

Theoretical evaluation of neutron scattering intensities, partial structure factors, and diffusion coefficients of the alloy of the peculiar metal Bi in Cu-Bi alloy

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(Received 28 April 1989)

Lebowitz's solution of $C_{ij}^0(r)$ for binary mixtures and a perturbation with a square-well attractive tail has been applied to compute three partial structure factors $S_{11}(k)$, $S_{22}(k)$, and $S_{12}(k)$ of molten Cu-Bi alloy, which shows some sort of inhomogeneities in the distribution of the constituents. The scattering intensities at various concentrations and at different temperatures obtained from these partial structure factors compare satisfactorily with the experimental results. From the partial structure factors, the Bhatia-Thornton structure factors $S_{NN}(k)$, $S_{CC}(k)$, and $S_{NC}(k)$ are computed for the Cu-Bi melts at various concentrations. It is found that in the case of this peculiar alloy the potential parameters are found to be concentration dependent to generate the scattering intensities. The compressibilities at various concentrations of Cu have been computed from the Kirkwood-Buff formula while the diffusion coefficients have been computed from Helfand's linear-trajectory principle for mixtures.

I. INTRODUCTION

In recent years structural studies of binary liquid alloys were extended to systems showing some peculiar structural properties.^{1,2} These can be divided into two groups. The first group exhibits some short-range order in the liquid state due to its compound-forming nature—such as Mg-Sn (Ref. 3) and Li-Pb (Ref. 4) systems. The second group, such as Al-Sn,⁵ Al-In,^{6,2} and Na-Li,⁷ shows a tendency to segregation. So far model calculations have been made in simple systems while recently an attempt has been made where inhomogeneities exist in the distribution of the constituents.

The copper-bismuth liquid alloy consisting of the peculiar metal bismuth, which shows a conspicuous right-hand shoulder in the structure factor⁸ in the pure state, has been extensively studied by several authors.^{2,9–12} The volume of mixing of this alloy in the concentration range 30–70 at. % Cu differ from the ideal volume of mixing.¹³ From the structure factor,^{2,10} magnetic susceptibility,¹⁴ electrical resistivity,¹⁴ and ultrasonic studies Sauerwald¹² and Steeb *et al.*² pointed out that molten Bi-Cu alloys have a tendency to segregation.

As is well known, the determination of the partial structure factors of a binary alloy is experimentally difficult.^{15,16} In the case of a binary alloy three partial structure factors have to be evaluated while the experimental data that is generally available is from neutron and x-ray scattering only. Sometimes it is possible to get a different set of data by enriching with different isotopes of the metals, as different isotopes have different scattering lengths but complete enrichment is not always easy. Hence model calculations in statistical mechanics offer a way to obtain the three partial structure factors independently and a suitable model may be able to predict to a good accuracy the structure factors, scattering intensities, and other related properties. It has been shown^{17,18}

that the structural properties of liquid metals can be explained satisfactorily on the basis of a simple perturbation treatment of the Percus-Yevic (PY) equation with a square-well attractive tail. Already this treatment has been extended to the calculation of partial structure factors and scattering intensities for the simple liquid alloys.^{19,20} Presently it is applied to Cu-Bi alloy, which shows some sort of inhomogeneity in the distribution of the constituents of the alloy. The partial structure factors can also be used fruitfully to obtain the Bhatia-Thornton correlation functions, which in turn are related to various thermodynamic properties²¹ and the diffusion coefficient²² calculations through the use of Helfand's linear-trajectory approximation. It has been pointed out by McDonald and Klein²³ that the potential parameters can be density dependent. In the present case it is found that with the variation of the concentration of the peculiar metal Bi, the potential parameters have to be varied slightly to generate accurate structure factors.

II. THEORY

As an extension of the MSM approximation²⁴ for square-well mixtures, we have, in the random-phase approximation (RPA),²⁵

$$C_{ij}(r) = \begin{cases} C_{ij}^0(r), & 0 < r < \sigma_{ij} \\ -\beta\Phi_{ij}(r) = \beta\epsilon_{ij}, & \sigma_{ij} \leq r \leq A_{ij}\sigma_{ij} \\ 0, & r > A_{ij}\sigma_{ij} \end{cases} \quad (1)$$

Here $\beta = 1/k_B T$ and $C_{ij}^0(r)$ is the hard-sphere solution of the Percus-Yevic equation for a binary mixture as obtained by Lebowitz.²⁶ σ_{ii} , ϵ_{ii} , and A_{ii} stand for the hard-sphere diameter, depth, and breadth of the square-well potential used for the hard-sphere species i , respectively. The mixed parameters have been determined through Lorentz-Berthlot mixing rules.²⁷

We use the Lebowitz solution of hard-sphere mixtures for $C_{ij}(r)$ in the region $0 < r < \sigma_{ij}$ and the well-known mean spherical model approximation in the region $\sigma_{ij} \leq r \leq A_{ij}\sigma_{ij}$ to get the direct correlation function (DCF). The various connected expressions and also their Fourier transformations have been given extensively,^{19,20,27} hence they will not be repeated here.

