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Effect of temperature dependence ultrasonic velocities and attenuation of GaP nanowires

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Abstract The higher order elastic constants of the hexagonal wurtzite crystal structure of GaP nanowires have been evaluated using Lennard-Jones potential model at room temperature. The ultrasonic velocity increases with the temperature along particular orientation with the unique axis of crystals. Temperature variation of the thermal relaxation time and Debye average velocities is also calculated along the same orientation. The temperature dependency of the ultrasonic properties is discussed in correlation with elastic, thermal and electrical properties. It has been found that the thermal conductivity is the main contributor to the behaviour of ultrasonic attenuation as a function of temperature and the responsible cause of attenuation is phonon–phonon interaction. The mechanical properties of GaP nanowires at low temperature are better than at room temperature, because at low temperature it has low ultrasonic velocity and ultrasonic attenuation. A particularly interesting case is GaP, which is the only (Ga, In)-V semiconductor with an indirect gap in the bulk phase, and are indispensable in modern microelectronic industries.

Keywords Elastic properties · Thermal conductivity · Ultrasonic properties · GaP nanowires

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Introduction

In present times, novel crystal structures in III–V semiconductor nanowires (NWs) have generated immense scientific interest. The dimensions of these wires are too large to exhibit electron/phonon confinement [1, 2]. However, the modified crystal structure in such wires indicates new possibilities in the realm of nanoscience and nanotechnology [3, 4]. It is now well established that NWs of III–V semiconductors can be grown in wurtzite (WZ) phase along their length, though the corresponding bulk materials are in zinc-blende (ZB) phase [5].

All the III–V compounds are important semiconductor materials, and are indispensable in modern microelectronic industries. Despite persistent efforts of worldwide research in exploring the physical and chemical properties of these semiconductors, many aspects of them are still far from thoroughly understood. III–V semiconductors are an intensively studied group of materials due to their favorable optical and electronic properties with widespread applications. With the recent progress in nanowire growth, however, it became possible to fabricate hexagonal polytypes of several III–V compounds [6, 7]. The reason for this phenomenon is the fact that nanowires grow very often along the cubic [111] direction. In contrast to bulk material, in thin nanowires, the stacking sequence changes easily, and under particular growth conditions can be altered completely to the wurtzite (WZ) crystal structure [8].

Recently, the study of semiconductor nanowires has attracted tremendous attention because of their intriguing physical and chemical properties and technological applications in nanodevice design [9, 10]. As an important semiconductor material, gallium phosphide (GaP) has a wide band gap of 2.26 eV, and it can be used in light emission devices in the visible range. This is attractive for

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optoelectronic application, in particular since GaP has a negligible lattice mismatch with respect to Si, making it a perfect candidate for integration of optoelectronic elements with mature Si technology [11].

Structural stability and electronic properties of bare and hydrogenated GaP nanowires in zinc-blende and wurtzite phases have investigated by Mohammad et al. [12] using first-principles calculations based on density functional theory. The electronic properties of very thin gallium phosphide nanowires of different shapes have studied by ab initio method using the generalized gradient approximation [13].

Ultrasonic nondestructive testing is a resourceful technique that can be applied for investigation of several materials. This is useful for characterization of microstructures, appraisal of defects, and evaluation of objects properties. By virtue of this, ultrasonic measurements during fabrication and heat treatment allow ensuring the absence of unacceptable discontinuities and the presence of a particular microstructure with preferred properties. The interaction of ultrasound with microstructure is important for many material problems. Attenuation and backscattering reduce the delectability of flaws, especially in materials with coarse grains or complex microstructures such as platinum alloys. Further, quantification of these wave propagation properties provide information about the microstructure that can be used in materials description studies, e.g. nondestructive determination of grain size. Acoustic coupling constant (non-linearity parameter) plays vital role for conversion of acoustical energy into thermal energy via phonon–phonon interaction in materials science. This property of material is very interested for material scientists since last decade. Wave propagation velocity is another key factor in ultrasonic characterization, which in combination with attenuation can provide important tools in understanding, the inspectability of materials; for example, it can provide information about crystallographic texture. Ultrasonic velocity is directly related to elastic constants and the elastic constants, in particular, provide valuable information on the stability and stiffness of materials [14–16].

