Molecular Dynamics Study of Structural Relaxation and Dynamical Correlations in Amorphous and Liquid $\text{Zr}_{67}\text{Ni}_{33}$ Alloys

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Molecular dynamics simulations have been performed to study structural relaxation in the $\text{Zr}_{67}\text{Ni}_{33}$ alloy with pair functional potential. The time and space correlation of atoms are analyzed in real and reciprocal space for equilibrium liquid, supercooled liquid and amorphous state. The thermodynamically determined glass transition temperature is also characterized by the van Hove real-space correlation function. The details of atom motion in the amorphous state are analyzed using graphics of the atom trajectories. The cooperative jump motion and the slow translational motion are observed as minor motion in the amorphous state. The relaxation of the incoherent intermediate structure factor clearly reveals two relaxation processes below the melting point. The slow (α) relaxation is described by the stretched exponential function. The relaxation time shows non-Arrhenius type temperature dependence. The index increases with temperature. The fast (β) relaxation is fitted by the Debye type exponential function. For the equilibrium liquid state, the relaxation times of the α and β relaxations are of the same order. Therefore the relaxation is detected as one process.

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I. Introduction

If an equilibrium liquid is cooled to below its melting point, it enters the supercooled liquid regime. If it is cooled further at a cooling rate sufficient to suppress competitive crystallization, its viscosity increases drastically and it attains the solid state with a random structure⁽¹⁾. This is called the amorphous state. Ordinarily, a higher cooling rate is required to produce amorphous alloys in comparison with other materials, for example, oxide glasses, inorganic salts and organic polymers. The critical temperature for transition from the liquid to the amorphous state is called the glass transition temperature, T_g . It is experimentally detected as a change in specific heat.

The experiments for the liquid-glass transition of alloys have been mainly performed from a macroscopic viewpoint. The property of the supercooled liquid alloy is related to the production and the working technologies of the amorphous alloy⁽²⁾⁽³⁾. It is of interest to understand the microscopic behavior of the supercooled liquid from the industrial viewpoint as well as that of condensed matter physics. The microscopic dynamical properties are measured in a neutron inelastic scattering experiment as reciprocal space information. The dynamics are divided into two categories. One is the high energy vibrational motion and the other is the low energy diffusional motion. The former is related to the phonon and measured as the dynamic structure factor. The latter is related to the slow dynamics and evaluated as an intermediate scattering function. The dynamic structure factors of equilibrium liquid metal such as Rb ⁽⁴⁾⁽⁵⁾, and amorphous

alloys such as Mg-Zn ⁽⁶⁾⁽⁷⁾ and Zr-Ni ⁽⁸⁾⁽⁹⁾ have been reported.

Recently, the intermediate scattering functions of supercooled liquid for the $\text{Ca}_{0.4}\text{K}_{0.6}(\text{NO}_3)_{1.4}$ salt (CKN)⁽¹⁰⁾⁽¹¹⁾ and polyvinylchloride (PVC) polymer⁽¹²⁾ were measured by the neutron inelastic scattering experiment. The result shows the two-step relaxation: the slow (α) relaxation and fast (β) relaxation. However, since the neutron measurement requires a long time, which may cause the crystallization, it is rather difficult for a supercooled liquid alloy.

Molecular dynamics (MD) simulations have been applied to study amorphous and liquid materials⁽¹³⁾ and specially to the slow dynamics of the supercooled liquid after the report of the neutron experiment on CKN. The MD simulation has been applied to the idealized systems such as soft core⁽¹⁴⁾⁻⁽¹⁶⁾ and Lennard-Jones⁽¹⁷⁾⁽¹⁸⁾, and real materials such as CKN⁽¹⁹⁾ and alkali chloride glass⁽²⁰⁾. For the metallic system, the MD simulations on slow dynamics were performed only on the Ni-P system as the idealized case of the amorphous Pd-Ni-P alloy⁽²¹⁾.

