

# Computational Study of Vibrational Dynamics of Binary $\text{Mg}_{0.70}\text{Zn}_{0.30}$ Metallic Glass by a Pseudopotential Theory

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## Abstract

The vibrational dynamics of  $\text{Mg}_{70}\text{Zn}_{30}$  metallic glass has been studied at room temperature in terms of phonon eigen-frequencies of longitudinal and transverse modes employing three different approaches proposed by Hubbard-Beeby (HB), Takeno-Goda (TG) and Bhatia-Singh (BS). The well-recognized model potential is employed successfully to explain electron-ion interaction in the metallic glass; instead of using experimental values of the pair correlation function  $g(r)$ , which is generated from the computed pair potential. The present findings of phonon dispersion curve are found in fair agreement with available theoretical as well as experimental data. The thermodynamic properties obtained by HB and TG approaches are found much lower than those obtained by BS approach. The pseudo-alloy-atom (PAA) model is applied for the first time instead of Vegard's Law.

**Key Words:** Pair potential, Metallic Glasses, Phonon dispersion curves, Thermal properties, Elastic properties

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## 1. Introduction

The homovalent  $\text{Mg}_{0.70}\text{Zn}_{0.30}$  glass is one of the most important candidates of simple metallic glasses. The dynamical properties of  $\text{Mg}_{0.70}\text{Zn}_{0.30}$  glass have been studied theoretically by von Heimendl [1], using the equation of motion method; by Tomenek [2], using a model calculation; by Saxena et al. [3], using effective pair potential and Takeno-Goda (TG) approach [4, 5]; by Agarwal et al. [6, 7], using Bhatia-Singh (BS) approach [8]; and Agarwal-Kachhava [9, 10], using TG as well as BS approaches. Experimentally, the phonon dispersion curves (PDC) for  $\text{Mg}_{0.70}\text{Zn}_{0.30}$  glass was determined by Suck et al. [11] for a few wave vector transfers near  $q_P = 2.61 \text{ \AA}^{-1}$ , at which the first peak is found in static structure factor calculation. The atomic and electronic structure has been studied by Hafner-Jaswal [12] and Hafner et al. [13] using ab initio pseudopotential technique. Benmore et al. [14, 15] have calculated longitudinal excitations within the first pseudo-Brillouin zone using the neutron Brillouin technique at room temperature. The temperature dependence of the dispersion and damping coefficients of transverse excitations was studied by Bryk and Mryglod [16], using the method of generalized collective modes. Thakore et al. [17] and Vora et al. [18, 19] have also studied the PDC of  $\text{Mg}_{0.70}\text{Zn}_{0.30}$  glass.

In most of the above studies, the pseudopotential parameter is evaluated such that it generates a pair correlation function, which is in good agreement with experimental data available in the literature. In most of these studies, the Vegard's law was used to explain electron-ion interaction for binaries. But it is well known that PAA is a more meaningful approach to explain such kind of interactions in binary alloys and metallic glasses [17–22]. Hence, in the present article the PAA model is used to investigate the phonon dynamics of  $A_{1-x}B_x$  binary glassy system.

The three theoretical approaches proposed by Hubbard-Beeby (HB) [23], Takeno-Goda (TG) [4, 5] and Bhatia-Singh (BS) [8, 24] are used to generate the phonon dispersion curve (PDC). The local field correction function due to Sarkar et al [25] is employed for the first time to include the exchange and correlation effects in such study. Long wave-length limits of the phonon modes are then used to investigate the thermodynamic and elastic properties viz. isothermal bulk modulus ( $B_T$ ), modulus of rigidity (G), Young's modulus (Y), longitudinal sound velocity ( $v_L$ ), transverse sound velocity ( $v_T$ ) and Debye temperature ( $\theta_D$ ).

## 2. Theoretical Methodology

The pair potential  $V(r)$  is calculated from the relation given by Vora et al. [18, 19],

$$V(r) = \left( \frac{Z^2 e^2}{r} \right) + \frac{\Omega_O}{\pi^2} \int F(q) \left[ \frac{\text{Sin}(qr)}{qr} \right] q^2 dq. \quad (1)$$

Here,  $Z$  and  $\Omega_O$  are the valence and atomic volume of the glassy alloys, respectively.

