Ballistic thermal conductance of a graphene sheet

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We derive a formula to calculate the ballistic thermal conductance of a two-dimensional system directly from the dispersion relations of phonons and electrons. We apply the method to a graphene and investigate both the temperature and the Fermi energy dependences of the ballistic thermal conductance. The ballistic thermal conductance per unit length of a graphene becomes isotropic from the threefold rotational symmetry. In the intrinsic graphene where the Fermi energy crosses the Dirac point, the thermal conductance of electrons increases in proportion to T^2 with temperature, while the phonon conductance increases in proportion to $T^{1.5}$ due to the quadratic dispersion relation of the out-of-plane acoustic mode and prevails over the electron-derived conductance irrespective of temperature. As the Fermi energy is moved from the Dirac point for the gated graphenes, the thermal conductance of electrons increases monotonically and the temperature dependence changes from a T^2 dependence in the intrinsic graphene to a T-linear one at low temperatures. The electron thermal conductance of the gated graphenes dominates over the phonon contribution at low temperatures.

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I. INTRODUCTION

The exceptional electronic transport properties of low-dimensional graphitic structures have been extensively demonstrated in carbon nanotubes[1] and graphene ribbons.[2–4] With respect to the electronic transport properties, ballistic transport has been observed up to room temperature in carbon nanotubes. As for thermal transport, thermal conductivity of single-walled carbon nanotubes has been measured[5] and the observed T-linear temperature dependence below 50 K has been explained well by ballistic transport.[6]

Recently, a graphene, a single atomic layer of graphite, which is a two-dimensional form of carbon, is found to exhibit high crystal quality and to have ballistic electronic transport at room temperature. Indeed, spin valve devices[7] and field effect transistors[8] (FETs) have been fabricated, although the gate area of FETs is large yet and the FETs work at 4 K at present. On the other hand, the ballistic thermal conductance of a graphene is shown to give the lower limit of the ballistic thermal conductances per unit circumference length of carbon nanotubes.[9]

A graphene has peculiar features about its electronic structure and its phonon dispersion relation.[1,10] A graphene is a zero-gap semiconductor with a linear dispersion relation in the vicinity of the K point (the so-called Dirac point) where the intrinsic Fermi energy crosses. Recently, the Fermi energy was controlled by applying the gate bias voltage.[10] The ballistic electronic conductance of the gated graphene strip has been studied extensively and the existence of a minimum conductivity of order e^2/h at vanishing gate bias and an approximately linear increase with gate bias are found.[11] As for the phonon dispersion relation, the out-of-plane acoustic ZA branch has a quadratic dispersion in the vicinity of the Γ point in the Brillouin zone;[12,13] this is a characteristic feature of the phonon dispersions of layered crystals.[14]

Our purpose is to calculate the ballistic thermal conductance of graphenes in low temperatures and to clarify the dependences on both the temperature and the Fermi energy for gated graphenes.

II. FORMULATION

First, we formulate the ballistic thermal conductance of phonons in a two-dimensional (2D) system. The ballistic thermal current density of the 2D phonon system formed between a hot heat bath and a cold heat bath arranged in the x direction is described as the Landauer heat flux as follows:

$$\dot{Q}_{\rm ph} = \sum_m \int \frac{dk_y}{2\pi} \int_{v_x>0} \frac{dk_x}{2\pi} \hbar\omega_m(\boldsymbol{k}) v_{xm}(\boldsymbol{k}) \times [n(\omega_m(\boldsymbol{k}), T_{\rm hot}) - n(\omega_m(\boldsymbol{k}), T_{\rm cold})], \quad (1)$$

where m represents a phonon mode, $\omega_m(\boldsymbol{k})$ a phonon dispersion of wave vector $\boldsymbol{k}$, $v_{xm}(\boldsymbol{k}) = \frac{\partial\omega_m(\boldsymbol{k})}{\partial k_x}$ is a phonon group velocity in the x direction and $n(\omega_m(\boldsymbol{k}), T)$ the Bose-Einstein distribution function of phonons. Here, the transmission coefficient for each phonon mode is assumed to be unity. Changing the integration variable in Eq. (1) from k_x to $\omega_m(k_x, k_y)$ and assuming a linear dependence of $\dot{Q}_{\rm ph}$ on $\Delta T = T_{\rm hot} - T_{\rm cold}$, the thermal conductance per unit length, $\kappa_{\rm ph} = \dot{Q}_{\rm ph}/\Delta T$, can be written as

$$\kappa_{\rm ph} = \sum_m \frac{k_B^2 T}{2\pi a h} \times 2 \int_0^{k_y^{\max}} dk_y a \int_{x_m^{\min}(k_y)}^{x_m^{\max}(k_y)} dx \frac{x^2 e^x}{(e^x - 1)^2}. \quad (2)$$