We have performed the MD simulation of $\text{Zr}_{67}\text{Ni}_{33}$ alloys at various temperatures to investigate the fundamental nature of the equilibrium liquid, the supercooled liquid and the amorphous state⁽²²⁾⁻⁽²⁵⁾. We have reported on the thermodynamical value⁽²²⁾, microscopic energy fluctuation in random structure⁽²³⁾, statistical structure⁽²²⁾ and dynamic structure factor to investigate the phonon dispersion relation⁽²⁴⁾⁽²⁵⁾. We found the simulated glass transition temperature for this alloy to be 640 K⁽²²⁾, which close to the calorimetrically determined experimental value, 652 K⁽²⁶⁾. The simulated structure factor⁽²⁴⁾ and the vibra-

tional energy⁽²⁵⁾ distribution reproduce the experimental values well.

In this paper, we have performed the MD simulation of Zr₆₇Ni₃₃ alloys for a wide range of temperature from the equilibrium liquid to the amorphous state and analyzed self-motion and cooperative motion as a function of time. These motions have been determined from the van Hove correlation function in real space and the intermediate scattering functions in reciprocal space. The trajectories of the atoms have also been analyzed to clarify the details of their motion and calculate the diffusion coefficient for the macroscopic behavior of the dynamics has been calculated.

II. Molecular Dynamics Procedure

The binary mixture of 640 Zr atoms and 320 Ni atoms were adopted. For the interaction between atoms, the pair functional potential⁽²⁷⁾ is used. Specifically, the Massobrio, Pontikis and Martin (MPM) potential⁽²⁸⁾, fitted by the cohesion energy and the elastic constant is used. The total energy of the system is written as

$$E_{\text{pot}} = \sum_{\alpha=1}^m \sum_{i_{\alpha}}^{N_{\alpha}} \left[\sum_{\beta=1}^m \sum_{j_{\beta} (\neq i_{\alpha})}^{N_{\beta}} A_{\alpha\beta} \exp \left[-p_{\alpha\beta} \left(\frac{r_{i_{\alpha}j_{\beta}}}{d_{\alpha\beta}} - 1 \right) \right] - \left\{ \sum_{\beta=1}^m \sum_{j_{\beta} (\neq i_{\alpha})}^{N_{\beta}} \xi_{\alpha\beta}^2 \exp \left[-2q_{\alpha\beta} \left(\frac{r_{i_{\alpha}j_{\beta}}}{d_{\alpha\beta}} - 1 \right) \right] \right\}^{1/2} \right] \quad (1)$$

where $r_{i_{\alpha}j_{\beta}}$ is the distance between i_{α} and j_{β} atoms. The second functional term represents the many body interactions in metal. The pair functional potential allows for quantitative simulation of elastic and ductile properties of metallic crystal⁽²⁷⁾⁽²⁹⁾.

The modified Nosé constant-temperature (NVT) method⁽³⁰⁾ where the volume is varied with object temperature by the constant thermal expansion coefficient of $2.659 \times 10^{-5} \text{ K}^{-1}$. The density of the amorphous state is 7.06 g cm^{-3} at 298 K, in agreement with the experimental value⁽³¹⁾. The simulation has been performed with a periodic boundary condition and begun by constructing a $4 \times 4 \times 5$ C16 tetragonal crystal cells. The system is first heated up to 2000 K well above the melting temperature ($T_m = 1393 \text{ K}$)⁽³²⁾, to obtain a homogeneous equilibrium liquid state. The system is quasi-continuously quenched to various object temperatures with a quench rate of $2 \times 10^{14} \text{ K}^{-1}$. After subsequent isothermal annealing at the object temperature, statistical analyses are performed. The equations of motion are solved using the fifth-order predictor corrector algorithm of Gear with a time step of 1 fs. We define the equilibrium liquid as the state above $T_m (= 1393 \text{ K})$, the amorphous state as that below $T_g (= 640 \text{ K})$ and the supercooled liquid as the state between T_m and T_g .

