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Theoretical Investigation of Superconducting State Parameters of Binary Metallic Glasses $Cu_{1-c}Sn_c$

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Abstract

Superconducting state parameters (SSP) viz. electron-phonon coupling strength λ, Coulomb pseudopotential μ^*, transition temperature T_C, isotope effect exponent $\alpha$and effective interaction strength N_OV of seven $Cu_{1-c}Sn_c$ binary metallic glasses have been reported using Ashcroft's empty core (EMC) model potential for the first time. Five local field correction functions proposed by Hartree (H), Taylor (T), Ichimaru-Utsumi (IU), Farid et al. (F) and Sarkar et al. (S) are used in the present investigation to study the screening influence on the aforesaid properties. The T_C obtained from H-local field correction function are found in qualitative agreement with available experimental data and show linear nature with the concentration c of element Sn. A linear T_C equation is proposed by fitting the present outcomes for H-local field correction function, which is in conformity with other results for the experimental data. Also, the present results are found in qualitative agreement with other such earlier reported data, which confirms the superconducting phase in the metallic glasses.

Key Words: Pseudopotential; superconducting state parameters; binary metallic glasses.

1. Introduction

The field of electron correlation in condensed matter, especially superconductivity, is one of the dynamic areas in condensed matter physics which involves discoveries of new and existing phenomena, novel materials and devices for sophisticated technological applications. During the last few years, superconducting metallic glasses based on various simples as well as transition metals have been obtained and studied by various researchers. The study of the SSP of the metallic glasses may be of great help in deciding their applications; the study of the dependence of the transition temperature T_C on the composition of metallic glass is helpful in finding new superconductors with high T_C [1–19]. Though pseudopotential theory is found very successful in studying the various properties of the metallic glasses, there are very few scattered attempts to study the superconducting state parameters (SSP) of binary metallic glasses based on model potential [7–19]. The application of pseudopotential to a binary metallic glass involves the assumption of pseudoions with average properties, which are assumed to replace two types of ions in the binary systems, and a gas of free electrons is assumed to permeate through them. The electron-pseudoion is accounted for by the pseudopotential and the electron-electron interaction is involved through a dielectric screening function. For successful prediction of the superconducting properties of the metallic glasses, the proper selection of the pseudopotential and

screening function is very essential [3–19]. Very recently, Vora and co-workers [3–13] have studied the SSP of some metallic superconductors using single parametric model potential formalism.

Hence, the present work is a theoretical investigation of the SSP of seven $Cu_{1-c}Sn_c$ binary metallic glasses, specifically $Cu_{0.80}Sn_{0.20}$, $Cu_{0.61}Sn_{0.39}$, $Cu_{0.50}Sn_{0.50}$, $Cu_{0.40}Sn_{0.60}$, $Cu_{0.25}Sn_{0.75}$, $Cu_{0.16}Sn_{0.84}$ and $Cu_{0.10}Sn_{0.90}$, has been reported for the first time in the present study. Also, with copper being a good conductor that exhibits conditional superconducting nature, and with tin being a metallic element that also exhibits superconducting nature in normal condition, such class of glasses may be quite suitable for industrial applications. Hence, in the present article, we study the SSP viz. electron-phonon coupling strength λ, Coulomb pseudopotential μ^*, transition temperature T_C, isotope effect exponent $\alpha$and effective interaction strength $N_OV$of seven binary $Cu_{1-c}Sn_c$ (C =0.20, 0.39, 0.50, 0.60, 0.75, 0.84 and 0.90 at.%) metallic glasses on the basis of Ashcroft's empty core (EMC) potential [20].

The $Cu_{1-c}Sn_c$binary glass systems are based on conditional superconducting element and superconducting element of the periodic table. When they are combined together, they exhibit superconducting nature in the normal condition. In these systems, the transition temperature T_C roughly correspondence to a half filled d-band if the transition metals are taken to have singly s-like free electron, which is produced maximum range of the transition temperature T_C i.e. 2–7 K.