Here, a is the lattice constant of a graphene. $x_m^{\min}(k_y) = \hbar\omega_m^{\min}(k_y)/k_B T$ and $x_m^{\max}(k_y) = \hbar\omega_m^{\max}(k_y)/k_B T$, where $\omega_m^{\min}(k_y)$ and $\omega_m^{\max}(k_y)$ are the minimum and the maximum values of $\omega_m(\boldsymbol{k})$ at a fixed k_y, respectively. In Eq. (2), a monotonic change of $\omega_m(\boldsymbol{k})$ as a function of k_x is assumed in the region of $k_x > 0$ in the Brillouin zone and hence $\omega_m^{\min}(k_y)$ and $\omega_m^{\max}(k_y)$ take these extreme values at either $k_x = 0$ or the Brillouin zone edge. Otherwise, the integration over x is divided into the summation of each region of $v_{xm}(\boldsymbol{k}) > 0$ in the Brillouin zone. The indefinite integral $I_1(x)$ of the integration over x in Eq. (2) can be obtained analytically as

$$I_1(x) = -\frac{x^2 e^{-x}}{1-e^{-x}} + 2x\ln(1-e^{-x}) - 2\sum_{n=1}^{\infty}\frac{e^{-nx}}{n^2}. \quad (3)$$

The convergence of the last summation, $\Sigma_{n=1}^{\infty}\frac{e^{-nx}}{n^2}$, is good for $x \geqslant 0$ and it has a value of $\pi^2/6$ in the worst case of $x=0$. Hence, we can evaluate κ_{ph} by calculating actually the single integration over k_y in Eq. (2).

Second, we formulate the ballistic thermal conductance of electrons, $\kappa_{\mathrm{el}}=\dot{Q}_{\mathrm{el}}/\Delta T$. The heat flux[15] $\dot{Q}_{\mathrm{el}}$ of ballistic electrons can be written as

$$\dot{Q}_{\mathrm{el}} = 2\sum_m \int \frac{dk_y}{2\pi}\int_{v_x>0}\frac{dk_x}{2\pi}[\varepsilon_m(\boldsymbol{k})-\mu]v_{xm}(\boldsymbol{k}) \times [f(\varepsilon_m(\boldsymbol{k}),T_{\mathrm{hot}}) - f(\varepsilon_m(\boldsymbol{k}),T_{\mathrm{cold}})], \quad (4)$$

where $\varepsilon_m(\boldsymbol{k})$ is the energy dispersion of electrons, μ is the Fermi energy, and $f(\varepsilon_m(\boldsymbol{k}),T)$ is the Fermi distribution function. The extra factor of 2 arises from the spin degeneracy of each band. Here, the transmission coefficient of each state is assumed to be unity. The similar transformation as the case of phonon leads to the following expression of κ_{el}:

$$\kappa_{\mathrm{el}} = 2\sum_m \frac{k_B^2 T}{2\pi a h} 2\int_0^{k_y^{\max}} dk_y a \int_{x_m^{\min}(k_y)}^{x_m^{\max}(k_y)} dx \frac{x^2 e^x}{(e^x+1)^2}. \quad (5)$$

Here, $x_m^{\min}(k_y)=[\varepsilon_m^{\min}(k_y)-\mu]/k_BT$ and $x_m^{\max}(k_y)=[\varepsilon_m^{\max}(k_y)-\mu]/k_BT$, where $\varepsilon_m^{\min}(k_y)$ and $\varepsilon_m^{\max}(k_y)$ are the minimum and maximum values of $\varepsilon_m(\boldsymbol{k})$ at a fixed k_y, respectively. The indefinite integral $I_2(x)$ of the integration over x in Eq. (5) can be obtained analytically. In the case of $x>0$,

$$I_2(x) = -\frac{x^2 e^{-x}}{1+e^{-x}} - 2x\ln(1+e^{-x}) - 2\sum_{n=1}^{\infty}(-1)^{n-1}\frac{e^{-nx}}{n^2}. \quad (6)$$

The convergence of the last summation, $\Sigma_{n=1}^{\infty}(-1)^{n-1}\frac{e^{-nx}}{n^2}$, in Eq. (6) is very good and it has a value of $\pi^2/12$ at $x=0$. In another case of $x<0$, the analogous equation can be obtained by replacing x by $-x$ in Eq. (6).

