Phonon Dynamics and Acoustic and Thermodynamic Properties of Pt$_{57.5}$Cu$_{14.7}$Ni$_{5.3}$P$_{22.5}$ Bulk Metallic Glass

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Abstract

The phonon dispersion curves for Pt$_{57.5}$Cu$_{14.7}$Ni$_{5.3}$P$_{22.5}$ bulk metallic glass (BMG) are computed employing various dielectric screenings using the simple model given by Bhatia and Singh. The force constants $\beta$ and $\delta$ for computing the dispersion curves are calculated from the elastic constants i.e. bulk modulus ($B$) and shear modulus ($G$) along with the calculated value of force constant $\kappa_e$ of the material of the glass for the first time. The results of the phonon dispersion curves show appropriate behavior in the long wavelength region in detail for both the longitudinal and transverse modes and give insight regarding the acoustic and thermal properties of the BMG. The transverse sound velocity and the longitudinal velocities with various dielectric screening are calculated from the dispersion curves in the long wavelength region. The corresponding thermodynamic property (Debye temperature) is calculated for different dielectric screenings. The theoretical results predicted are in a good agreement with the reported data in the literature for the Pt$_{57.5}$Cu$_{14.7}$Ni$_{5.3}$P$_{22.5}$ BMG and may be used for correlating other properties.

Keywords: Bulk metallic glass; Phonon dispersion; Dielectric screening; Elastic constant.

1. Introduction

Bulk metallic glasses have emerged with attractive mechanical and thermal properties having a wide range of technological applications [1,2]. The understanding of phonon dynamics and micro-structural configuration of metallic glasses is essential for understanding their acoustic, elastic and thermal properties [2–7]. Experimentally, neutron scattering has been used to study the phonon frequencies of metallic glasses [8,9]. The theoretically computed phonon dispersion curves both for longitudinal and transverse modes have also been reported by computer simulation and recursion techniques [8-13]

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for a variety of metallic glasses. Three main theoretical approaches, namely Hubbard and Beeby [14], Takeno and Goda [15] and that of Bhatia and Singh [4] are widely used for computing phonon frequencies of metallic glasses. Pratap et al. [16] has computed phonon frequencies of Cu_{57}Zr_{43} metallic glass using both Hubbard and Beeby and that of Takeno and Goda approaches. Vora and Gandhi [13] have reported phonon dispersion curves of Pd_{90}Ni_{10}Cu_{90}P_{21} using Takeno and Goda approach. The simple model approach by Bhatia and Singh [4] has been applied for computing phonon dispersion of various metallic glasses - Cu_{57}Zr_{43} by Agarwal et al. [7], for Zr–Ni alloys by Lad and Pratap [17] and by Agarwal for Zr-Ti-Cu-Ni-B [10] and Cu_{60}Zr_{20}Hf_{10}Ti_{10} [11] bulk metallic glasses (BMGs).

Pt_{57.5}Cu_{14.7}Ni_{5.3}P_{22.5} BMG due to its unique properties has potential industrial applications in electrochemical devices such as micro fuel cells, energy conversion/storage and sensors [18,19]. The large ductility and high fracture toughness properties of Pt_{57.5}Cu_{14.7}Ni_{5.3}P_{22.5} BMG are experimentally studied by Schroers and Johnson [20]. Chen et al. [21,22] reported the influence of annealing temperature on the hardness, elastic modulus and surface morphology of Pt_{57.5}Cu_{14.7}Ni_{5.3}P_{22.5} BMG. Similar studies on fracture toughness variation of the BMG under consideration were also reported by Ketkaew et al. [23]. However, understanding and controlling the properties of bulk metallic glasses in general is limited by the lack of sound theoretical understanding for the disordered and non-equilibrium glassy materials.