2. Computational Methodology

The most exhaustive study of the relationship between microscopic theory and observed superconducting transition temperature T_C was made by McMillan [21]. His work was based on the Eliashberg gap equation [22], which are extensions of the original BCS theory [23]. The formulation of the McMillan [21] has been reanalyzed particularly for the case of the strong coupling superconductors, by Allen and Dynes [24]. For a theoretical estimation of the material properties λ and μ^*, the essential ingredients are the electron-ion pseudopotential, dielectric screening and the phonon frequencies. While the phonon spectra are available experimentally from the inelastic scattering of the neutrons for number of the metallic glasses, otherwise it can be calculated from the Debye temperature relation given by Butler [25]. Since the theoretical prediction of the SSP depends on the appropriate representation of the electron-ion interaction potential, it is interesting to study the pseudopotential dependence of the electron-phonon coupling strength parameterλ and hence of T_C. According the McMillan [21], the electron-phonon coupling strength is given by

$$\lambda = 2\int_0^\infty d\omega \left[\alpha^2(\omega) F(\omega)/\omega\right], \tag{1}$$

where $\alpha^2(\omega) F(\omega)$ is the spectral function, which when appropriately evaluated in the plane wave approximation for the scattering on the Fermi surface yields [3–19],

$$\lambda = \frac{m_b\,\Omega_0}{4\pi^2\,k_F\,M\,\langle\omega^2\rangle}\int_0^{2k_F} q^3\,|V(q)|^2\,dq. \tag{2}$$

Here, m_b is the band mass, $M$the ionic mass, Ω_O is the atomic volume, k_F is the Fermi wave vector, $V(q)$ is the screened pseudopotential and $\langle\omega^2\rangle$ is the averaged square phonon frequency, of the binary glassy alloy. The averaged square phonon frequency $\langle\omega^2\rangle$ is calculated using the relation given by Butler [25], $\langle\omega^2\rangle^{1/2} = 0.69\,\theta_D$, where θ_D is the Debye temperature of the metallic glasses.

Using $X = q/2k_F$ and $\Omega_O = 3\pi^2 Z\Big/(k_F)^3$, we get equation (2) in the form,

$$\lambda = \frac{12m_bZ}{M\langle\omega^2\rangle}\int_0^1 x^3\,|W(X)|^2\,dX, \tag{3}$$

where Z and $W(X)$ are the valence of the metallic glasses and the screened Ashcroft's empty core pseudopotential [20] for binary mixture, respectively.

The well known screened Ashcroft's empty core (EMC) model potential [20] used in the present computations of the SSP of binary metallic glasses is of the form

$$W(X) = \frac{-2\pi Z}{\Omega_O X^2 k_F^2 \varepsilon(X)} \cos(2k_F X r_C), \tag{4}$$

where r_C is the parameter of the model potential of binary metallic glasses. The Ashcroft's empty core (EMC) model potential is a simple one-parameter model potential [22], which has been successfully found for various metallic complexes [9–19]. When used with a suitable form of dielectric screening functions, this potential has also been found to yield good results in computing the SSP of metallic glasses [9–19]. Therefore, in the present work we use the Ashcroft's empty core (EMC) model potential with Hartree (H) [26], Taylor (T) [27], Ichimaru-Utsumi (IU) [28], Farid et al. (F) [29] and Sarkar et al. (S) [30] local field correction functions for the first time. The model potential parameter r_C may be obtained by fitting either to some experimental data or to realistic form factors or other data relevant to the properties to be investigated. In the present work, r_C is fitted in such a way that, the presently computed values of the transition temperature T_C of the metallic glasses obtained from all local field correction functions are found as close as possible with the experimental data of T_C for metallic glasses whichever are available in the literature [31]. After fitting the model potential parameter r_C, the same r_C is then used in the computation of the SSP of binary metallic glasses.

The Coulomb pseudopotential μ^* is given by [3–19]

$$\mu^* = \frac{\frac{m_b}{\pi k_F} \int_0^1 \frac{dX}{\varepsilon(X)}}{1 + \frac{m_b}{\pi k_F} \ln\left(\frac{E_F}{10\,\theta_D}\right) \int_0^1 \frac{dX}{\varepsilon(X)}}, \tag{5}$$

where E_F is the Fermi energy; m is the mass of the electron; θ_D is the Debye temperature; and $\varepsilon(X)$ is the modified Hartree dielectric function, which is written as [26]

$$\varepsilon(X) = 1 + (\varepsilon_H(X) - 1)\ (1 - f(X)). \tag{6}$$

Here, $\varepsilon_H(X)$ is the static Hartree dielectric function [26] and the expression of $\varepsilon_H(X)$ is given by [26]

$$\varepsilon_H(X) = 1 + \frac{m e^2}{2\pi k_F \hbar^2 \eta^2} \left(\frac{1-\eta^2}{2\eta} \ln \left| \frac{1+\eta}{1-\eta} \right| + 1 \right)\ ;\ \eta = \frac{q}{2k_F}, \tag{7}$$

while $f(X)$ is the local field correction function. In the present investigation, the local field correction functions due to H [26], T [27], IU [28], F [29] and S [30] are incorporated to see the impact of exchange and correlation effects. The details of all the local field corrections are below.

