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Computation of phonon dynamics of $Mg_{70}Zn_{30}$ metallic glass

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Abstract In the present paper, the computation of the phonon dynamics of binary $Mg_{70}Zn_{30}$ metallic glass is reported using the well-recognized model potential of Gajjar et al. The present study includes the phonon dispersion curves (PDC), elastic and thermodynamic properties such as longitudinal sound velocity υ_L, transverse sound velocity υ_T, Debye temperature θ_D, isothermal bulk modulus B_T, modulus of rigidity G, Poisson's ratio σ and Young's modulus Y and specific heat capacity C_V of the glass. Three theoretical models given by Hubbard–Beeby (HB), Takeno–Goda (TG) and Bhatia–Singh (BS) are used to compute the PDC. Five local field correction functions proposed by Hartree (H), Taylor (T), Ichimaru–Utsumi (IU), Farid et al. (F) and Sarkar et al. (S) are employed for the first time to study the effect of exchange and correlation in the aforesaid properties. The pseudo-alloy-atom (PAA) model is applied for the first time instead of Vegard's Law.

Introduction

The homovalent $Mg_{70}Zn_{30}$ glass is one of the most important candidates of simple metallic glasses. The dynamical properties of $Mg_{70}Zn_{30}$ glass have been reported theoretically by von Heimendl [1] using the equation of motion method, by Tomanek [2] using a model calculation, by Saxena et al. [3] using the effective pair potential with Takeno–Goda (TG) approach [4], by Agarwal et al. [5] using Bhatia–Singh (BS) approach [6] and by Agarwal–Kachhava [7] using Takeno–Goda (TG) and Bhatia–Singh (BS) [6] approaches. Experimentally, the phonon dispersion curves (PDC) of $Mg_{70}Zn_{30}$ glass was studied by Suck et al. [8] for a few wave vector transfers near $\mathbf{q}_P$ = 2.61 $Å^{-1}$, at which the first peak is found in static structure factor calculation. The atomic and electronic structure has been studied by Hafner–Jaswal [9] and Hafner et al. [10] using ab initio pseudopotential technique. Benmore et al. [11, 12] have reported longitudinal excitations of the glass within the first pseudo-Brillouin zone using the neutron Brillouin zone technique at room temperature. The temperature dependence of the dispersion and damping coefficients of transverse excitations was studied by Bryk and Mryglod [13] using method of generalized collective modes. Also, Thakore et al. [14] and Vora et al. [15] have been computed phonon dynamics of this glass with effective atom model with well known approaches.

In most of the above studies, the pseudopotential parameter is evaluated in such a way that it generates the comparable results of pair correlation function or PDC, which is found in good agreement with experimental data. 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 [15–18]. Hence, in the present article the PAA mdoel is used to investigate the phonon dynamics of $A_{1-X}B_X$binary glassy system.

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The three theoretical approaches given by Hubbard–Beeby (HB) [19], Takeno–Goda (TG) [4] and Bhatia–Singh (BS) [6, 20] are used for the computation of the PDC. Five different types of local field correction functions proposed by Hartree (H) [17], Taylor (T) [17], Ichimaru–Utsumi (IU) [17], Farid et al. (F) [17] and Sarkar et al. (S) [17] are employed for the first time to study the effect of exchange and correlation in the aforesaid properties. Long wavelength limits of the phonon modes are used to investigate the thermodynamic and elastic properties, viz. longitudinal sound velocity υ_L, transverse sound velocity υ_T, Debye temperature θ_D, isothermal bulk modulus B_T, modulus of rigidity G, Poisson's ratio σ and Young's modulus Y and specific heat capacity C_V of $Mg_{70}Zn_{30}$ glass.

Theoretical methodology

The effective pair potential $V(r)$ is calculated from the relation given by Vora et al. [15],

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

where, Z and Ω_O are the valence and atomic volume of the glassy alloys, respectively.

The energy wave number characteristics appearing in Eq. (1) is written as [15],

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

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

The well-recognized model potential $W_B(q)$ of Gajjar et al. [15–18] used in the present computation of phonon dynamics of binary metallic glass is of the form.

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

here $U = qr_c$. This form has feature of the Coulombic term outside the core and varying cancellation due to repulsive and attractive contributions to the potential within the core in real space. The detailed information of this potential is given in the literature [15–18].