The partial structure factors $S_{ij}(k)$ can be written in terms of $C_{ij}(k)$ (Ref. 28) as

$$S_{11}(k) = \{1 - \rho_{11}\hat{C}_{11}(k) - \rho_{11}\rho_{22}\hat{C}_{12}^2(k)/[1 - \rho_{22}\hat{C}_{22}(k)]\}^{-1}, \quad (2)$$

$$S_{22}(k) = \{1 - \rho_{22}\hat{C}_{22}(k) - \rho_{11}\rho_{22}\hat{C}_{12}^2(k)/[1 - \rho_{11}\hat{C}_{11}(k)]\}^{-1}, \quad (3)$$

$$S_{12}(k) = (\rho_{11}\rho_{22})^{1/2}\hat{C}_{12}(k) \times \{[1 - \rho_{11}\hat{C}_{11}(k)][1 - \rho_{22}\hat{C}_{22}(k)] - \rho_{11}\rho_{22}\hat{C}_{12}^2(k)\}^{-1}. \quad (4)$$

The total structure factor $S(k)$ in terms of the partial structure factors is obtained as²⁷

$$S(k) = \sum_{i=1}^2 \sum_{j=1}^2 (C_i C_j)^{1/2} \frac{f_i(k) f_j(k)}{C_1 f_1^2(k) + C_2 f_2^2(k)} S_{ij}(k). \quad (5)$$

From neutron diffraction the coherent scattering intensity $I_{\text{coh}}(k)$ is related to the total structure factor as follows:²⁷

$$S(k) = \frac{I_{\text{coh}}(k) - (\langle b^2 \rangle - \langle b \rangle^2)}{\langle b \rangle^2}. \quad (6)$$

Here

$$\langle b^2 \rangle = C_1 b_1^2 + C_2 b_2^2, \quad (7)$$

$$\langle b \rangle = C_1 b_1 + C_2 b_2, \quad (8)$$

where b_1 and b_2 are the coherent scattering lengths of species 1 and 2, while C_1 and C_2 are their atomic fractions.

The isothermal compressibility χ_T for the binary alloy is related to $C_{ij}(0)$ through the Kirkwood-Buff equation as²⁷

$$\rho \chi_T k_B T = [1 - C_1 \rho_{11} \hat{C}_{11}(0) - C_2 \rho_{22} \hat{C}_{22}(0) - 2\rho_{12} C_1 C_2 \hat{C}_{12}(0)]^{-1}. \quad (9)$$

The expressions for the long-wavelength limit $\hat{C}_{ii}(0)$ and $\hat{C}_{12}(0)$ have also been given already.^{19,20}

The Bhatia-Thornton²¹ correlation functions in terms of fluctuations in (i) the particle density $S_{NN}(k)$, (ii) in local concentration (in mole fraction) $S_{CC}(k)$, and (iii) in number and composition, $S_{NC}(k)$, are computed from the partial structure factors $S_{ij}(k)$.²⁷ The relevant equations are given below

$$S_{NN}(k) = C_1 S_{11}(k) + C_2 S_{22}(k) + 2(C_1 C_2)^{1/2} S_{12}(k), \quad (10)$$

$$S_{CC}(k) = C_1 C_2 [C_2 S_{11}(k) + C_1 S_{22}(k) - 2(C_1 C_2)^{1/2} S_{12}(k)], \quad (11)$$

$$S_{NC}(k) = C_1 C_2 [S_{11}(k) - S_{22}(k) + S_{12}(k)(C_2 - C_1)/(C_1 C_2)^{1/2}]. \quad (12)$$

III. RESULTS AND DISCUSSION

The molar volumes at various concentrations of Cu were determined by Gomez *et al.*¹³ and hence the exact number density of the melt is known. The input parameters used in the calculations are given in Table I and the coherent neutron scattering intensities of pure liquids are shown in Fig. 1 while those of the liquid alloys with 0.1–0.9 atomic fractions of Cu are shown in Figs. 2(a) and 2(b). A glance at Table I indicates that the potential parameters are concentration dependent. From the intensity measurements one can easily perceive the alloy [vide Figs. 2(a) and 2(b)] to be a peculiar one, especially in the range of 0.2–0.6 atomic fraction of Cu. In this connection it may be pointed out that Bi has different polymorphic forms in the solid state and perhaps this lingers even after the melting point. Hence the theoretical evaluation of partial structure factors becomes all the more important. One can observe from these figures a strong shoulder in between 2 and 3 \AA^{-1} . It is also impor-

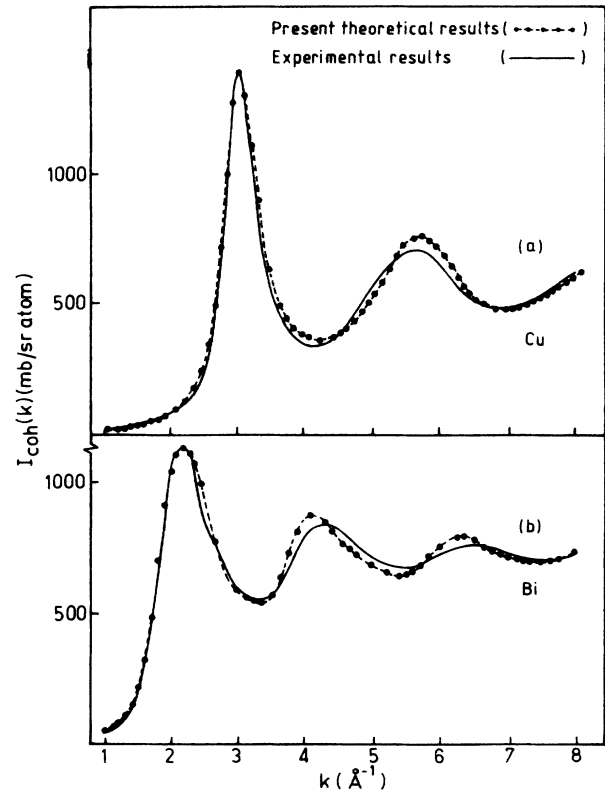


FIG. 1. Coherent scattering cross section $I_{\text{coh}}(k)$ vs k of (a) molten Cu at 1373 K, (b) molten Bi at 1073 K. Present theoretical results (\bullet) and experimental results (—).

