Electron–optical-phonon scattering in wurtzite crystals

B. C. Lee* and K. W. Kim
Department of Electrical and Computer Engineering, North Carolina State University, Raleigh, North Carolina 27695-7911

M. Dutta and M. A. Stroscio
U.S. Army Research Office, P.O. Box 12211, Research Triangle Park, North Carolina 27709-2211

(Received 17 March 1997)

We present Fröhlich-like electron–optical-phonon interaction Hamiltonian for wurtzite crystals in the long-wavelength limit based on the macroscopic dielectric continuum model and the uniaxial model. In general, the optical-phonon branches support mixed longitudinal and transverse modes due to the anisotropy. We calculate electron scattering rate due to the optical phonons in the bulk wurtzite GaN and demonstrate that the scattering rate due to transverselike phonon processes can be strongly enhanced over a range of angles with respect to the c axis. For the case of longitudinal-like modes, the anisotropy is small and the result is almost the same as that obtained with the cubic Fröhlich Hamiltonian. [S0163-1829(97)05428-3]

Recent advances in semiconductor optoelectronics[1] and electronics[2] based on wide band-gap nitride materials have resulted in substantial interest in the basic properties of wurtzite crystals. Particularly, a complete understanding of carrier-phonon interaction mechanisms and rates is essential to further progress in these fields. Since the wurtzite crystals have a different unit-cell structure (i.e., four atoms per unit cell) as well as lower symmetry compared to zinc-blende counterparts, phonon dynamics and carrier-phonon interactions in this material system may be substantially different from those with cubic symmetry. Clearly, there are many more distinct phonon branches (nine optical and three acoustic modes) in wurtzite materials. At the same time, the phonon modes may not be purely longitudinal nor transverse (the [0001] direction excepted). Current understanding of phonon dynamics and their interaction with carriers in wurtzite semiconductors is very primitive. In this paper, the macroscopic dielectric continuum model and the uniaxial model of Loudon[3] are applied to derive the Fröhlich Hamiltonian and the corresponding scattering rates for wurtzite materials.

We consider a uniaxial crystal in which only one group of three optical-phonon branches is infrared active. The wurtzite structure is a case in point since at the Γ point, only the $A_1(Z)$ and $E_1(X,Y)$ modes are infrared active among the nine optical-phonon modes. Our model can be extended to other uniaxial crystals which have larger numbers of polar modes since the method is the same. We take the c axis along the z direction and denote the perpendicular direction as $\perp$. Within the dielectric continuum approach, the optical-phonon modes in the no-retardation limit satisfy the classical electrostatic equations, i.e.,

$$\mathbf{E}(\mathbf{r}) = -\nabla\Phi(\mathbf{r}), \tag{1}$$

$$\mathbf{D}(\mathbf{r}) = \mathbf{E}(\mathbf{r}) + 4\pi\mathbf{P}(\mathbf{r}) = \epsilon_\perp(\omega)E_\perp(\mathbf{r})\hat{\rho} + \epsilon_z(\omega)E_z(\mathbf{r})\hat{\mathbf{z}}, \tag{2}$$

$$\nabla\cdot\mathbf{D}(\mathbf{r}) = 0, \tag{3}$$

where $\Phi(\mathbf{r})$ is the electrostatic potential due to the optical-phonon mode, $\mathbf{E}$ is the electric field, $\mathbf{D}$ is the displacement, $\mathbf{P}$ is the polarization field, and $\hat{\mathbf{z}}$ ($\hat{\rho}$) denotes the unit vector along the $\mathbf{z}$ ($\perp$) direction. The direction-dependent dielectric functions $\epsilon_\perp(\omega)$ and $\epsilon_z(\omega)$ are given by

$$\epsilon_\perp(\omega) = \epsilon_\perp^\infty \frac{\omega^2-\omega_{\perp L}^2}{\omega^2-\omega_\perp^2}, \tag{4}$$

$$\epsilon_z(\omega) = \epsilon_z^\infty \frac{\omega^2-\omega_{zL}^2}{\omega^2-\omega_z^2}, \tag{5}$$

