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Size and temperature dependent ultrasonic properties of thermoelectric nanowires

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ABSTRACT

Hicks and Dresselhauss proposed that conversion of bulk materials into low dimensional size might significantly enhance thermoelectric performance. PbTe nanowires have been suggested as promising thermoelectric materials. In the present paper, nonlinear second and third order elastic constants (SOECs/TOECs) for PbTe nanowires (NWs) are calculated using a simple interaction potential model. Finally, the temperature dependent ultrasonic attenuation is determined for different diameters of the PbTe NWs. Good correlation between the size and temperature dependent thermophysical properties and the ultrasonic attenuation has been established. The studies are important for the development of NWs for energy and thermoelectric cooling applications.

1. Introduction

Ever since the discovery of thermoelectricity, it has been a challenging task to enhance the thermoelectricity of available materials, which is characterised by dimensionless figure-of-merit ZT ($ZT=S^2\sigma T/k$) because S (Seebeck coefficient), σ (electric conductivity) and k (thermal conductivity) are independent for bulk materials. However, Hicks and Dresselhauss proposed that conversion of bulk materials into low dimensional size might significantly enhance thermoelectric performance [1]. Whether PbTe NWs are promising thermoelectric materials due to high ZT of bulk PbTe in the mid-temperature range (500–900 K) is investigated [2].

Several scientists have experimented with the mechanisms for the synthesis, electrical and thermal properties of PbTe based nanostructures [2–6]. Synthesising single nanostructures and characterising them is a challenging task due to their small dimensions experimentally. The theoretical ultrasonic studies are rather simple to predict the structural and thermophysical properties of the materials. In view of the above reasons, in this paper we report the calculation of nonlinear elastic properties (SOECs/TOECs) of single crystalline PbTe NWs and finally, the temperature dependent ultrasonic properties such as attenuation and velocity for two diameters of the NWs are determined to predict their microstructural/thermophysical properties.

2. Theory for high order elastic constants and ultrasonic properties

We have calculated the temperature dependent SOECs/TOECs following the procedure used in our previous work based on the simple interaction potential model [7]. This theoretical approach was originally developed for the bulk materials. Akhieser damping and thermoelectric relaxation are the two dominant processes that give rise to the appreciable ultrasonic attenuation at high temperatures due to phonon–phonon interaction. Both types of processes for the ultrasonic attenuation may be observed in nanostructured fcc crystals [7]. The thermal relaxation time for longitudinal ultrasonic wave is twice than that of shear wave given as follows:

$$\tau=\tau_S=0.5\tau_L=\frac{3k}{C_V V_D^2} \tag{1}$$

where k is thermal conductivity and C_V is the specific heat per unit volume; V_D is the Debye average velocity of longitudinal and shear ultrasonic wave velocities given as follows:

$$\frac{3}{{V_D}^3}=\frac{1}{V_L^3}+\frac{2}{V_S^3} \tag{2}$$

where V_L and V_S are longitudinal and shear ultrasonic wave velocities. The expression for ultrasonic attenuation due to

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thermoelastic relaxation is given as follows:

$$(\alpha/f^2)_{Th} = \frac{4\pi^2\langle\gamma_i^j\rangle^2 kT}{2\rho V_L^5} \quad (3)$$

where ρ is density of the material, T is the absolute temperature, α is the ultrasonic attenuation, f is the frequency of the ultrasonic wave, $\langle\gamma_i^j\rangle$ is the average Grüneisen number (i and j are the mode and direction of the propagation, respectively) has been calculated using SOECs and TOECs. The expression for ultrasonic attenuation due to Akhieser damping is given as follows:

$$(\alpha/f^2)_{Akh} = \frac{4\pi^2 E_0 D\tau}{6\rho V^3} \quad (4)$$

where V is the velocity of the ultrasonic waves, and D is the acoustic coupling constant that gives the measure of the acoustic energy converted into thermal energy given by

$$D = 9\langle(\gamma_i^j)^2\rangle - 3\langle\gamma_i^j\rangle^2\frac{C_V T}{E_0} \quad (5)$$

where E_0 is the thermal energy density.

The total ultrasonic attenuation coefficient over frequency square $(\alpha/f^2)_{\text{Total}}$ is given by

$$\left(\frac{\alpha}{f^2}\right)_{\text{Total}} = (\alpha/f^2)_{Th} + (\alpha/f^2)_{Akh} = (\alpha/f^2)_{Th} + (\alpha/f^2)_{Akh.long} + (\alpha/f^2)_{Akh..shear}. \quad (6)$$

3. Results and discussion

The calculated values of elastic constants using the lattice parameter (6.55 Å) [8] are presented in Table 1. The calculated bulk modulus $\{B=(C_{11}+2C_{12})/3\}$ of PbTe NWs is 36.9 GPa which matches the value 37.7 GPa described in the literature [9] well. Thus, the theory for the calculation of SOECs/TOECs for PbTe NWs is well established. All the calculated physical properties are presented in Table 2.