TABLE I. Input data and potential parameters used in the evaluation of partial structure factors of Cu-Bi melt.

Temp. (K)	Atomic fraction of Cu	σ_{11} (Å)	σ_{22} (Å)	ϵ_{11}/k_B (K)	ϵ_{22}/k_B (K)	A_{11}	A_{22}
1073.0	0.00	2.21	3.00	300.0	640.0	1.70	1.90
870.0	0.10	2.21	2.95	300.0	600.0	1.70	1.87
973.0	0.20	2.21	2.86	300.0	660.0	1.70	1.88
1035.0	0.30	2.22	2.67	300.0	640.0	1.70	1.90
1080.0	0.40	2.21	2.60	300.0	640.0	1.75	1.90
1120.0	0.50	2.21	2.70	300.0	660.0	1.70	1.95
1160.0	0.60	2.28	2.80	300.0	640.0	1.65	1.92
1190.0	0.70	2.24	3.03	300.0	590.0	1.64	1.90
1200.0	0.80	2.26	3.08	300.0	590.0	1.60	1.90
1275.0	0.90	2.22	3.00	300.0	640.0	1.70	1.95
1373.0	1.00	2.22	3.00	300.0	640.0	1.70	1.90

tant to note that there is a right-hand shoulder in the concentration range of 0.2–0.4 atomic fraction of Cu while peculiarly the shoulder is observed on the left-hand side at concentrations of 0.5 and 0.6. Thus Bi, which is a peculiar metal by itself,⁸ is showing its effect on the scattering intensities. It is very gratifying to note that the present computations could bring sufficiently good agree-

ment between experiment and theory where we use the Lebowitz solution of hard-sphere mixtures. In the range of 0.2–0.6 atomic fraction of Cu where inhomogeneities are large, the potential parameters are found to change significantly to reproduce faithfully the intensities around the first peak. At low concentrations of Cu or high concentration of Bi (0.1 and 0.2 atomic fractions of Cu) the