The energy wave number characteristics appearing in the Equation (1) is written as [18, 19]

$$F(q) = \frac{-\Omega_O q^2}{16\pi} |W_B(q)|^2 \frac{[\varepsilon_H(q) - 1]}{\{1 + [\varepsilon_H(q) - 1][1 - f(q)]\}}. \quad (2)$$

Here,  $W_B(q)$ ,  $\varepsilon_H(q)$ ,  $f(q)$  are the bare ion potential, the Hartree dielectric response function and the local field correction function to introduce the exchange and correlation effect, respectively.

The well recognized model potential  $W_B(q)$  used in the present computation of phonon dynamics of binary metallic glasses is of the form [18–22]

$$W_B(q) = \frac{-4\pi e^2 Z}{\Omega_O q^2} \left[ \begin{array}{l} \left\{ -1 + \frac{12}{U^2} + \frac{U^2}{1+U^2} + \frac{6U^2}{(1+U^2)^2} + \frac{18U^2}{(1+U^2)^3} - \frac{6U^4}{(1+U^2)^3} \right\} \cos(U) \\ + \frac{24U^2}{(1+U^2)^4} - \frac{24U^4}{(1+U^2)^4} \\ + \\ \left\{ \frac{6}{U} - \frac{12}{U^3} + \frac{U}{1+U^2} + \frac{3U}{(1+U^2)^2} - \frac{3U^3}{(1+U^2)^2} + \frac{6U}{(1+U^2)^3} \right\} \sin(U) \\ - \frac{18U^3}{(1+U^2)^3} + \frac{6U}{(1+U^2)^4} - \frac{36U^3}{(1+U^2)^4} + \frac{6U^5}{(1+U^2)^4} \\ + \\ 24U^2 \exp(1) \left\{ \frac{U^2-1}{(1+U^2)^4} \right\} \end{array} \right]. \quad (3)$$

Here,  $U = qr_c$ . This form has the feature of a Coulomb term outside the core and varying cancellation due to repulsive and attractive contributions to the potential within the core in real space [18–22]. The parameter  $r_c$  is adjusted such that the calculated values of  $g(r)$  agree with the experimental value of  $g(r)$  as close as possible.

A quantity which is equally important as the pair potential while studying a disorder system is the pair correlation function  $g(r)$ . In the present study the pair correlation function  $g(r)$  can be computed from the relation [18, 19],

$$g(r) = \exp \left[ \left( \frac{-V(r)}{k_B T} \right) - 1 \right]. \quad (4)$$

Here,  $k_B$  is the Boltzmann's constant and  $T$  is the room temperature of the amorphous system.

The three approaches for studying of phonons in amorphous alloys proposed by Hubbard -Beeby (HB) [20], Takeno-Goda (TG) [4] and Bhatia-Singh (BS) [6, 21] have been employed for studying the longitudinal and transverse phonon frequencies in the Mg-based glass.

According to the Hubbard-Beeby (HB) [23], the expressions for longitudinal and transverse phonon frequencies are

$$\omega_L^2(q) = \omega_E^2 \left[ 1 - \frac{\sin(q\sigma)}{q\sigma} - \frac{6 \cos(q\sigma)}{(q\sigma)^2} + \frac{6 \sin(q\sigma)}{(q\sigma)^3} \right] \quad (5)$$

$$\omega_T^2(q) = \omega_E^2 \left[ 1 - \frac{3 \cos(q\sigma)}{(q\sigma)^2} + \frac{3 \sin(q\sigma)}{(q\sigma)^3} \right], \quad (6)$$

where  $\omega_E^2 = \left(\frac{4\pi\rho}{3M}\right) \int_0^\infty g(r) V''(r) r^2 dr$  is the maximum frequency.