III. Results

1. van Hove correlation function

The behavior of the displacement caused by diffusion

and thermal vibration was estimated from the van Hove correlation function⁽³³⁾. Its self part for the α specimen is defined as

$$G_{\alpha}^s(r, t) = \frac{1}{N_{\alpha}} \sum_{i_{\alpha}=1}^{N_{\alpha}} \langle \delta(|r_{i_{\alpha}}(t) - r_{i_{\alpha}}(0)| - r) \rangle \quad (2)$$

where $r_{i_{\alpha}}(t)$ denotes the position of a particle i of α species at time t , N_{α} is the total number of α atoms and $\langle \dots \rangle$ denotes the time average. This equation gives the probability of deviation at time t from the origin at $t=0$. In a liquid state, the van Hove self correlation function is expected to rapidly approach to the hydrodynamics limit, i.e., the following Gaussian shape is predicted⁽³⁴⁾:

$$G^s(r, t) \approx \frac{1}{(4\pi Dt)^{3/2}} \exp \left(\frac{-r^2}{4Dt} \right) \quad (3)$$

where D is the self diffusion coefficient.

The standard distinct part of the van Hove correlation function for α - β pair is defined as

$$G_{\alpha\beta}^d(r, t) = \frac{N}{N_{\alpha}N_{\beta}} \sum_{i_{\alpha}=1}^{N_{\alpha}} \sum_{j_{\beta}=1 (\neq i_{\alpha})}^{N_{\beta}} \langle \delta(|r_{i_{\alpha}}(t) - r_{j_{\beta}}(0)| - r) \rangle. \quad (4)$$

The following form is used to express the distinct part of the van Hove correlation function for comparison with the pair distribution function.

$$g_{\alpha\beta}(r, t) = \frac{1}{4\pi r^2 \rho_0} G_{\alpha\beta}^d(r, t) \quad (5)$$

where ρ_0 is number density. This equation gives the time duration of the atom configuration around the reference atom.

The decay of the van Hove self correlation function for the Zr atom at various temperatures is shown in Fig. 1. The time 0.1 ps is on the order of half the thermal

Fig. 1 The decay of the van Hove self correlation function for Zr atoms at 100, 500, 700, 1000 and 1400 K.

vibration period. The displacement at 0.1 ps caused by the thermal vibration is rather symmetrical. The peak width at 0.1 ps increases with temperature. In the equilibrium liquid state at 1400 K, the profile broadens with time. It represents the hydrodynamic diffusional motion. In a supercooled liquid state at 1000 K, the shape of the van Hove function is similar to that in the equilibrium liquid state but its deviation distance is smaller. In an amorphous state at 100 K, the profile of the van Hove function essentially does not change after 0.1 ps. The major motion at 100 K is the thermal vibration around the fixed point. The overall profile in the amorphous state at 500 K is similar to that in the supercooled liquid

Fig. 2 The decay of the magnified van Hove self correlation function for Zr atoms at 100, 500 and 700 K.

state at 700 K. Figure 2 shows the magnified profile of the van Hove self correlation function for Zr atom. Weak shoulder (500 K) and weak subpeak (100 K) observed at the skirts indicate jump motion. This jump motion has been reported in the MD simulation on the soft core⁽¹⁵⁾⁽¹⁶⁾, Lennard-Jones⁽¹⁷⁾, CKN⁽¹⁹⁾, and Ni-P alloy⁽²¹⁾ systems. In the case of the soft core⁽¹⁶⁾ and the Lennard-Jones⁽¹⁷⁾ systems, the subpeak appears at the nearest neighbor atom distance. Whereas, in the present Zr-Ni amorphous alloy, the subpeak also appears at half the nearest neighbor atom distance. These differences are caused by the interatomic potential. The soft core and the Lennard-Jones systems have a non- or weakly attractive part in their potential. The jump motion occurs more frequently in the soft core system than in the present Zr-Ni system.