In this work, we compute the phonon dispersion curves of Pt_{57.5}Cu_{14.7}Ni_{5.3}P_{22.5} BMG using the simple model given by Bhatia and Singh [4] employing various dielectric screenings. This simple model proposed by Bhatia and Singh assumes a central force, effective between nearest neighbours and a volume-dependent force due to conduction electrons. Bhatia and Singh had fixed the values of force constants $\delta$ and $\beta$ using the value of longitudinal and transverse sound velocities along with the calculated value of force constant $\kappa_e$ as no experimental data on elastic constants were available, though they have suggested to fix these parameters from elastic constants. Subsequent theoretical study of phonon dispersion employing Bhatia and Singh approach using force constant method has also derived the force constants values in a similar way [7,10-12]. In the study presented in this paper, we fix the value of force constants $\delta$ and $\beta$ used in the computation of dispersion curves by using the value of bulk modulus ($B$) and shear modulus ($G$) of the BMG under consideration for the first time along with the calculated value of force constant $\kappa_e$. The dielectric screening due to conduction electrons is quite significant in the long wavelength region of the dispersion curves and to study the effects of various dielectric screenings on the dispersion curves in longitudinal mode, we employ the dielectric screening functions [24] due to Bhatia and Singh (BS) [4], Hartree (H), Hubbard (HB), Geldart and Vosko (GV), self-consistent screening due to Shaw (SCS) [25], and Overhauser (OH) [26]. The longitudinal sound velocities ($V_L$) are computed for different dielectric screenings and the transverse sound velocity ($V_T$) is obtained which is without dielectric screening from dispersion curves in the long wavelength region. The corresponding Debye temperature ($\theta_D$) is finally obtained for various dielectric screenings.
2. Theory

The equations for the longitudinal phonon frequencies ($\omega_L$) and transverse phonon frequencies ($\omega_T$) as given by Bhatia and Singh [4] are written as

\[
\omega_L^2 = \frac{2N}{\rho a^2} \left[ \beta I_0 + \delta I_2 \right] + \frac{\kappa_e k_F^2 q^2 [G(q r_s)]^2}{\rho [q^2 + k_F^2 \epsilon(q)]}, \tag{1}
\]

and

\[
\omega_T^2 = \frac{2N}{\rho a^2} \left( \beta + \frac{1}{2} \delta \right) I_0 - \frac{1}{2} \delta I_2, \tag{2}
\]

where $q$ is the momentum wave vector, $a$ is the distance between nearest neighbours and $N$ is the coordination number; $\beta$, $\delta$ and $\kappa_e$ are force constants. $\beta$ and $\delta$ are defined in terms of the first and second derivatives of inter-atomic potential $W(r)$ at $r = a$, as

\[
\beta = \frac{\rho a^2}{2M} \left[ \frac{dW(r)}{dr} \right]_{r=a}, \tag{3}
\]

\[
\delta = \frac{\rho a^3}{2M} \left[ \frac{d^2W(r)}{dr^2} \right]_{r=a}. \tag{4}
\]

In Eq. (1), the relevant force constant $\kappa_e$ due to the conduction electrons based on the Thomas–Fermi model can be written as

\[
\kappa_e = 4\pi n_e n_i z e^2 / K_F^2, \tag{5}
\]

where $e$ is the electron charge, $n_e = n_i z$ is mean electron density and $n_i$ is the ionic number density; $z = \sum_i C_i z_i$ is the mean valence of the glassy system, where $C_i$ and $z_i$ are the concentration and valence of the $i$th constituent; $K_F = (4 k_F / \pi a_0)$ is the Thomas–Fermi screening length, where $a_0$ is the Bohr radius and $k_F = (3\pi^2 n_e)^{1/3}$ is the Fermi wave number.

The $[G(q r_s)]^2$ in Eq. (1) is the shape factor to take into account the cancellation effects of kinetic and potential energies inside the core of the ions and is of the form

\[
[G(q r_s)]^2 = \left[ \frac{3[\sin(q r_s) - (q r_s) \cos(q r_s)]}{(q r_s)^3} \right]^2, \tag{6}
\]

where $r_s = [3/(4\pi n_i)]^{1/3}$ is the radius of the Wigner–Seitz sphere.

To study the effects of various dielectric screenings on the longitudinal phonon dispersion curve, we employ various dielectric screenings namely Bhatia and Singh (BS), Hartree (H), Hubbard (HB), Geldart and Vosk (GV), self-consistent screening due to Shaw (SCS) and Overhauser (OH). The dielectric screening function $\epsilon(q)$ in Eq. (1) takes the following forms for various dielectric screenings.

