

Scaling of the optical phonon frequency in diamond-like elements and III-V group semiconductor compounds

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In this work, the homology in the vibrational properties of solids with diamond and zincblende crystal structure is studied. Introducing the Tolpygo's dimensionless frequency we scale the phonon dispersion curves along high-symmetry directions for diamond-like elements and some III-V semiconductor compounds (AB compounds); also, starting from the diamond-like structure element phonon frequencies, we calculate the optical phonon frequencies for such AB compounds. The agreement between theoretical and experimental data is outstanding considering the simplicity of the calculations needed to obtain these results.

Keywords: Anharmonicity; diamond structure; phonon dispersion curves; Raman scattering; optical-phonon channels of decay.

En este artículo, estudiamos la homología en las propiedades vibracionales de los sólidos con estructuras tipo diamante y zincblenda. Por medio de la frecuencia adimensional de Tolpygo, escalamos las curvas de dispersión de fonones tanto para elementos con estructura de diamante, así como para algunos compuestos semiconductores de los grupos III-V; además, a partir de las frecuencias de los fonones en elementos con estructura de diamante, obtenemos las frecuencias del fonón óptico en compuestos AB. Considerando la sencillez de los cálculos empleados, la concordancia entre los resultados teóricos y los valores experimentales es notable.

Descriptor: Anarmonicidad; estructura de diamante; curvas de dispersión de fonones; esparcimiento Raman; canales de decaimiento del fonón óptico.

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1. Introduction

The homology among vibrational properties of elements having the diamond structure have been studied since Tolpygo's pioneering work [1]. In that paper, the so-called dimensionless or reduced frequency is introduced to study the scaling of germanium and silicon dispersion curves. From these results, it was evident that the scaled curves had a similar behavior showing slight differences. In other words, the behavior of these elements featured some kind of universality and, no matter which element with diamond structure we have, the corresponding dispersion curves can be obtained by scaling the curves from another one with the same crystal structure.

In a series of papers about this problem, Nilsson and Nelin [3] studied the homology between silicon and germanium by using Tolpygo's dimensionless frequency formalism. They built the silicon and germanium reduced dispersion curves in high-symmetry directions that had been obtained previously by Tolpygo, and also calculated the specific heat. Analyzing their results, Nilsson and Nelin suggested that the homology between the vibrational properties in silicon and germanium could be extended to diamond and gray tin (α -Sn).

Zincblende structure like diamond, has FCC translational symmetry and the same tetrahedral bond angle, but a basis with two different atoms per unit cell. Semiconductor compounds of the type AB (where A, and B are elements belonging to the III-V and II-VI groups of the periodical chart) like GaAs, AlAs, GaP, ZnSe, CdTe are well-known examples of materials with this crystalline structure that have been widely studied in recent years.

The similarities between the two crystalline structures strongly suggest that some kind of scaling on the vibrational properties in diamond and zincblende structure must exist. The main goal of this work is to show that, at least with respect to dispersion curves and optical-phonon frequencies, this scaling really exists and might be investigated in more detail.

In this paper, we study the scaled dispersion curves along high-symmetry directions of some III-V semiconductor compounds. Also, from diamond-like and GaAs reduced optical phonon-frequencies, we calculate the optical phonon frequencies for AB compounds and give the comparison between theoretical and experimental results. Finally, we give some concluding remarks, and outline future directions in this work.

TABLE I. Calculated phonon frequencies for AB compounds from a diamond-like element. Experimental data were taken from Refs. 6 and 13.

Compound	Group	Lattice constant (Å)	Raman frequency (cm ⁻¹) Experimental data	Raman frequency (cm ⁻¹) using Eq. (5)
AIP	III-V	5.45		509.6
AlAs	III-V	5.66	403.0	410.6
AlSb	III-V	6.14	345.0	344.8
AlN	III-V	4.42		875.0
GaP	III-V	5.45	402.8	417.3
GaAs	III-V	5.65	297.3	304.9
GaSb	III-V	6.10	240.3	245.8
GaN	III-V	4.52	742.0	757.0
InP	III-V	5.87	351.0	350.1
InAs	III-V	6.06	243.3	245.3
InSb	III-V	6.48	197.0	194.3