The distinct part of the van Hove correlation functions for Zr-Ni pairs at various temperatures is shown in Fig. 3. The van Hove correlation function at $t=0$ is equivalent to the pair distribution function. The shift of the distinct parts from 0 to 0.1 ps is clearly detected at the first peak but not at the second or third peak. Above the glass transition temperature, the profile loses the correlation and becomes flat with time due to the effect of long range hydrodynamic diffusion. The rate of change increases with temperature. On the other hand, below the glass transition temperature, the profile beyond the third peak does not change even after 10 ps. This means that there is a less long range diffusional motion, i.e., the atom configuration is frozen. The distinct parts of the van Hove correlation function for Zr-Zr and Ni-Ni pairs at 1000 K are shown in Fig. 4. The decay of the Ni-Ni pair correlation is fast in comparison with other pairs.

Fig. 3 The distinct part of the van Hove correlation functions for Zr-Ni pairs at 100, 500, 700, 1000 and 1400 K.

Fig. 4 The distinct part van Hove correlation function for Zr-Zr and Ni-Ni pairs at 1000 K.

2. Trajectory of the atom

The van Hove correlation function gives the averaged information of all the atoms in the system. This information indicates the predominant dynamics, but not minor motions. To clarify the minor motion, a three-dimensional computer graphic image has been produced. Figure 5 shows the trajectory of the atoms at 100 K during 10 ps. The white lines indicate the boundary of the MD cell. Figure 5(a) is the right bottom of the cell and (b) is the right top of the cell. They are continuously connected by the periodic boundary condition. The trajectory of Zr is indicated by ultramarine to sea-green colors with increasing time. That of Ni is indicated by red to yellow. Most of the atoms show the small amplitude thermal vibration. They are fundamentally fixed in space. However, two types of large displacements are observed on some atoms. One is jump motion indicated by thin white arrows, the other is slow translational motion indicated by thick white arrows. The jump motion occurs cooperatively at the same time for plural atoms. The jump length is shorter than the first neighbor distance and the jumping time is about 0.4 ps, which is twice the thermal vibration cycle. In the present simulation 6 jumps are observed during 10 ps. Thus the waiting time of jump motion for one atom is estimated to be of the order of 2 ns, which is one thousand times the thermal vibration cycle. The slow translational motion is the sum of continuous translational motion and thermal vibration. Cooperativity is not evident for the slow translational motion. In the MD simulation, we observe the so-called as-quenched

Fig. 5 The trajectory of the atoms in amorphous state at 100 K during 10 ps: (a) the right bottom of the MD cell and (b) the right top of the MD cell.

state of the amorphous alloy. After a long time isothermal annealing, such minor motions should occur more rarely.

3. Intermediate scattering function

The intermediate scattering function⁽³⁴⁾ shows the time duration of the density correlation in reciprocal space. It is also estimated from the Fourier transformation of the van Hove correlation function. Its incoherent part (self part) is calculated as

$$F_{\alpha}^s(Q, t) = \int_0^{r_c} G_{\alpha}^s(r, t) \frac{\sin(Qr)}{Qr} dr \quad (6)$$

Fig. 6 The time variations of the incoherent intermediate scattering function of Zr and Ni atoms at various temperatures.

The computation of the distinct part is time consuming. To save time, the self part is calculated.

The time variations of the intermediate scattering function of Zr and Ni atoms at various temperatures are shown in Fig. 6 in a logarithmic scale. These results are obtained at the wave number $Q = 25.8 \text{ nm}^{-1}$, i.e., the first peak of the structure factor. The behavior is similar to the neutron measurement results of CKN⁽¹⁰⁾⁽¹¹⁾. In the equilibrium liquid state, at 2000 and 1400 K, the relaxation occurs in one step. On the other hand, in the supercooled liquid (1000 and 700 K) and the amorphous (500 and 100 K) state, relaxation with two steps is observed. The second (slow) decay corresponds to α relaxation and the first (fast) decay to β relaxation.