The H-screening function [26] is purely static, and it does not include the exchange and correlation effects and is thus expressed as

$$f(X) = 0. \tag{8}$$

Taylor (T) [27] has introduced an analytical expression for the local field correction function, which satisfies the compressibility sum rule exactly. This is the most commonly used local field correction function and covers the overall features of the various local field correction functions proposed before 1972. According to Taylor (T) [27],

$$f(X) = \frac{q^2}{4k_F^2} \left[1 + \frac{0.1534}{\pi k_F^2} \right]. \tag{9}$$

The Ichimaru-Utsumi (IU) local field correction function [28] is a fitting formula for the dielectric screening function of the degenerate electron liquids at metallic and lower densities, which accurately reproduces the

Monte-Carlo results as well as it also, satisfies the self consistency condition in the compressibility sum rule and short range correlations. The fitting formula is

$$f(X) = A_{IU}Q^4 + B_{IU}Q^2 + C_{IU} + \left[A_{IU}Q^4 + \left(B_{IU} + \frac{8\,A_{IU}}{3}\right)Q^2 - C_{IU}\right]\left\{\frac{4-Q^2}{4Q}\ln\left|\frac{2+Q}{2-Q}\right|\right\}. \quad (10)$$

On the basis of Ichimaru-Utsumi (IU) local field correction function [28], Farid et al. (F) [29] have given a local field correction function of the form

$$f(X) = A_FQ^4 + B_FQ^2 + C_F + \left[A_FQ^4 + D_FQ^2 - C_F\right]\left\{\frac{4-Q^2}{4Q}\ln\left|\frac{2+Q}{2-Q}\right|\right\}. \quad (11)$$

Based on equations (10) and (11), Sarkar et al. (S) [30] have proposed a simple form of local field correction function of the form

$$f(X) = A_S\left\{1 - \left(1 + B_SQ^4\right)\exp\left(-C_SQ^2\right)\right\}, \quad (12)$$

where $Q = 2X$. Parameters A_{IU}, B_{IU}, C_{IU}, A_F, B_F, C_F, D_F, A_S, B_S and C_S are the atomic volume dependent parameters of IU, F and S-local field correction functions. The mathematical expressions of these parameters are narrated in the respective papers of the local field correction functions [28–30].

After evaluating λ and μ^*, the transition temperature T_C and isotope effect exponent α are investigated from the McMillan's formula [3–19, 26]:

$$T_C = \frac{\theta_D}{1.45}\,exp\left[\frac{-1.04\,(1+\lambda)}{\lambda - \mu^*\,(1+0.62\lambda)}\right], \quad (13)$$

$$\alpha = \frac{1}{2}\left[1 - \left(\mu^*\,\ln\frac{\theta_D}{1.45T_C}\right)^2\frac{1+0.62\lambda}{1.04\,(1+\lambda)}\right]. \quad (14)$$

The effective interaction strength $N_OV$represents combined effect of the electronic density of states at the Fermi surface and pairing potential arising from the electron-phonon interaction, respectively [32]. The expression for the effective interaction strength N_OV is given by [2–12, 32]

$$N_OV = \frac{\lambda - \mu^*}{1 + \frac{10}{11}\lambda}. \quad (15)$$

3. Results and Discussion

The values of the input parameters relevant to the pure components of the $Cu_{1-c}Sn_c$ metallic glassy system are narrated in Table 1. The input parameters of the pure metallic components are taken from the literature [33, 34]. The values of the input parameters for the $Cu_{1-c}Sn_c$ metallic glasses under investigation are obtained from the relevant values for the two components by using pseudo-alloy-atom (PAA) model, the following definitions are adopted [3–13] and assembled in Table 1,

$$Z = (1-C)(Z_A) + C\,(Z_B), \quad (16)$$

$$M = (1-C)(M_A) + C\,(M_B), \quad (17)$$

$$\Omega_O = (1-C)(\Omega_{OA}) + C\,(\Omega_{OB}), \quad (18)$$

$$\theta_D = (1-C)(\theta_{DA}) + C\,(\theta_{DB}), \quad (19)$$

where A and B are denoted the first (Cu) and second (Sn) pure metallic components and c is the fractional concentration of the second metallic component.