Our formulation for the ballistic thermal transport coefficient in the two-dimensional system does not require calculation of a transmission function like the usual formulation.[9] It should be mentioned that the ballistic thermal conductance of a graphene does not depend on the transport direction and is isotropic from the threefold rotational symmetry of a graphene, contrary to that of a graphene strip. In the following numerical calculation of κ_{ph} and κ_{el}, the dispersion relations of $\omega_m(\boldsymbol{k})$ and $\varepsilon_m(\boldsymbol{k})$ are obtained according to a book by Saito *et al.*[1]

III. NUMERICAL RESULTS

First, we present the ballistic thermal conductance of phonons, κ_{ph}, in Fig. 1. The contribution of the low-energy mode is dominant at low temperatures and hence the ballistic phonon conductance below about 20 K is mainly determined by the lowest out-of-plane acoustic mode (ZA branch) which has a quadratic dispersion.[16] The contributions of the in-plane acoustic modes (TA and LA branches) which have linear dispersions cannot be neglected in a temperature range above 20 K.

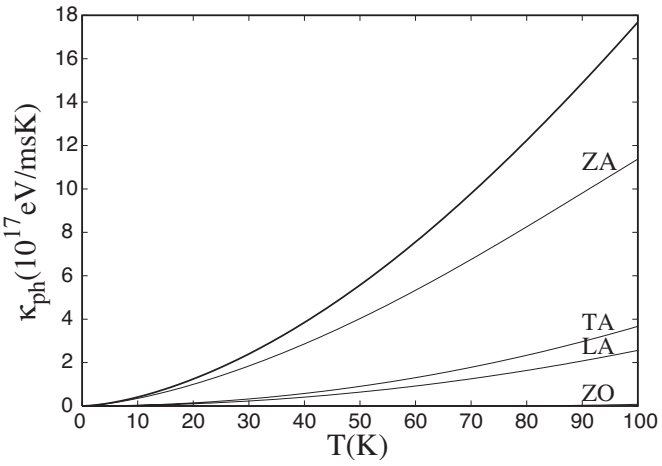

FIG. 1. Phonon-derived thermal conductance κ_{ph} (thick solid line) as a function of temperature and its decomposition (thin solid line) into each mode.

Next, we present the ballistic thermal conductance of electrons, κ_{el}, for some values of the Fermi energies in a rigid band model, $\mu=\varepsilon(K)+\mu_0$, in Fig. 2. A graphene has an isotropic linear dispersion relation in the vicinity of the K and K' points (the so-called Dirac point), where the top of bonding π band and the bottom of antibonding π^* band contact each other at the intrinsic Fermi level of $\varepsilon(K)$. Hence, the π and π^* bands have the same contributions to the ballistic thermal conductance in the case of an intrinsic graphene. For the gated graphenes, on the other hand, π and π^* bands have different contributions to the ballistic thermal conductance but the ballistic thermal conductance does not depend on the sign of the Fermi level shift μ_0, although it depends on the absolute value of μ_0. As the Fermi level increases above or decreases below the Dirac point, the ballistic thermal conductance of electrons increases monotonically and its temperature dependence changes gradually as seen in Fig. 2.

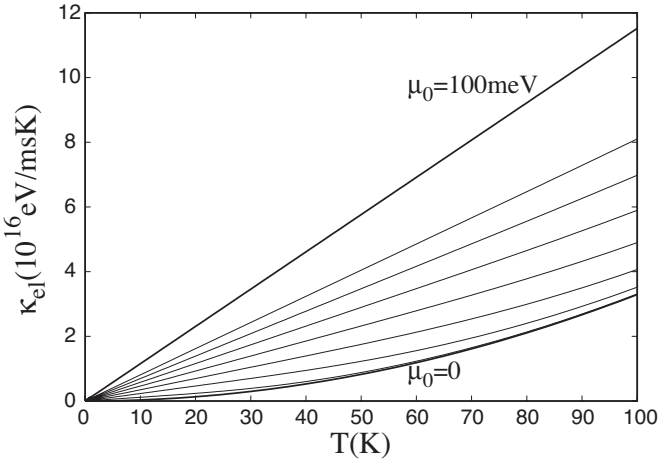

FIG. 2. Electron-derived thermal conductance κ_{el} at $\mu_0=0$ and 100 meV (thick solid line) and at $\mu_0=10,20,30,40,50,60,70$ meV (thin solid line) successively from the bottom, as a function of temperature.

