Ultrasonic behaviour of velocities and higher order elastic constants in Zener alloys

P. K. Yadawa[*] and Devraj Singh

Department of Applied Physics, AMITY School of Engineering and Technology,
Bijwasan, New Delhi-110 061, India

*Email: pkyadawa@aset.amity.edu

ABSTRACT

The ultrasonic properties of the hexagonal closed packed solid Ag-Zn alloys (Zener alloys) have been studied at room temperature for their characterization. For the investigations of ultrasonic properties, we have also computed second order elastic constants using Lennard–Jones potential. The velocities V_1 and V_2 have maxima and minima respectively with 45^0 with unique axis of the crystal, while V_3 increases with the angle from unique axis. The inconsistent behaviour of angle dependent velocities is correlated to the action of second order elastic constants. Debye average sound velocities of these alloys are increasing with the angle and has maximum at 55^0 with unique axis at room temperature. Hence when a sound wave travels at 55^0 with unique axis of these alloys, then the average sound velocity is maximum. Achieved results have been discussed and compared with available experimental and theoretical results.

Keywords: Alloys; elastic properties; ultrasonic properties.

1. Introduction

The existence of glasslike excitations in a wide variety of discovered crystals further increases the puzzle of their origin. It is a very good investigation for classifying the various types of disorder in crystal that lead to these excitations. Since all of these crystals distortions by elastic dipoles and possibly interactions between such dipoles, can be envisioned, we decided to investigate the question of whether alloys showing Zener relaxation may also have glasslike excitations. They are a large class of alloys that have been extensively studied over the last four decades at higher temperatures after it had discovered by Zener that in - brass (Cu: Zn) an internal friction peak exists above room temperature (typically around ~ 700K) [1, 2]. These relaxations are believed to arise from a thermally activated stress-induced reorientation of elastic dipoles consisting of solute atom pairs present in the solid solution in a nearest neighbour configuration. Typically activation energies with these relaxations are in the range of 1 eV [3]. The Ag-Zn alloys are the prototype of the noble-metal–divalent-metal (Zn, Cd) Hume-Rothery systems. The crystal structures and the solubility limits of Hume-Rothery intermediate phases were proposed [4] to correlate with the number of the valence electrons per atom (1 for the noble metal and 2 for Zn, Cd). Thus, an extensive study of the Ag-Zn binary alloys can promote a general description and comprehension of the properties of this important class of material. The interdiffusion coefficients and the ratio of the intrinsic diffusion coefficients in Ag-Zn alloys have been determined by Shimozaki et al [5]. Magyari-Kope et al. [6] investigated the elastic properties of the random Ag-Zn binary alloys in fcc and bcc phases.

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 [7, 8].

In the present investigation, the ultrasonic properties like orientation dependent sound velocities, higher order elastic constants for the alloys $Ag_{1.0}Zn_{0.0}$: alloy-1; $Ag_{0.9}Zn_{0.1}$: alloy-2; $Ag_{0.8}Zn_{0.2}$: alloy-3; $Ag_{0.7}Zn_{0.3}$: alloy-4; $Ag_{0.6}Zn_{0.4}$: alloy-5; $Ag_{0.5}Zn_{0.5}$: alloy-6; $Ag_{0.4}Zn_{0.6}$: alloy-7; $Ag_{0.3}Zn_{0.7}$: alloy-8; $Ag_{0.2}Zn_{0.8}$: alloy-9 are evaluated and discussed.

2. Theory

2.1 Higher order elastic constants

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

$$C_{IJ} = \frac{\partial^2 U}{\partial e_I \partial e_J}; \qquad I \ or \ J = 1,......6 \qquad (1)$$

$$C_{IJK} = \frac{\partial^3 U}{\partial e_I \partial e_J \partial e_K}; I \ or \ J \ or \ K = 1,......6 \qquad (2)$$

where, U is elastic energy density , $e_I = e_{ij}$ (i or j = x, y, z, I=1, …6) is component of strain tensor. Eqs. (1)- (2) leads six second and ten third order elastic constants (SOEC and TOEC) for the hexagonal closed packed structured materials [9].

$$
\begin{aligned}
C_{11} &= 24.1 p^4 C' & C_{12} &= 5.918 p^4 C' \\
C_{13} &= 1.925 p^6 C' & C_{33} &= 3.464 p^8 C' \\
C_{44} &= 2.309 p^4 C' & C_{66} &= 9.851 p^4 C'
\end{aligned} \right\} \qquad (3)
$$

Where $C' = \chi \ a \ / \ p^5$; $\chi = (1/8)[\{nb_0 (n-m)\}/\{ a^{n+4} \}]$;

m, n=integer quantity; b_0=Lennard Jones parameter. $p=c/a$: axial ratio; *c* is the height of the unit cell and *a* be the basal plane distance.