Table 1. Input parameters and other constants.

Metallic glass	Z	r_C (au)	Ω_O (au)^3	M (amu)	θ_D (K)
$Cu_{0.80}Sn_{0.20}$	1.60	0.3931	100.08	74.58	313.40
$Cu_{0.61}Sn_{0.39}$	2.17	0.6723	119.41	85.05	285.28
$Cu_{0.50}Sn_{0.50}$	2.50	0.7278	130.61	91.12	269.00
$Cu_{0.40}Sn_{0.60}$	2.80	0.7517	140.79	96.63	254.20
$Cu_{0.25}Sn_{0.75}$	3.25	0.8083	156.06	104.91	232.00
$Cu_{0.16}Sn_{0.84}$	3.52	0.8563	165.22	109.87	218.68
$Cu_{0.10}Sn_{0.90}$	3.70	0.8647	171.32	113.18	209.80

The theoretical work related to the SSP of the $Cu_{1-c}Sn_c$ metallic glasses is not available in the literature so far. Nobody have reported the SSP of these metallic glasses either theoretically or experimentally, to our knowledge. Only Mizutani [31] has reported experimental transition temperature T_C of these systems. Therefore, we have followed his work in the present computation.

The graphical representation of the model potential parameter r_C with the concentration c of Sn are plotted in Figure 1. It is noticed from the Figure 1 that the linear nature is observed for the model potential parameter r_C with the concentration c of Sn. The graphical analyses of the SSP of $Cu_{1-c}Sn_c$ systems are plotted in the Figures 2–6 with the experimental data [31].

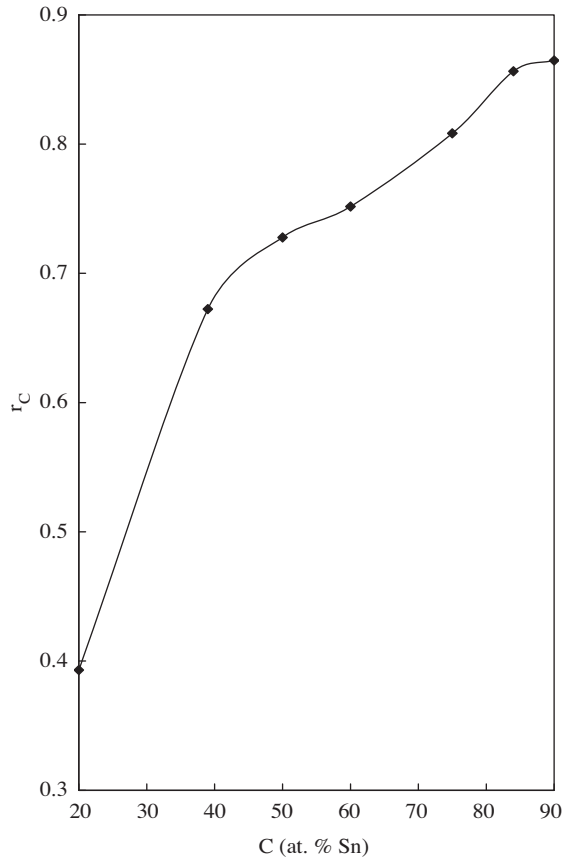

Figure 1. Variation of the model potential parameter r_C with Sn-concentration c (in at.%).

