

# Elastic Moduli and Phonon Dispersion Curves for Amorphous Metals and Alloys

Ram N. Singh and Issam Ali

**Abstract**—Changes in elastic constants on amorphization are obtained from the corresponding cubic crystal values following a theoretical scheme based on equations of motion of deformation and dispersion relations. The long wavelength limit of the dispersion relations for an amorphous system enables to connect the bulk moduli to force constants. Bulk, shear and Young moduli and Poisson ratio are obtained for amorphous Ag, Al, Au, Cr, Cu, Fe, Ge, Ir, K, Li, Mo, Na, Nb, Ni, Pb, Pd, Pt, Si, Ta, Th, V and W. We also make a connection of elastic properties to thermal properties through Debye theory. Finally, we present phonon dispersion curves computed for selected amorphous metals (Si, Pb, Ni, Al, Au, Th and Fe) and amorphous alloys (ZrCo and Zr67Ni33).

**Index Terms**—Amorphous metals, amorphous alloys, elastic constants, phonon dispersion.

## I. INTRODUCTION

Ordinarily, metallic liquids solidify immediately into crystalline phases below melting temperature. However, crystallization can be avoided by fast cooling rates, which is very significant for the synthesis of an amorphous phase for metallic systems. Since the pioneering work of Klement *et al.* [1], a good deal of progress [2]-[6] has been made to enhance the attainability of the amorphous phase at reduced cooling rates and at temperatures close to the glass transition temperature. Retaining the disordered structure of liquids in the amorphous metallic state, while maintaining its stability, is an outcome of delicate balance of mechanical and thermal forces.

The mechanical and thermal properties of amorphous metals are strikingly different than ordinary glasses. Unlike oxide glasses, the amorphous metals are soft, plastically deformable and endowed with high tearing strength. Though the density at amorphization is only 1-2% less than the corresponding crystalline phase, the bulk modulus decreases by about 7%. Young and shear moduli, however, are lowered by ~20-40%. The velocity of the longitudinal sound wave ( $v_L$ ) in amorphous metallic systems is found to be ~16% less than the corresponding crystalline phase whose transverse sound velocity ( $v_T$ ) increases by ~9% on amorphization. Also,  $v_L/v_T \approx \sqrt{3}$ , which, in general, is not true for crystalline metals.

In this work we have used a simple central force model to discuss the elastic deformation of amorphous metals. The effective interaction between ions is modelled through a

central pairwise potential effective between the nearest neighbors only. The volume-dependent force due to conduction electrons has been included using the Thomas-Fermi method. Relevant relations and results for the elastic constants, bulk modulus, shear modulus, Young modulus, Poisson ratio and velocities of sound for amorphous metals are presented in Section II. The evaluation of dispersion curves and connection of elastic properties to thermal properties through Debye temperature are discussed in Section III. Phonon dispersion curves for ZrCo and Zr67Ni33 are also presented. This is followed by conclusion in Section IV.

## II. ELASTIC DEFORMATION OF AMORPHOUS METALS

Amorphous metals are often treated as elastically isotropic deformed systems. Macroscopically, deformations are homogeneous, but at the microscopic levels there are local stresses which cause inhomogeneous deformations. The force ( $F = F_{ion} + F_{electron}$ ) consists of contributions from ions and electrons. Bhatia and Singh [7] suggested a simple theoretical approach where ionic interactions are evaluated with a central pairwise potential, and the force acting on ions due to electrons is modelled through a volume-dependent term using the Thomas-Fermi method.

In an amorphous material, the nearest neighbor atoms around each atom may be regarded (ignoring local stresses) as continuously distributed over a spherical surface of appropriate radius  $a$ . It facilitates to solve the equation of motion where the sum over  $n$  neighbors can be replaced by an integral,  $\sum_n \dots = \left(\frac{z}{4\pi}\right) \int \dots d\Omega$ ,  $z$  is the coordination number) over the surface of a sphere of volume  $\Omega$ . For the longitudinal and transverse wave frequencies  $\omega_L$  and  $\omega_T$  for the elastic wave propagation in amorphous materials, one has [7].