2.2 Ultrasonic Velocity

The anisotropic behaviour of the material can be understood with the knowledge of ultrasonic velocity because the velocity is related to the second order elastic constants [9]. On the basis of mode of atomic vibration, there are three types of velocities (longitudinal, quasi shear and shear) in acoustical region [10]. These velocities vary with the direction of propagation of wave from the unique axis of hexagonal structured crystal [11]. The ultrasonic velocities as a function of angle between direction of propagation and unique axis for hexagonal structured materials are [12]:

$$V_1^2 = \{C_{33}\cos^2\theta + C_{11}\sin^2\theta + C_{44}$$
$$+ \{[C_{11}\sin^2\theta - C_{33}\cos^2\theta$$
$$+ C_{44}(\cos^2\theta - \sin^2\theta)]^2$$
$$+ 4\cos^2\theta\sin^2\theta(C_{13}+C_{44})^2\}^{1/2}\}/2\rho \quad (4)$$
$$V_2^2 = \{C_{33}\cos^2\theta + C_{11}\sin^2\theta$$
$$+ C_{44} - \{[C_{11}\sin^2\theta - C_{33}\cos^2\theta$$
$$+ C_{44}(\cos^2\theta - \sin^2\theta)]^2$$
$$+ 4\cos^2\theta\sin^2\theta(C_{13}+C_{44})^2\}^{1/2}\}/2\rho \quad (5)$$
$$V_3^2 = \{C_{44}\cos^2\theta + C_{66}\sin^2\theta\}/\rho \quad (6)$$

where V_1, V_2 and V_3 are longitudinal, quasi shear and pure shear wave ultrasonic velocities. Variables ρ and θ represent the density of the material and angle with the unique axis of the crystal respectively. The Debye temperature (T_D) is an important physical parameter for the characterization of materials, which is well related to the Debye average velocity (V_D).

$$T_D = \frac{\hbar V_D (6\pi^2 n_a)^{1/3}}{k_B} \quad (7)$$

here
$$V_D = \left\{\frac{1}{3}\left(\frac{1}{V_1^3} + \frac{1}{V_2^3} + \frac{1}{V_3^3}\right)\right\}^{-1/3}$$

$$(8)$$

where $\hbar$ is quantum of action and is equal to Planck's constant divided by 2π; k_B is Boltzmann Constant; n_a is atom concentration.
The above formulae have been used for the evaluation of ultrasonic velocity and related parameters for the selected materials.

3. Results and Discussion

The unit cell parameters 'a', axial ratio 'p' and calculated values of second order elastic constants of the chosen alloys are presented in table 1. The value of m and n for chosen materials are 6 and 7. The value of b_0 is 1.0×10^{-67} erg cm^7 for these alloys. The compute oriented dependent ultrasonic velocities of these alloys are visualized in figures. 1-4. As we know that the elastic constants of a solid provide

valuable insight into the nature of atomic bonding forces and are also related to hardness and are used for the determination of the ultrasonic velocity. table 1 depicts that values of SOEC are more or less as obtained by others theoretically [13] and experimentally [14].