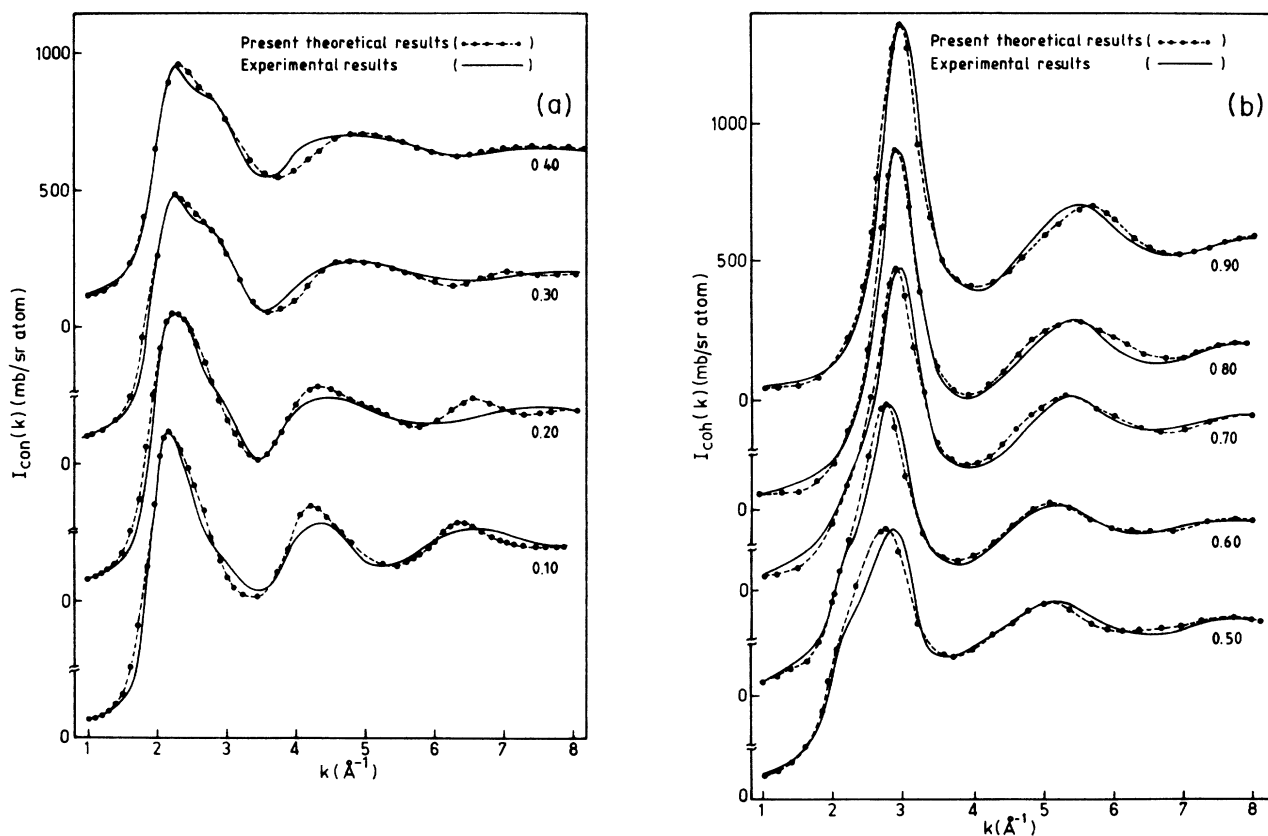


FIG. 2. (a) Coherent scattering cross section $I_{\text{coh}}(k)$ vs k for Cu-Bi melt for different atomic fractions of Cu. Present theoretical results (—●—) and experimental results (—). (b) Coherent scattering cross section $I_{\text{coh}}(k)$ vs k for Cu-Bi melt for different atomic fractions of Cu. Present theoretical results (—●—) and experimental results (—).

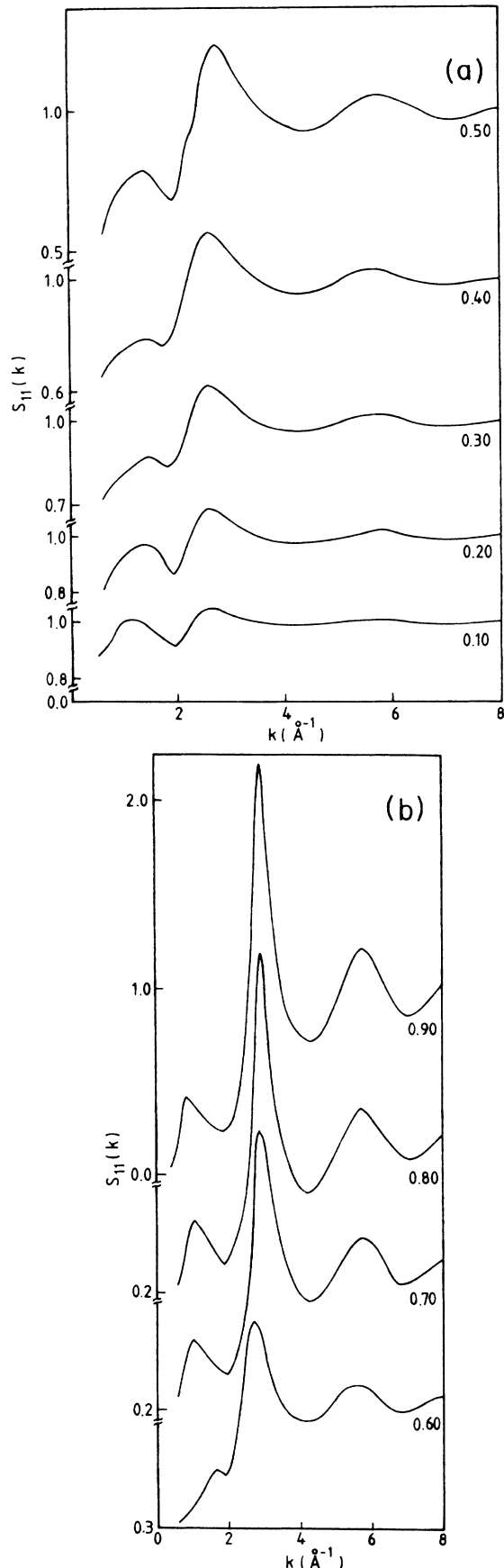


FIG. 3. (a) $S_{11}(k)$ vs k for different atomic fractions of copper. (b) $S_{11}(k)$ vs k for different atomic fractions of copper.

calculated second peak is observed to be slightly higher than the experimental value.

In the present calculations we give I_{coh} of pure Cu and Bi, and the associated parameters are not expected to be constant in this peculiar alloy. Further, it is important to observe that the parameters of Cu in general are found to be constant, as expected for a simple metal with a symmetric first peak. As the concentration of Cu increases, the first peak becomes symmetric. $S_{11}(k)$, the partial structure factor of copper, already shows a premaximum characteristic of a complex alloy containing the peculiar metal bismuth that perturbs the uniform distribution of Cu atoms in the momentum space. However, it may be observed that this premaximum broadens as the Bi content increases. Further, it may also be seen that as the concentration of Cu approaches that of pure metal the principal peak height approaches that of pure Cu, while at low concentrations as expected the height of the principal peak becomes very small [vide Figs. 3(a) and 3(b)].