Following Takeno-Goda (TG) [4, 5], the wave vector  $q$ -dependent longitudinal and transverse phonon frequencies are written

$$\omega_L^2(q) = \left(\frac{4\pi\rho}{M}\right) \int_0^\infty dr g(r) \left[ \left\{ r V'(r) \left( 1 - \frac{\sin(qr)}{qr} \right) \right\} + \left\{ r^2 V''(r) - r V'(r) \right\} \left( \frac{1}{3} - \frac{\sin(qr)}{qr} - \frac{2 \cos(qr)}{(qr)^2} + \frac{2 \sin(qr)}{(qr)^3} \right) \right], \quad (7)$$

$$\omega_T^2(q) = \left(\frac{4\pi\rho}{M}\right) \int_0^\infty dr g(r) \left[ \left\{ r V'(r) \left( 1 - \frac{\sin(qr)}{qr} \right) \right\} + \left\{ r^2 V''(r) - r V'(r) \right\} \left( \frac{1}{3} + \frac{2 \cos(qr)}{(qr)^2} + \frac{2 \sin(qr)}{(qr)^3} \right) \right]. \quad (8)$$

According to modified BS approach [8], the phonon frequencies of longitudinal and transverse branches are given by Shukla and Campanha [24]:

$$\omega_L^2(q) = \frac{2N_C}{\rho q^2} (\beta I_0 + \delta I_2) + \frac{k_e k_{TF}^2 q^2 |G(qr_S)|^2}{q^2 + k_{TF}^2 \varepsilon(q)} \quad (9)$$

$$\omega_T^2(q) = \frac{2N_C}{\rho q^2} \left( \beta I_0 + \frac{1}{2} \delta (I_0 - I_2) \right). \quad (10)$$

Other details of the constants used in this approach have been narrated in literature [8, 24]. Here,  $M$ ,  $\rho$  are the atomic mass and the number density of the glassy component, while  $V'(r)$  and  $V''(r)$  be the first and second derivative of the effective pair potential, respectively.

In the long-wavelength limit of the frequency spectrum the both frequencies, i.e. transverse and longitudinal, are proportional to the wave vectors and obey the relationships

$$\omega_L \propto q \text{ and } \omega_T \propto q,$$

$$\omega_L = v_L q \text{ and } \omega_T = v_T q, \quad (11)$$

where  $v_L$  and  $v_T$  are the longitudinal and transverse sound velocities of the glassy alloys, respectively. For the three approaches, HB, TG and BS,  $v_L$  and  $v_T$  are expressed as follows:

$$\text{HB approach [23]: } \begin{cases} v_L(\text{HB}) = \omega_E \sqrt{\frac{3\sigma^2}{10}} & (12) \\ v_T(\text{HB}) = \omega_E \sqrt{\frac{\sigma^2}{10}} & (13) \end{cases}$$

$$\text{TG approach [4, 5]: } \begin{cases} v_L (\text{TG}) = \left[ \left( \frac{4\pi\rho}{30M} \right) \int_0^\infty dr g(r) r^3 \{rV''(r) - 4V'(r)\} \right]^{1/2} & (14) \\ v_T (\text{TG}) = \left[ \left( \frac{4\pi\rho}{30M} \right) \int_0^\infty dr g(r) r^3 \{3rV''(r) - 4V'(r)\} \right]^{1/2} & (15) \end{cases}$$

$$\text{BS approach [8, 24]: } \begin{cases} v_L (\text{BS}) = \left[ \frac{N_C}{\rho} \left( \frac{1}{3}\beta + \frac{1}{5}\delta \right) + \frac{k_e}{3} \right]^{1/2} & (16) \\ v_T (\text{BS}) = \left[ \frac{N_C}{\rho} \left( \frac{1}{3}\beta + \frac{1}{15}\delta \right) \right]^{1/2} & (17) \end{cases}$$