The three theories for studying of phonons in amorphous alloys proposed by Hubbard–Beeby (HB) [19], Takeno–Goda (TG) [4] and Bhatia–Singh (BS) [6, 20] have been employed for studying the longitudinal and transverse phonon frequencies in $Mg_{70}Zn_{30}$ glass.

According to the HB, the expressions for longitudinal and transverse phonon frequencies are [19],

$$\omega_L^2(q) = \omega_E^2\left[1-\frac{\sin(q\sigma)}{q\sigma}-\frac{6\cos(q\sigma)}{(q\sigma)^2}+\frac{6\sin(q\sigma)}{(q\sigma)^3}\right] \quad (4)$$

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

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

Following to TG [4], the wave vector (q) dependent longitudinal and transverse phonon frequencies are written as

$$\omega_L^2(q) = \left(\frac{4\pi\rho}{M}\right)\int_0^\infty dr\, g(r)\left[\left\{rV'(r)\left(1-\frac{\sin(qr)}{qr}\right)\right\} + \left\{r^2V''(r)-rV'(r)\right\}\left(\frac{1}{3}-\frac{\sin(qr)}{qr}-\frac{2\cos(qr)}{(qr)^2}+\frac{2\sin(qr)}{(qr)^3}\right)\right], \quad (6)$$

$$\omega_{\mathrm{T}}^2(q) = \left(\frac{4\pi\rho}{M}\right)\int_0^\infty dr\, g(r)\left[\left\{rV'(r)\left(1-\frac{\sin(qr)}{qr}\right)\right\} + r^2V''(r) - rV'(r)\right\}\left(\frac{1}{3}+\frac{2\cos(qr)}{(qr)^2}+\frac{2\sin(qr)}{(qr)^3}\right)\Bigg]. \tag{7}$$

According to modified BS approach [6, 20], the phonon frequencies of longitudinal and transverse branches are give by

$$\omega_{\mathrm{L}}^2(q) = \frac{2N_{\mathrm{C}}}{\rho q^2}(\beta I_0 + \delta I_2) + \frac{k_{\mathrm{e}}k_{\mathrm{TF}}^2q^2|G(qr_{\mathrm{S}})|^2}{q^2+k_{\mathrm{TF}}^2\varepsilon(q)} \tag{8}$$

$$\omega_{\mathrm{T}}^2(q) = \frac{2N_{\mathrm{C}}}{\rho q^2}\left(\beta I_0 + \frac{1}{2}\delta(I_0 - I_2)\right). \tag{9}$$

Other details of the constants used in this approach were already narrated in literature [6, 20]. Here M, ρ 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, transverse and longitudinal sound velocities υ_{L} and υ_{T} are computed. The isothermal bulk modulus B_{T}, modulus of rigidity G, Poisson's ratio σ, Young's modulus Y and Debye temperature θ_{D} are found using the expressions [15],

$$B_{\mathrm{T}} = \rho_{\mathrm{M}}\left(\upsilon_{\mathrm{L}}^2 - \frac{4}{3}\upsilon_{\mathrm{T}}^2\right), \tag{10}$$

$$G = \rho_{\mathrm{M}}\upsilon_{\mathrm{T}}^2. \tag{11}$$

With ρ_{M} is the isotropic number density of the solid.

$$\sigma = \frac{1-2\left(\frac{\upsilon_{\mathrm{T}}^2}{\upsilon_{\mathrm{L}}^2}\right)}{2-2\left(\frac{\upsilon_{\mathrm{T}}^2}{\upsilon_{\mathrm{L}}^2}\right)}, \tag{12}$$

$$Y = 2G(\sigma+1), \tag{13}$$

$$\theta_{\mathrm{D}} = \frac{\hbar\omega_{\mathrm{D}}}{k_{\mathrm{B}}} = \frac{\hbar}{k_{\mathrm{B}}}2\pi\left[\frac{9\rho}{4\pi}\right]^{1/3}\left[\frac{1}{\upsilon_{\mathrm{L}}^3}+\frac{2}{\upsilon_{\mathrm{T}}^3}\right]^{(-1/3)}, \tag{14}$$

here ω_{D} is the Debye frequency.

The low temperature specific heat C_{V} is obtained from Kovalenko and Krasny [21],

$$C_V = \frac{\Omega_{\mathrm{O}}\hbar^2}{k_{\mathrm{B}}T^2} \sum_{\lambda=L,T}\int\frac{d^3q}{(2\pi)^3}\frac{\omega_\lambda^2(q)}{\left[\exp\left(\frac{\hbar\omega_\lambda(q)}{k_{\mathrm{B}}T}\right)-1\right]\left[1-\exp\left(-\frac{\hbar\omega_\lambda(q)}{k_{\mathrm{B}}T}\right)\right]}. \tag{15}$$

The basic feature of temperature dependence of C_V is determined by the behaviour of $\omega_\lambda(q)$.