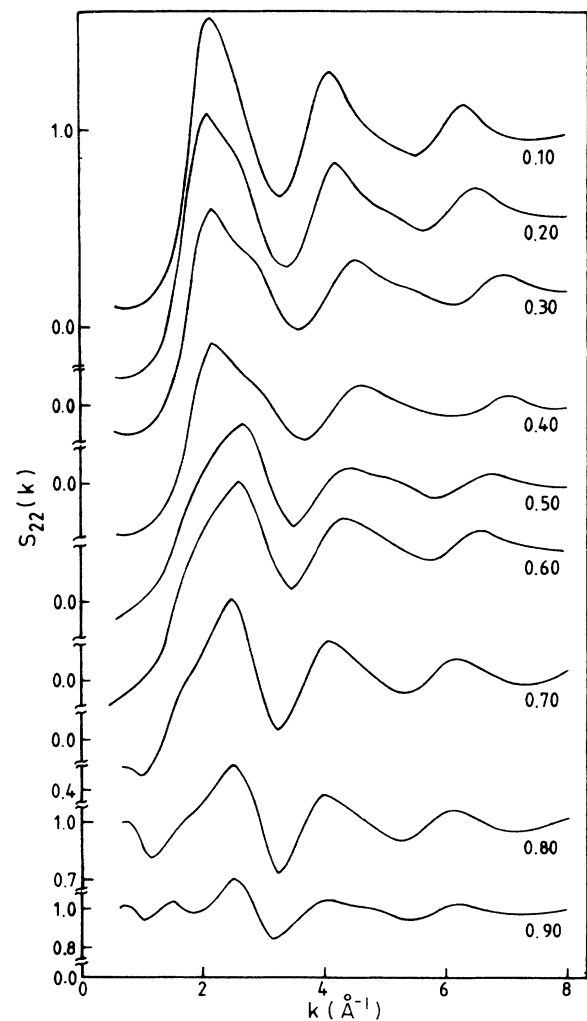


FIG. 4. $S_{22}(k)$ vs k for different atomic fractions of copper.

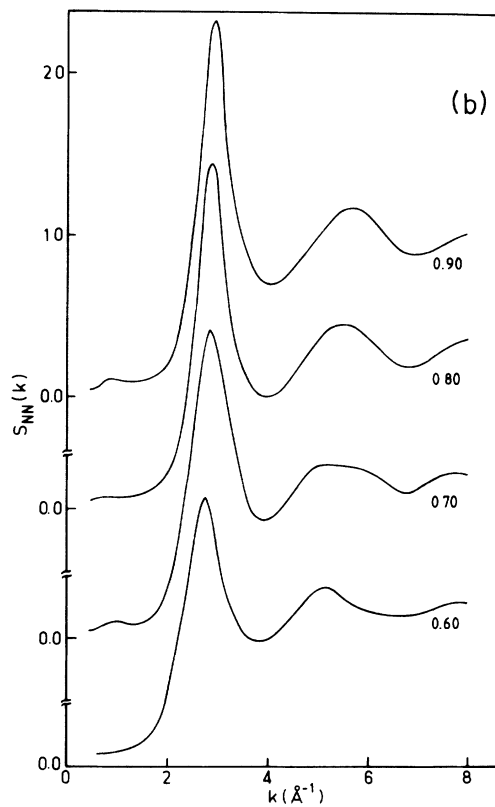
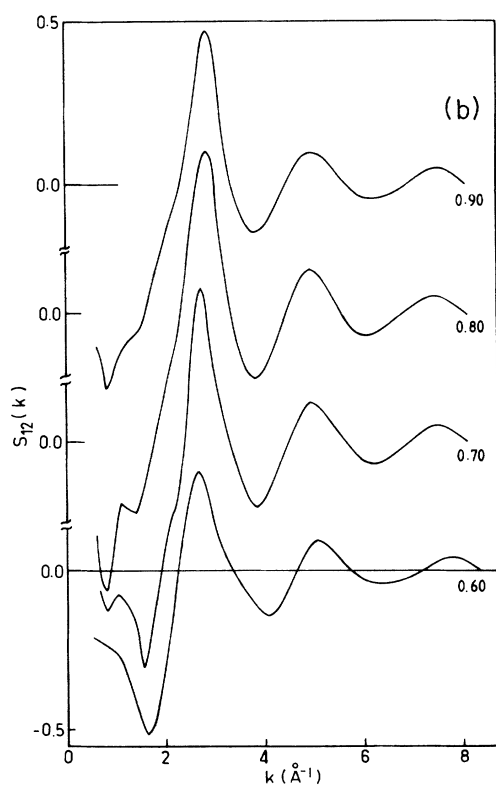
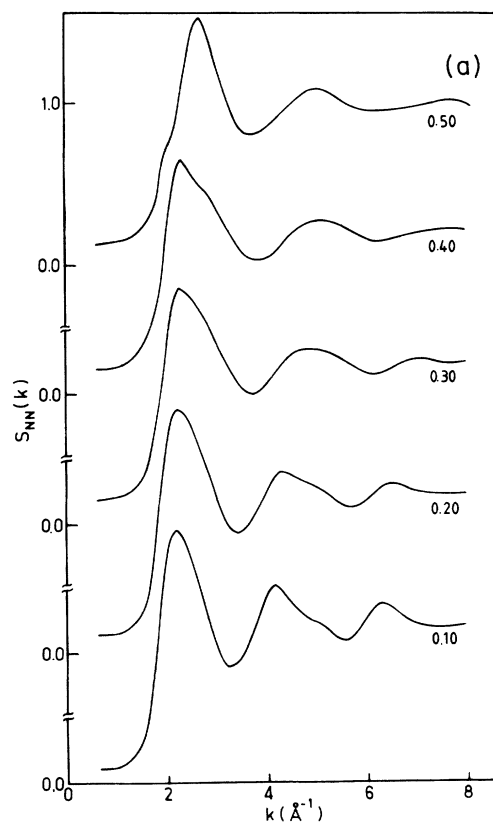
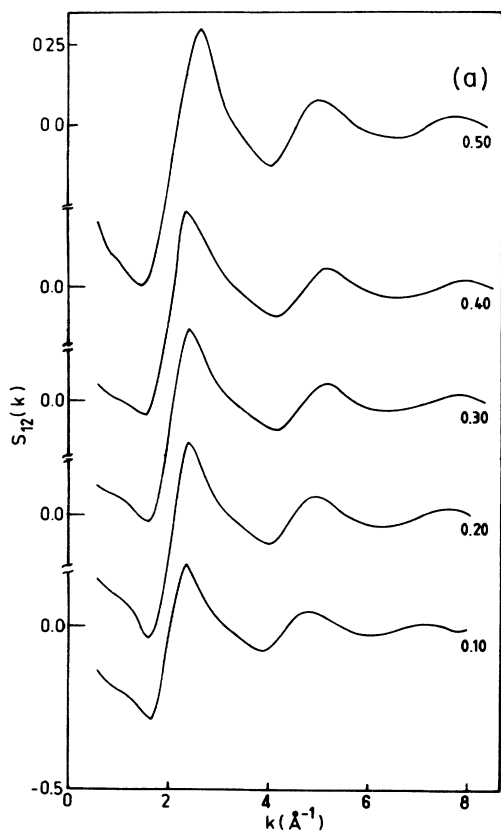