Bhatia and Singh [4] used as

\[
\epsilon(q) = \frac{1}{2} \left[ 1 + \frac{k_F}{q} \left( 1 - \frac{q^2}{4 k_F^2} \right) \ln \left[ \frac{q + 2 k_F}{q - 2 k_F} \right] [1 - f(q)] \right], \tag{7}
\]

where $f(q)$ is given by Hubbard [27] as
\[ f(q) = \frac{1}{2} q^2 / \left( q^2 + k_F^2 + \frac{1}{2} K_F^2 \right) . \]  

Hartree dielectric screening [24] takes the form of 
\[ \epsilon_H(q) = 1 + Q_0(q) , \]  
where \( Q_0(q) = (K_{TF}^2 / q^2) f(x) \) with \( x = q / k_F \) 
and \( f(x) = 0.5 + ((4 - x^2) / 8x) \ln \left( \frac{x+2}{2-x} \right) . \)

The HB, GV SCS and OH dielectric screening functions [24] are given by 
\[ \epsilon(q) = 1 + Q_0(q) / (1 - f(q)Q_0(q)) , \]  
where \( f_{HB}(q) = 0.5q^2 / (q^2 + k_F^2 + K_{TF}^2) . \) 
\[ f_{GV}(q) = 0.5q^2 / (q^2 + v k_F^2) \] with \( v = 2 / [1 + 1.153(K_{TF}^2 / 4k_F^2)] . \) 
\[ f_{SCS}(q) = A[1 - \exp(-B(q/k_F)^2)] \] with \( A = 1 \) and \( B = 0.535 . \) 
and \( f_{OH}(q) = 0.275(q/k_F)^2 / [1 + 2.5(q/k_F)^2 + 0.9375(q/k_F)^4]^{1/2} . \)

In Eqs. (1) and (2) 
\[ I_n = \int_0^{\pi} \sin \theta \cos^n \theta \left[ \sin^2 \left( \frac{1}{2} qa \cos \theta \right) \right] d\theta , \]  
where \( \theta \) is the angle between the unit vector along the displacement of the wave and the vector joining the atom at the origin to one of its nearest neighbours, 
so that with \( x = qa \), \( I_0 \) and \( I_2 \) are respectively, 
\[ I_0(x) = 1 - \frac{\sin x}{x} , \]  
\[ I_2(x) = \frac{1}{3} - \sin x \left[ \frac{1}{x} - \frac{x}{x^3} \right] - \frac{2 \cos x}{x^2} , \]  
for the limiting case \( q \to 0 \), \( x \to 0 \); \( I_0(x) \approx \frac{1}{6} x^2 \) and \( I_2(x) \approx \frac{1}{10} x^2 \). Substituting these values in Eqs. (1) and (2), we have the longitudinal and transverse sound velocities respectively, \( V_L(0) = \omega_L / q \) and \( V_T(0) = \omega_T / q \) in low momentum region \( (q \to 0) \) as 
\[ \rho V_L^2(0) = N \left( \frac{1}{3} \beta + \frac{1}{5} \delta \right) + \kappa_e , \]  
\[ \rho V_T^2(0) = N \left( \frac{1}{3} \beta + \frac{1}{15} \delta \right) . \]

In terms of the elastic moduli of the glassy material [4] 
\[ C_{11} = \rho V_L^2(0) = B + \frac{4}{3} G , \]  
\[ C_{44} = \rho V_T^2(0) = G . \]

The longitudinal sound velocities \( (V_L) \) for various dielectric screenings are obtained from the \( \omega_L-q \) dispersion curves in the long wavelength region. Similarly, the transverse sound velocity \( (V_T) \) is obtained from the \( \omega_T-q \) dispersion curve in the long wavelength region. The value of the Debye temperature \( (\theta_D) \) is obtained using the relation [28]
\[ \theta_B = \frac{h}{k_B} \left( \frac{9n_i}{4\pi} \right)^{1/3} \left( \frac{1}{v_L^3} + \frac{2}{v_T^3} \right)^{-1/3}. \]  \hspace{1cm} (22)

3. Calculations

The experimental values of \( B, G \) and \( \rho \) are taken from Schroers and Johnson [20] for \( \text{Pt}_{57.5}\text{Cu}_{14.7}\text{Ni}_{5.3}\text{P}_{22.5} \) as \( 198.7 \times 10^9 \, \text{Nm}^{-2} \), \( 33.3 \times 10^9 \, \text{Nm}^{-2} \) and \( 15.02 \times 10^3 \, \text{kgm}^{-3} \) respectively. The value of \( n_i \) calculated using the relation \( \rho = n_i M \) is found to be \( 6.876 \times 10^{28} \, \text{m}^{-3} \). Since the BMG under consideration is of FCC structure, \( n_i \) can be written as

\[ a^3 n_i = \sqrt{2}. \]  \hspace{1cm} (23)