$$\rho\omega_L^2(q) = \frac{2z}{a^2}(\beta I_1 + \delta I_2) + \frac{\kappa_e K_{TF}^2 q^2 G^2(qr_s)}{q^2 + K_{TF}^2 g(q)} \quad (1)$$

$$\rho\omega_T^2(q) = \frac{2z}{a^2} \left[ \beta I_1 + \frac{\delta}{2} (I_1 - I_2) \right] \quad (2)$$

The second term on the right hand side of (1) takes into account the electrostatic contributions.  $\beta$ ,  $\delta$  and  $\kappa_e$  are force constants parameters and can be determined using the observed elastic data.  $K_{TF}$  is the reciprocal of the Thomas-Fermi screening length.  $G(q)$  stands for exchange and correlation effects of the electron screening through the modified Hartree dielectric function. The shape factor  $G(qrs)$  ( $rs$  is the Wigner-Seitz radius) takes into account [8] the cancellation effects occurring between the kinetic and potential energies inside the ion cores, making the effective

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potential weak in the core.  $\beta$  and  $\delta$  are related to the first and second derivatives, respectively, of the ionic pairwise potential  $\phi(r)$  at  $r = a$ , the nearest neighbour distance,

$$\beta = \frac{\rho a^2}{2M} \left[ \frac{1}{r} \frac{\partial \phi(r)}{\partial r} \right]_{r=a}, \delta = \frac{\rho a^3}{2M} \left[ \frac{\partial}{\partial r} \left( \frac{1}{r} \frac{\partial \phi}{\partial r} \right) \right]_{r=a} \quad (3)$$

where  $\rho = niM$  ( $ni$  is the nearest neighbor density) and  $M$  denotes the mass of the ion. The integrands I1 and I2 stand for

$$I_1 = 1 - \frac{\sin(qa)}{qa} \quad (4)$$

$$I_2 = \frac{1}{3} - \sin(qa) \left[ \frac{1}{qa} - \frac{2}{(qa)^3} \right] - \frac{2\cos(qa)}{(qa)^2} \quad (5)$$

#### A. Elastic Constants

Equations (1) and (2) are used to obtain relations for elastic constants. There are only two (camp11 and camp44) independent elastic constants for isotropic materials. For  $q \rightarrow 0$ , (1) and (2) give

$$\rho v_L^2 = \frac{z\beta}{3} + \frac{z\delta}{5} + \kappa_e \quad (6)$$

$$\rho v_T^2 = \frac{z\beta}{3} + \frac{z\delta}{15} \quad (7)$$

where  $v_L (= \omega_L/q)$  and  $v_T (= \omega_T/q)$  are longitudinal and transverse wave velocities.  $v_L$  and  $v_T$  can readily be related to elastic constants. Bhatia and Singh [9] expressed the equation of motion for displacement  $\vec{s}$  for elastically isotropic solids as

$$\rho \frac{\partial^2 \vec{s}}{\partial t^2} = c_{amp11} \text{grad div } \vec{s} + c_{amp44} \nabla^2 \vec{s} \quad (8)$$

For shear waves ( $\text{div } \vec{s} = 0$ ), (8) gives

$$v_T = \left( \frac{c_{amp44}}{\rho} \right)^{1/2} \quad (9)$$

For longitudinal waves, the deformation produced is not accompanied by rotation ( $\text{curl } \vec{s} = 0$ ), hence, noting the vector identity,  $\text{grad div} = \text{curl curl} + \nabla^2$ , one gets

$$v_L = \left( \frac{c_{amp11}}{\rho} \right)^{1/2} \quad (10)$$

Further, the force constants on the right hand side of (6) and (7) can readily be related [10] to the elastic constants  $c_{ij}$  of cubic structure materials, i.e.,

$$c_{amp11} = c_{11} - \frac{2}{5} Pa \quad (11)$$

$$c_{amp44} = c_{44} - \frac{1}{5} Pa \quad (12)$$

where  $Pa = c_{11} - c_{12} - 2c_{44}$  is the elastic anisotropy of a cubic material. In the special case where the system is in equilibrium under central forces alone ( $\kappa_e = \beta = 0$ ), [6] and [7] yield

$$\frac{v_L}{v_T} = \sqrt{3} \quad (13)$$

A similar result to (13) has also been obtained by Grest *et al.* [11] by computer simulations of a glass in which the atoms interact via central forces of the Lennard-Jones potential.

