STRUCTURE AND VIBRATIONAL DYNAMICS OF $Mg_{70}Zn_{30}$ METALLIC GLASS

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ABSTRACT

For the two-component metallic Mg70Zn30 glass, an Ashcroft form effective pair potential is suggested using the Wigner-Seitz radius concept. This potential is used to investigate the system's vibrational dynamics further. The calculated phonon frequencies, which align with the findings of the Molecular Dynamics (MD) and Neutron Inelastic Scattering (NIS) experiments, demonstrate the reliability of our research. The longitudinal and transverse sound velocities obtained from the elastic portion of the two phonon modes are used to investigate the thermodynamic and elastic characteristics of this glass.

INTRODUCTION

The amorphous solid phases identified as metallic glasses are formed when ultra-rapid cooling of liquid alloys prevents segregation and crystallization. These non-crystalline materials have been the focus of much recent research because of their unique physical characteristics of shared scientific and technological interest[1,2]. A crucial aspect of this research is the comprehensive understanding of atomic structure and vibrational dynamics based on interatomic forces. This understanding is essential to comprehend the thermodynamic and transport aspects of solids. Despite many-body effects, the accuracy of volume energies and pair potentials can be considered quite high, as phonon frequencies computed using this method align with the experiment within a few percent[3]. Detailed calculations have also demonstrated their utility in forecasting phase changes in metals[4] and alloys[5,-7], as well as associated changes in binding energy and density.

Calculations of the cohesive, structural, dynamic, and thermodynamic properties of the crystalline and liquid metals[5] and alloys[8,9] have thoroughly examined the accuracy and dependability of effective inter-atomic potentials obtained for simple metals and their alloys using pseudopotential theory. Based on these pseudo potential derived forces, a theoretical investigation of the atomic structure[9,10] and the dynamics of the metallic glass $Mg_{70}Zn_{30}$ have been performed. When compared with the neutron scattering experimental results[11], the calculated structure is found to be not just accurate but very realistic, further validating the precision of our research.

This work proposes an effective pair potential in the Ashcroft form using the Wigner-Seitz radius notion. The obtained potential has been used to study the vibrational dynamics of $Mg_{70}Zn_{30}$ glass utilizing the theory of random phase approximation (RPA). Neutron inelastic scattering and molecular dynamics data were compared with the calculated results. The thermal and elastic characteristics have also been investigated using longitudinal and transverse velocities obtained from the slope of the elastic part of two phonon modes.

THEORETICAL DESCRIPTION

A simple metallic alloy $A_{1-x}B_x$ can be considered as one component metallic fluid with effective mass, effective number density as

$$M^{eff} = xM_A + (1-x)M_B \quad \ldots (1)$$

$$\rho^{eff} = x\rho_A + (1-x)\rho_B \quad \ldots (2)$$

Where suffixes A and B represent the parameters of two individuals of the metallic glass. Similarly, the effective pair potential of the glass may be written in the usual form as:

$$V_{eff}(r) = \left(\frac{Z^{eff}}{r}\right)^2 - 2\left(\frac{(Z^{eff})^2}{\pi}\right)\int_0^\infty dq \frac{Sin(qr)}{qr} F_N(q) \quad \ldots (3)$$

Where $F_N(q) = \frac{q^4\Omega^2}{16\pi^2 Z^2 e^4}\left[1 - \frac{1}{\varepsilon(q)}\right]|W_b(q)|$ ... (4)

and $Z^{eff} = Z_A C_A + Z_B C_B$ is the effective valence of the glass, C_A and C_B are concentrations of A and B, respectively, and are obtained in terms of concentration fraction and molar volumes V_A and V_B as:

$$C_A = \frac{xV_A}{xV_A+(1-x)V_B} \quad \text{and} \quad C_B = \frac{(1-x)V_B}{xV_A+(1-x)V_B} \quad \ldots (5)$$

The Fourier transform of the Ashcroft Pseudopotential[12] is

$$W_b(q) = -\frac{4\pi Z^{eff} e^2}{q^2\Omega} Cos\,(q.r_c^{eff} \quad \ldots (6)$$

The dielectric response function $\varepsilon(q)$ is given as

$$\varepsilon(q) = 1 - \frac{(4\pi e^2/q^2)\chi(q)}{1+(4\pi e^2/q^2)\chi(q).G(q)} \quad \ldots (7)$$

