Studies on Acoustical Dissipation and Related Parameters in Vth Group Mononitrides

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ABSTRACT

Second and third order elastic moduli of fifth group mononitrides (viz. VN, NbN and TaN) have been evaluated using Born model for ionic solids. Using the calculated values of second and third order elastic moduli, temperature dependence of acoustic attenuation for longitudinal and shear modes of propagation along <100>, <110> and <111> directions of propagation have been studied in a wide temperature range (50 K-500 K). Gruneisen parameters and nonlinearity constants have also been calculated for longitudinal and shear waves along different directions of propagation and polarization. Results have been discussed and it has been found that attenuation contribution due to thermoelastic mechanism is negligible compared to the phonon-phonon interaction mechanism. It has also been observed that VN, which is hardest among the series has least attenuation, while, TaN which has the least hardness among the members of the transition metal nitride series has highest attenuation.

INTRODUCTION

Acoustical attenuation, Gruneisen parameters, nonlinearity parameters and thermal relaxation time can be used to obtain insight into the materials microstructure and related physical properties of solids. The study of temperature dependence of sound attenuation in crystals is a powerful tool for understanding the mechanism of interaction of sound wave with the crystal lattice. Predominant causes for acoustic attenuation in solids are phonon-phonon interaction, electron-phonon interaction, thermoelastic loss and dislocation damping. At high temperatures, electron mean free path is not comparable to phonon mean free path, thus attenuation due to electron-phonon interaction is not present at higher temperatures. Therefore, phonon-phonon interaction, thermoelastic loss and dislocation damping are the dominant processes that will give rise the appreciable attenuation beyond 50 K and above. Experimental / theoretical studies of NaCl-type (B1) compounds reveal that dislocation mechanism yields a negligible contribution to sound attenuation in NaCl-type crystals and contribution to thermoelastic loss too, is negligibly small. Therefore, the ultrasonic attenuation is mainly dominant by phonon-phonon interaction mechanism [1-3].

Transition metal nitrides (TMNs) are very important compounds having the unique chemical and physical properties such as superior hardness, high melting temperature etc. TMNs are chemically very stable and extremely hard. Due to these properties, TMNs are widely used in industry as cutting tools. These have potential applications in information storage technology (for coating the magnetic sheets), high power energy industry, and optoelectronics [4, 5]. Metal

nitrides are gaining more and more importance in the high temperature art. They are thermodynamically most stable and technically most interesting nitrides. Many transition metal nitrides have stimulated commercial interest because of their extreme hardness, wear and corrosion resistance and thermal and electrical properties. Several studies [4-10] have been performed on these compounds, but results on acoustical studies are not available on these TMNs. Therefore, in the present investigation, acoustical behaviour of TMNs has been studied. This paper describes a theoretical computational approach of temperature dependence of acoustical dissipation in B1-type V[th] group mononitrides. The principal goal of this work is to show the anharmonicity due to acoustic wave propagation in transition metal nitrides over a wide temperature range and compute related parameters contributing to acoustical dissipation. The calculations have been performed in the framework of Born theory of ionic solids [11] for NaCl (B1) type compounds. This approach is capable of giving good results for evaluating the elastic moduli and hence acoustical dissipation of wide variety of ionic solids. It has been shown that ultrasonic attenuation in these transition metal nitrides is related to the hardness of the material. Larger the hardness, smaller is attenuation.