The calculated values of the electron-phonon coupling strength λ for seven $Cu_{1-c}Sn_c$ binary metallic glasses, using five different types of the local field correction functions with EMC model potential, are shown in Figure 2 with other experimental data [31]. It is noticed from the Figure 2 that, λ vales are quite sensitive

to the local field correction functions. It is noticed from the present study that, the percentile influence of the various local field correction functions with respect to the static H-screening function on the electron-phonon coupling strength λ is 23.98–63.33%. Also, the H-screening yields lowest values of λ, whereas the values obtained from the F-function are the highest. It is also observed from the Figure 2 that, λ goes on increasing from the values of 0.5255→1.2101 as the concentration c of Sn is increased from 20→90 at.%. The increase in λ with concentration c of Sn shows a gradual transition from weak coupling behaviour to intermediate coupling behaviour of electrons and phonons, which may be attributed to an increase of the hybridization of sp-d electrons of Sn with increasing concentration c. This may also be attributed to the increase role of ionic vibrations in the Sn-rich region. The theoretical data of the λ is not available for the further comparisons.

The computed values of the Coulomb pseudopotential μ^*, which accounts for the Coulomb interaction between the conduction electrons, obtained from the various forms of the local field correction functions are tabulated in Figure 3, which shows the weak dependence of μ^* on the local field correction functions. It is observed from the Figure 3 that for all metallic glasses, the μ^* lies between 0.1268 and 0.1561, which is in accordance with McMillan [21], who suggested $\mu^* \approx 0.13$ for transition metals. The percentile influence of the various local field correction functions with respect to the static H-screening function on μ^* for the metallic glasses is observed in the range of 4.79–10.87%. Again the H-screening function yields lowest values of theμ^*, while the values obtained from the F-function are the highest. Here also, as the concentration c of Sn increases the present results of μ^* decreases. The theoretical or experimental data of the μ^* is not available for the further comparisons.

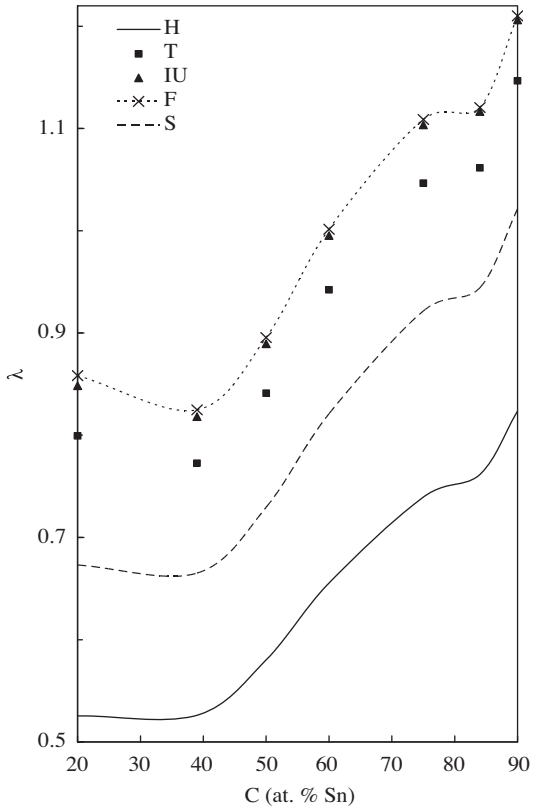

Figure 2. Variation of electron-phonon coupling strength λ with Sn-concentration c (in at.%).

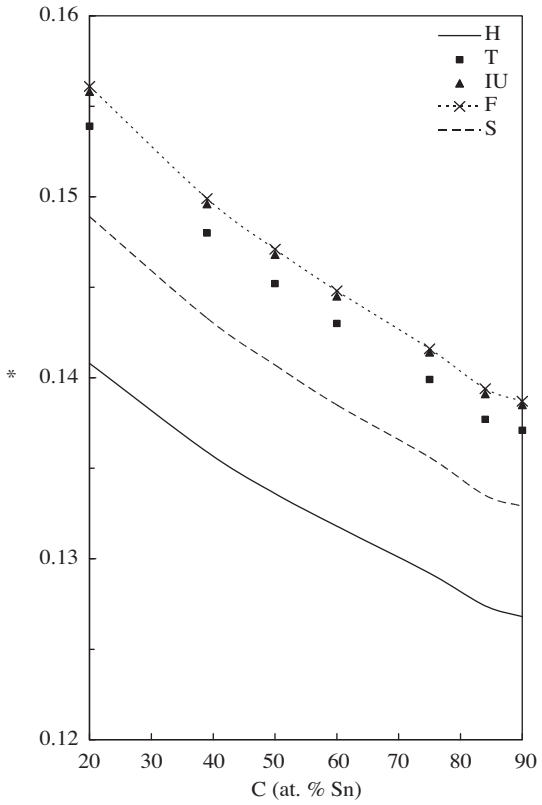

Figure 3. Variation of coulomb pseudopotential μ^* with Sn-concentration c (in at.%).

