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Dynamics of a bulk metallic glass $Cu_{60}Zr_{20}Hf_{10}Ti_{10}$

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Abstract

Computations of the frequencies of the longitudinal and transverse phonon modes in a quaternary bulk metallic glass ($Cu_{60}Zr_{20}Hf_{10}Ti_{10}$) based on a simple model approach have been presented. The model assumes a central force, effective between the nearest neighbours and a volume dependent force. Both types of excitations of phonons are computed for the bulk metallic glass for the first time both for self-consistent screening of conduction electrons with and without the inclusion of correlation effects. Phonon frequency expressions reproduce the main characteristic features of the dispersion curves.

Keywords: Phonon frequency; Dispersion relation; Bulk metallic glass

1. Introduction

With the advent of metallic glasses [1], a new era was opened in the field of materials science. Metallic glasses show an excellent combination of their mechanical, chemical, magnetic and electrical properties [2–5], which is far superior to the crystalline alloys of identical composition. The cooling rate required for these materials was very high, of the order of $10^6\,K\,s^{-1}$, thereby restricting the specimen geometry to thin ribbons, foils and powders, where at least one-dimension was small enough, to the order of microns, to permit such a high cooling rate. The promising attributes of metallic glasses have led scientists and engineers to dream of new alloys that would form glasses at low cooling rates like oxide glasses, which would enable their production in bulk form. This dream was turned into a reality by the pioneering investigations of Inoue et al. [6]. Turnbull's criterion [7] has played a key role in the development of metallic glasses including metallic glasses in bulk, i.e. bulk metallic glasses (BMGs). BMGs have emerged over the past 16 years with attractive properties and technological promise [8,9]. Several significant contributions have come from Johnson's group at Caltech, USA [10,11], where the pioneering investigations of Duwez gave birth to the field of metallic glass [1]. The acoustic, elastic and thermal properties of metallic glasses are closely related to their binding nature and vibrational characteristics [12–14]. The acoustic and elastic properties as well as vibrational features in the metallic glasses are poorly understood. A fundamental understanding of micro-structural configuration in amorphous solids is not as developed as in the crystalline solids. The bulk metallic glasses (BMGs) make them in the form suitable for measurements of elastic wave propagation. If one arbitrarily defines the millimeter scale as 'bulk', the first bulk metallic glass was the ternary Pd–Cu–Si alloy prepared by Chen [15]. Agarwal and Kachhava [16,17] computed phonon frequencies and their dependence on dielectric screening of the ternary metallic glass $Pd_{77.5}Si_{16.5}Cu_6$ for the first time. The simple model [18] is extended here for the quaternary BMG $Cu_{60}Zr_{20}Hf_{10}Ti_{10}$ for the first time for the computation of phonon eigen-frequencies both for the transverse and the longitudinal modes of vibration with and without including correlation effects. This simple model assumes a central force, effective between nearest neighbours and a volume dependent force due to conduction electrons. This system is assumed to be a mixed structure of fcc and hcp structure in the proportionate portion to the constituents.

The understanding of the vibrational dynamics is a prerequisite to any understanding of thermodynamic, transport

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and other properties of condensed systems on a microscopic level [19]. In our present work, we would like to study one of the dynamical properties–dispersion relation. There are three main theoretical approaches to derive the phonon frequencies of the metallic glasses. In one approach, Hubbard and Beeby [20] have derived expressions for the longitudinal and transverse phonon frequencies on the basis of the physical argument that the product of the static pair correlation function and the second-order derivative of the potential is peaked near the hard-core radius. They studied the collective motion in liquids as a generalization of phonon theory of solids using random phase approximation. In another approach, Takeno and Goda [21] have expressed phonon eigen-frequencies in terms of many body correlation functions of atoms and inter-atomic potentials in amorphous solids. Apart from these two approaches as a third approach Bhatia and Singh [18] have proposed a phenomenological model to obtain the longitudinal and transverse phonon dispersion in a one-component metallic glass and amorphous materials. All these three approaches have been widely utilized to study the phonon dispersion and collective excitations in metallic glasses; however, the simple model approach given by Bhatia and Singh [18] is found better for providing significant understanding of the structure of the amorphous binary system of transition metals by Agarwal et al. [22] for $Cu_{57}Zr_{43}$ and by Lad and Pratap [23] for Zr–Ni alloys. Pratap et al. [24] have recently derived phonon frequencies of the $Cu_{57}Zr_{43}$ system using the other two approaches; still the simple model approach under consideration [22] seems to be closer to the results obtained by Kobayashi and Takeuchi [25] using a recursion method. These observations have motivated the author to extend this approach to the quaternary system.