Ultrasonic attenuation is very important physical parameter to characterize the material, which is well related to several physical quantities like thermal conductivity, specific heat, thermal energy density and higher order elastic constants [17]. The elastic constants provide valuable information about the bonding characteristic between adjacent atomic planes and the anisotropic character of the bonding and structural stability [18, 19].

Given the aforementioned potential applications, the complete knowledge of mechanical and physical properties of GaP nanowires, however, is still lacking. For example, the non-linear elastic mechanical properties such as higher order elastic constants, attenuation, etc. has not been addressed yet, therefore, in present work we predict the ultrasonic properties of GaP nanowires at different temperatures. The ultrasonic attenuation coefficient, acoustic coupling constants, higher order elastic constants, thermal relaxation time and ultrasonic wave velocities for GaP for unique direction of propagation of wave are calculated as a function of temperature. The calculated ultrasonic parameters are discussed with related thermophysical properties for the characterization of the chosen nanowires.

Theory

In the present investigation, the theory is divided into two parts:

Second-and third order elastic constants

The second (C_{IJ}) and third (C_{IJK}) order elastic constants of material are defined by following expressions.

$$C_{IJ} = \frac{\partial^2 U}{\partial e_I \partial e_J}; \quad I \text{ or } J = 1,\ldots,6 \tag{1}$$

$$C_{IJK} = \frac{\partial^3 U}{\partial e_I \partial e_J \partial e_K}; \quad I \text{ or } J \text{ or } K = 1,\ldots,6 \tag{2}$$

where, U is elastic energy density, $e_I = e_{ij}$ (i or $j = x, y, z$, $I = 1, \ldots, 6$) is component of strain tensor. Equations (1) and (2) leads six second and ten third order elastic constants (SOEC and TOEC) for the hexagonal structure materials [20].

$$\left.\begin{array}{ll} C_{11} = 24.1\,p^4 C' & C_{12} = 5.918\,p^4 C' \\ C_{13} = 1.925\,p^6 C' & C_{33} = 3.464\,p^8 C' \\ C_{44} = 2.309 p^4 C' & C_{66} = 9.851\,p^4 C' \end{array}\right\} \tag{3a}$$

$$\left.\begin{array}{ll} C_{111} = 126.9p^2 B + 8.853p^4 C' & C_{112} = 19.168p^2 B - 1.61p^4 C' \\ C_{113} = 1.924p^4 B + 1.155p^6 C' & C_{123} = 1.617p^4 B - 1.155p^6 C' \\ C_{133} = 3.695p^6 B & C_{155} = 1.539p^4 B \\ C_{144} = 2.309p^4 B & C_{344} = 3.464p^6 B \\ C_{222} = 101.039p^2 B + 9.007p^4 C' & C_{333} = 5.196p^8 B \end{array}\right\} \tag{3b}$$

where $p = c/a$: axial ratio; $C' = \chi a/p^5$; $B = \psi a^3/p^3$; $\chi = (1/8)[\{nb_0\,(n-m)\}/\{\,a^{n+4}\}]\,\psi = -\chi/\{6\,a^2\,(m+n+6)\}$; m, n = integer quantity; b_0 = Lennard-Jones parameter.

Ultrasonic attenuation and allied parameters

The predominant causes for the ultrasonic attenuation in a solid at room temperature are phonon–phonon interaction (Akhieser loss) and thermoelastic relaxation mechanisms. The ultrasonic attenuation coefficient $(A)_{Akh}$ due to

phonon–phonon interaction and thermoelastic relaxation mechanisms is given by the following expression [21].

$$(A/f^2)_{Akh} = 4\pi^2 \left(3E_0 <(\gamma_i^j)^2> - <\gamma_i^j>^2 C_V T\right) \tau/2\,\rho\,V^3 \tag{4}$$

$$(A/f^2)_{Th} = 4\pi^2 <\gamma_i^j>^2 kT/2\rho V_L^5 \tag{5}$$

where, f frequency of the ultrasonic wave; ρ density of the material; V: ultrasonic velocity for longitudinal and shear wave; V_L longitudinal ultrasonic velocity; E_0 thermal energy density; γ_i^j Grüneisen number (i, j are the mode and direction of propagation).