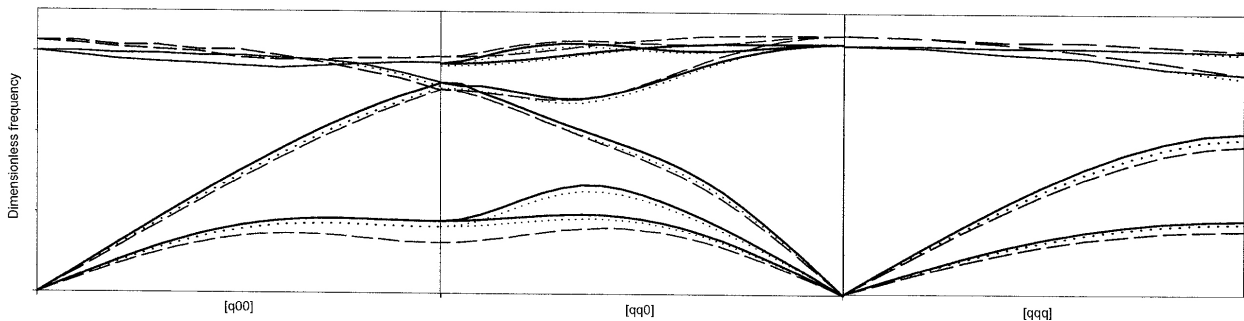


FIGURE 1. Calculated dimensionless dispersion curves of Si, Ge, and α -Sn (Dotted: Ge, Short dashed: α -Sn, solid: Si)

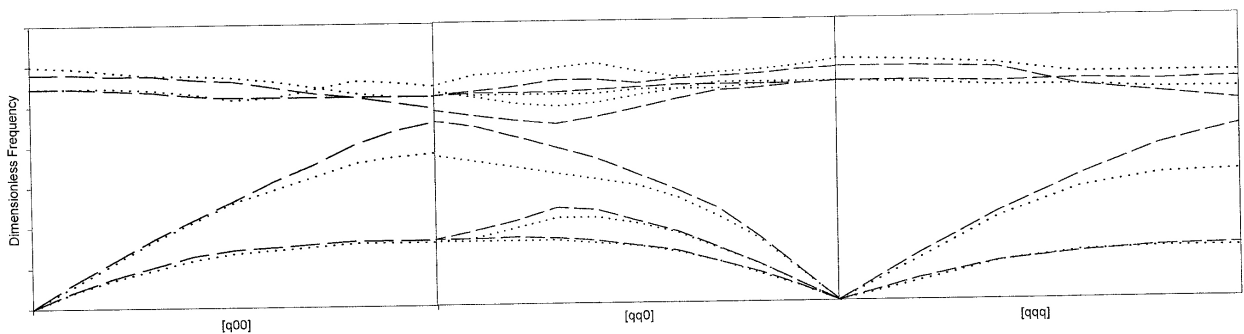


FIGURE 2. Calculated dimensionless dispersion curves of GaAs and GaP (GaAs: Dashed, GaP: Dotted)

2. Dimension-less frequency and scaled dispersion curves on high-symmetry directions: diamond-like elements

The main idea on Tolpygo paper is to introduce the dimension-less frequency Ω_{qj} instead of vibrational frequencies in such a way that element-independent vibrational properties (like frequencies, linewidths and specific heats) could be obtained. The procedure is as follows: the vibrational frequencies (with wave vector and labeling the branch, namely

optical or acoustical) are transformed by

$$\Omega_{qj} = 2\pi \left(\frac{\mu a^3}{e^2} \right)^{\frac{1}{2}} \nu_{qj} \tag{1}$$

where

$$\mu = \frac{m_1 m_2}{(m_1 + m_2)} \tag{2}$$

is the reduced mass, m_1 and m_2 are the masses of atoms in the lattice, a is the lattice constant and e is the electronic charge. This is the only combination of μ , a , e resulting in a dimension-less Ω_{qj} .

In a previous work [5], we have presented a simple four parameter model in order to study the optical-phonon lifetime for crystals having the diamond structure. This four parameter model consists of nearest neighbor, angle-bending, and dipole-dipole interactions. Thus, in that paper, among other results, we calculated the dispersion curves in high-symmetry directions for silicon, germanium, gray tin and diamond obtaining a very good agreement between theoretical and experimental data especially on the boundary zones. From these dispersion curves, it is more than evident that the scaling previously mentioned in Eq. (1) could be applied to diamond-like structure elements. The scaled dispersion curves are shown in Fig. 1; at least as a first approximation the silicon, germanium, and α -tin dispersion curves along high-symmetry directions fit very well, especially the acoustic branches. Another interesting feature is that the phonon density of states in silicon, germanium and gray tin are very similar in shape, undoubtedly a fundamental fact to understand why this scaling is actually working for these diamond-like elements. Diamond, however, has a very different behavior showing marked differences from the other diamond-like elements, and for clearness it's not shown here.