The acoustic coupling constants (D_L and D_S) are given in Table 2 and it is clear that D_L and D_S decrease with temperature and also $D_L > D_S$. Thus the conversion of ultrasonic energy into thermal energy for longitudinal ultrasonic waves is greater than that for shear ultrasonic waves. The plots of thermal conductivity [2] and the calculated thermal relaxation time are shown in Fig. 1(a) and (b), respectively.

When an ultrasonic wave propagates through crystalline material, the equilibrium of phonon distribution is disturbed. The time taken for re-establishing equilibrium of the thermal phonons is called the thermal relaxation time (τ) shown in Fig. 1(b). It is clear from Fig. 1(b) that τ for $d=182$ nm NWs decreases with temperature and increases with the diameter of PbTe NWs. However, no effect of the diameter size on τ is observed at 50 K. For diameter of the NW $d=277$ nm, τ increases from 50 K to 100 K and decreases beyond 100 K. In fact, the phonon–phonon interaction mechanism for ultrasonic attenuation is not prominent at temperatures <100 K. Therefore, τ increases from 50 K to 100 K.

Table 1
SOECs and TOECs ($\times 10^{10}$ Pa) versus temperature of PbTe NWs.

T(K)	C_{11}	C_{12}	C_{44}	C_{111}	C_{112}	C_{123}	C_{144}	C_{166}	C_{456}
50	7.94	0.60	0.71	−192.71	−1.81	0.23	1.36	−2.53	1.36
100	8.10	0.51	0.71	−193.47	−1.20	−0.90	1.36	−2.53	1.36
150	8.28	0.41	0.71	−194.82	−0.55	−2.02	1.36	−2.53	1.36
200	8.48	0.31	0.71	−196.36	0.11	−3.14	1.37	−2.53	1.36
250	8.69	0.22	0.71	−197.98	0.77	−4.27	1.37	−2.54	1.36
300	8.90	0.12	0.71	−199.65	1.43	−5.40	1.37	−2.54	1.36

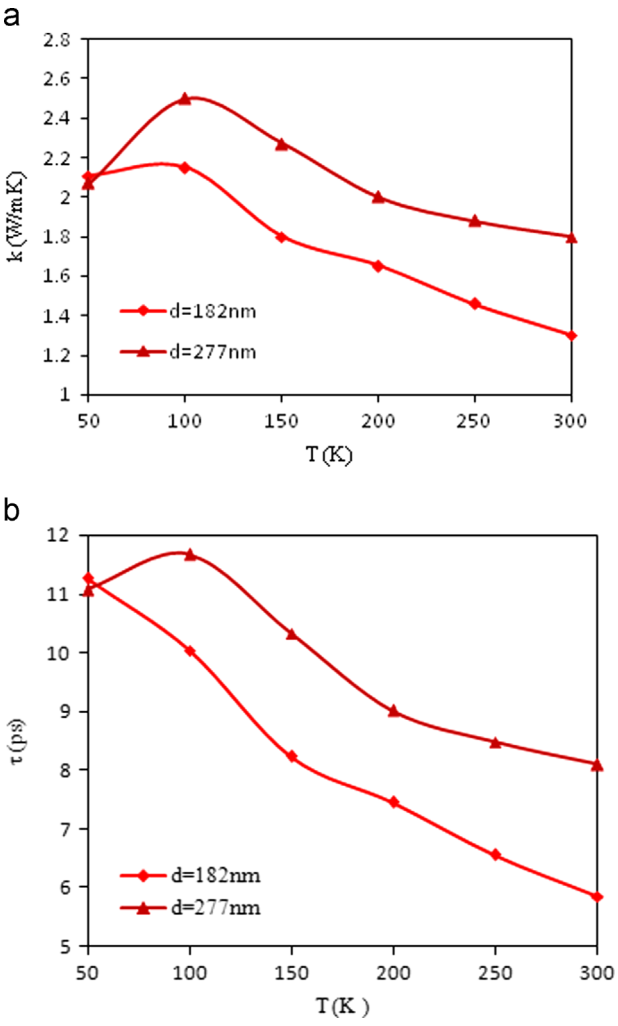

Fig. 1. (a) Thermal conductivity versus temperature and (b) thermal relaxation time versus temperature for different diameters of PbTe NWs.

Table 2
Specific heat per unit volume, thermal energy density, ultrasonic velocities and nonlinearity parameters of PbTe NWs at different temperatures.

T (K)	C_V ($\times 10^5$ J K^{-1} m^{-3})	E_0 ($\times 10^7$ J m^{-3})	V_L ($\times 10^2$ m s^{-1})	V_S ($\times 10^2$ m s^{-1})	V_D ($\times 10^2$ m s^{-1})	D_L	D_S
50	5.17	1.39	31.07	9.264	10.43	48.90	1.262
100	5.90	4.21	31.38	9.266	10.44	48.00	1.260
150	6.03	7.19	31.75	9.267	10.44	46.82	1.259
200	6.09	10.21	32.13	9.268	10.45	45.70	1.259
250	6.09	13.27	32.52	9.270	10.45	44.68	1.258
300	6.11	16.30	32.91	9.271	10.45	43.77	1.257