These relaxations are fitted by a sum of the exponential functions.

$$F^s(Q, t) = (1 - A) \exp \left[- \left(\frac{t}{\tau_{\text{Deb}}} \right)^\delta \right] + A \exp \left[- \left(\frac{t}{\tau_{\text{rel}}} \right)^\beta \right] \quad (7)$$

Table 1 Different parameters in sum of the exponential functions of eqs. (7) and (8), obtained by fitting to the incoherent intermediate scattering function MD data.

	T (K)	a	β	τ_{rel} (ps)	τ_{Deb} (ps)
Zr	100	0.00025	0.55	42000	0.12
	500	0.00031	0.61	650	0.12
	700	0.00034	0.63	180	0.12
	1000	0.00039	0.88	5.9	0.15
	1400	0.00039	0.95	0.91	0.18
	2000	0.00039	0.97	0.40	0.18
Ni	100	0.00031	0.51	30000	0.11
	500	0.00038	0.60	450	0.11
	700	0.00043	0.61	130	0.13
	1000	0.00045	0.83	4.1	0.14
	1400	0.00045	0.91	0.68	0.15
	2000	0.00045	0.93	0.30	0.15

where we define A as the Debye-Waller factor

$$A = \exp(-aT) \quad (8)$$

As shown by the solid lines in Fig. 6, both relaxations are expressed well by eq. (7). The fitted parameters of eq. (7) are listed in Table 1. The α relaxation is fitted by the stretched exponential function with the exponent $\beta (< 0)$. The exponent β increases with increasing temperature. The relaxation time of the α relaxation, τ_{rel} shows non-Arrhenius-type dependence on temperature. The α relaxation time of Zr atom is longer than that of Ni atom. Judging from the relaxation time, the α relaxation is related to the hydrodynamic diffusion of the atom. The β relaxation is described by the exponential function with a fixed exponent $\delta (= 1.20)$. This behavior is similar to the experimental result for PVC⁽¹²⁾, which shows Debye type β relaxation ($\delta = 1$). On the other hand, the experimental result for CKN shows the power function. Since the β relaxation time, τ_{Deb} , is almost constant for various temperatures and equivalent to half the Einstein period, it is related to the thermal vibration⁽²⁵⁾.

4. Diffusion constant and non-Gaussian parameter

In order to analyze the self diffusion coefficient, the mean square displacement (MSD) of the atom is calculated as

$$\Delta r^2 = |r(t) - r(0)|^2. \quad (9)$$

The calculated MSD for the Zr atom is shown in Fig. 7. In the equilibrium liquid state the MSD shows good linearity (a), but not at low temperature (b). Below the glass transition temperature, the total displacement is on the order of the initial displacement by thermal vibration. In the Einstein formula, the MSD is equal to $6Dt$ at $t \rightarrow \infty$. In the present simulation, the self diffusion coefficient has been determined from the slope of the MSD from 30 to 70 ps. The estimated value above the glass transition temperature is followed by the Vogel-Fulcher relation rather than the Arrhenius relation, as shown in Fig. 8. This close to the temperature dependence of the viscosity for fragile glass system. The Stokes-Einstein formula⁽³⁵⁾ gives the inversely proportional rela-

Fig. 7 The mean square displacement (MSD) for Zr atoms: (a) at high temperatures and (b) at low temperatures.

tion between the diffusion coefficient and the viscosity. The lighter atom, Ni, diffuses faster than Zr. Since the Einstein formula is not satisfied below the glass transition temperature, D is overestimated. Massobrio has pointed out that the lowest value of the diffusion coefficient calculated by the Einstein formula in the conventional MD is on the order of $\approx 10^{-11} \text{ m}^2 \text{ s}^{-1}$ ⁽³⁶⁾.