where $\omega_\perp$ (ω_z) is the lattice dispersion frequency, and $\omega_{\perp L}$ (ω_{zL}) is the longitudinal-optical (LO) phonon frequency, $\epsilon_\perp^\infty$ (ϵ_z^∞) is the high-frequency dielectric constant perpendicular to (along) the z axis. Thus, the static dielectric constants are $\epsilon_\perp^0 = \epsilon_\perp^\infty\omega_{\perp L}^2/\omega_\perp^2$ and $\epsilon_z^0 = \epsilon_z^\infty\omega_{zL}^2/\omega_z^2$. The macroscopic equations of motion for uniaxial materials give relations between $\mathbf{E}(\mathbf{r})$ and $\mathbf{P}(\mathbf{r})$ and the relative displacement of an ion pair $\mathbf{u}(\mathbf{r})$,[3–5]

$$\frac{d^2u_\perp(\mathbf{r})}{dt^2} = -\omega_\perp^2 u_\perp(\mathbf{r}) + \frac{1}{\sqrt{4\pi\mu n}}\sqrt{\epsilon_\perp^0-\epsilon_\perp^\infty}\,\omega_\perp E_\perp(\mathbf{r}), \tag{6}$$

$$\frac{d^2u_z(\mathbf{r})}{dt^2} = -\omega_z^2 u_z(\mathbf{r}) + \frac{1}{\sqrt{4\pi\mu n}}\sqrt{\epsilon_z^0-\epsilon_z^\infty}\,\omega_z E_z(\mathbf{r}), \tag{7}$$

$$P_\perp(\mathbf{r}) = \sqrt{\mu n/4\pi}\sqrt{\epsilon_\perp^0-\epsilon_\perp^\infty}\,\omega_\perp u_\perp(\mathbf{r}) + \frac{\epsilon_\perp^\infty-1}{4\pi}E_\perp(\mathbf{r}), \tag{8}$$

$$P_z(\mathbf{r}) = \sqrt{\mu n/4\pi}\sqrt{\epsilon_z^0-\epsilon_z^\infty}\,\omega_z u_z(\mathbf{r}) + \frac{\epsilon_z^\infty-1}{4\pi}E_z(\mathbf{r}), \tag{9}$$

where μ is the reduced mass and n is the number of unit cells in the unit volume. If we assume a harmonic dependence in space and time, we can obtain $\Phi(\mathbf{r})$, $\mathbf{E}(\mathbf{r})$, and $\mathbf{P}(\mathbf{r})$ upon solving Eqs. (1)–(9). We denote the phonon wave vector as $\mathbf{q}$ and define θ as the angle between $\mathbf{q}$ and z (c) axis. One frequency has a trivial solution $\omega=\omega_\perp$ and $\mathbf{E}(\mathbf{r})=0$; this is

the case of so-called ordinary phonon. The phonon frequencies for extraordinary phonons are obtained from[3]

$$\epsilon_{\perp}(\omega)\sin^2\theta+\epsilon_z(\omega)\cos^2\theta=0. \tag{10}$$

We normalize each orthonormal mode according to[6]

$$[\sqrt{\mu n}\mathbf{u}(\mathbf{q})]\cdot[\sqrt{\mu n}\mathbf{u}(\mathbf{q})]=\hbar/2\omega V, \tag{11}$$

where $\mathbf{u}(\mathbf{q})$ is the Fourier transform of $\mathbf{u}(\mathbf{r})$ and V is the crystal volume. Then, the electron–optical-phonon interaction in second quantized form is

$$H=\sum_{\mathbf{q}} -e\Phi(\mathbf{q})e^{i\mathbf{q}\cdot\mathbf{r}}(a_{\mathbf{q}}+a^{\dagger}_{-\mathbf{q}}), \tag{12}$$

where $\Phi(\mathbf{q})$ is the Fourier transform of $\Phi(\mathbf{r})$ and $a^{\dagger}_{-\mathbf{q}}$ and $a_{\mathbf{q}}$ are the creation and annihilation operator, respectively. Using the expression in Ref. 6, the interaction Hamilton for a uniaxial crystal is