Substituting the value of \( n_i \) in Eq. (23) gives \( a = 2.74 \times 10^{-10} \, \text{m} \). The calculated value of mean valence of the BMG using the relation \( z = \sum_i C_i z_i \) is 2.08. The value of \( \kappa_e \) is calculated using Eq. (5) with \( z = 2.08 \) for \( \text{Pt}_{57.5}\text{Cu}_{14.7}\text{Ni}_{5.3}\text{P}_{22.5} \) is found to be \( 162.198 \times 10^9 \, \text{Nm}^{-2} \). The values of force constants \( \beta \) and \( \delta \) are obtained by substituting the values of \( B, G \) and \( \kappa_e \) in Eqs. (18-21) taking \( N = 12 \) for FCC structure. Hence, all the values of the quantity in dispersion relations in Eqs. (1) and (2) are known. The parameters used for computing phonon dispersion curves are listed in Table 1.

| Pt\textsubscript{57.5}Cu\textsubscript{14.7}Ni\textsubscript{5.3}P\textsubscript{22.5} | \hline
| \( \rho \) (10\textsuperscript{3} \text{kgm}^{-3}) | 15.02 |
| \( n_i \) (10\textsuperscript{28} \text{m}^{-3}) | 6.876 |
| \( \kappa_e \) (10\textsuperscript{9} \text{Nm}^{-2}) | 152.198 |
| \( k_F \) (10\textsuperscript{10} \text{m}^{-1}) | 1.618 |
| \( a \) (10\textsuperscript{-10} \text{m}) | 1.514 |
| \( K_F^2 \) (10\textsuperscript{20} \text{m}^{-2}) | 3.893 |
| \( \beta \) (10\textsuperscript{9} \text{Nm}^{-2}) | 2.313 |
| \( \delta \) (10\textsuperscript{9} \text{Nm}^{-2}) | 36.006 |
| \( z \) | 2.08 |

4. Results and Discussion

The phonon dispersion relation in Eq. (1) is applied to \( \text{Pt}_{57.5}\text{Cu}_{14.7}\text{Ni}_{5.3}\text{P}_{22.5} \) BMG with different dielectric screening functions viz. BS, H, HB, GV, SCS, and OH to get the longitudinal phonon frequencies. Similarly, Eq. (2) is applied to obtain the transverse phonon frequency which is without dielectric screening. The phonon dispersion curves for \( \text{Pt}_{57.5}\text{Cu}_{14.7}\text{Ni}_{5.3}\text{P}_{22.5} \) BMG are shown in Fig. 1. From the Fig. 1, the longitudinal (\( \omega_T-q \)) and transverse (\( \omega_L-q \)) dispersion curves show linear dispersion curves in the long wavelength region.
As seen from Fig. 1, $\omega_L$-$q$ dispersion curves depend on the dielectric screening employed. The position of the first peak for $\omega_L$-$q$ curves is found at $q = 1.2 \times 10^{10}$ m$^{-1}$ for BS and H form of dielectric screenings, while for HB, GV, SCS and OH form of dielectric screenings it is found at $q = 1.3 \times 10^{10}$ m$^{-1}$. The height of the peak is also dependent on the type of dielectric screenings employed. The difference in $\omega_L$ values increases as $q$ starts increasing and it gradually widens and becomes prominent at the first maxima (peak) of the curve. The peak heights of SCS, GV, HB, OH, H and BH form of dielectric screenings are in the ascending order. For SCS the peak height is lowest while it is the highest for BS form of screening. The overall difference between SCS and BS is 19.56%. With further increase in $q$, the $\omega_L$ values start decreasing and the $\omega_L$-$q$ curves converge at the $q$ value corresponding to the first minima of $\omega_L$-$q$ curves obtained at $q = 2.8 \times 10^{10}$ m$^{-1}$ and it is independent of the dielectric screenings. Beyond this $q$ value, $\omega_L$ values are independent of dielectric screenings employed. The first peak position of $\omega_T$-$q$ curve is obtained at $q = 2.0 \times 10^{10}$ m$^{-1}$ which is at higher $q$ value than the first peak position of $\omega_L$-$q$ curves for all dielectric screenings. The dispersion curve of $\omega_T$-$q$ increases with wave number $q$ and gets saturated around the first peak with a small variation. With further increase in $q$ value, the damping of transverse phonons becomes prominent.