3. Dimension-less frequency and scaled dispersion curves along high-symmetry directions: AB compounds

Following the same procedure outlined above, we can obtain the scaled phonon dispersion curves along high-symmetry directions for AB compounds like GaAs, AlAs, GaP, GaSb, AlSb by using Eq. (1); in this case, reduced mass is given by $\mu = (m_A m_B)/(m_A + m_B)$. Dispersion curves for GaAs-GaP along high-symmetry directions for this type of compounds are shown in Fig. 2. The agreement among scaled curves corresponding to different III-V and II-VI semiconductor compounds is quite good, a fact that has been marginally mentioned before [12].

4. Optical-phonon frequencies of AB compounds from diamond-like elements

From the optical phonon frequencies of diamond-like elements calculated using the four parameter model [5], we can obtain the optical-phonon frequencies for an AB compound, particularly for the longitudinal optical phonons. First, taking the optical-phonon frequency ν_{0d} from a diamond-like element, we apply it to the transformation given by Eq. (1) obtaining Ω_{0d} , the reduced optical-phonon frequency. The second step is to invert this relationship; thus, we obtain

$$\nu_{0,AB} = \frac{1}{2\pi} \left(\frac{e^2}{\mu_{AB} \alpha_{AB}^3} \right)^{1/2} \Omega_{0d}, \quad (3)$$

where Ω_{0d} is the reduced optical-phonon frequency from a diamond-like structure element, and the reduced mass is cal-

culated for a unit cell with two atoms, labeled A and B. Finally, upon substitution of Ω_{0d} , we have

$$\nu_{0,AB} = \left(\frac{\mu_d \alpha_d^3}{\mu_{AB} \alpha_{AB}^3} \right)^{\frac{1}{2}}, \quad (4)$$

therefore

$$\omega_{0,AB} = \left(\frac{\mu_d \alpha_d^3}{\mu_{AB} \alpha_{AB}^3} \right)^{\frac{1}{2}} \omega_{0d}. \quad (5)$$

Table I presents the comparison between theoretical calculations obtained from (5) and experimental results reported in several references. Although the agreement is very good, with this approach we obtain only one of the two optical phonon frequencies present in an AB compound.

Gallium and aluminum nitride (GaN and AlN, respectively) like other III-V nitrides, have received an increasing research interest because of their applications in electronic devices, such as blue light-emitting diodes, and semiconductor optical devices ranging from IR to green wavelengths. At normal ambient conditions, GaN and AlN have wurtzite crystalline structure. Under certain conditions, epitaxial layers of zincblende GaN have been experimentally observed [6] yielding two longitudinal optical-phonon frequencies at 733cm^{-1} and 742cm^{-1} . On the other hand, theoretical calculations based on first-principles pseudopotentials [7] had led to TO phonon frequencies at the center zone; other calculations involving a two-parameter Keating, model [9], give the phonon dispersion curves for zincblende GaN and AlN. Following the same procedure described above, we obtain the Raman mode phonon-frequencies in GaN and AlN; the results obtained in this work are compared with those of Refs. 6 and 9, (Table I). The agreement between them is remarkable and highlights the predictive nature of the dimensionless frequency approach.

5. Discussion and concluding remarks

i) Dispersion curves

Based upon Tolpygo dimensionless frequency, we have shown that, phonon dispersion curves for elements with diamond structure scale very well, so that with the available data for one of these elements (Si, Ge or α -Sn), we could in principle obtain the dispersion curves for the remaining of them. Moreover, analyzing the reduced dispersion curves of AB compounds, we conclude that the homology present in diamond-like materials is also in the former semiconductor compounds. Therefore, we could take advantage of this scaling and use it to calculate other vibrational properties for this kind of compounds.

ii) Optical phonon frequencies

The agreement found between calculated and experimental optical-phonon frequencies suggest that, because of the similarity in crystal structure, not only

these frequencies but line widths and specific heats also must obey some rule of scaling. In other words, by using appropriate transformations and a set of data as a starting point (in this case, the optical-phonon frequency of diamond-like elements or AB compounds)

we could, in principle and with a precision subjected to the accuracy of dispersion curves, calculate another vibrational property for a solid with diamond or zinblende structure.

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