METHOD OF CALCULATION

Theory of second and third order elastic Moduli

Second and third order elastic moduli (SOEM and TOEM) play a unique role in evaluation of the acoustic attenuation and related properties.
SOEM and TOEM at any required temperature can be evaluated by adding vibrational contribution to elastic constants to second and third order elastic constants [11- 15] at absolute zero viz. C_{ij}^{0} & C_{ijk}^{0} i.e.

$$C_{ij}(T) = C_{ij}^{0} + C_{ij}^{vib.}$$ (1)

$$C_{ijk}(T) = C_{ijk}^{0} + C_{ijk}^{vib.}$$ (2)

Where $C_{ij}^{vib.}$ and $C_{ijk}^{vib.}$ are vibrational contribution to elastic constants. Explicit expressions for $C_{ij}^{vib.}$ and C_{ijk}^{vib} are given in ref [1-3]

Theory of acoustic attenuation

At very low temperatures (liquid helium temperatures), the attenuation resulting from phonon-phonon coupling falls to negligible values as the phonon population is frozen out leaving the residual attenuation. In the next higher temperature range (≈ 4.2 K $\leq T \leq \Theta_D/10$, where Θ_D is Debye temperature), the thermal-phonon lifetime is large (satisfying the condition

$\omega\tau >> 1$, where ω is angular frequency of acoustic wave and τ is thermal relaxation time) and sound attenuation occurs due to general crystal impurities, imperfect polishing and is slightly temperature dependent, since a small number of thermal phonons are present for sound scattering depending on the material. Theoretical evaluation of the phonon-phonon interaction process for $\omega\tau >> 1$ was first carried out by Landau and Rummer for transverse ultrasonic waves and qualitative agreement with experiment was found but the calculated attenuation was too small that can be treated as negligible. The mean free path of the thermal phonons is much larger than the sound wave length in this case [16, 17].

Akhieser treated the attenuation for the case $\omega\tau << 1$ and temperatures $\Theta_D/10 \leq T \leq \Theta_D$ as a combination of phonon-phonon interaction, heat flow losses and viscous damping. In the region $\omega\tau << 1$, it is appropriate to consider the ultrasound wave as causing a modulation of a rapidly relaxing ensemble of phonons. Finally, at high temperatures ($T \geq \Theta_D$) for the condition $\omega\tau << 1$, the attenuation becomes temperature independent [18].

The physical basis for obtaining attenuation coefficient is that the elastic constants contributed by the thermal phonons relax. The phonon contribution to the unrelaxed elastic constants is evaluated by taking into consideration the change in energy of the thermal phonons due to applied instantaneous strain. The frequency of each mode ν_i is changed by $\dfrac{\partial \nu_i}{\nu_i} = -\gamma_i^{\ j} S_j$, where $\gamma_i^{\ j}$ is generalized Gruneisen parameter & S_j is instantaneous strain. It is assumed that all the phonons of a given direction of propagation and polarization have equal change in frequency. Then phonons of ith branch and jth mode suffer a change in temperature $\dfrac{\Delta T_i}{T_0} = -\gamma_i^{\ j} S_j$ (T is the temperature). A relaxed elastic constant is obtained after there is phonon-phonon coupling among various branches and the ΔT_i relax to a common temperature change ΔT, given by $\dfrac{\Delta T}{T} = -\langle \gamma_i^{\ j} \rangle S_j$; where $\langle \gamma_i^{\ j} \rangle$ is the average value of $\gamma_i^{\ j}$. Thermal relaxation time is given as [16, 17]:

$$\tau = \tau_S = \frac{\tau_L}{2} = \frac{3k}{C_v <V>^2}$$ (3)

Where k is thermal conductivity, C_v is specific heat per unit volume and $<V>$ is Debye average velocity.

Acoustic attenuation is directly related to Gruneisen parameter ($\gamma_i{}^j$), acoustic coupling constant (β), thermal relaxation time (τ), thermal energy density (E_o) and ultrasonic velocity. Ultrasonic attenuation due to phonon-phonon interaction in Akhiezer regime (ωτ « 1) is given by [19, 20]

$$(\alpha / f^2)_{p-p} = \frac{2\pi^2 \beta_L E_0 \tau_L}{3 d V_L{}^3} \qquad (4)$$

$$(\alpha / f^2)_{p-p} = \frac{2\pi^2 \beta_S E_0 \tau_S}{3 d V_S{}^3} \qquad (5)$$

Non-linearity coupling constant [17, 18]

$$\beta = 9 \left\langle \left(\gamma_i{}^j \right)^2 \right\rangle - \frac{3 \left\langle \gamma_i{}^j \right\rangle^2 C_v T}{E_0} \qquad (6)$$

$\left\langle \left(\gamma_i{}^j \right)^2 \right\rangle$ and $\left\langle \gamma_i{}^j \right\rangle^2$ are square average & average square Gruneisen parameters, V_l and V_s are sound velocities for longitudinal waves and shear waves respectively and d is density.