$$H=\sum_{\mathbf{q}}\left[\frac{4\pi e^2\hbar V^{-1}}{(\partial/\partial\omega)[\epsilon_{\perp}(\omega)\sin^2\theta+\epsilon_z(\omega)\cos^2\theta]}\right]^{1/2}\frac{1}{q}\,e^{i\mathbf{q}\cdot\mathbf{r}}(a_{\mathbf{q}}+a^{\dagger}_{-\mathbf{q}})$$

$$=\sum_{\mathbf{q}}\sqrt{2\pi e^2\hbar/V\omega}\,\frac{(\omega_{\perp}^2-\omega^2)(\omega_z^2-\omega^2)}{[(\epsilon_{\perp}^0-\epsilon_{\perp}^{\infty})\omega_{\perp}^2(\omega_z^2-\omega^2)^2\sin^2\theta+(\epsilon_z^0-\epsilon_z^{\infty})\omega_z^2(\omega_{\perp}^2-\omega^2)^2\cos^2\theta]^{1/2}}\frac{1}{q}\,e^{i\mathbf{q}\cdot\mathbf{r}}(a_{\mathbf{q}}+a^{\dagger}_{-\mathbf{q}}). \tag{13}$$

The transition probability from electron state $\mathbf{k}$ to $\mathbf{k}'$ per unit time, $W(\mathbf{k},\mathbf{k}')$, is calculated from the Fermi golden rule as

$$W(\mathbf{k},\mathbf{k}')=\frac{2\pi}{\hbar}\,|M_{\mathbf{k}-\mathbf{k}'}|^2\,\delta(E_{\mathbf{k}'}-E_{\mathbf{k}}\pm\hbar\omega_{\mathbf{k}-\mathbf{k}'}), \tag{14}$$

where $E_{\mathbf{k}}$ is the electron energy, $M_{\mathbf{q}}$ (where $\mathbf{q}=\mathbf{k}-\mathbf{k}'$ is a transferred momentum) is a transition matrix element based on the Hamiltonian of Eq. (13), $\hbar\omega_{\mathbf{q}}$ is transition energy, and the upper (lower) sign corresponds to phonon emission (absorption). We also define the scattering rate $W(\mathbf{k})$ as

$$W(\mathbf{k})=\sum_{\mathbf{k}'} W(\mathbf{k},\mathbf{k}'). \tag{15}$$

In this work, ϵ_z^{∞} and $\epsilon_{\perp}^{\infty}$ are taken to be equal; this is quite a good assumption because ϵ^{∞} is due to electrons. We consider two limiting cases in which the solutions of Eq. (10) are expressed in simple forms.

(i) $|\omega_{\perp L}-\omega_{zL}|,|\omega_{\perp}-\omega_z|\ll|\omega_{\perp L}-\omega_{\perp}|,|\omega_{zL}-\omega_z|$. Infrared-active phonon frequencies of wurtzite structures satisfy this condition.[3] The solutions of Eq. (10) are $\omega=\Omega_L$ or Ω_T with[3]

$$\Omega_L^2=\omega_{zL}^2\cos^2\theta+\omega_{\perp L}^2\sin^2\theta, \tag{16}$$

$$\Omega_T^2=\omega_z^2\sin^2\theta+\omega_{\perp}^2\cos^2\theta. \tag{17}$$

Ω_L and Ω_T correspond to the LO-like and transverse-optical (TO) -like modes, respectively. For each of these modes, the matrix elements may be calculated based on Eq. (13). For $\omega=\Omega_T$, we have

$$|M_{\mathbf{q}}^T|^2=\frac{2\pi e^2\hbar}{Vq^2\Omega_T}\,\frac{(\omega_{\perp}^2-\omega_z^2)^2\sin^2\theta\,\cos^2\theta}{(\epsilon_{\perp}^0-\epsilon_{\perp}^{\infty})\omega_{\perp}^2\cos^2\theta+(\epsilon_z^0-\epsilon_z^{\infty})\omega_z^2\sin^2\theta}\times(n_{\mathrm{ph}}+\tfrac{1}{2}\pm\tfrac{1}{2}), \tag{18}$$