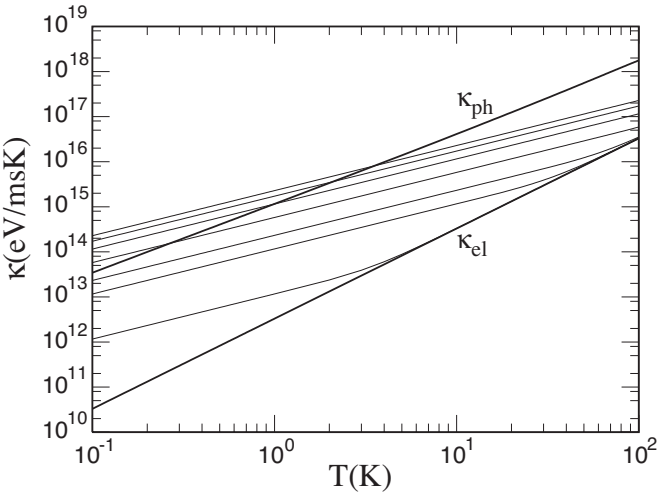

FIG. 3. Phonon-derived thermal conductance κ_{ph}, electron-derived thermal conductance κ_{el} at $\mu_0=0$ (thick solid line), and electron-derived thermal conductance at $\mu_0=1,10,20,50,100,150,200$ meV (thin solid line) successively from the bottom, as a function of temperature in the logarithmic scale.

Finally, the ballistic thermal conductances of both the phonons and the electrons are shown in the logarithmic scale in Fig. 3, to clarify each temperature dependence and to compare their magnitudes with each other. It can be seen that the electron contribution can dominate over the phonon contribution at low temperatures in the case of $\mu_0 > 50$ meV.

IV. DISCUSSION AND CONCLUSION

At first, we discuss the asymptotic temperature dependence of κ_{ph} in the low-temperature regime. The asymptotic behavior can be calculated from Eqs. (2) and (3), but more easily by using the transmission function $T(\omega)$ (Ref. 9):

$$\kappa_{ph} = \int_0^\infty T(\omega)\hbar\omega(dn/dT)d\omega/2\pi a = \frac{k_B^2 T}{ha}\int_0^\infty dx T\left(\frac{k_B T x}{\hbar}\right)x^2\frac{e^x}{(e^x-1)^2}, \tag{7}$$

where $T(\omega)$ can be given as

$$T(\omega) = \sum_m T_m(\omega) = \sum_m \frac{1}{2\pi}\int_{\omega_m(\boldsymbol{k})=\omega} dk_y a. \tag{8}$$

Here, the transmission coefficient of each phonon mode is assumed to be unity. At low temperatures, only low-energy modes can be excited and the transmission function can be calculated easily. The ZA branch for the out-of-plane modes has a quadratic dispersion relation, and its transmission function $T(\omega)$ is proportional to $\sqrt{\omega}$. Hence, its contribution to κ_{ph} has a $T^{1.5}$ dependence at low temperature. On the other hand, TA and LA branches of the in-plane acoustic modes have linear dispersions and their transmission functions are proportional to ω. Hence, their contributions to κ_{ph} have a T^2 dependence at low temperature.

Next, we discuss the asymptotic temperature dependence of κ_{el} in the low-temperature regime. By using the transmission function $T_m(\varepsilon)$, it can be written as

$$\kappa_{el} = \frac{4k_B^2 T}{ha}\sum_m \int_{-\infty}^\infty dx T_m(k_B T x)x^2\frac{e^x}{(e^x+1)^2}. \tag{9}$$