Debye average velocity is given by [19, 20]

$$\frac{3}{(<V>)^3} = \frac{1}{V_L{}^3} + \frac{2}{V_S{}^3} \qquad (7)$$

Propagation of sound wave through crystal produces compression and rarefactions as a result heat is transmitted from compressed region (at higher temperature) to rarefied region (at lower temperature) and hence thermoelastic loss occurs, which is given by [19, 20].

$$(\alpha / f^2)_{Th} = \frac{4\pi^2 \left\langle \gamma_i{}^j \right\rangle^2 KT}{2 \rho V_L{}^5} \qquad (8)$$

The loss due to movement of dislocations can be obtained in terms of viscous drag coefficient given as [21]:

$$\Pi_{screw} = 0.071 \text{€} \qquad (9a)$$

$$\Pi_{edge} = 0.053\text{€}(1-\sigma^2) + 0.0079/(1-\sigma^2) (\text{€}/\lambda) \psi \qquad (9b)$$

here $\psi = \text{€}_1 - (4/3) \text{€}_s$, $\text{€}_1 = E_0 \beta_l \tau / 3$, $\text{€}_s = E_0 \beta_s \tau / 3$

$\lambda = (C_{11} + 2C_{12})/3$, $\text{€} = (C_{11} - C_{12} + C_{44})/3$

And $\sigma = C_{12}/(C_{11} + C_{12})$

here λ, €, ψ, σ and € are the bulk modulus, shear modulus, phonon viscosity, Poisson's ratio and compressional viscosity respectively and C_{ij}'s are second order elastic constants.

The diffusion coefficient (D_L and D_S for longitudinal and shear wave) measures the ability of thermal phonons to absorb energy from a sound wave of velocity V. The expressions for D_L and D_S are given by [22, 23]

$$D_L = V_L{}^2 \tau \qquad (10a)$$

$$D_S = V_S{}^2 \tau \qquad (10b)$$

Where V_L and V_S are the sound velocity for longitudinal and shear waves and τ is thermal relaxation time.

RESULTS AND DISCUSSION

Second and third order elastic moduli (SOEM and TOEM) have been computed using input parameters (nearest neighbour distance, r = 2.9246 A°, 3.1070A° and 3.1303A° [4] and hardness parameters, q = 0.4162A°, 0.3887A° and 0.4020A° (calculated using best fit values of bulk modulus) for VN, NbN and TaN respectively) as given in Tables 1 and 2. Using present approach, these calculated values of C_{11}, C_{12} and C_{44} for VN are about 24.4%, 31.8% and 34.5%, respectively overestimated, while using other approaches these elastic moduli are overestimated upto 38% compared to the experimental data.

Table 1. Calculated, experimental and other work second order elastic constants (10^{12} dynes/cm^2) of Vth group transition metal nitrides with B1 structure

	C_{11}	C_{12}	C_{44}
VN			
Present	5.33	1.78	1.79
Experiment	5.33[b]	1.35[b]	1.33[b]
Other work	6.52[b], 7.38[b]	1.63[b], 1.86[b]	1.37[b] 1.19[b]
NbN			
Present	6.08	1.31	1.33
Experiment	6.08[a] 5.56[b]	1.34[a] 1.52[b]	1.17[a] 1.25[b]
Other work	7.39[a], 7.18[b]	1.61[a], 1.25[b]	0.75[a], 0.97[b],
TaN			
Present	5.51	1.30	1.31
Experiment	6.79[c]	1.50[c]	1.90[c]
Other work	7.88[a], 8.86[c]	1.67[a], 1.62[c],	1.20[a], 2.66[c]

[a]Reference [2].
[b]Reference [5].
[c]Reference [6].