Results and discussion

The input parameters and other related constants used in the present computations are $Z = 2.00$, $\Omega_{\mathrm{O}} = 2.0472\times 10^{-23}$ cm^3, $r_{\mathrm{C}} = 5.8665\times10^{-9}$ cm, $N_{\mathrm{C}} = 12.00$, $M = 5.871\times10^{-23}$ gm and $\rho_{\mathrm{M}} = 2.9699$ gm/cm^3. All the input parameters are taken from the literature [15].

The comparison of presently computed pair potentials and other such theoretical results [3, 5] are displayed in Fig. 1. From the Fig. 1, it is seen that the first zero for $V(r = r_0)$ due to Hartree function occurs at $r_0 = 10$ au, while under the exchange and correlation influence, the zero shifts towards at $r_0 \le 4.7$ au. The well width and the $V_{\min}(r)$ position of the pair potentials are also affected by the behaviour of the screening. It is also noticed that the well depth of presently computed pair potentials are moved towards

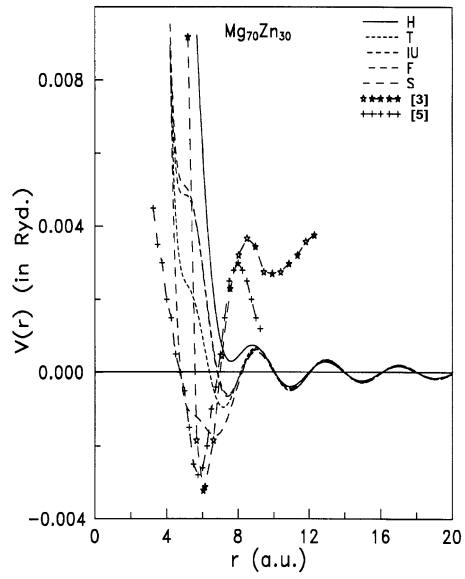

Fig. 1 Pair potential for $Mg_{70}Zn_{30}$ glass

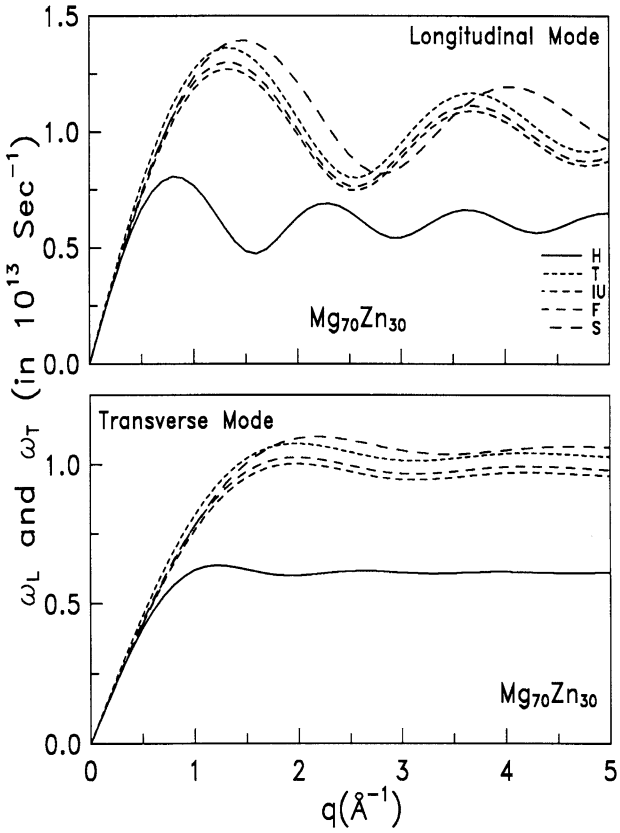

Fig. 2 Screening influence on phonon dispersion curves of $Mg_{70}Zn_{30}$ glass

higher *r*-values and also show lower as compared to the other theoretical results [3, 5]. Also, the depth position of the pair potential is shifted towards the higher *r*-values due to S to H-screening. All the potentials show oscillatory behaviour at larger *r*-values. The result of Saxena et al. [3] and Agarwal et al. [5] shows significant oscillations and potential energy remains positive in the large *r*-region. Thus, Coulomb repulsive potential part dominates the oscillations due to ion–electron–ion interactions in their studies.