#### B. Elastic Moduli

TABLE I: ELASTIC CONSTANTS AND DEBYE TEMPERATURES FOR SELECTED AMORPHOUS METALS;  $C_{11}$  AND  $C_{44}$  (AMORPHOUS PHASE, GPA);  $v_L$  AND  $v_T$  (MS<sup>-1</sup>);  $B$  AND  $E$  (AMORPHOUS PHASE, GPA);  $\Theta_A$  (AMORPHOUS PHASE, K);  $\Theta_L$  (LIQUID PHASE, K);  $\Theta_C$  (CRYSTALLINE PHASE, K)

	$c_{11}$	$c_{44}$	$v_L$	$v_T$	$B$	$E$	$\nu$	$\Theta_a$	$\Theta_l$	$\Theta_c$
Ag	143	32.2	3688	1752	99.8	87.3	0.354	154.5	164.1	224
Al	113	26.3	6454	3118	77.5	70.8	0.348	276.6	294	428
Au	202	32.3	3233	1293	159	90.6	0.405	115.2	121.6	165
Cr	318	117	6643	4030	162	283	0.209	416.3	459.9	630
Cu	219	58.5	4931	2548	141	154	0.318	268.8	244.4	343
Fe	279	85.4	5957	3297	165	219	0.279	346.2	364.7	470
Ge	150	56.5	5300	3249	75.0	135	0.199	244.6	273	374
Ir	650	221	5371	3134	355	549	0.242	298.9		420
K	5.91	1.74	2669	1450	3.59	4.50	0.291	59.2	71.6	91
Li	19.7	5.69	6079	3268	12.1	14.8	0.297	253.9	275.2	344
Mo	434	124	6519	3490	268	323	0.299	319.6		450
Na	8.18	2.80	2902	1698	4.45	6.94	0.240	97.2	158	158
Nb	225	39.6	5118	2150	172	110	0.393	185.6		275
Ni	291	78.4	5270	2968	187	206	0.316	324.9	324.7	450
Pb	56.2	10.1	2227	945	42.7	28.1	0.390	62.8	81	105
Pd	264	53.3	4684	2103	193	146	0.374	200.4	204.7	274
Pt	370	65.1	4146	1740	283	181	0.393	164.1	168.5	240
Si	194	70.8	9115	5501	100	172	0.214	441.0	471	645
Ta	290	70.7	4172	2059	196	189	0.339	176.3		240
Th	103	34.0	2968	1704	57.7	85.2	0.254	106.7		163
V	224	52.8	6136	2976	154	142	0.346	287.4		380
W	512	153	5151	2815	308	294	0.287	255.5		400

The bulk modulus ( $B_a$ ), shear modulus ( $G_a$ ), Young modulus ( $E_a$ ) and the Poisson ratio ( $\nu_a$ ) for isotropic elastic materials are related [9] to the elastic constants, i.e.

$$B^a = c_{amp\ 11} - \frac{4}{3}c_{amp\ 44} \quad (14)$$

$$G^a = c_{amp\ 44} \quad (15)$$

$$E^a = \frac{c_{amp\ 44}(3c_{amp\ 11} - 4c_{amp\ 44})}{c_{amp\ 11} - c_{amp\ 44}} \quad (16)$$

$$\nu = \frac{c_{amp\ 11} - 2c_{amp\ 44}}{2(c_{amp\ 11} - c_{amp\ 44})} \quad (17)$$