For NbN these second order elastic moduli (C_{11} and C_{12}) are about 3.23% and 13.81% overestimated respectively while C_{44} is about 6.4% underestimated compared to experimental data. Our second order elastic modulii C_{11}, C_{12} and C_{44} for TaN are about 26.21%, 13.33% and 31.05% respectively underestimated compared to the experimental data while, values obtained by others have larger deviation. Thus our values of second order elastic moduli are in good agreement with available experimental and theoretical data [5, 8, and 9]. Minor disagreement can be attributed to the fact that a precise determination of the elastic properties of refractory compounds is difficult from the viewpoint of both theoretical calculation and experimental measurement due to the availability of good sample quality and difficulty in growing single crystal of reasonable quality. No experimental data are available for TOEC of these TMNs, but our calculated values of TOEC for LiF using present approach are in reasonable relation to those observed experimentally for LiF [24]. These TOEC values may be considered as a good prediction of the experimental observation. Therefore, our calculated values of SOEM and TOEM for these TMNs are satisfactory.

Debye temperatures (Θ_D) [10], phonon energy density (E_o), specific heat (C_v) [25] and Debye average velocity, (<V>) are given in Table (3). Debye average velocity is found to decrease with increasing molecular weight. This shows that larger the mass of the cation lower the Debye average velocity.

Square average Gruneisen number (($< \gamma_i^{j2} >_l$) and ($< \gamma_i^{j2} >_s$)) and average square Gruneisen parameter (($< \gamma_i^j >^2{}_l$) and ($< \gamma_i^j >^2{}_s$)) for longitudinal and shear waves, nonlinearity coupling constants (β_l, β_s and βs*) and viscous drag coefficient (Π_{screw} and Π_{edge}) have been given in Table (4) and (5). These values are SOEM and TOEM dependent. Therefore, on the basis of good agreement of SOEM and TOEM, the values of γ_i^j, β and Π are valid.

Table 2. Third order elastic constants (10^{12} dynes/cm^2) of the materials at 300K

	C_{111}	C_{112}	C_{123}	C_{144}	C_{166}	C_{456}
VN	-75.62	-14.47	5.36	5.35	-14.58	5.34
NbN	-92.25	-10.83	4.21	4.20	-10.96	4.19
TaN	-82.41	-10.62	4.08	4.07	-10.75	4.07
LiF	-6.75	-2.60	1.01	1.01	-2.80	0.986
Ref. [11]	-5.97	-2.61	0.91	0.97	-2.64	0.880

At a given temperature, along a particular direction of propagation, acoustic attenuation (α/f^2) has been found to increase from VN to TaN (Table 3) which can be explained on the basis of the fact that for a given temperature, larger the

value of Debye temperature, smaller will be thermal energy density (since E_o depends on Θ_D). Thus, less phonon-phonon interaction hence small attenuation. Debye temperature depends on SOEM values through the sound velocity [23]. Therefore, increase in attenuation ((α/f^2)$_l$, (α/f^2)$_s$ and (α/f^2)$_{th}$) from VN to TaN is indirectly governed by SOEM values.

Table 3. Debye temperature $\Theta_{D,(K)}$ phonon energy density E_O (10^8erg/ mole K), specific heat C_v (10^7erg/mole K) and Debye average velocity, <V>(10^5cm/sec) at 300 K.

.	Θ_D (K)	E_o	C_v	<V>
VN	772	2.6264	1.7286	6.5375
NbN	730	2.9335	1.4956	4.8848
TaN	432	3.2581	1.5829	3.5355

Temperature variation of acoustical attenuation due to p-p interaction for longitudinal and shear wave {(α/f^2)$_l$ and (α/f^2)$_s$} calculated using equations. (4) and (5) along [100], [110] and [111] directions of propagation are shown in Figs. (1) and (2).