The Grüneisen number for hexagonal structured crystal along <001> orientation or $\theta = 0°$ is direct consequence of second and third order elastic constants. $D = 3(3E_0 <(\gamma_i^j)^2> - <\gamma_i^j>^2 C_V T)/E_0$ is known as acoustic coupling constant, which is the measure of acoustic energy converted to thermal energy. When the ultrasonic wave propagates through crystalline material, the equilibrium of phonon distribution is disturbed. The time for re-establishment of equilibrium of the thermal phonon distribution is called thermal relaxation time (T) and is given by following expression:

$$\tau = \tau_S = \tau_L/2 = 3k/C_V V_D^2 \tag{6}$$

Here T_L and T_S are the thermal relaxation time for longitudinal and shear wave. k and C_V are the thermal conductivity and specific heat per unit volume of the material, respectively. The Debye average velocity (V_D) is well related to longitudinal (V_L) and shear wave (V_{S1}, V_{S2}) velocities. The expressions for ultrasonic velocities are given in our previous papers [22].

Results and discussion

Higher order elastic constants

The lattice constants i.e. the unit cell parameters 'a' (basal plane parameter) and 'p' (axial ratio) for GaP, are 3.8418 Å and 1.649 [23], respectively. The value of m and n for GaP nanowires are 6 and 7. The value of b_0 for GaP is 1.0×10^{-63} erg cm^7. The second order elastic constants (SOEC) and third order elastic constants (TOEC) have been calculated for GaP using Eq. (3a) and bulk modulus (B) are presented in Table 1.

The elastic constants of the material are important, since they are related to hardness and therefore of interest in applications where mechanical strength and durability are important. Also, the second order elastic constants are used for the determination of the ultrasonic attenuation and related parameters. The calculated value of C_{12} is few different than some other [26] results. Actually Wang et al. [26] have based ab initio periodic Hartree–Focke (HF) calculations to evaluate elastic constants, which is quite different from present approach which is used Lennard-Jones potential model. Although obtained order of SOEC are of the same as given in Table 1. The bulk moduli of hexagonal GaP nanowires are evaluating using the second order elastic constants with the formula $B = 2(C_{11} + C_{12} + 2C_{13} + C_{33}/2)/9$. It is obvious from Table 1 that, there is good agreement between the present and reported elastic constants and bulk modulus of GaP nanowires [24–27]. Thus our theoretical approach for the calculation of second order elastic constants for hexagonal structured GaP nanowires at room temperature is well justified. To satisfy the conditions of the mechanical stability following criteria [28]: $C_{11} > C_{12}$|; $(C_{11} + C_{12}) C_{33} > 2C_{13}^2$; $C_{44} > 0$; $C_{66} > 0$ on the elastic constants must be fulfilled. As our calculated results for elastic constants reasonably satisfy aforesaid criteria for the GaP nanowires which indicate that this nanowire is mechanically stable. However, third order elastic constants are not compared due to lack of data in the literature but the negative third order elastic constants are found some previous papers for hexagonal structure materials [29, 30]. Hence applied theory for the evaluation of higher order elastic constants at room temperature is justified.

Ultrasonic velocity and allied parameters

The temperature dependent thermal conductivity of GaP nanowires have been taken from the previous work [25]. The value of C_V and E_0 are evaluated using tables of physical constants and Debye temperature. The value of temperature dependent density (ρ), specific heat per unit volume (C_V), thermal energy density (E_0) thermal

Table 1 SOEC, TOEC and bulk modulus (B) in the unit of GPa of GaP at room temperature

	C_{11}	C_{12}	C_{13}	C_{33}	C_{44}	C_{66}	B
GaP	182.5	44.8	39.6	194.1	47.6	71.6	89.6
[24, 26] GaP	178	56.3	41.2	198.3	51.2		88 [25]

	C_{111}	C_{112}	C_{113}	C_{123}	C_{133}	C_{344}	C_{144}	C_{155}	C_{222}	C_{333}
GaP	−2976	−472	−101	−129	−655	−614	−151	−101	−2355	−2506

Table 2 Density (ρ: in 10^3 kg m^{-3}), specific heat per unit volume (C_V: in 10^5 J m^{-3} K^{-1}), thermal energy density (E_0: in 10^6 J m^{-3}), thermal conductivity (k: in W cm^{-1} K^{-1}) and acoustic coupling constant (D_L, D_S) of GaP

Temp.	ρ (10^3 kg m^{-3})	C_V (10^5 J m^{-3} K^{-1})	E_0 (10^6 J m^{-3})	K (W cm^{-1} K^{-1})	D_L	D_S
50	4.165	1.312	1.74	6.8	49.5	1.46
100	4.160	4.945	17.53	4.0	50.9	1.46
150	4.155	7.279	47.05	2.4	51.6	1.46
200	4.149	8.393	87.57	1.8	52.2	1.46
250	4.141	8.981	134.29	1.5	52.6	1.46
300	4.138	9.372	176.56	1.1	52.7	1.46