The computed values of the elastic constants for amorphous metals are tabulated in Table I. The values of the  $c_{ij}$  of cubic crystalline materials are taken from [12]. The table suggests that amorphous metals differ considerably from one another and possess a wide range of elastic moduli. Such diverse values of elastic properties make the amorphous metals interesting for particular technical applications. The elastic anisotropy  $Pa$  is mostly negative except for Nb, Cr, Mo and W. The highest negative values of  $Pa$  are found for Ir, Fe, Ni and Cu, whereas Na and K have the lowest negative  $Pa$ . Highest values of bulk ( $Ba$ ), shear ( $Ga$ , or  $c_{amp44}$ ) and Young ( $Ea$ ) moduli are found for amorphous metals like Ir, W and Mo, while alkali group metals (Na and K) possess the lowest values of  $Ba$ ,  $Ga$  and  $Ea$ . The Poisson ratio is found to vary from 0.2 (for Ge) to 0.4 (for Au). We may recall that Poisson originally argued for a universal value of  $\nu = 1/4$  for all materials. This makes  $c_{amp12} = c_{amp44}$ . Most materials have Poisson ratio values ranging from zero to 0.5. A perfectly incompressible material deformed elastically at small strains would have the Poisson ratio of exactly 0.5. Materials like steel and rigid polymers exhibit values around 0.3.

It is well known [13] that when an amorphous material crystallizes, the shear modulus increases by about 30-40%, while the corresponding increase in the bulk modulus  $Ba$  is only 4-6%. We can, with the use of the present formalism, provide an estimate to the changes  $\Delta B$ , of the bulk modulus, and  $\Delta G$ , of the shear modulus. First, we note that at equilibrium, the sum of the ionic ( $p_i$ ) and electronic ( $p_e$ ) pressures must be zero,

$$p_i + p_e = 0 \quad (18)$$

With  $p_i (= \partial U_i / \partial \Omega, U_i$  is the ionic energy and  $\Omega$  stands for volume) and  $p_e (= -f \kappa e d\Omega / \Omega)$  (18) can be expressed further in terms of potential parameters as

$$\frac{\beta'}{3} + \frac{3A}{5}\Omega^{-5/3} = 0 \quad (19)$$

where  $\beta' = z\beta$ . In deriving (19), we have taken the free electron value of  $\kappa e (= A\Omega^{-5/3}, A$  being a constant). Making use of (6), (7), (14), (15), we get

$$\frac{\Delta G}{G} = \frac{n}{5} \left[ 3 \frac{\Delta B}{B} - \frac{1}{m} \frac{\Delta \Omega}{\Omega} \right] \quad (20)$$

where  $n = B/G$  and  $m = B/\kappa e$ . For a typical change of  $\Delta \Omega / \Omega = -0.02$ ,  $\Delta B/B \approx 0.06$  on crystallization and for  $m = 1.5$  one has

$$\frac{\Delta G}{G} = 0.04n \quad (21)$$

As  $n$  ranges from 2 to 5 for different materials [13], (21) suggests that  $\Delta G/G$  on crystallization changes from 8% to 20%. This latter result can further be improved by considering local atomic distortions and local stresses.

### III. DISPERSION CURVES FOR AMORPHOUS METALS AND ALLOYS

The study of phonon dispersion curves (frequency  $\omega$  versus wave number  $q$ ) for amorphous metals has been subject of considerable interest [14], [15] of both theoretical and experimental investigations.

The dispersion curves exhibit certain broad features. Longitudinal phonons show oscillatory behavior with prominent maximum frequency. The position of the first frequency minimum roughly coincides with the first peak in the structure factor. On the other hand, the maximum frequency of the transverse phonons is smaller and appears at a larger  $q$  than that of the longitudinal phonons. The oscillatory behavior of the transverse phonons is quite insignificant beyond the first peak.

We can successfully explain the features of elastic waves in amorphous systems through the formalism of Section II. For simplicity, we assume that the system is in equilibrium under central forces alone ( $\beta = \kappa e = 0$ ). Equations (1) and (2) then simplify to

$$\omega_L^2 = \frac{2z\delta}{a^2\rho} I_2(qa) \quad (22)$$

$$\omega_T^2 = \frac{z\delta}{a^2\rho} [I_1(qa) - I_2(qa)] \quad (23)$$

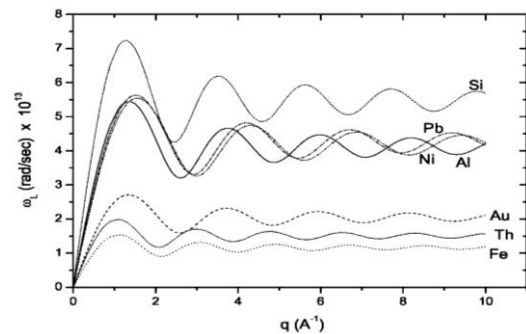


Fig. 1. Longitudinal frequency  $\omega_L$  vs.  $q$  for selected amorphous metals.