Table 4. Square Average and average square Gruneisen number for longitudinal $< \gamma_i^{j2} >_l$, $< \gamma_i^j >^2{}_l$ and shear $< \gamma_i^j >^2{}_s$, $< \gamma_i^j >^2{}_{s*}$ Waves, Nonlinearitycoupling constants β_l, β_s at 300K

	$< \gamma_i^{j2} >_l$	$< \gamma_i^j >^2{}_l$	$< \gamma_i^j >^2{}_s$	$< \gamma_i^j >^2{}_{s*}$	β_l	β_s	β_{s*}
VN							
[100]	5.35	3.10	1.29	----	28.27	11.62	----
[110]	5.68	3.96	10.06	10.08	25.73	90.61	90.80
[111]	22.17	8.68	5.26	4.25	143.89	47.42	38.27
NbN							
[100]	5.18	2.65	0.70	----	30.17	6.32	----
[110]	4.73	3.24	8.44	11.43	22.40	75.99	102.90
[111]	47.36	12.32	5.86	5.47	349.65	52.82	49.26
TaN							
[100]	5.17	2.73	0.79	----	33.37	7.11	----
[110]	4.82	3.35	8.76	11.12	27.20	78.87	100.12
[111]	40.47	11.36	5.72	5.04	309.39	51.51	45.41

[l] for longitudinal wave
[S] for shear wave, polarized along [001]
[S*] for shear wave, polarized along [$1\bar{1}0$]

Temperature variation of acoustical attenuation due to thermoelastic mechanism, obtained using equation (8) is shown in Fig. (3). Temperature variation of diffusion coefficient (D) for longitudinal and shear wave is shown in Fig. (4) (This gives the energy of thermal phonons absorbed from sound wave). The overall features of the temperature variation of (α/f^2) for all compounds of the series studied are similar. Rate of change of acoustic attenuation due to phonon-phonon interaction $((\alpha/f^2)_l$ and $(\alpha/f^2)_s)$ with temperature is small in the temperature range 50-100 K, since in the temperature range $(\approx 4.2\ K \leq T \leq \Theta_D/10)$ the energy density E_o is small enough hence it creates less phonon-phonon interaction due to which rate of transfer of energy from acoustical phonons to thermal phonons is small, resulting small attenuation due to p-p interaction. In the temperature range $\Theta_D/10 \leq T \leq \Theta_D$, thermal energy density (E_0) increases rapidly (Fig.6) so that phonon-phonon interaction increases at faster rate, thus rate of increase in attenuation is large in the temperature range 100 K-500 K.

Acoustic dissipation due to thermoelastic loss, $(\alpha/f^2)_{th}$ increases up to room temperature and then becomes nearly constant due to conversion of acoustic energy into thermal energy. Conversion of acoustic energy into thermal energy is given by acoustic coupling constant.

Table 5. Viscosity due to screw and edge dislocation of V groupmononitrides at 300K for longitudinal (in cp) and shear(in mp.) waves.

|		Π_{screw}	Π_{edge}
VN			
Long.	[100]	0.24	0.20
	[110]	0.22	0.11
Shear	[100]	0.99	0.87
	[110]	7.78	5.61
NbN			
Long.	[100]	0.37	0.30
	[110]	0.27	0.13
Shear	[100]	0.78	0.81
	[110]	9.40	6.47
TaN			
Long.	[100]	0.75	0.63
	[110]	0.61	0.34
Shear	[100]	1.61	1.68
	[110]	17.9	12.50