Fig. 1 V_L vs angle with unique axis of crystal

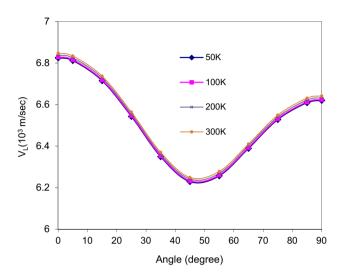**Fig. 2** V_{S1} vs angle with unique axis of crystal

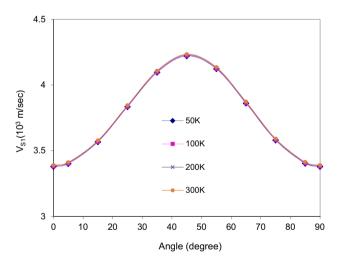**Fig. 3** V_{S2} vs angle with unique axis of crystal

Fig. 4 V_D vs angle with unique axis of crystal

conductivity (k) and calculated acoustic coupling constants (D_L and D_S) are presented in Table 2.

The computed orientation dependent ultrasonic wave velocities and Debye average velocities at room temperature are shown in Figs. 1, 2, 3 and 4. Figures 1, 2 and 3 show that the velocities V_L and V_{S1} have minima and maxima, respectively at 45° with the unique axis of the crystal while V_{S2} increases with the angle from the unique axis. The combined effect of SOEC and density is reason for abnormal behaviour of angle dependent velocities.

The nature of the angle dependent velocity curves in the present work is found similar as that for heavy rare-earth metals, laves-phase compounds, lanthanides metals and third group nitrides [14, 17, 22, 29]. Thus the angle dependencies of the velocities in GaP nanowires are justified.

Debye average velocities (V_D) of GaP nanowires are increasing with the angle and have maxima at 55° at 300 K (Fig. 4). Since V_D is calculated using V_L, V_{S1} and V_{S2} [14, 21], therefore the angle variation of V_D is influenced by the constituent ultrasonic velocities. The maximum V_D at 55° is due to a significant increase in longitudinal and pure shear (V_{S2}) wave velocities and a decrease in quasi-shear (V_{S1}) wave velocity. Thus it can be concluded that when a sound wave travels at 55° with the unique axis of this nanowire then the average sound wave velocity is maximum.

The calculated thermal relaxation time is visualised in Fig. 5. The angle dependent thermal relaxation time curves follow the reciprocal nature of V_D as $\tau \propto 3K/C_V V_D^2$. This implies that T for chosen nanowire is mainly affected by the thermal conductivity. The 'T' for hexagonal structured materials is the order of pico second [17, 29]. Hence the calculated thermal relaxation time justifies the hexagonal structure of chosen compound. The minimum T for wave propagation along $\theta = 55°$ implies that the re-establishment time for the equilibrium distribution of thermal phonons will be minimum for propagation of wave along this direction. Thus the present average sound velocity directly correlates with the Debye temperature, specific heat and thermal energy density of GaP nanowires.

Ultrasonic attenuation due to phonon–phonon interaction and thermal relaxation phenomena

In the evaluation of ultrasonic attenuation, it is supposed that wave is propagating along the unique axis [<001>

direction] of GaP nanowires. The attenuation coefficient over frequency square $(A/f^2)_{Akh}$ for longitudinal $(A/f^2)_L$ and shear wave $(A/f^2)_S$ are calculated using Eq. (4) under the condition $\omega T \ll 1$ at different temperature. The thermoelastic loss over frequency square $(A/f^2)_{Th}$ is calculated with the Eq. (5). The values of temperature dependent $(A/f^2)_L$, $(A/f^2)_S$, $(A/f^2)_{Th}$ and total attenuation $(A/f^2)_{Total}$ are presented in Figs. 6, 7.

In the present investigation, the ultrasonic wave propagates along the unique axis of the crystal, the Akhieser type of loss of energy for longitudinal and shear wave and thermo elastic loss increases with the temperature and maximum at 250 K of the nanowires (Figs. 6, 7). $(A/f^2)_{Akh}$ is proportional to D, E_0, τ and V^{-3} (Eqs. 4, 6). The E_0 is increasing and V is decreasing with the temperature (Figs. 1, 2, 3, 4). Hence, Akhieser loss in GaP nanowires is predominantly affected by the thermal energy density E_0 and the thermal conductivity.