In the long-wavelength limit of the frequency spectrum, transverse and longitudinal sound velocities  $v_L$  and  $v_T$  are computed. The isothermal bulk modulus  $B_T$ , modulus of rigidity  $G$ , Poisson's ratio  $\sigma$ , Young's modulus  $Y$  and the Debye temperature  $\theta_D$  are found using the expressions [18, 19],

$$B_T = \rho \left( v_L^2 - \frac{4}{3}v_T^2 \right), \quad (18)$$

$$G = \rho v_T^2, \quad (19)$$

where  $\rho$  is the isotropic number density of the solid. We now have

$$\sigma = \frac{1 - 2 \left( \frac{v_T^2}{v_L^2} \right)}{2 - 2 \left( \frac{v_T^2}{v_L^2} \right)}, \quad (20)$$

$$Y = 2G(\sigma + 1), \quad (21)$$

$$\theta_D = \frac{\hbar\omega_D}{k_B} = \frac{\hbar}{k_B} 2\pi \left[ \frac{9\rho}{4\pi} \right]^{1/3} \left[ \frac{1}{v_L^3} + \frac{2}{v_T^3} \right]^{(-1/3)}, \quad (22)$$

where  $\omega_D$  is the Debye frequency.

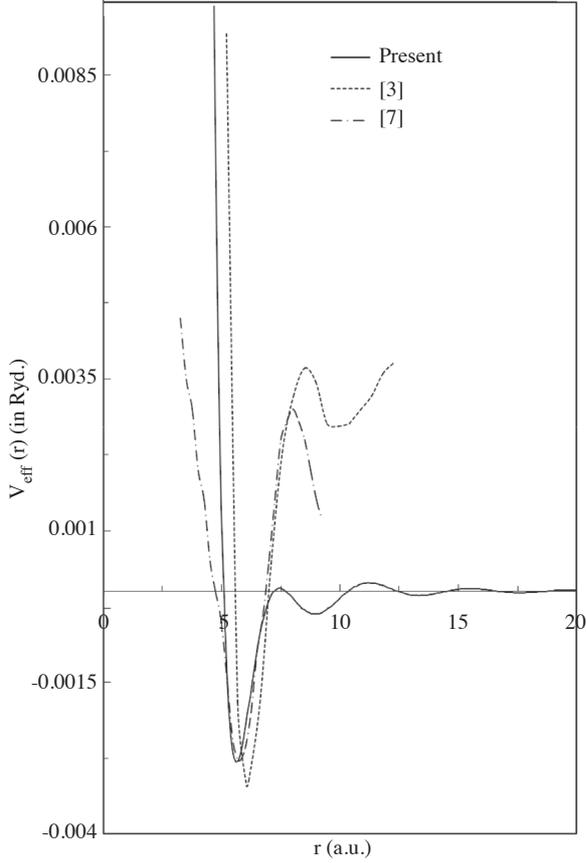
### 3. Results and Discussion

The input parameters and other related constants used in the present computations are:  $Z = 2.00$ ,  $\Omega_O = 2.0472 \times 10^{-23} \text{ cm}^3$ ,  $r_C = 4.8665 \times 10^{-9} \text{ cm}$ ,  $N_C = 12.00$ ,  $M = 5.871 \times 10^{-23} \text{ gm}$  and  $\rho = 2.9699 \text{ gm/cm}^3$ .

