier temperature for $n = 2 \times 10^{19}$ cm$^{-3}$. Each line of a different line type denotes a contribution to the total ELR from a distinct carrier-phonon scattering mechanism. Thin lines indicate the results in the absence of carrier screening.

![Fig. 2. Energy loss rates per electron-hole pair of GaN in wurtzite structure as a function of carrier temperature for $n = 2 \times 10^{19}$ cm$^{-3}$. Each line of a different line type denotes a contribution to the total ELR from a distinct carrier-phonon scattering mechanism. Thin lines indicate the results in the absence of carrier screening.](image)

### IV. SUMMARY

In this paper, we describe the ELRs of the hot electron-hole plasma in GaN and ZnO, and show that the energy losses of electron-hole plasmas in these materials are due
dominantly to hole-phonon couplings. At high carrier
temperatures, the carriers lose energy via both long-
range polar and short-range non-polar optical-phonon
scatterings. As the carrier temperature decreases, the
energy loss via carrier-optical phonon couplings dimin-
ishes quickly and the carrier energy relaxation is domi-
nated by the piezoacoustical coupling to the lattice at rel-
atively higher temperatures, compared to that in GaAs.
At low carrier temperatures, only acoustic modes are
important. The long-range interactions via polar opti-
cal and piezoacoustical scattering are reduced rapidly at
high carrier densities. The energy-loss behaviors of GaN
and ZnO are rather similar in general, but the effect of
hole effective mass discrepancy is rather significant. The
present results would be verified by, for example, fem-
tosecond optical measurements. By comparing the the-
etrical ELR with an experimentally determined ELR,
one can obtain useful information on physical quantities
such as various scattering matrix elements and deforma-
tion potentials.

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