2. Theory

The longitudinal phonon frequencies (ω_L) and transverse phonon frequencies (ω_T) are, respectively, given by [18]:

$$\omega_L^2 = \frac{2N}{\rho a^2}[\beta I_0 + \delta I_2] + \frac{\kappa_e^2 K_{TF}^2 q^2 (G(qr_s))^2}{\rho(q^2 + K_{TF}^2 \varepsilon(q))}, \quad (1)$$

and

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

where N is the coordination number, a is the nearest neighbour distance and $\rho = n_i M$ is the mean atomic density in which M is the mean atomic mass defined as:

$$\begin{aligned} M = {} & 0.60 \text{ times the atomic mass of Cu} \\ & +0.20 \text{ times the atomic mass of Zr} \\ & +0.10 \text{ times the atomic mass of Hf} \\ & +0.10 \text{ times the atomic mass of P} \end{aligned} \quad (3)$$

and n_i is the ion density. For hcp structure, $n_i = 4/(\sqrt{3}a^3 y)$ with $y = c/a$ ratio and for fcc structure, $n_i = \sqrt{2}/a^3$. In the quaternary glassy system under consideration, Cu is of fcc structure with concentration C_A while other three constituents Zr, Hf and Ti are of hcp structure with concentrations C_B, C_C and C_D, respectively, in pure form. Then, n_i can be written as:

$$n_i = C_A \frac{\sqrt{2}}{a^3} + \frac{4}{a^3\sqrt{3}}\left(\frac{C_B}{y_B} + \frac{C_C}{y_C} + \frac{C_D}{y_D}\right). \quad (4)$$

β, δ and κ_e are force constants. β and δ are defined in the terms of the inter-atomic potential $W(r)$, as:

$$\beta = \frac{\rho a^2}{2M}\left[\frac{1}{r}\frac{dW(r)}{dr}\right]_{r=a}, \quad (5)$$

$$\delta = \frac{\rho a^3}{2M}\left[\frac{d}{dr}\left(\frac{1}{r}\frac{dW(r)}{dr}\right)\right]_{r=a}. \quad (6)$$

The conduction electron screening to the inter-atomic potential is represented by the Thomas–Fermi screening length defined in terms of charge e and mass m of electron as:

$$K_{TF}^2 = \left(\frac{16\pi^2 m e^2}{h^2}\right)\left(\frac{3n_e}{\pi}\right)^{1/3}. \quad (7)$$

Then, the relevant force constant κ_e due to the conduction electrons on the basis of the Thomas–Fermi model can be derived as:

$$\kappa_e = \frac{4\pi n_e n_i z e^2}{K_{TF}^2}, \quad (8)$$

where n_e is the electron density so that $n_e = n_i z$ and z is the mean valence of the glassy system. In Eq. (1), $\varepsilon(q)$ is the self-consistent dielectric screening function [26]:

$$\varepsilon(q) = \frac{1}{2}\left[1 + \frac{k_F}{q}\left(1 - \frac{q^2}{4k_F^2}\right)\ln\left|\frac{q + 2k_F}{q - 2k_F}\right|\right], \quad (9)$$

where $k_F = (3\pi^2 n_e)^{1/3}$ is the Fermi wave number.

To incorporate the correlation effects in $\varepsilon(q)$, (9) is modified to the form:

$$\varepsilon(q) = \frac{1}{2}\left[1 + \frac{k_F}{q}\left(1 - \frac{q^2}{4k_F^2}\right)\ln\left|\frac{q + 2k_F}{q - 2k_F}\right|\right]\left[1 - f(q)\right], \quad (10)$$

where $f(q)$ is given by Hubbard [27]:

$$f(q) = \frac{1}{2}\frac{q^2}{q^2 + k_F^2 + \frac{1}{2}K_{TF}^2}. \quad (11)$$

The cancellation effects of kinetic and potential energies inside the core of the ions, making the effective potential weak in the core, give a shape factor, $[G(qr_s)]^2$, to be multiplied