The extra factor of 4 compared to the phonon case arises from both the spin degeneracy and the contributions of K and K' points in the Brillouin zone. If we set $\mu=\varepsilon(K)+\mu_0$ and $\varepsilon_m(\boldsymbol{k})\approx\varepsilon(K)\pm v_F k$ in the vicinity of the K point in the Brillouin zone for π^* and π bands, the transmission function can be obtained as $T(\varepsilon)=\pm\frac{a}{\pi}\frac{[\varepsilon-\varepsilon(K)]}{v_F}$, respectively. Thus, κ_{el} can be obtained analytically for $\mu_0>0$:

$$\kappa_{el} \simeq \frac{4k_B^2 T}{ha}\frac{a}{\pi v_F}\left[2k_B T\frac{9}{2}\frac{\pi^3}{25.8} + 2\,\mu_0 I_2(x)\Big|_0^{\mu_0/(k_B T)} - 2k_B T I_3(x)\Big|_0^{\mu_0/(k_B T)}\right], \tag{10}$$

where $I_3(x)$ is the indefinite integral of $\frac{x^3e^x}{(e^x+1)^2}$ and is given by the equation

$$I_3(x) = -\frac{x^3e^{-x}}{1+e^{-x}} - 3x^2\ln(1+e^{-x}) - 6x\sum_{n=1}^\infty(-1)^{n-1}\frac{e^{-nx}}{n^2} - 6\sum_{n=1}^\infty(-1)^{n-1}\frac{e^{-nx}}{n^3}. \tag{11}$$

In the derivation of Eq. (10), the following equation was used:

$$\int_0^\infty \frac{x^3e^x}{(1+e^x)^2}dx = 6\sum_{n=1}^\infty\frac{(-1)^{n-1}}{n^3} = 6\frac{3}{4}\frac{\pi^3}{25.8}. \tag{12}$$

When $\mu_0=0$, the second and third terms in the brackets of Eq. (10) vanish and κ_{el} is proportional to T^2. When $\mu_0 \gg k_B T$, on the other hand, the first and third terms in the brackets of Eq. (10) cancel each other and κ_{el} is proportional to T.

In Fig. 3, κ_{ph} has a $T^{1.5}$ dependence below about 20 K as pointed out by Mingo and Broido,[9] but it deviates a little upward from the dependence above 20 K. The temperature dependence of κ_{el} changes with increasing μ_0 from the T^2 dependence of an intrinsic graphene to a T-linear dependence above $\mu_0>50$ meV. In the case of $\mu_0=1$, 10, and 20 meV, the electron-derived thermal conductance changes from a T-linear dependence to a T^2 dependence with increasing temperature can be seen. The calculated temperature dependences in Fig. 3 can be explained well by Eq. (10).

Next, we discuss the relation between κ_{el} and the ballistic electronic conductance S per unit length of a graphene. In the diffusive electronic transport of metal, it is well known that the Wiedemann-Franz law is satisfied between the electronic conductivity σ and the thermal conductivity κ of electrons:[15]

$$\frac{\kappa}{\sigma T} = L, \tag{13}$$

where L is called the Lorentz number and it takes a constant value of $L_0 = \frac{1}{3}\pi^2\left(\frac{k_B}{q}\right)^2$.[15] If we evaluate the ballistic electronic conductance S per unit length of a graphene sheet by assuming a small chemical potential difference between two parallel gates, S is given by

$$S = \frac{q^2}{ah}\frac{4}{2\pi}\int_{k_y>0} dk_y a\left[\frac{1}{e^{x_m^{\min}(k_y)}+1} - \frac{1}{e^{x_m^{\max}(k_y)}+1}\right]. \tag{14}$$

Here, the transmission coefficient is assumed to be unity. In calculations of the electronic conductance of a graphene strip between two gates,[11] the authors assumed a deep electrostatic potential in the gate region and the transmission coefficient took a value less than unity. At low temperatures, S can be calculated similarly to $\kappa_{\rm el}$ for $\mu_0 > 0$:

$$S \simeq \frac{4q^2}{ha}\frac{a}{\pi v_F}\left\{2k_BT\ln 2 + 2\mu_0\frac{1}{e^x+1}\Big|_{\mu_0/(k_BT)}^{0} - 2k_BT\left[\frac{xe^{-x}}{1+e^{-x}} + \ln(1+e^{-x})\Big|_{\mu_0/(k_BT)}^{0}\right]\right\}. \tag{15}$$

In the metallic case of $\mu_0 \gg k_BT$, the Wiedemann-Frantz law holds in the ballistic case between $\kappa_{\rm el}$ and S with the same Lorentz number L_0 as the diffusive transport case. In the case of $\mu_0 = 0$, however, the Lorentz number L becomes $L = 2.37L_0$. It means that the heat flux flows more easily than the charge flux caused by the linear dispersion relation around the Dirac point.