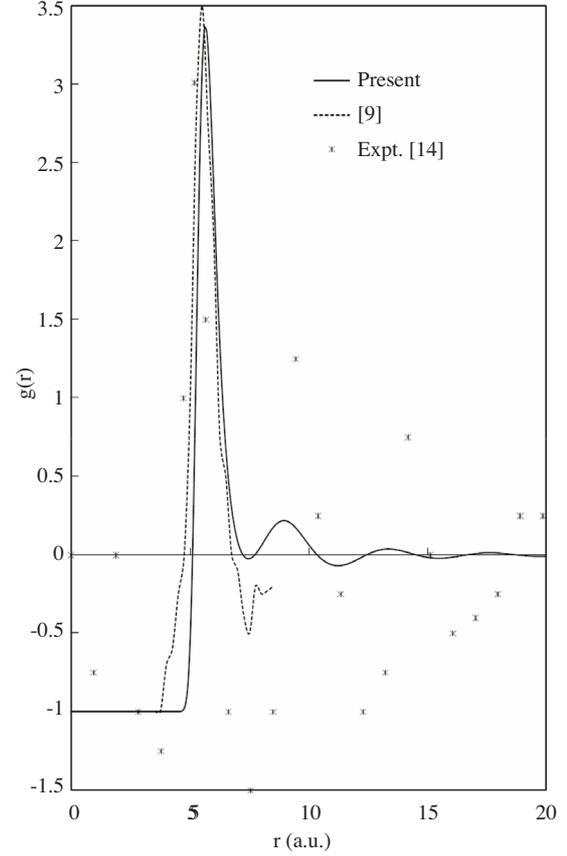
The comparison of presently computed pair potential of  $\text{Mg}_{0.70}\text{Zn}_{0.30}$  metallic glass and other such theoretical results [3, 6, 7] are displayed in Figure 1. From Figure 1 it is seen that the first zero for  $V(r = r_0)$  due to Sarkar et al.'s  $S$  local field correction function [25] occurs at  $r_0 = 5.8 \text{ au}$ . The well width and the  $V_{\min}(r)$  position of the pair potential are also affected by the behavior of the screening. It is also noticed that the well depth of presently computed pair potential is moved towards lower  $r$ -values and also shows lower as depth compared to the other theoretical results [3, 6, 7]. The presently computed results of the pair potential are found in qualitatively good agreement with the theoretical reported data [3, 6, 7]. The pair potential  $V(r)$  of Saxena et al. [3] and Agarwal et al. [6, 7] are highly oscillatory. Such oscillation for large  $r$ -region is not present in the computation and  $V(r)$  converges very rapidly to zero for higher  $r$ -values. The result of Saxena et al. [3] and Agarwal et al. [6] shows significant oscillations and potential energy remains positive in the large  $r$  region. Thus, the Coulomb repulsive potential part dominates the oscillations due to ion-electron-ion interactions in their studies.

The presently computed  $g(r)$  of  $\text{Mg}_{0.70}\text{Zn}_{0.30}$  metallic glass is displayed in Figure 2 along with experimental [14] and theoretical data [9]. An excellent agreement of presently computed  $g(r)$  with experimental

data [11] is seen around the first peak. The structural information obtained from the  $g(r)$  of  $Mg_{0.70}Zn_{0.30}$  metallic glass is listed in Table 1. The ratio  $r_2/r_1$  of the position of the second peak ( $r_2$ ) to that of the first peak ( $r_1$ ) is close to the  $c/a$  ratio in close-packed hexagonal structure, i.e.  $c/a = 1.59$ . This means that the short range order of near neighbor in the amorphous state is affected more or less by the atomic arrangement of crystalline state. The ratio  $r_3/r_1$  of the position of the third peak ( $r_3$ ) to that of the first peak ( $r_1$ ), i.e. the ratio of the third atomic shell radius to the nearest-neighbor distance, is also calculated and found 2.14.



**Figure 1.** Pair Potential of  $Mg_{0.70}Zn_{0.30}$  metallic glass.



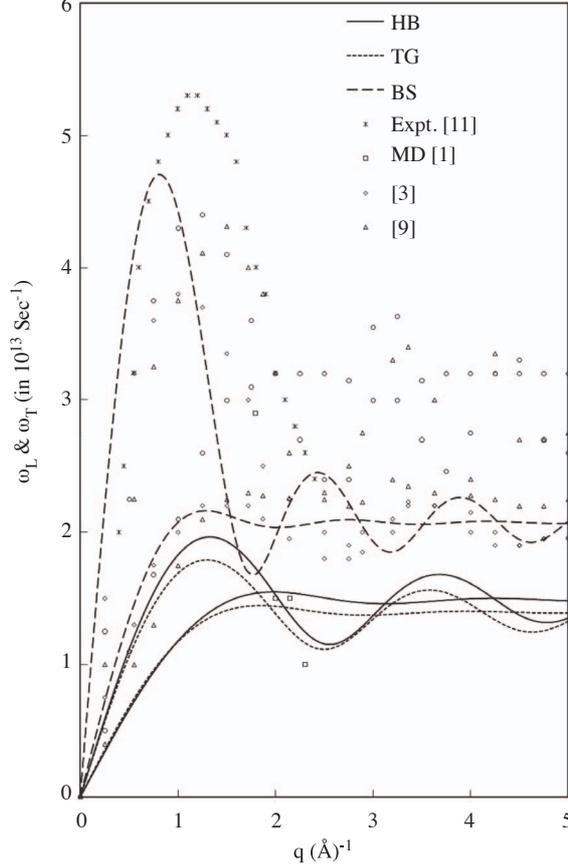
**Figure 2.** Pair Correlation Function ( $g(r)$ ) of  $Mg_{0.70}Zn_{0.30}$  metallic glass.

