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LATTICE DYNAMICS OF METALLIC GLASS $Ca_{70}Mg_{30}$ ON THE MODEL OF BHATIA AND SINGH

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The lattice dynamical studies of the metallic glass $Ca_{70}Mg_{30}$ by Bhatia and Singh on their model contained two shortcomings, firstly the electron–ion interaction matrix was wrong and secondly, the numerical value of the bulk modulus of the electron gas was accepted arbitrarily. By modifying the electron–ion dynamical matrix and determining all the model parameters from the experimental data, we made a fresh study of the lattice dynamics of $Ca_{70}Mg_{30}$ and compared it to the earlier studies of Bhatia and Singh and also with experimental phonons.

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1. Introduction

In recent past there has been an extensive interest in the experimental [1, 2] and theoretical [3–8] studies of lattice dynamics of metallic glasses. As concerns experiment, the dynamical structure factor, $S(q, w)$, usually predicts the longitudinal phonons dispersion relation. Suck et al. [1], in a study of the experimental dynamical structure factor of $Mg_{70}Zn_{30}$, have indeed obtained information on transverse modes too. On the other hand, the theoretical studies predict both the longitudinal and transversal phonons. Out of all the models of metallic glasses developed so far, three of them, namely that of Takeno and Goda [6], Hubbard and Beeby [7] and that of Bhatia and Singh [8] are based on similar grounds i.e. quasicrystalline picture or on liquid-like collective behavior. While the models of Hubbard and Beeby [7] and that of Takeno and Goda [6] are quite general and are based on long-range interatomic interactions, the model of Bhatia and Singh [8] considers the interatomic interaction effective between first nearest neighbours.

The model of Bhatia and Singh [8] is a very simple and an ad hoc model of metallic glass. This model is an extension of the model of cubic metals developed

by Bhatia [9]. In order to describe the picture of a metallic glass in Bhatia's model of metals, Bhatia and Singh [8] considered that a two-component metallic glass, for example $Ca_{70}Mg_{30}$, is virtually a binary metallic alloy Ca_xMg_{1-x}. This model thus took account of the great mass difference between Ca and Mg atoms. Bhatia and Singh [8], like Bhatia [9] for cubic metals, retained the interionic interactions in metallic glass effective between the first nearest neighbours giving rise to two model parameters, β and δ. The electron–ion interaction was considered to be the same as that used in metals. Such an interaction resulted in another model parameter, k_e, the bulk modulus of the electron gas. Bhatia and Singh have applied their model to study phonons in metallic glass $Ca_{70}Mg_{30}$. This model was adopted to study the phonons in metallic glasses $Mg_{70}Zn_{30}$, $Cu_{57}Zr_{43}$ and $Pd_{77.5}Si_{16.5}Cu_6$ by Agarwal and Kachhava [10, 11].

On going through the work of Bhatia and Singh [8], we noticed two drawbacks in their model:

1. The development of the electron–ion dynamical matrix is erroneous. They left one important function to be multiplied in this matrix.
2. They chose arbitrarily the model parameter k_e in order to determine the remaining two parameters, β and δ, from two experimental data. This in turn gave a doubt of validity of all three model parameters.

We have thus made a revised study of the lattice dynamics of metallic glass $Ca_{70}Mg_{30}$ in the model of Bhatia and Singh [8] after incorporating the requisite modifications. In Sec. 2 we present the modified form of the electron–ion dynamical matrix. In Sec. 3 we present a new scheme of determining model parameters. In Sec. 4 we present the numerical results of the phonon dispersion relations and comparison of them with the theoretical works of Hafner [5] and Bhatia and Singh [8] and with the experimental phonons from the work of Suck et al. [1, 2]. In that section we also present important comments and conclusions.

2. Modified form of the electron–ion dynamical matrix

Bhatia and Singh [8] wrote the expression for the electron–ion dynamical matrix for cubic metals as given by

$$D_{\alpha\alpha}^{i-e}(q) = \frac{(4\pi n_i n_e z e^2) q^2}{q^2 + k_{TF}^2}. \tag{1}$$

In Eq. (1), n_i, n_e are the ionic and electronic number density, z is the valence, e is the electronic charge, q is the phonon wave vector, k_{TF} is the Thomas–Fermi wave vector.