The variation of the computed values of the transition temperature T_C for $Cu_{1-c}Sn_c$ metallic glasses with the atomic concentration c of Sn, using five different types of the local field correction functions with EMC potential are shown in Figure 4. From the Figure 4 it can be noted that, the static H-screening function yields lowest T_C whereas the F-function yields highest values of T_C. The graph also includes the experimental values due to Mizutani [31]. It is seen that T_C is quite sensitive to the local field correction functions, and the results of T_C by using H-screening are found in qualitative agreement with the experimental data [31] for the $Cu_{1-c}Sn_c$ metallic glasses under investigation, as the relevant curves for H-screening almost overlaps the experimental curves. It is noticed from the present study that, the percentile influence of the various local field correction functions with respect to the static H-screening function on the electron-phonon coupling strength T_C is 47.17–376.35%. The calculated results of the transition temperature T_C for $Cu_{0.80}Sn_{0.20}$, $Cu_{0.61}Sn_{0.39}$, $Cu_{0.50}Sn_{0.50}$, $Cu_{0.40}Sn_{0.60}$, $Cu_{0.25}Sn_{0.75}$, $Cu_{0.16}Sn_{0.84}$ and $Cu_{0.10}Sn_{0.90}$ metallic glasses deviate in the range of 0.04–376.54%, 0.01–312.25%, 0.02–221.54%, 0.03–158.03%, 0.05–114.23%, 0.03–100.90% and 0.01–86.71% from the experimental findings [31], respectively. The theoretical data of the T_C is not available for the further comparisons.

It is also seen from the graphical nature, T_C increases linearly with increasing Sn-concentration c. The composition dependence can be described by linear regression of the data obtained for H-screening for different values of the concentration c, which yields

$$T_C\,(K) = 0.0811\,c - 0.4299. \tag{20}$$

The graph of the fitted T_C equation is displayed in Figure 4, which indicates that T_C drops and increases almost linearly with increasing Sn content with a slope $dT_C/dc = 0.0811$. Wide extrapolation predicts a $T_C = 0.4299$ K for the hypothetical case of amorphous pure Sn.

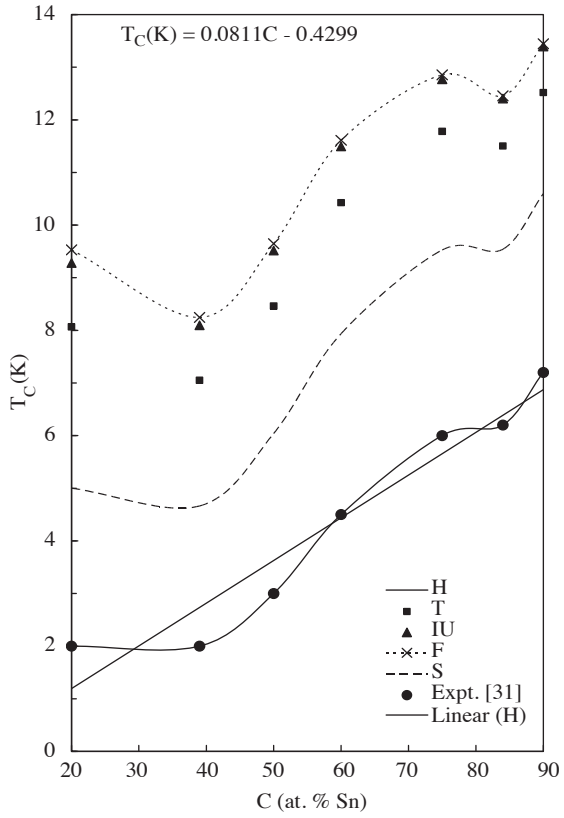

Figure 4. Variation of transition temperature T_C with Sn-concentration c (in at.%).

The presently computed values of the T_C are found in the range, which is suitable for further exploring the applications of the metallic glasses for usage like lossless transmission line for cryogenic applications. It is evident that the calculated λ values systematically increase with increase valence of the glassy systems. Also, the computed values of the T_C tend to increase with increase valence, indicating the general validity of the present theory. This is the important conclusion that can be drawn from the above discussion.