The function G(q) is called the local field function and accounts for exchange and correlation among the conduction electrons. For the present study, we have chosen the function G(q) given by Hubbard and Sham[13]:

$$G(q) = \frac{q^2}{2\left[q^2+\nu\left(K_F^{eff}\right)^2\right]} \quad \ldots (8)$$

$$\nu = \frac{2}{\left(1+0.153/\pi K_F^{eff}\right)} \quad \ldots (9)$$

$$K_F^{eff} = (3\pi^2 Z^{eff}\rho^{eff})^{1/3} \quad \ldots (10)$$

In the above expressions, r_c^{eff} is called the ionic core radius of the bimetallic glass and is a disposable parameter. It is obtained using the relation:

$$x\frac{4}{3}\pi r_A^3 + (1-x)\frac{4}{3}\pi r_B^3 = \frac{4}{3}\pi r_{glass}^3 \quad \ldots (11)$$

Where r_A, r_B, and r_{glass} are the Wigner -Seitz radii of A, B, and glass, respectively. The Wigner-Seitz radius of the glass is related to the effective core radius by the relation[14]

$$r_C^{eff} = \frac{0.51 r_{glass}}{(Z^{eff})^{1/3}} \quad \ldots (12)$$

The phonon eigen frequencies are meaningful parameters for studying the collective excitations in the system. The expressions for longitudinal and transverse phonon eigen frequencies in random phase approximation (RPA) are given as[15]

$$F_l^2(q) = W_E^2\left[1 - \frac{3Sin(q\sigma)}{q\sigma} - \frac{6Cos(q\sigma)}{q\sigma} + \frac{8Sin(q\sigma)}{q\sigma}\right] \quad \ldots (13)$$

$$F_t^2(q) = W_E^2\left[1 + \frac{3Cos(q\sigma)}{q\sigma} - \frac{3Sin(q\sigma)}{q\sigma}\right] \quad \ldots (14)$$

Where

$$W_E = \left[\frac{4\pi\rho^{eff}}{3M^{eff}}\int_0^\infty dr r^2\, g_{eff}(r)\frac{\partial^2}{\partial r^2}V_{eff}(r)\right]^{1/2} \quad \ldots (15)$$

and "σ" is the hardcore diameter. The hardcore diameter of the alloy is obtained from the expression[16]:

$$\sigma = \frac{\pi}{6}[N_A\sigma_A^3 + N_B\sigma_B^3] \quad \ldots (16)$$

$$\eta = \frac{\pi}{6}\left(\frac{1}{\Omega}\right)(\sigma_{glass})^3 \quad \ldots (17)$$

Where $N_A = {}^{C_A}/_{\Omega}$ and $N_B = {}^{C_B}/_{\Omega}$ and "Ω" is the atomic volume of the glass.

RESULTS AND DISCUSSION

Using the Wigner-Seitz radius concept and Eq. 3, the effective pair potential of $Mg_{70}Zn_{30}$ glass is calculated. Fig.1 displays both the potentials: (i) computed potential using the concept of effective atom and (ii) the potential that results from treating the metallic glass as a three-component system. As reported elsewhere[17], the three-component system comprises the bare ions of two distinct species, AA and BB, and a mixed ion, AB, submerged in a

homogeneous electron gas. Despite minor quantitative changes, Fig. 1 shows the two potentials are in excellent qualitative agreement. This substantial agreement between two potentials justifies that metallic glass should be treated as a single-component system of effective atoms while determining the pair potential.

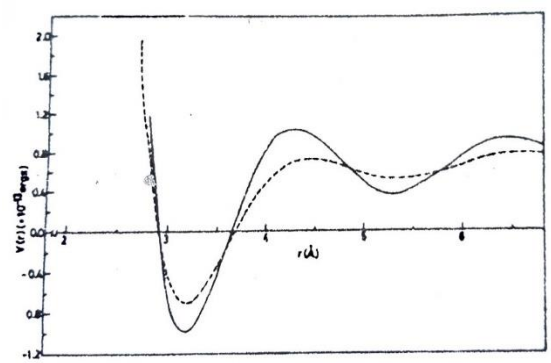

Fig.1: Effective pair potential of $Mg_{70}Zn_{30}$ glass: present computed results-------- results of Saxena et al.

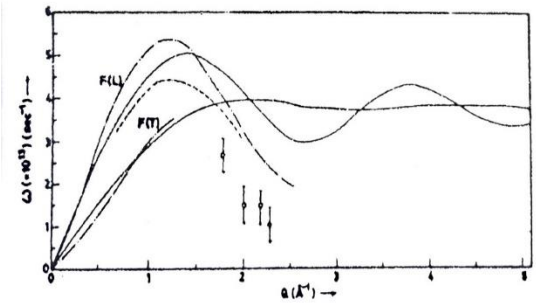

Fig.2: Longitudinal F(L) and Transverse F(T) phonon dispersion curves of $Mg_{70}Zn_{30}$ glass: _____ present computed results, ---------- Molecular Dynamics results, §§§§ Neutron inelastic scattering results of Suck et al. and - - - - - - Model calculation of Tomanek.