FIG. 5. (a) $S_{12}(k)$ vs k for different atomic fractions of copper. (b) $S_{12}(k)$ vs k for different atomic fractions of copper.

FIG. 6. (a) $S_{NN}(k)$ vs k for different atomic fractions of copper. (b) $S_{NN}(k)$ vs k for different atomic fractions of copper.

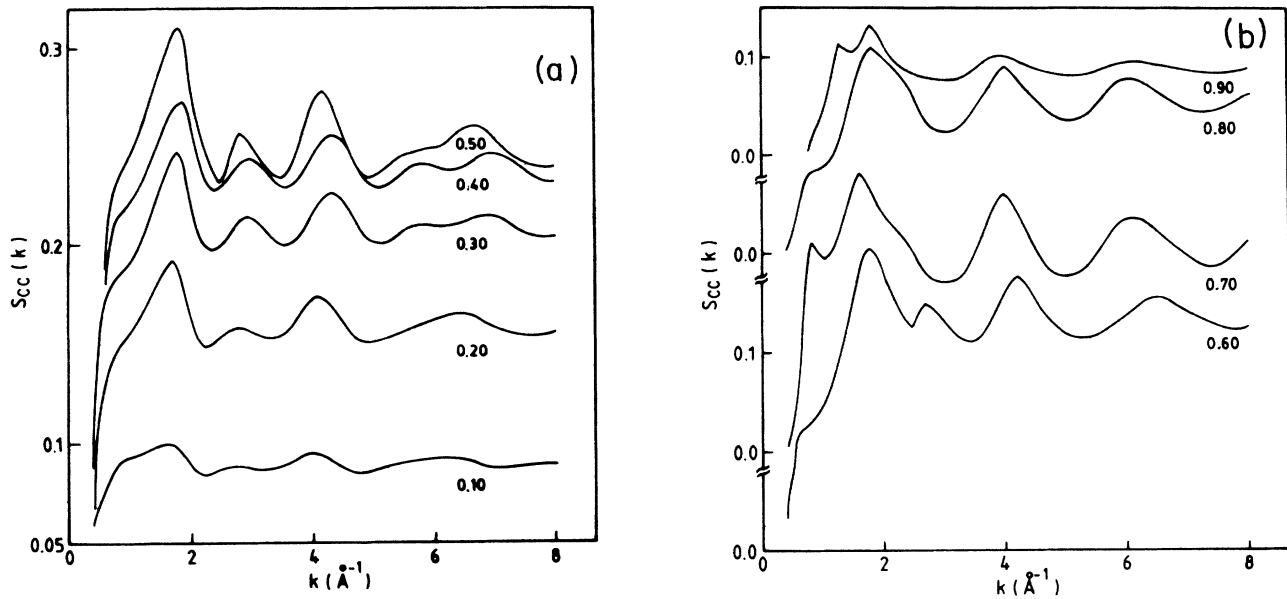


FIG. 7. (a) $S_{CC}(k)$ vs k for different concentration of copper. (b) $S_{CC}(k)$ vs k over different concentration of copper.