and for $\omega=\Omega_L$, the matrix element is

$$|M_{\mathbf{q}}^L|^2\simeq\frac{2\pi e^2\hbar}{Vq^2\Omega_L}\left[\frac{\sin^2\theta}{(1/\epsilon_{\perp}^{\infty}-1/\epsilon_{\perp}^0)\omega_{\perp L}^2}+\frac{\cos^2\theta}{(1/\epsilon_z^{\infty}-1/\epsilon_z^0)\omega_{zL}^2}\right]^{-1}(n_{\mathrm{ph}}+\tfrac{1}{2}\pm\tfrac{1}{2}). \tag{19}$$

Here the upper (lower) sign corresponds to phonon emission (absorption), and $n_{\mathrm{ph}}=[\exp(\hbar\omega/k_BT)-1]^{-1}$ ($\omega=\Omega_T,\Omega_L$) is the phonon occupation number where k_B is the Boltzmann constant and T is the temperature. Note that, in general, matrix element $M_{\mathbf{q}}^T$ is not zero because the TO-like mode is not purely transverse in uniaxial material; accordingly, this branch does interact with electrons unlike isotropic materials. When θ is equal to zero or $\pi/2$, this becomes a purely transverse mode and there is no interaction with electrons. For the isotropic case ($\omega_{\perp}=\omega_z$ and $\omega_{\perp L}=\omega_{zL}$), the matrix element $M_{\mathbf{q}}^T$ is zero and $M_{\mathbf{q}}^L$ reduces to the well-known Fröhlich interaction as expected.

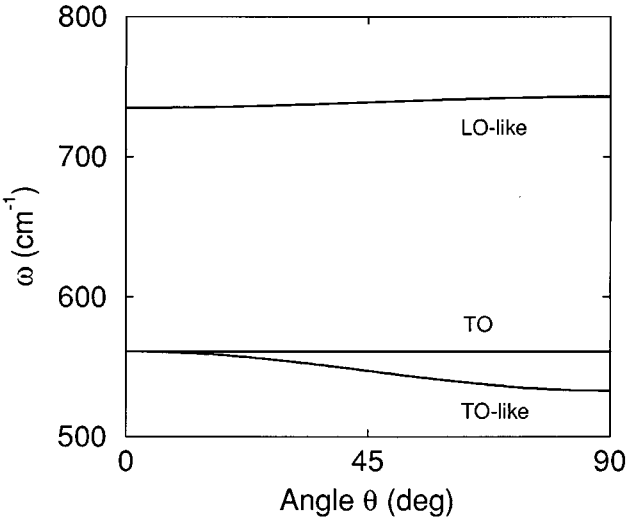

FIG. 1. Angular variation of infrared-active phonon frequencies in bulk wurtzite GaN. In this figure, θ is the angle between the phonon wave vector $\mathbf{q}$ and the c axis.

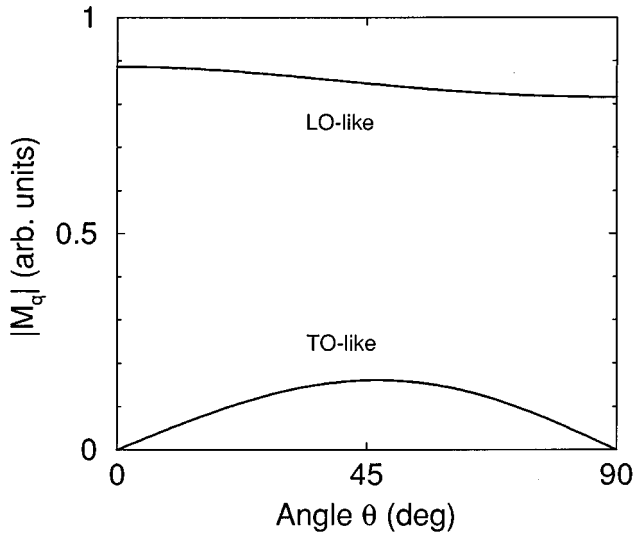

FIG. 2. Angular variation of the matrix element $|M_{\mathbf{q}}|$ for optical-phonon absorption in bulk wurtzite GaN. In this figure, θ is the angle between the phonon wave vector $\mathbf{q}$ and the c axis.