Table 1. Lattice parameter 'a' (in Å), 'p' and Second order elastic constants (in 10^{10}Nm^{-2}) of AgZn alloys at room temperature.

a	p
1.656	1.645
1.64	1.64
1.641	1.646
1.63	1.645
1.619	1.63
1.609	1.613
1.598	1.598
1.582	1.579
1.572	1.574

| Alloys	C_{11}	C_{12}	C_{13}	C_{33}	C_{44}	C_{66}
1	8.265	2.030	1.786	8.699	2.143	3.241
2	8.808	2.163	1.892	9.158	2.270	3.454
3	9.047	2.222	1.958	9.545	2.348	3.548
4	9.682	2.378	2.093	10.19	2.510	3.797
5	10.46	2.568	2.219	10.61	2.662	4.100
6	11.24	2.761	2.336	10.94	2.802	4.409
7	12.15	2.984	2.479	11.39	2.973	4.766
8	13.60	3.340	2.709	12.15	3.249	5.334
9	14.54	3.570	2.877	12.83	3.451	5.701
8[13]	11.0	5.6	6.3	12.9	2.7	
8[14]	13.0	6.5	6.4	15.8	4.1	

Hence our theoretical approach to evaluate elastic constants is justified and applicable for Zener alloys. It can be also seen from table 1, that the SOEC are found to be increasing from alloys: 1-9 due to large grain size of hcp Zn metals. The SOEC of these alloys are less than pure Zn metals [15] and pure Ag metals [16].
Figures.1–3 clear that velocities V_1 and V_2 have minima and maxima respectively at 45° with the unique axis of the crystal, while V_3 increases with the angle from the unique axis. Indistinct activity of angle dependent velocities is correlated with activity of SOEC. The tendency of angle

dependent velocity curves (figures. 1-4) reveals the same nature of angle dependent curves for hexagonal wurtzite structured materials GaN, AlN, InN, CdS, CdSe and laves-phase compounds [9, 11, 12, 17, 18]. This shows velocity of our theoretical approach for evaluations of ultrasonic velocities.
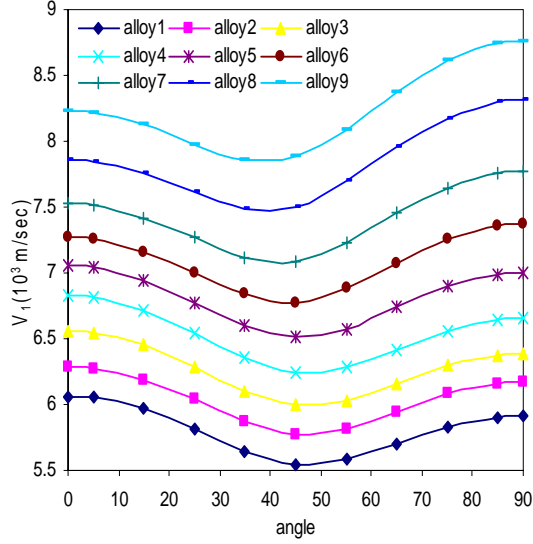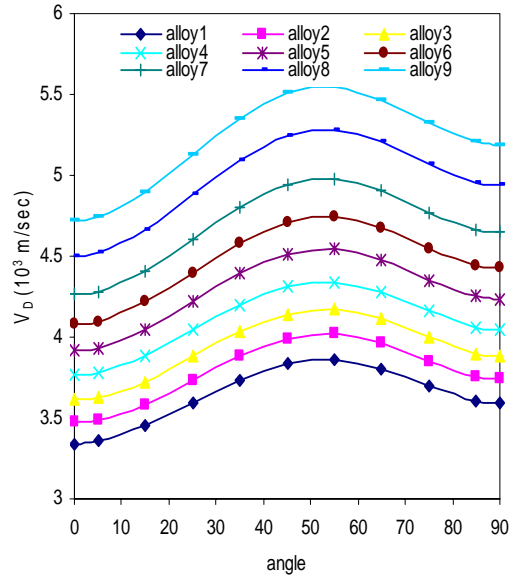