Table 6. Ultrasonic attenuation due to phonon-phonon interaction and thermoelastic loss (10^{-19} dB sec^{2}cm^{-1}) along different directions of propagation and Vickers's hardness (GPa) for VN, NbN and TaN at 300 K

| Comp.	$(\alpha/f^2)_l$	$(\alpha/f^2)_s$	$(\alpha/f^2)_{Th}$	Vickers's Hardness
VN [100]	2.73	5.78	0.13	14.4
[110]	2.49	45.03	0.16	
[111]	13.92	23.58	0.37	
NbN[100]	4.04	8.23	0.10	13.3
[110]	3.00	98.86	0.12	
[111]	46.87	68.72	0.47	
TaN [100]	12.10	22.23	0.23	11.0
[110]	9.86	246.35	0.29	
[111]	112.19	160.91	0.99	

Acoustic coupling constant for longitudinal wave along different directions of propagation is shown in figure (6), which increases upto room temperature and then becomes nearly constant.
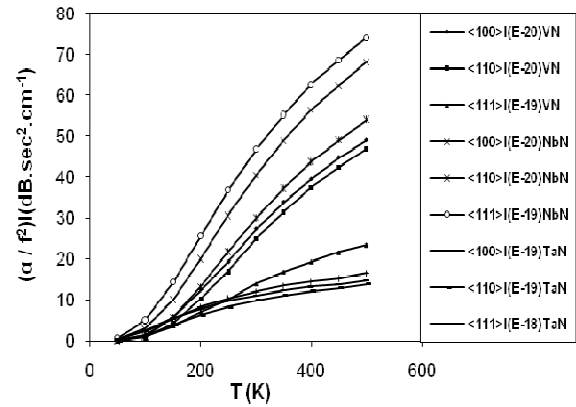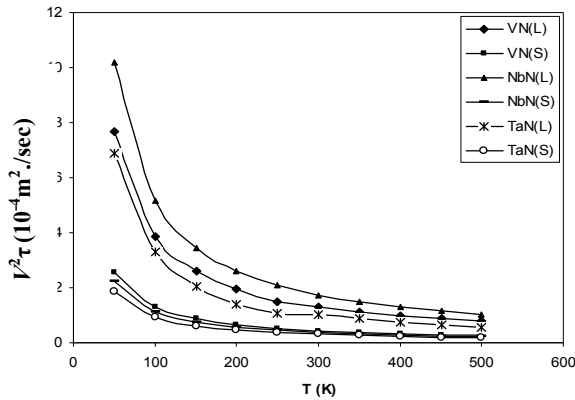

Figure 1. Temperature variation of $(\alpha/f^2)_l$ along diffrent directions for Vth group mononitrides
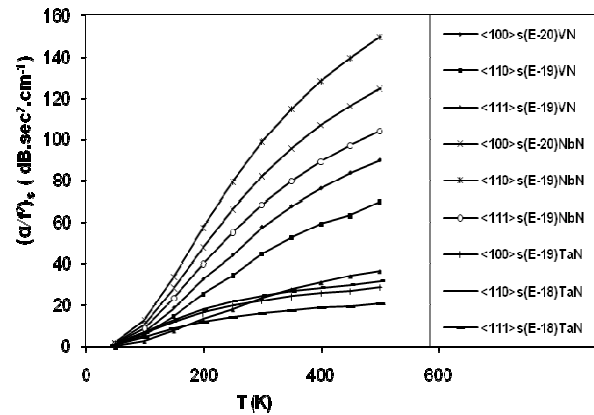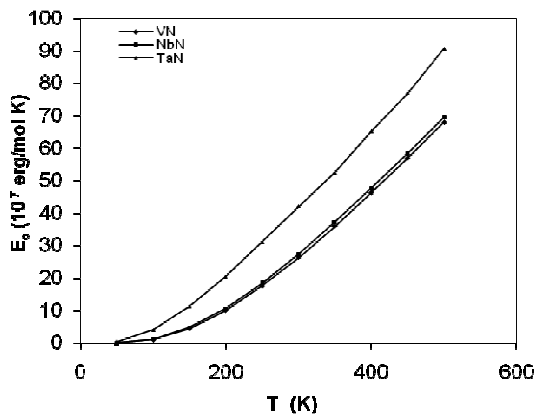