The discrepancy due to the hydrodynamics limit of the diffusion is estimated by the non-Gaussian parameter⁽¹⁵⁾⁽³⁷⁾ as

$$A(t) = \frac{3\langle |r(t) - r(0)|^4 \rangle}{5\langle |r(t) - r(0)|^2 \rangle^2} - 1 \quad (10)$$

where the parentheses represent an average over atoms and time. The calculated profiles for the Zr atom are shown in Fig. 9. The non-Gaussian parameter has the maximum at first and converges to zero in the equilibri-

Fig. 8 Self diffusion coefficient for Zr and Ni atoms estimated from the MSD.

Fig. 9 The non-Gaussian parameter for Zr atoms at various temperatures.

um liquid state (2000 and 1400 K). This indicates the existence of the hydrodynamic diffusion described by eq. (3). A large deviation of the non-Gaussian parameter from zero is observed in the supercooled liquid and amorphous states. The jump diffusion and the slow translational motions cause the non-Gaussian behavior even though they are minor motion.

IV. Discussion and Conclusive Remarks

Here, we discuss the jump motion in the amorphous

state in comparison with diffusion in the crystal. The amorphous material shows the cooperative atom motion while the crystal shows the single atom motion. In the case of the crystal, an atom can jump to a new site only when it is a vacant site. In some sense, there is no apparent vacancy in the amorphous structure. Since the potential of each atom has a wide range of fluctuation, the jump motion can occur at a lower potential site like a vacancy. Since the jump motion strongly affects the potential of a specific neighbor atom, the neighbor atom may jump simultaneously. This may cause a multiple cooperative jump motion.

In the liquid state, all atoms move over a long range by the hydrodynamic diffusion. The amorphous state has been intuitively understood as a frozen random structure. However, the present MD result yields the following description for the amorphous state. The predominant motion in the amorphous state is the thermal vibration of atoms around fixed positions. The minor motion comprises jump motion and slow translational motion. These long range displacements occur only for some atoms in the amorphous state. These irregular motions cause the non-Gaussian behavior of the diffusional motion and the so-called macroscopic relaxation of the amorphous alloy⁽³⁸⁾.

In the field of the metallurgy, the macroscopic thermodynamical experiment has usually been performed to investigate the physical state of matter. From the microscopic viewpoint, the dynamics are estimated as follows. In the equilibrium liquid state, the relaxation time of the hydrodynamic diffusion is comparable to that of the thermal vibration, and atom displacement is described well by the Gaussian approximation. In the supercooled liquid state, the relaxation time of hydrodynamic diffusion is longer than that of thermal vibration and the non-Gaussian parameter shows a finite value. The effect of the glass transition is observed in the van Hove correlation function. In the amorphous state, the long range configuration of the distinct part does not change, and jump motion is detected in the self part. In the supercooled liquid state, a change of the long range configuration in the distinct part occurs, and the jump motion in the self part cannot be separated from hydrodynamic diffusion.