Fig. 2 displays the phonon eigen frequencies for the longitudinal and transverse phonon modes as determined by Eqs. (13) and (14) respectively. The effective pair correlation function, $g_{eff}(r)$, is taken from the molecular dynamics plus potential energy mapping results[18]. The computed results were compared with the Molecular Dynamics results of Heimendahl[19]. A reasonably good agreement is observed between the two curves. The same figure also displays the results of model calculation[19] along with Neutron Inelastic Scattering (NIS) data of Suck et al[20].

It can be noted from the Fig. 2 that oscillations predominate in the longitudinal phonon mode. The MD results[19] reasonably agree with the phonon eigen frequencies of the longitudinal and transverse phonon modes. The Molecular Dynamics results of the transverse branch are known only up to 1.2Å^{-1}. Nevertheless, the calculated result is monoatomic and exhibits no oscillations. For longitudinal excitations, the dispersion relations of Molecular Dynamics show a clear maximum at Q_p~1.21Å^{-1}, almost half the wave vector value of the first peak in the static structure factor (Q=2.54Å-1). The subsequent minimum is not identifiable as the peak in the structural factor becomes too broad. The stiffness of the amorphous solid against sheer deformations is reflected in the well-defined vibrational energy for transverse waves for small momenta. The dispersion relations for transverse excitations do not have a peak. The calculated findings indicate that the longitudinal branch at Q~Q_P has a minimum. The current longitudinal frequencies are qualitatively consistent with the neutron inelastic scattering experiment's four points. However, in the long wavelength limit, the longitudinal and transverse modes behave elastically and follow the relationships.

$F_1=C_lq$ and $F_t= C_tq$

Where C_l and C_t are the longitudinal and transverse sound velocities, respectively, in the glass, the values of C_l and C_t derived from the elastic part of the two phonon modes C_l=4.5x10^{5}cm/sec and C_t= 2.9x10^{5}cm/sec are in agreement with the values obtained from the Brillouin Scattering experiment[21] (C_l = 5.1x10^{5}cm/sec and C_t= 2.5x10^{5}cm/sec). The thermal and elastic properties of $Mg_{70}Zn_{30}$ glass have been studied using the calculated values of longitudinal and transverse sound velocities. Debye temperature has been calculated using the relation[22]

$$\theta_D = \frac{h}{k_B} 2\pi \left(\frac{9\eta}{4\pi}\right)^{1/2} \left[\frac{1}{c_l^2} + \frac{2}{c_t^2}\right]^{-1/3} \quad \ldots (18)$$

The results have been compared with the obtained through the expression[23]

$$\frac{1}{\theta_D^2} = \frac{\rho_A}{\theta_{D_A}^2} + \frac{\rho_b}{\theta_{D_b}^2} \quad \ldots (19)$$

Where p_A and p_B are the atomic concentrations and Θ_A and Θ_B are the Debye temperature[24] of the pure components of the metals in the alloy. The value of Debye temperature calculated using Eq.18 ($\theta_D = 305K$) is in excellent agreement with the value ($\theta_D = 307K$) computed through Eq.19. Besides, the isothermal bulk modulus B_r of an isotropic solid is given by

$B_r = \boldsymbol{\rho}(C_l^2 - 4/3C_t^2)$ …(20)

where $\boldsymbol{\rho}$ is the density of the isotropic solid. For $Mg_{70}Zn_{30}$ metallic glass, the value using Eq.20 is $B_r = 2.587 \times 10^{11}$ dynes/cm^2. This is approximately 28% lower than the B_r averaged value over the crystalline metals $B_r = 3.58 \times 10^{11}$dynes/cm^2. The theoretical bulk modulus for the pure metals is $B_r = 3.58 \times 10^{11}$dynes/cm^2(Mg) and $B_r = 3.58 \times 10^{11}$dynes/cm^2(Zn). This B_T(Zn) value has been calculated from the experimental data on isothermal compressibility. Both results suggest that the longitudinal elastic modes are softer in the glassy phase.

CONCLUSION

The current analysis concludes that the present potential treats the system as if it is comprised of an effective atom and produces relatively reasonable outcomes than the traditional potential. It simplifies the computations. The computed potential is then used to study the system's vibrational dynamics, and the obtained results are consistent with those obtained experimentally. The elastic properties calculated from the long wavelength limit of the phonon modes indicate softer modes in the glassy phase.

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