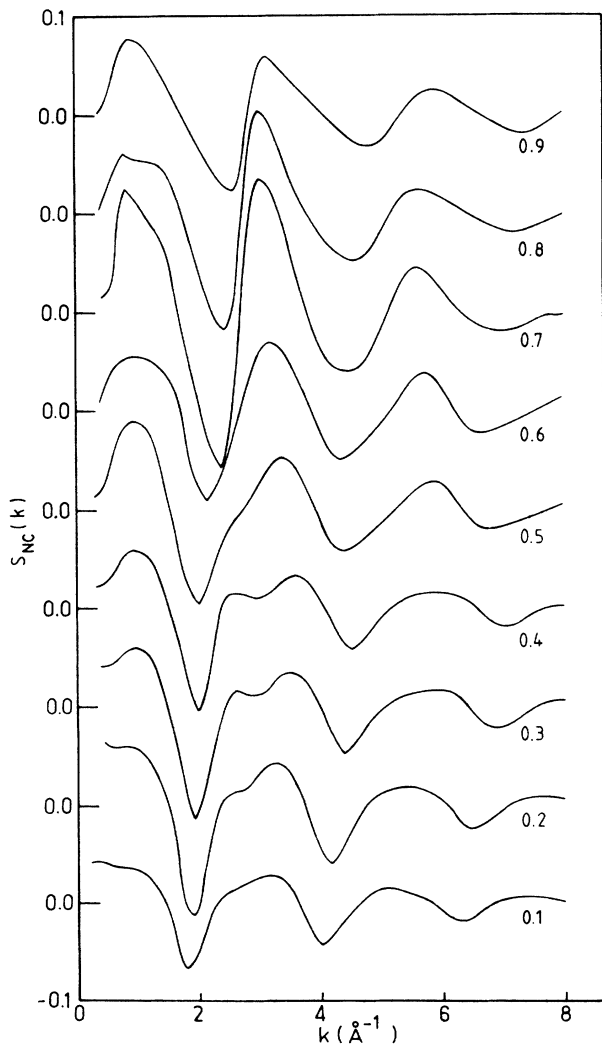


FIG. 8. $S_{NC}(k)$ vs k for different atomic fractions of copper.

$S_{22}(k)$ is depicted in Fig. 4 and it shows an interesting trend. Thus up to 0.4 we observe a right-hand shoulder while it shows a clear left-hand shoulder from 0.6 atomic fraction of Cu, and at 0.9 the $S_{22}(k)$ at low k values is rather erratic. This may be due to the segregation tendency of Bi which has different allotropic modifications at low temperatures. $S_{12}(k)$ is shown in Fig. 5. The $S_{NN}(k)$ (Fig. 6) shows the usual features. A slight shoulder is observed at 0.4 and 0.5 atomic fractions of Cu. A valley is observed in $S_{CC}(k)$ (Fig. 7) with a small peak between a 0.1 and a 0.5 atomic fraction of Cu and a satellite starts appearing with a 0.6 atomic fraction of Cu and becomes conspicuous as the concentration of Cu increases to a 0.9 atomic fraction. The $S_{NC}(k)$ (Fig. 8) becomes zero at large k and shows a large negative value at a 0.7 atomic fraction of Cu. The peak between 2 and 4 \AA^{-1} splits into two between a 0.2 and a 0.5 atomic fraction of Cu. The splitting of the peak between $k=2$ and 4 \AA^{-1} is due to increased concentration of Bi, which appears to have a great segregation tendency. In Table II we give the compressibilities and $S_{ij}(0)$ for different atomic fractions of Cu. The present computed values appear to be high compared with the available experimental results,⁹ especially at high concentrations of Bi, i.e., up to a 0.5 atomic fraction of Cu. However, there is good agreement at high concentrations of Cu, i.e., 0.6 to 1.

IV. COMPUTATION OF DIFFUSION COEFFICIENT

In this section we give an outline of the method in the evaluation of diffusion coefficients from the computed partial structure factors using Helfand's linear-trajectory principle applied to mixtures.^{20,27} The diffusion coefficient D_i is connected to the friction constant ξ_i by

TABLE II. Partial structure factors in long-wavelength limit and isothermal compressibilities.

Atomic fraction of Cu	Temp. (K)	$S_{11}(0)$	$S_{22}(0)$	$S_{12}(0)$	χ_T (10^{-12} cm ² /dyn)
0.0	1073.0	1.0000	0.0863	0.0000	22.44
0.1	870.0	0.7781	0.0836	-0.0889	26.90
0.2	973.0	0.6526	0.1288	-0.1235	35.43
0.3	1035.0	0.6062	0.2741	-0.1963	53.67
0.4	1080.0	0.5272	0.3400	-0.2133	52.11
0.5	1120.0	0.4771	0.2930	-0.2425	29.81
0.6	1160.0	0.2976	0.2125	-0.1851	11.62
0.7	1190.0	0.1682	0.2937	-0.1993	4.43
0.8	1200.0	0.0776	0.2555	-0.1090	3.75
0.9	1275.0	0.0608	0.3921	-0.1191	3.52
1.0	1373.0	0.0277	1.0000	-0.0000	2.03

$$D_i = \frac{k_B T}{\xi_i}, \quad (13)$$

where k_B is the Boltzmann constant. The friction coefficient ξ_i of the species i can be written as

$$\xi_i = \xi_i^H + \xi_i^S + \xi_i^{SH}. \quad (14)$$

In Eq. (14) ξ_i^H , ξ_i^S , and ξ_i^{SH} are the friction coefficients due to repulsive core interaction, soft interactions, and the cross effects between the hard and soft forces in the potential, respectively. The relevant equations are

$$\xi_i^H = \sum_{j=1}^2 \frac{8}{3} \sigma_{ij}^2 g_{ij}(\sigma_{ij}) \rho_j (2\pi \mu_{ij} k_B T)^{1/2}, \quad (15)$$

$$\xi_i^S = - \sum_{j=1}^2 (\rho_j / 3) \left[\frac{2\pi \mu_{ij}}{k_B T} \right]^{1/2} \times \frac{1}{(2\pi)^2} \int_0^\alpha k^3 \hat{V}_{ij}^S(k) \hat{G}_{ij}(k) dk, \quad (16)$$

and

$$\xi_i^{SH} = - \sum_{j=1}^2 \frac{2\rho_j}{3} g_{ij}(\sigma_{ij}) \left[\frac{2\mu_{ij}}{\pi k_B T} \right]^{1/2} \times \int_0^\alpha dk [k \sigma_{ij} \cos(k \sigma_{ij}) - \sin(k \sigma_{ij})] \hat{V}_{ij}^S(k). \quad (17)$$