Figure 1. V_1 vs angle with unique axis of crystal
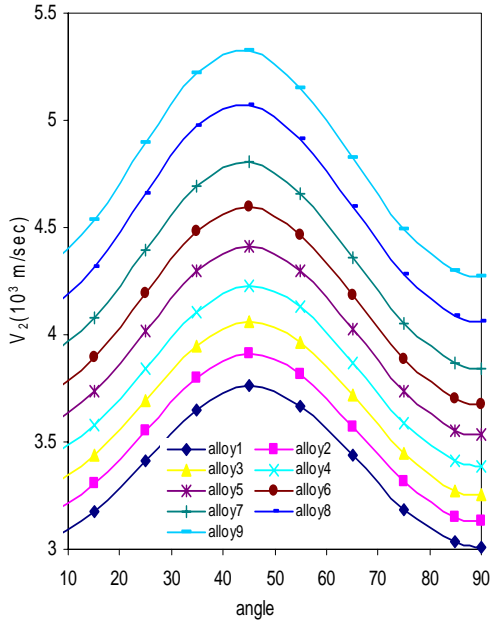
Figure 2. V_2 vs angle with unique axis of crystal
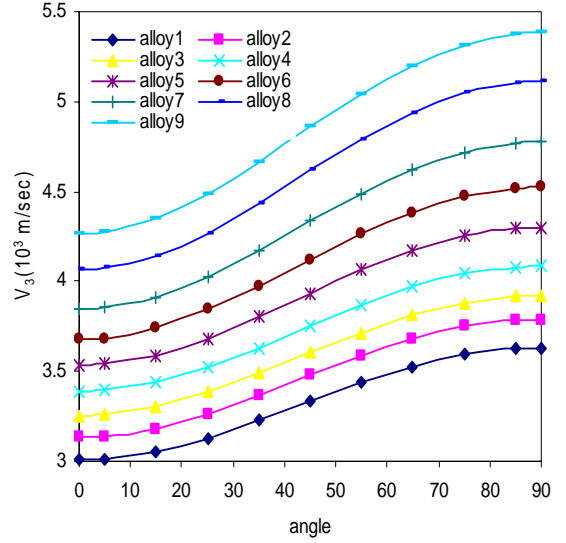
Figure 3. V_3 vs angle with unique axis of crystal

Figure 4. V_D vs angle with unique axis of crystal

Figures 1–4 depict that the magnitudes of ultrasonic velocities increase from alloy: 1 to alloy: 9 as boost up of zinc amount. The longitudinal ultrasonic velocity along <001> direction ($\theta=0°$ with unique axis) is largest for alloy: 9, because alloy: 9 have highest zinc amount as well as highest C_{33} value. As we know that shear wave is also known surface wave. Therefore the ultrasonic velocities V_2 and V_3 are the surface wave velocity. It is clear from the values of V_2 and V_3 for ultrasonic wave propagation along $\theta=0°$ are the same, while these are different for other directions as visualized in Figs. 2-3. Hence <001> direction is the direction of symmetry for chosen alloys. Debye average velocities (V_D) of these alloys are increasing with the angle and have maxima at 55° at 300 K (figure. 4). Since V_D is calculated using V_1, V_2 and V_3 with eq. (8), therefore the temperature variation of V_D follows the combined effect of temperature variation of

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V_1, V_2 and V_3. The maximum in V_D at 55° is due to a significant increase in longitudinal and pure shear wave velocities and a decrease in quasi-shear wave velocity. Thus it can be concluded that when a sound wave travels at 55° with the unique axis of these crystals then the average sound wave velocity is maximum. Since the Debye average velocity is calculated using the constituent velocities V_1, V_2 and V_3, hence a good resemblance in V_D implies that our calculated velocities are correct.

4. Conclusions

Based on the above discussion is worthwhile to state that:

- Present method to evaluate second order elastic constants involving many body interaction potential for hexagonal wurtzite crystal structured materials (alloys) is correct.
- Although the nature of angle dependency of acoustic velocity in these alloys is quite similar to that of other hexagonal materials and alloys, but they favour instability due to lower anisotropy.
- All elastic constants and density are mainly the affecting factor for anomalous behaviour of ultrasonic velocity in these alloys.
- The <001> direction is the direction of symmetry for these alloys as they have the same quasi-shear and pure shear wave velocities.
- The elastic constant and velocity data of these alloys directly correlate their energy density and density of state, while the average sound velocity is a direct consequence of enthalpy of formation, Debye temperature, specific heat and thermal energy density of these alloys.
- The mechanical and ultrasonic properties of these alloys will not be better than pure Ag and Zn due to their low SOEC and ultrasonic velocity.

The achieved results in present investigation can be used for further investigation using other methods such as polarizing microscope, X-rays, surface tension, NMR and various transport phenomena. These results may expand future prospects for the applications of the Zener alloys.

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