Figure 2. Temperature variation of $(\alpha/f^2)_s$ along different directions for Vth group mononitrides
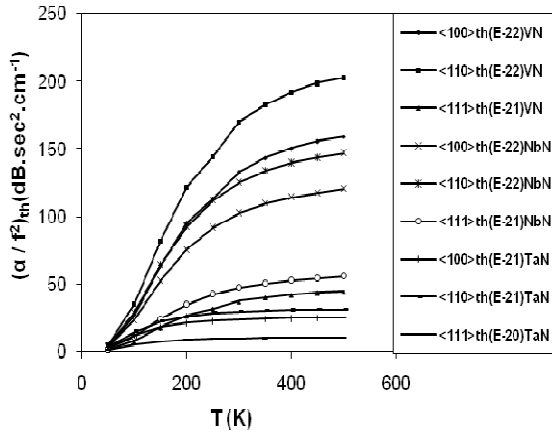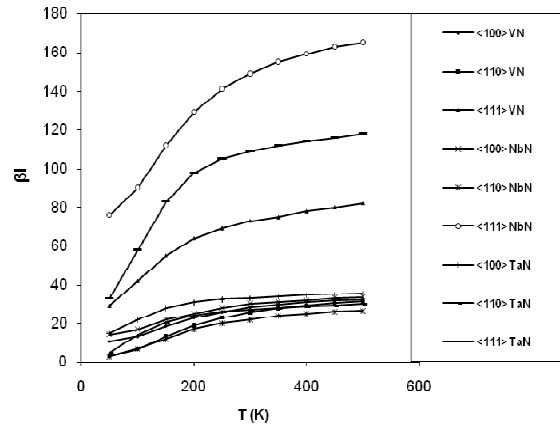
Figure 3. Temperature variation of $(\alpha/f^2)_{th}$ along different directions for V^{th} group mononitrides

Figure 4. Temperature variation of diffusion coefficient (D) for longitudinal and shear wave for V^{th} group mononitrides

Figure 5. Temperature variation of energy density, E_0 for V^{th} group mononitrides

Figure 6. Temperature variation acoustic coupling (β_l) constant for longitudinal wave along different directions of propagation for V^{th} group mononitrides

From Table 6, it can be seen that (α/f^2) due to thermoelastic loss is about 1% (negligible) of (α/f^2) due to phonon-phonon interaction. Thus phonon-phonon interaction mechanism is mainly responsible for acoustic dissipation.

In Table 6, (α/f^2) due to phonon-phonon interaction and Thermoelastic loss alongwith hardness [27] of these transition metal nitrides has been shown. It can be seen that ultrasonic attenuation along all the directions of propagation is least for VN which is the hardest in the group and is the largest for TaN, which has smallest hardness. This can be explained on the basis that less phonon-phonon interaction will occur; therefore less energy will be dissipated from propagating sound wave. Hence (α/f^2) will be small. However, for less hard metal nitride lattice vibrations occur easily resulting enhanced phonon-phonon interaction causing larger attenuation

CONCLUSION

Our values of second and third order elastic moduli are in good agreement with available experimental/theoretical values. The Debye temperature and elastic moduli are basic parameters through which acoustic characterization of different type of materials can be made. Acoustic attenuation is negligible in the temperature range, $(\approx 4.2K \leq T \leq \Theta_D /10)$ and increases rapidly in temperature range, $(\Theta_D/10 \leq T \leq \Theta_D)$. The attenuation due to thermo elastic loss is only 1% to the attenuation due to phonon-phonon interaction; therefore, phonon-phonon interaction is the dominant mechanism for acoustic attenuation in these transition metal nitrides. Thus ultrasonic attenuation is mainly governed by the loss due to phonon-phonon interaction phenomena in these transition metal nitrides. Acoustic attenuation increases with molecular

weight in these TMNs. Also, it has been observed that ultrasonic attenuation for VN is the smallest (which is hardest) and for Tan it is largest which has least hardness in the group.

ACKNOWLEDGEMENT

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