Therefore, the ultrasonic attenuation increases due to the reduction in the thermal conductivity. Thus ultrasonic attenuation is mainly governed by the phonon–phonon interaction mechanism. A comparison of the ultrasonic attenuation could not be made due to lack of experimental data in the literature.

Figures 6 and 7 indicate that the thermoelastic loss is very small in comparison to Akhieser loss and ultrasonic attenuation for shear wave $(A/f^2)_S$ is much greater than that of longitudinal wave $(A/f^2)_L$. This reveals that ultrasonic attenuation due to phonon–phonon interaction along shear wave is governing factor for total attenuation $((A/f^2)_{Total} = (A/f^2)_{Th} + (A/f^2)_L + (A/f^2)_S)$. The total attenuation is mainly affected by thermal energy density and thermal conductivity. Thus it may predict that at 50 K the GaP nanowires behaves as its purest form and is more ductile as evinced by minimum attenuation while at 250 K GaP nanowires is least ductile. Therefore impurity will be least
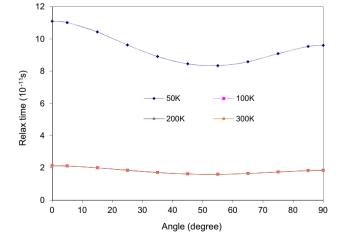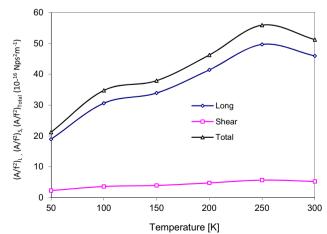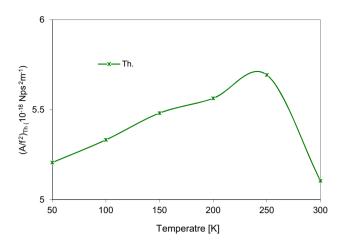
Fig. 5 Relaxation time vs angle with unique axis of crystal

Fig. 6 $(A/f^2)_{Total}$ and $(A/f^2)_{Akh}$ vs temperature of GaP

Fig. 7 $(A/f^2)_{Akh}$ vs temperature of GaP

in the GaP at low temperature (50 K). The minimum ultrasonic attenuation for GaP at 50 K justifies its quite stable hexagonal structure state. The total attenuation of GaP nanowires is much larger than InAs nanowires and third group nitrides due to their large thermal conductivity and acoustic coupling constants [29, 31, 32]. This implies that the interaction between acoustical phonon and quanta of lattice vibration for GaP nanowires is large in comparison to third group nitrides and InAs nanowires.

Summary and conclusions

In summary, our theory, which has evaluated higher order elastic constants for hexagonal GaP nanowires using interaction potential model is justified. For elastic properties, our results indicate that the GaP nanowire is brittle in nature and the obtained results for elastic constants thoroughly satisfy the established criterion of the mechanical stability. Also the order of thermal relaxation time for GaP nanowires is found in picoseconds, which justifies their hexagonal structure. The re-establishment time for the equilibrium distribution of thermal phonons will be minimum for the wave propagation along $\theta = 55°$ due to being smallest value of T along this direction. The acoustic coupling constant of GaP nanowires for longitudinal wave are found much larger than third group nitrides and InAs nanowires. Hence the conversion of acoustic energy into thermal energy will be large for GaP. This shows general suitability of chosen material. The ultrasonic attenuation for shear wave is much greater than that of longitudinal wave. This reveals that ultrasonic attenuation due to phonon–phonon interaction along shear wave is governing factor for total attenuation and total attenuation is a governing factor for thermal conductivity and thermal energy density.

The mechanical properties of GaP nanowires at low temperature (50 K) are better than at high temperature (250 K), because at low temperature it has low ultrasonic attenuation.

Thus obtained results in the present work can be used for further investigations, general and industrial applications. Our theoretical approach is valid for temperature dependent ultrasonic characterization of GaP nanowires. The ultrasonic behavior in GaP nanowires as discussed above shows important microstructural characteristic feature, which are well connected to thermoelectric properties of the material. These results, together with other well-known physical properties, may expand future prospects for the application and study of GaP nanowires.

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