The sound velocities of the transverse and longitudinal modes are estimated for this BMG in the long wavelength region ($q \to 0$) of the dispersion curves. For estimating the sound velocities of transverse and longitudinal modes, we have taken the data range ($q = 0.1 \times 10^{10}$ m$^{-1}$ to $q = 0.4 \times 10^{10}$ m$^{-1}$) for all the screenings. The values of sound velocities estimated from the dispersion curves for the transverse ($V_T$) and longitudinal ($V_L$) modes are listed in Table 2.
Table 2. The longitudinal ($V_L$) and transverse ($V_T$) sound velocities along with Debye temperature ($\theta_D$) for Pt$_{57.5}$Cu$_{14.7}$Ni$_{5.3}$P$_{22.5}$ BMG employing different dielectric screenings.

| Dielectric screenings | $V_L$ ($10^5$ cms$^{-1}$) | $V_T$ ($10^5$ cms$^{-1}$) | $\theta_D$ (K) |
|-----------------------|-----------------------------|-----------------------------|----------------|
| BS                    | 4.413                       | 1.457                       | 202.2          |
| H                     | 4.379                       | 202.1                       |
| HB                    | 4.112                       | 201.9                       |
| GV                    | 3.904                       | 201.6                       |
| SCS                   | 3.592                       | 201.1                       |
| OH                    | 4.035                       | 201.8                       |
| Experimental          | 4.000 [20]                  | 1.481 [20]                  | 206 [2]        |

The value of transverse velocity computed from the slope of the dispersion curve, which is without any dielectric screening in the long wavelength region is $V_T = 1.457 \times 10^5$ cms$^{-1}$. The computed transverse sound velocity is 1.6 % less than the experimental value of $1.481 \times 10^5$ cms$^{-1}$ reported by Schroers and Johnson [20]. Similarly, the values of longitudinal velocities ($V_L$) computed for various dielectric screening are listed in Table 2. The screenings due to various dielectric screening functions have an effect on the longitudinal sound velocity. However, the values of longitudinal sound velocities computed from the dispersion curves are close to the experimental value of $4.000 \times 10^5$ cms$^{-1}$ [20] with the dielectric screening due to OH showing nearest to the experimental value. The corresponding Debye temperature, $\theta_D$ (K) is computed using Eq. (22) by substituting the value of transverse sound velocity which is without dielectric screening and the longitudinal sound velocity values for various dielectric screenings as listed in Table 2. There is not much variation in the computed values of $\theta_D$ for different dielectric screening function and it is very close to the reported value of 206 K [2].

The position of the first minimum of $\omega_L$-$q$ curves lie where the first peak ($q_p$) of the static structure factor, $S(q)$ is predicted. The position of the first minimum of $\omega_L$-$q$ curves lies at $q = 2.8 \times 10^{10}$ m$^{-1}$ and is independent of the dielectric screenings. Since no experimental data for the static structure factor of Pt$_{57.5}$Cu$_{14.7}$Ni$_{5.3}$P$_{22.5}$ BMG is available, we predicted from our theoretical phonon dispersion curves to be around $q = 2.8 \times 10^{10}$ m$^{-1}$. Moreover, the computed value of $q_p$ for the system under consideration is slightly less than $2k_F$, where $k_F = 1.618 \times 10^{10}$ m$^{-1}$ is the calculated value of the BMG as shown in Table 1. The ratio of $2k_F/q_p=1.15$ for this BMG is in good agreement with the stability of metallic glasses [29]. The structure factor provided key structural information on the atomic network for amorphous matters and a universal relationship exists between the elastic properties and the wavelength $\lambda$ (i.e. $2\pi/q_p$) [3]. Since the experimental data for phonon frequencies are rare and the limitation of the experimental techniques for describing the micro-structure of metallic glasses, it is expected that theoretical computation of phonon dispersion curves will give insight on understanding the structural information and elastic properties of metallic glasses.
4. Conclusion

We theoretically investigated the phonon dispersion curves of $\text{Pt}_{57.5}\text{Cu}_{14.7}\text{Ni}_{5.3}\text{P}_{22.5}$ BMG employing various dielectric screenings using the Bhatia and Singh approach. In computing the dispersion curves we have fixed the values of force constants $\delta$ and $\beta$ using the experimental values of bulk modulus ($B$) and shear modulus ($G$) along with the calculated value of force constant $\kappa_e$ of the BMG under consideration. This method of fixing force constants is applied for the first time. From the computed dispersion curve, the acoustic and thermodynamic properties along with the position of the first peak of static structure factor can be predicted. The values of sound velocities and Debye temperature computed from the phonon dispersion curves of $\text{Pt}_{57.5}\text{Cu}_{14.7}\text{Ni}_{5.3}\text{P}_{22.5}$ BMG using this method is in good agreement with the experimental data. Thus, in view of the excellent agreement with the experimental data, we plead for the reliability of the results computed by this method and we are presently working on applying this method for computing phonon dispersion curves of other BMGs.

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