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REFERENCES

- (1) C. A. Angell: J. Phys. Chem. Solids, **49** (1988), 863.
- (2) T. Zhang, A. P. Tsai, A. Inoue and T. Masumoto: Sci. Rep. RITU, **A36** (1992), 261.
- (3) T. Masumoto: Mater. Sci. Eng., **A179/180** (1994), 8.
- (4) R. Winter, C. Pilgrim and F. Hensel: J. de Physique IV, **1** (1991), C5-45.
- (5) J. R. D. Copley and J. M. Rowe: J. Phys. Soc. Japan, **32** (1974), 49.
- (6) J. B. Suck, H. Rudin, H. J. Guntherodt and H. Beck: J. Phys., **C13** (1980), L1045.
- (7) J. B. Suck, P. A. Egelstaff, R. A. Robinson, D. S. Sivia and A. D. Taylor: J. Non-Cryst. Solids, **150** (1992), 245.
- (8) J. B. Suck, H. Rudin and H. J. Guntherodt: *Dynamic structure factor of the metallic glass Ni₂₄Zr₇₆*, ed. by S. Steeb and H. Warlimonts, 5th International Conference on Rapidly Quenched Metals, North-Holland, Würzburg, (1985), p. 471.
- (9) J. B. Suck: *Dynamics of disordered materials*, ed. by D. Richter, A. J. Dianoux, W. Petry and J. Teixeira, Springer-Verlag, Berlin, (1989), p. 182.
- (10) W. Knaak, F. Mezei and B. Farago: Europhys. Lett., **7** (1988), 529.
- (11) F. Mezei, W. Knaak and B. Farago: Phys. Rev. Lett., **58** (1987), 571.
- (12) J. Colmenero, A. Arbe and A. Alegria: Phys. Rev. Lett., **71** (1993), 2603.
- (13) F. Yonezawa: Solid State Physics, **45** (1991), 179.
- (14) J. L. Barrat and J. N. Roux: J. Non-Cryst. Solids, **131-133** (1991), 255.
- (15) H. Miyagawa and Y. Hiwatari: Phys. Rev., **A40** (1989), 6007.
- (16) J. N. Roux, J. L. Barrat and J. P. Hansen: J. Phys. Condensed Matter, **1** (1989), 7171.
- (17) G. Wahnstrom: Phys. Rev., **A44** (1991), 3752.
- (18) G. Wahnstrom: J. Non-Cryst. Solids, **A131-133** (1991), 109.
- (19) G. F. Signorini, J. L. Barrat and M. L. Klein: J. Chem. Phys., **92** (1990), 1294.
- (20) K. Kinugawa, K. Kadono and H. Tanaka: J. Non-Cryst. Solids, **150** (1992), 281.
- (21) L. J. Lewis: Phys. Rev., **B44** (1991), 4245.
- (22) T. Aihara Jr., K. Aoki and T. Masumoto: Scripta Metall., **28** (1993), 1003.
- (23) T. Aihara Jr., K. Aoki and T. Masumoto: Mater. Sci. Eng., **A179-180** (1994), 256.
- (24) T. Aihara Jr. and T. Masumoto: J. Phys. Condensed Matter, **7** (1995) 1525.
- (25) T. Aihara Jr., K. Aoki and T. Masumoto: Mater. Trans., JIM, **36** (1995), 399.
- (26) Z. Altounian, T. Guo-hua and J. O. Ström-Olsen: J. Appl. Phys., **54** (1983), 3111.
- (27) A. E. Carlsson: Solid State Phys., **43** (1990), 1.
- (28) C. Massobrio, V. Pontikis and G. Martin: Phys. Rev., **B41** (1991), 10486.
- (29) M. S. Daw, S. M. Foils and M. I. Baskes: Mater. Sci. Rep., **9** (1993), 251.
- (30) S. Nosé: J. Chem. Phys., **81** (1984), 511.
- (31) Z. Altounian and J. O. Ström-Olsen: Phys. Rev., **B27** (1983), 4149.
- (32) T. B. Massalsky, Ed., *Binary alloy phase diagrams*, 2nd Ed., Vol. 3, ASM International, Materials Park, OH, (1991), p. 2890.
- (33) L. van Hove: Phys. Rev., **95** (1954), 249.
- (34) J. P. Hansen and I. R. McDonald: *Theory of simple liquids*, 2nd Ed., Academic Press, London, (1986), Chapter 8.
- (35) R. B. Bird, W. E. Stewart and E. W. Lightfoot: *Transport phenomena*, John Wiley & Sons, New York, (1960), p. 514.
- (36) C. Massobrio, V. Pontikis and G. Cicotti: Phys. Rev., **B39** (1989), 2640.
- (37) B. R. A. Nijboer and A. Rahman: Physica, **32** (1966), 451.
- (38) A. Inoue, T. Masumoto and H. S. Chen: J. Mater. Sci., **20** (1985), 4057.