ρ_j is the number density of species j . $\hat{G}_{ij}(k)$ and $\hat{V}_{ij}^S(k)$ are the Fourier transforms of the total correlation function $[g_{ij}(r) - 1]$ and the soft part of the potential $V_{ij}^S(r)$, respectively.²⁷ Further, μ_{ij} is the reduced mass and is given by

$$\mu_{ij} = m_i m_j / (m_i + m_j). \quad (18)$$

In Tables III and IV we give the various contributions of the friction constants and also self- and mutual-diffusion coefficients. From Table III we note that soft part and soft-hard part predominate over the hard-sphere part, unlike in simple alloys. This is due to the high value of ϵ_{Bi}/k_B of Bi, which shows a segregation tendency and attraction towards its own species—hence the predominance of ξ^S and ξ^{SH} . The same tendency is also observed in the case of Mg-Bi alloy.²⁷

It is important to note that the self-diffusion coefficients of both Cu and Bi attain a maximum at a 0.4 atomic fraction of Cu. Similar behavior is also observed in Mg-Bi alloy.²⁷ This composition may correspond to maximum segregation and cross-correlation effects may be at a minimum. Peculiarly enough, the compressibility is almost maximum around this composition.

However, it may be noted that the ratio of D_{Cu}/D_{Bi} is nearly constant; the maximum variation is around 16%. The maximum volume change is about 7.4%. This shows that the alloy forms a regular solution in spite of the

TABLE III. Various friction coefficients for liquid Cu-Bi alloys.

Concentration (atomic frac- tion of Cu)	Temp. (K)	ξ_i^{SH}/k_B (10^5 s/cm ²)		$\xi_i^S/k_B T$ (10^5 s/cm ²)		$\xi_i^{SH}/k_B T$ (10^5 s/cm ²)	
		Cu	Bi	Cu	Bi	Cu	Bi
0.1	870.0	0.094 84	0.196 20	0.092 753	0.292 104	0.142 29	0.367 42
0.2	973.0	0.080 74	0.160 43	0.073 055	0.245 439	0.112 05	0.297 32
0.3	1035.0	0.066 03	0.116 94	0.057 863	0.180 646	0.084 74	0.210 58
0.4	1080.0	0.062 61	0.106 29	0.049 493	0.144 667	0.073 76	0.175 82
0.5	1120.0	0.074 23	0.130 08	0.051 129	0.155 338	0.082 38	0.200 75
0.6	1160.0	0.104 01	0.182 73	0.050 763	0.160 305	0.105 78	0.256 49
0.7	1190.0	0.151 41	0.294 97	0.057 185	0.179 642	0.140 40	0.354 10
0.8	1200.0	0.164 04	0.312 71	0.056 523	0.156 839	0.144 86	0.364 90
0.9	1275.0	0.147 85	0.271 52	0.052 231	0.139 015	0.118 15	0.304 47

TABLE IV. Calculated self- and mutual-diffusion coefficients for liquid Cu-Bi alloy at different concentrations and temperatures.

Atomic fraction of Cu	Temp.	D_{Cu} (10^{-5} cm ² /s)	D_{Bi} (10^{-5} cm ² /s)	$D_{\text{Cu-Bi}}$ (10^{-5} cm ² /s)	$D_{\text{Cu}}/D_{\text{Bi}}$
0.1	870.0	3.03	1.17	2.84	2.59
0.2	973.0	3.76	1.42	3.29	2.64
0.3	1035.0	4.79	1.96	3.95	2.44
0.4	1080.0	5.38	2.34	4.17	2.30
0.5	1120.0	4.81	2.05	3.44	2.34
0.6	1160.0	3.83	1.67	2.54	2.30
0.7	1190.0	2.87	1.21	1.70	2.37
0.8	1200.0	2.74	1.20	1.51	2.28
0.9	1275.0	3.14	1.40	1.57	2.25

peculiar segregation tendency of Bi and the difference in atomic weights. The present theoretical investigations completely support the conclusions drawn by Steeb *et al.*^{2,11}

V. CONCLUSIONS

The existence of the peculiar metal Bi in the Cu-Bi alloy exerts its influence in the potential parameters. Thus it is found that they are concentration dependent. By slight adjustment of the parameters it is possible to generate the total structure factors faithfully. The partial structure factor of Bi, namely $S_{22}(k)$, shows right-hand and left-hand shoulders in different ranges of concentration and is attributed to the segregation tendency of the

metal. It is observed that in spite of the segregation tendency of the metals, the diffusion coefficient ratio of the metals is nearly a constant while the volume changes are also small. These are characteristics of regular solution.

ACKNOWLEDGMENTS

One of the authors (A.S.) is thankful to the Department of Science and Technology, Government of India, for financial assistance for this work, while R.V.G.R. is grateful to the University Grants Commission, New Delhi. The authors are thankful to Professor S. Steeb of Max-Planck-Institut für Metallforschung, Stuttgart, for providing them with various reprints and data.

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