Figure 5 depicts the variation of α with Sn-concentration c increases (or as the Cu concentration decreases). The computed values of the α show a weak dependence on the dielectric screening, its value is being lowest for the H-screening function and highest for the F-function. Since the theoretical or experimental value of α has not been reported in the literature so far, the present data of α may be used for the study of ionic vibrations in the superconductivity of amorphous substances. Since H-local field correction function yields the best results for λ and T_C, it may be observed that α values obtained from this screening provide the best account for the role of the ionic vibrations in superconducting behaviour of this system. The most important feature noted here is that as the concentration c of Sn increases the present results of α increases sharply.

The values of the effective interaction strength $N_OV$are depicted in Figure 6 for different local field correction functions. It is observed that the magnitude of N_OV shows that the metallic glasses under investigation lie in the range of weak coupling superconductors. The values of the N_OV also show a feeble dependence on dielectric screening, its value being lowest for the H-screening function and highest for the F-screening function. The theoretical or experimental data of the N_OV is not available for the further comparisons. The variation of present values of the N_OV show that, the metallic glasses under consideration fall in the range of weak coupling superconductors. Here also, as the concentration c of Sn increases the present results of N_OV increases. The effective interaction strength N_OV represents combined effect of the electronic density of states at the Fermi surface, which is related to the Coulomb pseudopotential μ^*

and pairing potential arising from the electron-phonon interaction related to the electron-phonon coupling strength λ, respectively [32]. Hence, the graphical nature of the effective interaction strength N_OV is same as those of the electron-phonon coupling strength λ.

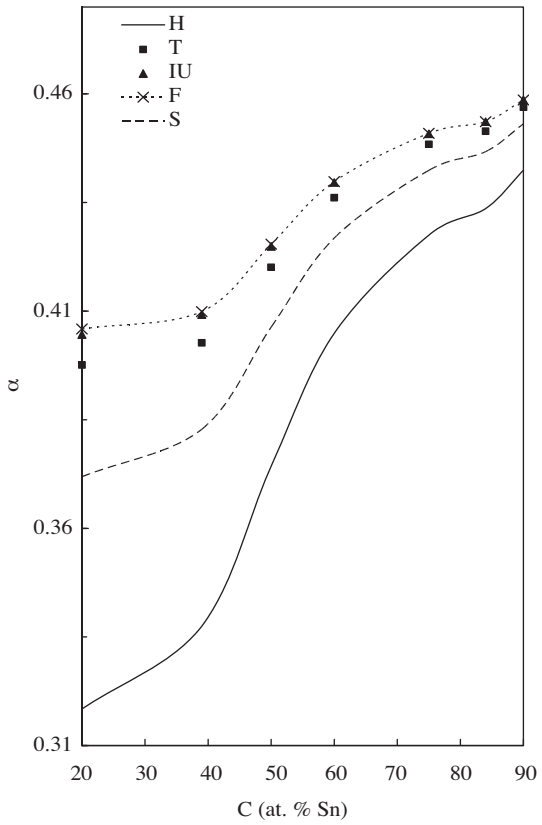

Figure 5. Variation of isotope effect exponent α with Sn-concentration c (in at.%).

The effect of local field correction functions plays an important role in the computation of λ andμ^*, which makes drastic variation on T_C, α and N_OV. The local field correction functions due to IU, F and S are able to generate consistent results regarding the SSP of the binary metallic glasses as those obtained from more commonly employed H and T functions. Thus, the use of these more promising local field correction functions is established successfully. The computed results of α and N_OV are not showing any abnormal values for $Cu_{1-c}Sn_c$ metallic glasses.

The main difference of the local field correction functions are played in important role in the production of the SSP of ternary metallic glasses. The Hartree (H) dielectric function [26] is purely static and it does not include the exchange and correlation effects. Taylor (T) [27] has introduced an analytical expression for the local field correction function, which satisfies the compressibility sum rule exactly. The Ichimaru-Utsumi (IU) local field correction function [28] is a fitting formula for the dielectric screening function of the degenerate electron liquids at metallic and lower densities, which accurately reproduces the Monte-Carlo results as well as it also satisfies the self consistency condition in the compressibility sum rule and short range correlations. Therefore, H-local field correction function gives the qualitative agreement with the experimental data [31] with EMC model potential and found suitable in the present case. On the basis of Ichimaru-Utsumi (IU) local field correction function [28], Farid et al. (F) [29] and Sarkar et al. [30] have given a local field correction function. Hence, F-function represents same characteristic nature. Also, the SSP computed from Sarkar et al. [30] local field correction are found in qualitative agreement with the available experimental data [31].