Utilizing Eq. (8) of their paper for the definition of k_e, we have

$$k_e = \frac{4\pi n_i n_e z e^2}{k_{TF}^2}. \tag{2}$$

With the help of Eq. (2), Eq. (1) is transformed to

$$D_{\alpha\alpha}^{i-e}(q) = \frac{k_e k_{TF}^2 q^2}{q^2 + k_{TF}^2}. \tag{3}$$

By dividing numerator and denominator of Eq. (3) by k_{TF}^2, we have

$$D_{\alpha\alpha}^{\mathrm{i-e}}(q) = \frac{k_{\mathrm{e}}q^2}{1+q^2/k_{\mathrm{TF}}^2}. \tag{4}$$

Equation (4) of this paper is the same as that given by Shukla and Salzberg [12, 13] for the modified Bhatia model [8] for cubic metals. The expression for the electron–ion interaction matrix in the original model of Bhatia [13] was quite cumbersome. It should be noted that Eq. (4) is valid for Thomas–Fermi screening, i.e. when one uses the dielectric screening of Thomas–Fermi given by

$$\varepsilon_{\mathrm{TF}}(q) = 1 + \frac{k_{\mathrm{TF}}^2}{q^2}. \tag{5}$$

When one applies the dielectric screening function of Lindhard [14] and that of Langer and Vosko [15], as is the case with the work of Bhatia and Singh [8], k_{TF}^2 should be replaced by $k_{\mathrm{TF}}^2\tilde{g}(q)$. Bhatia and Singh [8] have also pointed it out very clearly. Thus, Eq. (4) is replaced by

$$D_{\alpha\alpha}^{\mathrm{i-e}}(q) = \frac{k_{\mathrm{e}}q^2}{1+q^2/k_{\mathrm{TF}}^2\tilde{g}(q)} = \frac{k_{\mathrm{e}}k_{\mathrm{TF}}^2q^2\tilde{g}(q)}{q^2+k_{\mathrm{TF}}^2\tilde{g}(q)}. \tag{6}$$

A look at Eq. (12) of the paper of Bhatia and Singh [8] shows that it does not contain $\tilde{g}(q)$ term in the numerator. For $q \to 0$, $\tilde{g}(q) = 1$. Thus this term has no influence for $q \to 0$ but for higher q, $\tilde{g}(q)$ drops off rapidly and has a great influence in the numerical calculation. The multiplication of $G^2(qr_{\mathrm{s}})$ in Eq. (5) is the practice [16] very common in the study of metals and we would like to adhere to it.

3. New scheme of determining model parameters

The explicit expressions for the calculation of the longitudinal and transversal phonon frequencies are given by [8]

$$\rho\omega_{\mathrm{L}}^2 = \frac{2\nu}{a^2}(\beta I_0 + \delta I_2) + \frac{k_{\mathrm{e}}k_{\mathrm{TF}}^2q^2\tilde{g}(q)G^2(qr_{\mathrm{s}})}{q^2+k_{\mathrm{TF}}^2\tilde{g}(q)}, \tag{7}$$

$$\rho\omega_{\mathrm{T}}^2 = \left|\frac{2\nu}{a^2}\right|\left(\beta I_0 + \frac{1}{2}\delta(I_0 - I_2)\right) \tag{8}$$

where

$$\delta = \frac{\rho a^3}{2M}\frac{\mathrm{d}}{\mathrm{d}r}\left[\frac{1}{r}\frac{\mathrm{d}W}{\mathrm{d}r}\right], \qquad \beta = \frac{\rho a^2}{2M}\left[\frac{1}{r}\frac{\mathrm{d}W}{\mathrm{d}r}\right]$$

$$I_0 = 1 - \frac{\sin x}{x}, \qquad I_2 = \frac{1}{3} - \left(\frac{1}{x} - \frac{2}{x^3}\right)\sin x - \frac{2}{x^2}\cos x.$$

The expressions for δ, β, I_0 and I_2 are from the paper of Bhatia and Singh [8].

By expanding Eqs. (6) and (7) in $q \to 0$ limit, the explicit expressions for the longitudinal and transversal sound velocities, $v_{\mathrm{L}}(0)$ and $v_{\mathrm{T}}(0)$, are given by

$$\rho v_{\mathrm{L}}^2(0) = \nu\left(\frac{\beta}{3} + \frac{\delta}{5}\right) + k_{\mathrm{e}}, \tag{9}$$

$$\rho v_T^2(0) = \nu \left(\frac{\beta}{3} + \frac{\delta}{15} \right). \tag{10}$$