Finally, we remark on the relation between the ballistic thermal conductances of a graphene and a carbon nanotube. The ballistic phonon-derived thermal conductance of carbon nanotubes exhibits the universal features of quantized thermal conductance of $\kappa_{\rm ph} = 4\pi^2 k_B^2 T/3h$ at low temperatures, independent of the radius or the atomic geometry.[6] Furthermore, the ballistic electron-derived thermal conductance of metallic carbon nanotubes has also the same universal quantized value as the phonon-derived conductance at low temperatures. Therefore, the thermal conductance per unit circumference length of carbon nanotubes decreases with an increase of the radius and the thermal conductance of a graphene per unit length gives the lower limit as pointed out by Mingo and Broido.[9] On the other hand, the ballistic thermal conductance of graphenes is sensitive to the energy dispersion relations of electrons and phonons. Hence, the phonon conductance depends on the phonon mode and the electron-derived conductance depends on the Fermi energy. The electron-derived conductance becomes much smaller than the phonon-derived conductance for the intrinsic graphene as shown in Fig. 2, contrary to carbon nanotubes.

In summary, we derived a formula to calculate the ballistic thermal conductance in a two-dimensional system directly from the dispersion relations of phonons and electrons and applied the method to graphenes. The ballistic thermal conductance per unit length of a graphene becomes isotropic. In the intrinsic graphene, the thermal conductance of phonons increases in proportion to $T^{1.5}$ at low temperatures and prevails over that of electrons irrespective of temperature. In the gated graphenes with a Fermi energy shift much larger than k_BT, the thermal conductance of electrons increases proportional to T and dominates over the phonon contribution at low temperatures.

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[1] R. Saito, G. Dresselhaus, and M. S. Dresselhaus, *Physical Properties of Carbon Nanotubes* (Imperial College Press, London, 1998).
[2] M. Fujita, K. Wakabayashi, K. Nakada, and K. Kusakabe, J. Phys. Soc. Jpn. **65**, 1920 (1996).
[3] K. Nakada, M. Fujita, G. Dresselhaus, and M. S. Dresselhaus, Phys. Rev. B **54**, 17954 (1996).
[4] T. Yamamoto, K. Watanabe, and K. Mii, Phys. Rev. B **70**, 245402 (2004).
[5] J. Hone, M. Whitney, C. Piskoti, and A. Zettl, Phys. Rev. B **59**, R2514 (1999).
[6] T. Yamamoto, S. Watanabe, and K. Watanabe, Phys. Rev. Lett. **92**, 075502 (2004).
[7] E. W. Hill, A. K. Geim, K. Novoselov, F. Schedin, and P. Blake, IEEE Trans. Magn. **42**, 2694 (2006).
[8] C. Berger, Z. Song, T. Li, X. Li, A. Y. Ogbazghi, R. Feng, Z. Dai, A. N. Marchenkov, E. H. Conrad, P. N. First, *et al.*, J. Phys. Chem. **108**, 19912 (2004).
[9] N. Mingo and D. A. Broido, Phys. Rev. Lett. **95**, 096105 (2005).
[10] K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, M. I. Katsnelson, I. V. Grigorieva, S. V. Dubonos, and A. A. Firsov, Nature (London) **438**, 197 (2005).
[11] J. Tworzydlo, B. Trauzettel, M. Titov, A. Rycerz, and C. W. J. Beenakker, Phys. Rev. Lett. **96**, 246802 (2006).
[12] R. Al-Jishi and G. Dresselhaus, Phys. Rev. B **26**, 4514 (1982).
[13] N. Mounet and N. Marzari, Phys. Rev. B **71**, 205214 (2005).
[14] H. Zabel, J. Phys.: Condens. Matter **13**, 7679 (2001).
[15] J. M. Ziman, *Electrons and Phonons* (Oxford University Press, New York, 1963).
[16] Actually, the set of force constants in Ref. 1 does not give a quadratic dispersion relation in the vicinity of Γ point for the ZA branch, and the force constant $\phi_{to}^{(2)}$ was modified from −0.40 to −0.38 in units of 10^4 dyn/cm to give a quadratic dispersion relation.