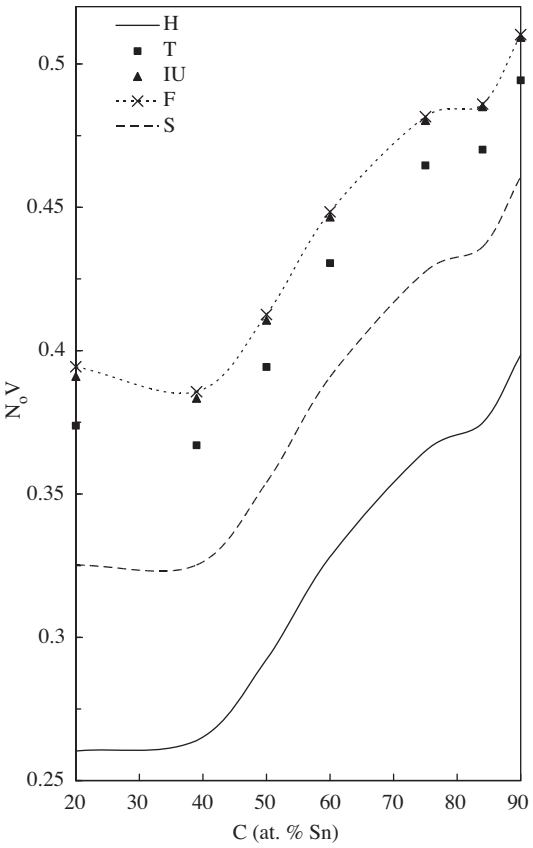

Figure 6. Variation of effective interaction strength (N_OV) with Sn-concentration c (in at.%).

According to Matthias' rules [35, 36], the metallic glasses having Z <2 do not exhibit superconducting nature. Hence, $Cu_{0.80}Sn_{20}$ metallic glass is expected to be non-superconducting, yet it exhibits superconducting nature in the present case. When we go from $Z = 1.60$ to $Z = 3.70$, the electron-phonon coupling strength λ changes with lattice spacing a. Similar trends are also observed in the values of T_C for most of the metallic glasses. Hence, a strong dependency of the SSP of the metallic glasses on the valence Z is found.

Lastly, we would like to emphasize the importance of involving a precise form for the pseudopotential. It must be confessed that although the effect of pseudopotential in strong coupling superconductor is large, yet it plays a decisive role in weak coupling superconductors i.e. those substances which are at the boundary dividing the superconducting and nonsuperconducting region. In other words, a small variation in the value of electron-ion interaction may lead to an abrupt change in the superconducting properties of the material under consideration. In this connection we may realize the importance of an accurate form for the pseudopotential.

4. Conclusions

Lastly we concluded that, the H-local field correction when used with EMC model potential provide the best explanation for superconductivity in the $Cu_{1-c}Sn_c$ system. The values of the electron-phonon coupling strength λ and the transition temperature T_C show an appreciable dependence on the local field correction functions, whereas for the Coulomb pseudopotential μ^*, isotope effect exponent $\alpha$and effective interaction strength N_OVa weak dependence is observed. The magnitude of the λ, $\alpha$and N_OV values shows that $Cu_{1-c}Sn_c$ metallic glasses are weak to intermediate superconductors. In the absence of theoretical or

experimental data for SSP, the presently computed values of these parameters may be considered to form reliable data for $Cu_{1-c}Sn_c$ metallic glasses, as they lie within the theoretical limits of the Eliashberg-McMillan formulation. The linear nature of T_C shows that, it decreases with increase of Sn content in $Cu_{1-c}Sn_c$ metallic glasses. It is also concluded that, the $Cu_{1-c}Sn_c$ metallic glasses are favoured superconductors. The comparisons of presently computed results of the SSP of $Cu_{1-c}Sn_c$ metallic glasses with available experimental findings are highly encouraging, which confirms the applicability of the EMC model potential and different forms of the local field correction functions.

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