The screening influence on the phonon frequencies of $Mg_{70}Zn_{30}$ glass are displayed in Fig. 2, where the results of HB approach are reported. The first minimum in the longitudinal branch is seen around at $q \approx 1.6$ Å^{-1} for H, $q \approx 2.6$ Å^{-1} for T, IU and F and $q \approx 2.8$ Å^{-1} for S local field correction functions. At the first peak on ω_L the screening influence is found 68.88% for T, 57.58% for IU, 61.12% for F and 72.76% for S function with respect to static Hartree function. At $q \approx 1.0$ Å^{-1} point, such screening variation on ω_T due to T, IU, F and S screening is 31.82%, 23.06%, 25.82% and 25.88%, respectively. The presently computed results of the PDC due to T, IU and F functions are lying between those due to H and S-screening.

The PDC calculated from the HB, TG and BS approaches with S-local field correction function are shown in Fig. 3. It is noticed from the figure that the first minimum in the longitudinal branch is seen around at $q \approx 1.8$ Å^{-1} for BS, $q \approx 2.9$ Å^{-1} for TG and $q \approx 2.8$ Å^{-1} for HB approach. The first crossing position of ω_L and ω_T branches is observed at 2.2 Å^{-1} in HB, 1.7 Å^{-1} in TG and 1.7 Å^{-1} in BS approach. In comparison to the other reported data [3, 5], the present results are suppressed one with the others.

The PDC shows the existence of collective excitations at larger momentum transfer due to longitudinal phonons only and the instability of the transverse phonons due to the anharmonicity of the atomic vibrations in the metallic systems. Actually, Neutron Inelastic Scattering (NIS) experiments on $Mg_{70}Zn_{30}$ glass, by Suck et al. [8] have exposed vigorously low-lying short wavelength collective density excitation at wave vector transfer where the structure factor shows its main peak, which are called phonon–roton states [8]. The difference in the magnitude of the minimum around $2k_F$ seems to be due to the fact that the concept of roton has not been taken into account theoretically.

The anomalous behaviour of C_V is shown in Fig. 4 at low temperature region, on increasing the temperature, the specific heat capacity C_V for S local field correction function show high bump. The comparisons of presently computed outcomes for HB approach provide satisfactory results with the experimental data [3], while for TG and BS approaches the present results are found lower for higher temperature. The reason

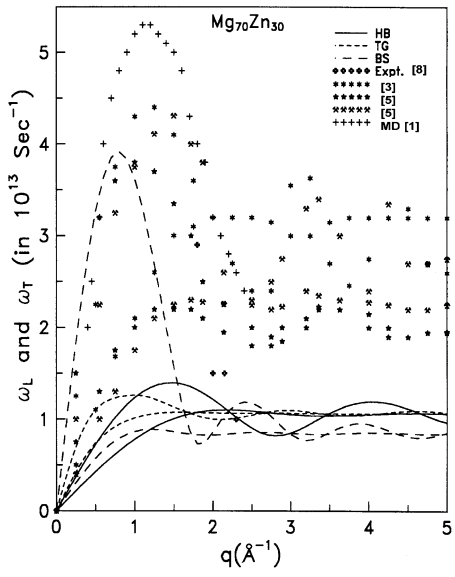

Fig. 3 Phonon dispersion curves for $Mg_{70}Zn_{30}$ glass using three approaches