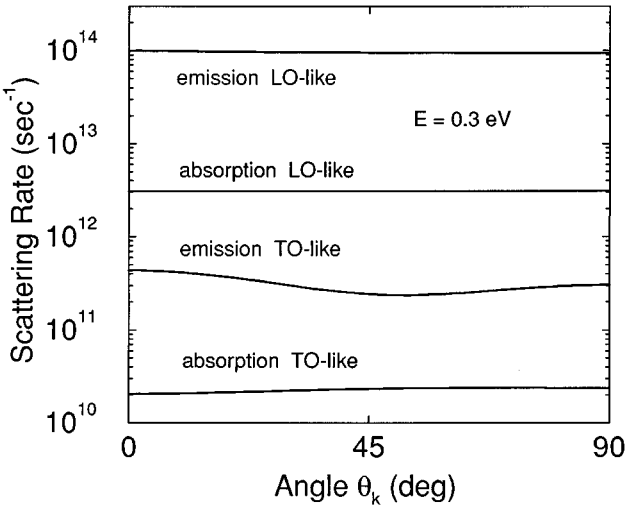

FIG. 3. Scattering rate $W(\mathbf{k})$ vs electron incident angle $\theta_{\mathbf{k}}$ with respect to the c axis. A constant electron energy of 0.3 eV is chosen as an example. Note that $\theta_{\mathbf{k}}$ (of this figure) and θ (of Figs. 1 and 2) represent different quantities.

(ii) $|\omega_{\perp L}-\omega_{zL}|,|\omega_{\perp}-\omega_{z}|\gg|\omega_{\perp L}-\omega_{\perp}|,|\omega_{zL}-\omega_{z}|$. For this case, the solutions of Eq. (10) are $\omega=\Omega_{\perp}$ or Ω_z with[3]

$$\Omega_{\perp}^2=\omega_{\perp}^2\cos^2\theta+\omega_{\perp L}^2\sin^2\theta, \tag{20}$$

$$\Omega_z^2=\omega_z^2\sin^2\theta+\omega_{zL}^2\cos^2\theta. \tag{21}$$

The matrix element $M_{\mathbf{q}}^{\perp}$ for $\omega=\Omega_{\perp}$ is

$$|M_{\mathbf{q}}^{\perp}|^2\simeq\frac{2\pi e^2\hbar}{Vq^2}\,\frac{\omega_{\perp L}^2}{\Omega_{\perp}}\left(\frac{1}{\epsilon_{\perp}^{\infty}}-\frac{1}{\epsilon_{\perp}^{0}}\right)\sin^2\theta(n_{\mathrm{ph}}+\tfrac{1}{2}\pm\tfrac{1}{2}) \tag{22}$$

and $M_{\mathbf{q}}^{z}$ for $\omega=\Omega_z$ is

$$|M_{\mathbf{q}}^{z}|^2\simeq\frac{2\pi e^2\hbar}{Vq^2}\,\frac{\omega_{zL}^2}{\Omega_z}\left(\frac{1}{\epsilon_z^{\infty}}-\frac{1}{\epsilon_z^{0}}\right)\cos^2\theta(n_{\mathrm{ph}}+\tfrac{1}{2}\pm\tfrac{1}{2}). \tag{23}$$