For metallic glasses, with special attention to $Ca_{70}Mg_{30}$ the only experimental data, available apart from phonon frequencies, are the sound velocities. Bhatia and Singh [8] had thus no other choice than to determine three model parameters from Eqs. (9) and (10). Had they accepted the theoretical value of k_e from Eq. (2), they would have calculated two parameters, β and δ from Eqs. (9) and (10). Bhatia and Singh [8] were against this procedure so they accepted the value of k_e for $Ca_{70}Mg_{30}$ equal to 14.2×10^{10} dyn/cm^2 against the theoretical value of $k_e = 29.4 \times 10^{10}$ dyn/cm^2 calculated by Eq. (2). They argued this by stating: "k_e should be taken as a parameter and not the free electron value, in actual calculation a 50% variation of k_e from the chosen value does not affect the results significantly". We would ask a question: if one accepts arbitrarily the value of k_e as about half of that given by the free electron value and then substitutes this value of k_e to determine the remaining two parameters from Eqs. (9) and (10), are not all the model parameters adjusted arbitrarily?

What we suggest is to utilize one of the longitudinal phonon frequencies available from the experimental data and then use Eqs. (7), (9) and (10) to determine all the three parameters β, δ, and k_e correctly.

4. Numerical computations, comments and discussions

We tried several values of the experimental frequency ω_L for different wave vectors. We thus obtained several sets of the model parameters. Several of them predicted almost similar phonon dispersion relations.

We selected that value of model parameters which gave k_e close to the free electron value and gave a good fit with the experimental phonons.

TABLE

Input data and output values of force constants for metallic glass $Mg_{70}Zn_{30}$.

Input data	Output values of the model parameters		
	Parameters	Our calculations	Bhatia & Singh
$v_L(0) = 5.2 \times 10^5$ m s^{-1}	β [10^9 N m^{-2}]	1.91	0.7
$v_T(0) = 2.51 \times 10^5$ m s^{-1}	δ [10^9 N m^{-2}]	1.19	7.2
$n_i = 4.89 \times 10^{24}$ m^{-3}	k_e [10^9 N m^{-2}]	23.7	14.2
$\omega_L = 2.6 \times 10^{13}$ Hz			
$q = 1.8 \times 10^{10}$ m			

In Table are given the experimental input data and the output values of the force constants. Also there are given the values of parameters obtained by Bhatia and Singh [8].

In Fig. 1 there are plotted the computed dispersion relations of $Ca_{70}Mg_{30}$. In this figure there are also plotted the theoretical results obtained by Bhatia

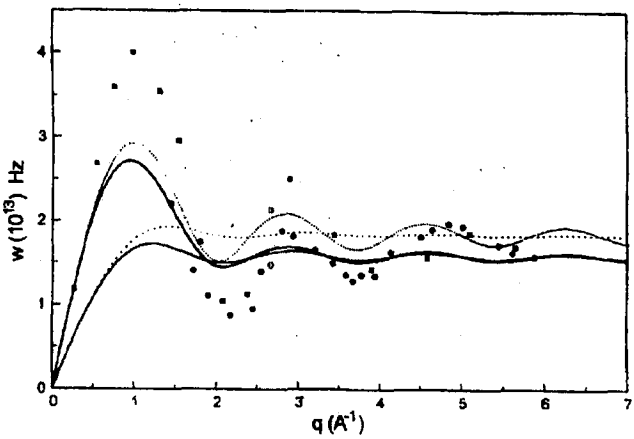

Fig. 1. Phonon dispersion relation in $Ca_{70}Mg_{30}$. Solid lines are our calculated results: □ □ □ experimental points; · · · Hafner's result; - - - Bhatia and Singh's result.

and Singh [8] without modifying the electron–ion dynamical matrix. In the same figure we also plotted the experimental phonons from the work of Suck et al. [1, 2] and the theoretical phonons from the work of Hafner [5]. A look at Fig. 1 reveals that our theoretical dispersion curves lie lower than that given by the work of Bhatia and Singh [8] and that of Hafner [5]. This shows that our results are in better agreement with the experimental longitudinal phonons in comparison with previous theoretical works. We can conclude saying that we determined all the three model parameters from the three experimental data. We also corrected the error introduced by Bhatia and Singh [8] in the electron–ion dynamical matrix.

We have achieved our basic aims:

1. We determined the corrected model parameters from the experimental data.
2. We corrected the electron–ion dynamical matrix used incorrectly by Bhatia and Singh [8] and all subsequent researchers who utilized their model.
3. We made a revised study of phonons in $Ca_{70}Mg_{30}$ on a new scheme obtaining better agreement with experiments than that given by Bhatia and Singh [8].

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