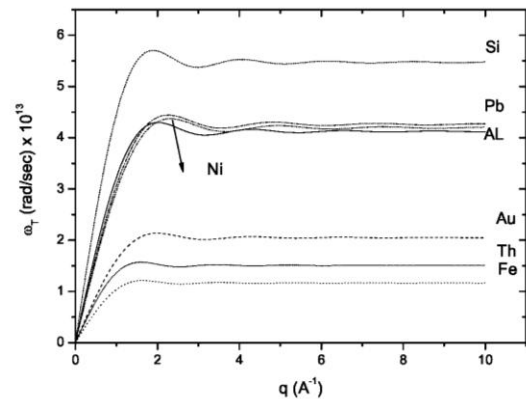


Fig. 2. Transverse frequency  $\omega_T$  vs.  $q$  for selected amorphous metals.

The values of  $z\delta$  can be obtained from the elastic constants

( $z\delta = 5\text{camp}11 = 15\text{camp}44$ ). The density  $\rho$  and the atomic mass  $M$  are used to calculate the atomic volume  $\Omega$ , which yields the nearest neighbor distance  $a$  ( $\Omega = 0.71a^3$ ). The computed values of  $\omega_L$  and  $\omega_T$  for selected amorphous metals (Al, Au, Fe, Ni, Si and Th) are plotted in Fig. 1 and Fig. 2, respectively. We observe that the  $\omega_L$ - $q$  curves exhibit oscillations whereas transverse phonons ( $\omega_T$ - $q$  curves) hardly show any oscillations. For  $q \rightarrow \infty$ ,  $\omega_L \approx \omega_T$ . On the other hand, for  $q \rightarrow 0$ ,  $\omega_L = \sqrt{3}\omega_T$ , which is almost exactly the value obtained in computer simulations by Grest *et al.* [11] for central forces. The position of the first minimum in the  $\omega_L$ - $q$  curve is found to be in the region where the static structure factor for liquid metals [16] has its first maximum. Though the nature of oscillations of  $\omega_L$  and  $\omega_T$  are similar for the different metals considered here, the magnitudes of the maxima differ considerably. The maximum value of  $\omega_L$  and  $\omega_T$  are found for amorphous Si followed by Pb, Ni, Al, Au, Th and Fe. The values of  $\omega_L$  and  $\omega_T$  in Si are almost 4.8 times higher than those of Fe.

A connection of elastic properties to thermal properties can be made through Debye theory. The long wavelength vibrational contribution to the heat capacity at low temperature,  $T$ , is given as

$$c_V = \frac{2\pi^2 k_B^4 T^3}{5\hbar^3 U_{av}^3} = \frac{12\pi^4 k_B n}{5} \left( \frac{T}{\Theta_{ampD}} \right)^3 \quad (24)$$

where  $k_B$  is the Boltzmann constant,  $\hbar = h/2\pi$  ( $h$  is Plank's constant) and  $n$  the number density ( $N/\Omega$ ). Equation (24), after some simplification, yields,

$$\Theta_{ampD} = 251.4 \left( \frac{\rho}{M} \right)^{1/3} U_{av} \quad (25)$$

with

$$U_{av} = \left[ \frac{1}{3} (v_L^{-3} + 2v_T^{-3}) \right]^{-1/3} \quad (26)$$

In (25),  $\rho$  is expressed in  $\text{g cm}^{-3}$ ,  $M$ , the atomic mass, in  $\text{g mol}^{-1}$ , and  $v_L$  and  $v_T$  in  $\text{km s}^{-1}$ .  $v_L$  and  $v_T$  can be obtained from the elastic constants  $c_{amp11}$  and  $c_{amp44}$ , respectively, using (10) and (9) and are tabulated in Table I.  $v_L$  and  $v_T$  are found to be maximum for Si ( $v_L = 9115 \text{ m s}^{-1}$  and  $v_T = 5501 \text{ m s}^{-1}$ ) and minimum for Pb ( $v_L = 2227 \text{ m s}^{-1}$  and  $v_L = 945 \text{ m s}^{-1}$ ). The computed values of  $\Theta_{ampD}$  from (25) are also tabulated in Table 1. For sake of comparison, we listed the Debye temperature,  $\Theta_{lmD}$ , computed [17] for liquid metals at melting temperature.  $\Theta_{cD}$  for crystalline phase [18] is also tabulated. It may be noted that the Debye temperature for amorphous metals are lower than the corresponding liquid and crystalline phases.