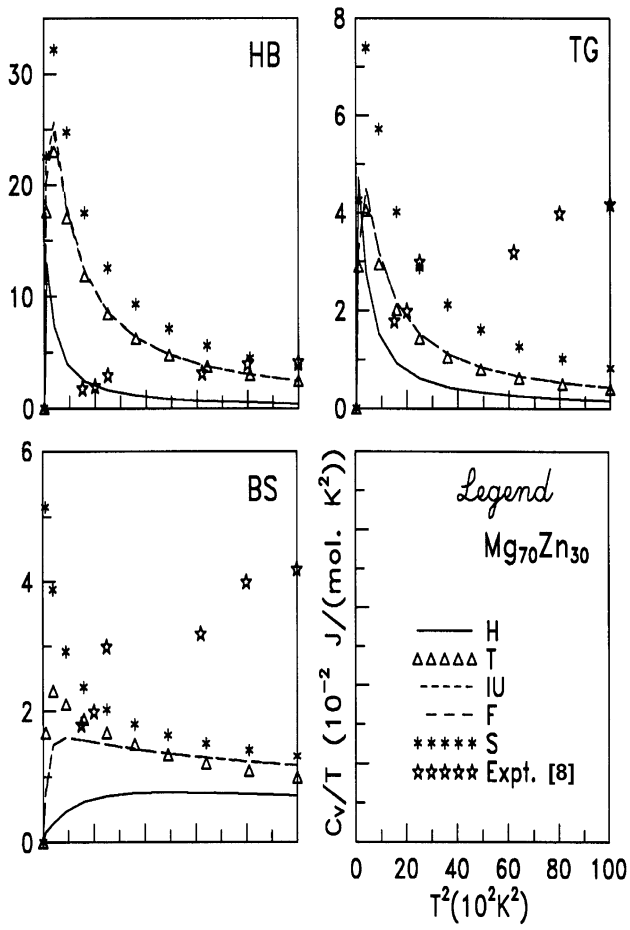

Fig. 4 The specific heat of $Mg_{70}Zn_{30}$ glass

behind the anomalous behaviour may be due to the low frequency modes modify the generalized vibrational density of states of the glass with that of the polycrystal. These modes are mainly responsible for the difference in the temperature dependence of the specific heat, which departs from the normal behaviour.

The thermodynamic and elastic properties computed from the long wavelength limit of the PDC for $Mg_{70}Zn_{30}$ metallic glass is tabulated in Table 1. Here, presently computed longitudinal and transverse sound velocities due to BS approach show good agreement with other such theoretical results [3]. These comparisons favour the present calculation and suggest that proper choice of dielectric screening is important part for explaining the thermodynamic and elastic properties of $Mg_{70}Zn_{30}$ glass.

For most of the metallic glasses, the thermodynamic and elastic data are not available in the literature either experimentally or theoretically. But the advantage of the present computation is generating the theoretical sets of data, which is very useful to provide important information regarding the particular glass.

Conclusions

Finally, it is concluded that the PDC generated form three approaches reproduce all the broad characteristics of dispersion curves. The well-recognized model potential of Gajjar et al. with more advanced F and S-local field correction functions generate consistent results. Hence the model potential is suitable for studying the phonon dynamics of $Mg_{70}Zn_{30}$ metallic glass, which confirms the applicability of the model potential in the aforesaid properties and supports the present approach of PAA. Such study on phonon dynamics of other binary liquid alloys and metallic glasses is in progress.

Table 1 Thermodynamic and elastic properties of $Mg_{70}Zn_{30}$ Glass

App.	SCR	$\upsilon_L \times 10^5$ cm/s	$\upsilon_T \times 10^5$ cm/s	$B_T \times 10^{11}$ dyne/cm^2	$G \times 10^{11}$ dyne/cm^2	σ	$Y \times 10^{11}$ dyne/cm^2	θ_D (K)
HB	**H**	1.5764	0.9101	0.4100	0.2460	0.2500	0.6150	110.02
	T	1.6449	0.9497	0.4464	0.2678	0.2499	0.6696	114.80
	IU	1.5348	0.8861	0.3886	0.2332	0.2500	0.5830	107.11
	F	1.5692	0.9060	0.4063	0.2438	0.2499	0.6094	109.52
	S	1.5300	0.8833	0.3862	0.2317	0.2500	0.5793	106.78
TG	**H**	2.3756	1.3877	0.9135	0.5719	0.2410	1.4195	167.57
	T	3.2780	1.9154	1.7384	1.0896	0.2408	2.7039	231.29
	IU	3.1384	1.8689	1.5422	1.0373	0.2253	2.5420	225.28
	F	3.2227	1.9174	1.6286	1.0919	0.2260	2.6773	231.15
	S	3.0669	1.7964	1.5155	0.9584	0.2389	2.3746	216.87
BS	**H**	6.8967	3.1327	10.2396	2.9146	0.3700	7.9861	384.55
	T	5.5587	1.3498	8.4549	0.5411	0.4687	1.5895	167.85
	IU	5.7818	1.7036	8.7788	0.8619	0.4525	2.5038	211.44
	F	5.7343	1.7163	8.5990	0.8748	0.4508	2.5384	212.98
	S	5.3762	0.8866	8.2727	0.2335	0.4860	0.6938	110.43
Others [3]		4.7,5.1	2.5, 2.6	–	–	–	–	305.21

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