**Table 1.** Peak positions and peak ratios in the pair correlation functions of  $Mg_{0.70}Zn_{0.30}$  metallic glass.

| Structural data                   | Present Results |
|-----------------------------------|-----------------|
| First peak position $r_1$ (a.u.)  | 5.8             |
| Second peak position $r_2$ (a.u.) | 9.2             |
| Third peak position $r_3$ (a.u.)  | 12.4            |
| Ratio $r_2/r_1$                   | 1.59            |
| Ratio $r_3/r_1$                   | 2.14            |

The presently calculated PDC due to the three discussed approaches are shown in Figure 3 along with available theoretical [1, 3, 9] and experimental data [11]. The longitudinal branch of PDC calculated via the BS approach is higher than those obtained by the other two theoretical approaches. The presently computed values of the PDC from HB and TG approaches show lower results in comparison with other reported values.

First minima in the longitudinal branch lie near to the value of  $q_P$  at which the structure factor  $S(\mathbf{q})$  shows its first peak [14]. It is noticed from Figure 3 that the first minimum in the longitudinal branch is seen around at  $q \approx 1.8 \text{ \AA}^{-1}$  for BS,  $q \approx 2.5 \text{ \AA}^{-1}$  for TG and HB approaches, respectively. The first crossing position of  $\omega_L$  and  $\omega_T$  branches is observed at  $2.0 \text{ \AA}^{-1}$  in HB,  $1.9 \text{ \AA}^{-1}$  in TG and  $1.6 \text{ \AA}^{-1}$  in BS approach. In comparison to the other reported data [1, 3, 9, 11], the present results are suppressed values.



**Figure 3.** Phonon Dispersion Curves of  $\text{Mg}_{0.70}\text{Zn}_{0.30}$  metallic glass.

The computed thermodynamics properties are listed in Table 2. The results due to HB and TG approaches are lower than these due to BS approach. As experimental data for these properties are not available in the literature, it is very difficult to make further comments. Here, longitudinal and transverse sound velocities due to BS approach show good agreement with other such theoretical outcome [3]. The comparison with other such results [3] favors the present calculation and suggests that proper choice of dielectric screening is important part in explaining the thermodynamic and elastic properties of  $\text{Mg}_{0.70}\text{Zn}_{0.30}$  glass.

**Table 2.** Thermodynamic and Elastic Properties of  $\text{Mg}_{0.70}\text{Zn}_{0.30}$  metallic glass.

| Prop.                                       | HB     | TG     | BS     | Others [3] |
|---|--------|--------|--------|------------|
| $v_L \times 10^5$ , cm/sec                  | 1.5590 | 1.4140 | 6.0506 | 4.7, 5.1   |
| $v_T \times 10^5$ , cm/sec                  | 1.0829 | 1.0513 | 1.9760 | 2.5, 2.6   |
| $B_T \times 10^{11}$ , dyne/cm <sup>2</sup> | 0.2671 | 0.1619 | 9.6723 | –          |
| $G \times 10^{11}$ , dyne/cm <sup>2</sup>   | 0.3612 | 0.3404 | 1.2026 | –          |
| $Y \times 10^{11}$ , dyne/cm <sup>2</sup>   | 0.7468 | 0.6005 | 3.8464 | –          |
| $\theta_D$ , K                              | 129.74 | 124.62 | 247.86 | 305.21     |

## 4. Conclusions

Finally, it is concluded that the PDC generated from three the approaches reproduce all the broad characteristics of dispersion curves. The well recognized model potential with more advanced S-local field correction function generates consistent results. Hence, the model potential is suitable for studying the vibrational dynamics of  $\text{Mg}_{0.70}\text{Zn}_{0.30}$  metallic glass, which confirms not only the applicability of the model potential in the aforesaid properties but also supports the present approach of PAA. Such study on phonon dynamics of other binary liquid alloys and metallic glasses is in progress.

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