The present approach can also be applied to amorphous binary metal alloys. As an example, we have presented the results of  $\omega_L$  and  $\omega_T$  for ZrCo and Zr<sub>67</sub>Ni<sub>33</sub> in Fig. 3 and Fig. 4, respectively. For ZrCo, the force constant  $\delta$  has been obtained by using the experimental values [19]  $\rho = 7.6 \text{ g cm}^{-3}$ ,  $c_{amp44} = 49.125 \text{ GPa}$  and  $a = 0.3201 \text{ nm}$ . For Zr<sub>67</sub>Ni<sub>33</sub>,  $\delta$  is determined from the experimental and molecular dynamics simulation results [20], [21]  $v_L = 4800 \text{ m s}^{-1}$  and  $\rho = 7.06 \text{ g}$

$\text{cm}^{-3}$ . Phonon frequencies  $\omega_L$  and  $\omega_T$  are higher in Zr<sub>67</sub>Ni<sub>33</sub> than that of ZrCo. Likewise the amorphous metals, alloys also exhibit oscillations in  $\omega_L$  but no substantial oscillations in  $\omega_T$  after the first peak.

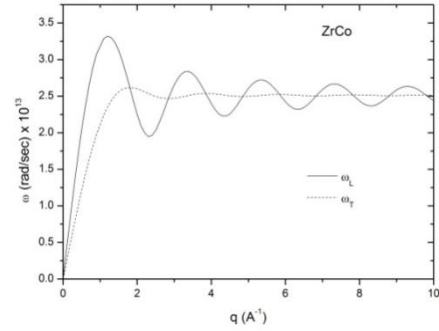


Fig. 3. Phonon curves for ZrCo alloy.

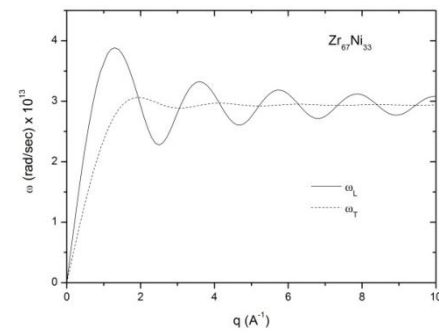


Fig. 4. Phonon curves for Zr67Ni33 alloy.

#### IV. CONCLUSION

A dispersion relation based on central pairwise potential for one-component amorphous metals is used to investigate the elastic moduli. The long range wavelength limit ( $q \rightarrow 0$ ) of the dispersion relation connects the elastic moduli to force constants and the velocities of longitudinal and transverse phonons in the amorphous system. It successfully explains the essential features of dispersion relations and the change of elastic moduli on amorphization as observed from experimental and simulation studies. The asymptotic ( $q \rightarrow \infty$ ) values of the phonon frequencies [ $\omega_L(q \rightarrow \infty) = \omega_T(q \rightarrow \infty)$ ] are quite different from one metal to another.

But, in the long wavelength limit ( $q \rightarrow 0$ ),  $\omega_L(q \rightarrow 0) = \sqrt{3}\omega_T(q \rightarrow 0)$ . The Debye temperature  $\Theta_{ampD}$  computed for a variety of metals are lower on average by 40% from the  $\Theta_{cD}$  of the crystalline phase. However,  $\Theta_{ampD}$  are lowered by about 6% to the values of  $\Theta_{lmD}$  of the liquid metals near the melting temperatures, with few exceptions like Cu, Na and Pb.  $\omega_L$ ,  $\omega_T$  and their asymptotic values are higher in Zr<sub>67</sub>Ni<sub>33</sub> than ZrCo. A further improvement is possible by including detailed electronic contributions with the proper exchange and correlation effects.

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