As an example, we calculate the electron scattering rate due to the optical-phonon scattering in bulk wurtzite GaN. The phonon frequencies of GaN are taken from the experimental results in Ref. 7: $\omega_{zL}=735\ \mathrm{cm}^{-1}$ for the A_1 phonon, $\omega_z=533\ \mathrm{cm}^{-1}$ for the A_1 phonon, $\omega_{\perp L}=743\ \mathrm{cm}^{-1}$ for the E_1 phonon, and $\omega_{\perp}=561\ \mathrm{cm}^{-1}$ for the E_1 phonon. The dielectric constants are $\epsilon_{\perp 0}=9.28$, $\epsilon_{z0}=10.1$, and $\epsilon_{\perp\infty}=\epsilon_{z\infty}=5.29$. For the electron effective mass, we take the value of $0.20\ m_0$ obtained from cyclotron resonance measurements,[8] where m_0 is the electron rest mass. The temperature is taken as $T=300$ K. First, we display the angular variation of the optical-phonon frequencies in Fig. 1. The LO-like mode ($\omega=\Omega_L$) is almost flat and the TO-like mode exhibits about a 5% anisotropy. In Fig. 2, we calculate $|M_{\mathbf{q}}|$ from Eq. (13) as a function of θ for the case of phonon absorption. As expected, electron–optical-phonon scattering is dominated by the LO-like mode. The matrix element for the LO-like mode exhibits about 9% anisotropy (18% anisotropy for the squared matrix element). For the TO-like mode, the anisotropy is very strong and electron-phonon interaction is enhanced over a range of angles θ near $\pi/4$ with respect to the c axis. However, the largest value of the matrix element for this mode is about 7% of that for LO-like mode. Figure 3 depicts the total scattering rate as a function of $\theta_{\mathbf{k}}$ which is the angle between the initial electron state $\mathbf{k}$ and the c axis. Unlike the isotropic case, the scattering rate cannot be obtained analytically because the phonon frequency ω in Eq. (14) is $\mathbf{q}$ dependent. Indeed, there is scattering due to the TO-like mode although its contribution is small. The total scattering rate due to LO-like phonon is insensitive to the angle $\theta_{\mathbf{k}}$. Thus, for the bulk GaN, using the cubic interaction Hamiltonian is a rather good approximation.

In summary, this paper presents a theory of electron–optical-phonon interaction in uniaxial crystals of the wurtzite type based on the macroscopic dielectric continuum model and the uniaxial model. Our calculation shows that the total Fröhlich scattering rates in such wurtzite materials manifest direction-dependent corrections that deviate about 10% from the rates obtained for the case of zinc-blende materials. These manifestations are particularly important in cases where the matrix elements for the TO-like modes are of interest.

The authors are grateful for many helpful discussions with S. Yu, Y. M. Sirenko, M. A. Littlejohn, and J. M. Zavada. This work was supported by the U.S. Army Research Office and the Office of Naval Research.

*Present address: Department of Physics, Inha University, Inchon, Korea.

[1] See, for example, S. Nakamura, M. Senoh, S. Nagahama, N. Iwasa, T. Yamada, T. Matsushita, Y. Sugimoto, and H. Kiyoku, Appl. Phys. Lett. **70**, 616 (1997); **69**, 1477 (1996).

[2] See, for example, M. A. Khan, J. N. Kuznia, A. R. Bhattarai, and D. T. Olson, Appl. Phys. Lett. **62**, 1786 (1993); M. A. Khan, M. S. Shur, J. N. Kuznia, Q. Chen, J. Burm, and W. Schaff, *ibid.* **66**, 1083 (1995).

[3] R. Loudon, Adv. Phys. **13**, 423 (1964); W. Hayes and R. Loudon, *Scattering of Light by Crystals* (Wiley, New York, 1978).

[4] M. Born and K. Huang, *Dynamical Theory of Crystal Lattices* (Clarendon Press, Oxford, England, 1954).

[5] A. Anselm, *Introduction to Semiconductor Theory* (Prentice-Hall, Englewood Cliffs, NJ, 1981).

[6] K. W. Kim and M. A. Stroscio, J. Appl. Phys. **68**, 6289 (1990).

[7] T. Azuhata, T. Sota, K. Suzuki, and S. Nakamura, J. Phys. Condens. Matter. **7**, L129 (1995).

[8] M. Drechsler, D. M. Hofmann, B. K. Meyer, T. Detchprohm, H. Amano, and I. Akasaki, Jpn. J. Appl. Phys. 1 **34**, L1178 (1995).