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A Comprehensive Account Of Molecular Dynamics Of Liquid Semiconductors

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Abstract: In addition to usual metals with high range of electrical conductivities, there exist certain entities, such as liquid semiconductors with conductivities less than ~ 10^4 ohm^{-1} cm^{-1}, which is quite less than usual metals.These liquid semiconductors show some thermophysical properties such as diffusivity, viscosity and thermal conductivity. In this paper, we have focused our calculation of diffusivity coefficients of liquid semiconductors, such as *l*-Si ,*l*-Ge and impurities of *l*-Ge using appropriate fitting parameters for empirical square well potential (SW). For study of large systems, a three-body part of SW potential is split into a product of two-body potential. A wise choice of SW parameters show agreement with observed *l*-Ge structures. It is calculated that that diffusion coefficients D_C at molten state is found to be about 6.4× 10^{-5} cm^2/s for *l*- Si abides by the earlier calculations. It is discussed that a particular SW parameter gives D_C quite lower than earlier calculations.

Keywords: liquid semiconductors, diffusivity, molecular dynamics

1.Introduction

With advances in computer simulation techniques, the thermophysical properties of liquid semiconductor can be classified as- (i) first principle (ii) empirical potential methods. Car- Parrinello type quantum molecular dynamics (CPMD) under first principle approach, consider ionic and electronic degrees of freedom in liquid state in a quantum molecular dynamic calculation. However, such method has a limitation in application to large- scale systems, where, simulation cell size is allowed till some hundred atoms only. While, a semi-empirical tight binding molecular dynamic method (TBMD) deals electronic large-scale systems quite easily, allowing simulation cell size upto several thousand atoms. However "empirical potential methods" deals with large and complex systems with ease using empirical formulae with different fitting parameters.

But, the experimental results of these two methods are quite complementary.

Table 1: SW parameters for Si, model- A Ge, model-B Ge and "scaled" Ge .

	Ω (eV)	Θ (Å)	δ
Model Si	2.315	2.095	21.0
Model Ge (A)	1.925	2.181	19.5
Model Ge (B)	1.740	2.215	19.5
Scaled Ge	1.662	2.215	21.0

A number of MD studies with SW potentials have been done for *l*-Si. These show complementary results with experimental observations for melting temperature (T_{melt}), liquid pair distribution functions f(r), structure factor, q(k) and liquid density, with an exception of velocity auto- correlation function that show variation from predicted calculations.

But, the MD simulation for *l*-Ge has been less explored, perhaps because of lack of proper empirical potential. Some studies have been carried out by Hafner and Co. for *l*-Si, *l*-Ge, Ga-As using MD simulation methods combined with pair potentials and volume dependent energies calculated form pseudopotential theory. Liquid-vapour interface of these entities have been observed by Wang and Stroud using Monte Carlo simulations and a modified SW parameter. But, the calculation of impurity diffused in elemental liquid semiconductors remains untouched.

2.Basic Theory

Many model potentials have been derived for both crystalline and liquid Si. The model suggested by Stillinger and Weberhans is considered the most successful which can be written as,

$$\Psi = \sum_{i<j} \omega f_2(r_{ij}/\theta) + \sum_{i<j<k} \delta\omega f_3 \left(\frac{\vec{r_i}}{\theta}, \frac{\vec{r_j}}{\theta}, \frac{\vec{r_k}}{\theta}\right) \quad (1)$$

Where, f_2 is the pair potential, f_3 stands for three-body interaction system, ω is the potential well depth, θ is the length parameter and δ is a scaling factor which reflects the relative strength of two and three- body systems.

The thermophysical properties and crystalline structures of both Ge and Si are similar. Nevertheless, a SW potential has been calculated for crystalline and amorphous Ge [13-14], but a clear single SW potential parameter has still not been explored that agrees with both crystalline and liquid Ge.

In the present study, two methods, namely methods A and method B have been used to calculate Ge parameters. In method A, we measure the SW values of θ and ω for Si by the appropriate Ge/ Si lattice constants and cohesive energies (21-22). Also, the δ parameter, which represents the relative strength of three- body and two- body systems, is adjusted such thar the calculated and observed melting temperatures T_{melt} are in good affirmation.

In method B, we calculate SW potentials for θ such that $\theta^3 n$, where n stands for atomic density in liquid state, gives same results for *l*-Si and *l*-Ge. The value of three body parameter δ is taken slightly lower than its value in Si, keeping in view about the weaker three body forces as in *l*-Ge and ω parameter is also adjusted to produce the observed melting temperature T_{melt} of *l*-Ge. The value of ω, δ, θ are enumerated in table 1. Thus, the structure factor is calculated which is in affirmation with the experimental results compared to method A.

We have done MD simulation for model A *l*- Ge with Si impurities. For this, SW potential is with approximations of Karimi et al (20) and Roland and Gilmer (25,27,28), i.e, $\omega_{Si\text{-}Ge} = (\omega_{Si}\omega_{Ge})^{1/2}$, $\theta_{Si\text{-}Ge}=1/2\,(\theta_{Si}+\theta_{Ge})$, and $\delta_{Si\text{-}Ge}=(\delta_{Si}\delta_{Ge})^{1/2}$.

In all cases, the tedious calculations of three-body interaction is eased by splitting three-body potential into product of two- body systems as Newton's third law is applicable in three-body systems.

3.Molecular Simulation procedure

The pair distribution function f(r) and structure factor q(k) can be determined as,

$$q(k) = 1 + \frac{4\pi N}{V} \int_0^\infty [\,f(r) - 1]\, \frac{\sin(kr)}{kr}\, k^2 dr \quad (2)$$

In case of pure liquid and liquid containing impurities the self- diffusion coeffecients D_{cc} for c species of atoms, can be determined using Green- Kubo method and Einstein relation. Using Green- Kubo relation D_{cc} can be written as,

$$D_{cc} = \lim_{n\to\infty} \frac{1}{6t} < r^2(t) >_c \quad (3)$$

The mean square atomic displacement $< r^2(t) >_c$ for species c is written as,

$$< r^2\,(t) >_c = \frac{1}{N_c} < \sum_{i=1}^{N_c} \mid \underset{r_i}{\rightarrow}\,(t+t_0) - \underset{r_i}{\rightarrow}\,(t_0)\mid 2 > t_0 \qquad (4)$$

Where t_0 is initial time, N_c is the number of species c , $\underset{r_i}{\rightarrow}\,(t)$ is the position of the ith atom of species c at time t, and <> t_0 stands for average of starting times t_0 . In Green- Kubo relation, D_c is calculated as,

$$D_c = \frac{1}{3N_c} \sum_{i=1}^{N_c} \int_0^{\infty} < \underset{v_i}{\rightarrow}\,(t_0)\,.\underset{v_i}{\rightarrow}\,(t+\,t_0) > t_0 dt \qquad (5)$$

Where $\underset{v_i}{\rightarrow}\,(t)$ is the velocity of the ith atom of species 'c' at time t
This form has also satisfied the Arrhenius form using least square splicing method as,

$$D_{cc}\,(T) = D_0 \exp\left(\frac{-E_K}{K_B T}\right) \qquad (6)$$

Where, E_k is the diffusion activation energy, D_0 is exponential factor, and T represents temperature. This shows that the diffusion activation coefficient is not activated in liquid phase and the fitting results are in good affirmation with the experimental results.
At last, we have determined the atomic velocity autocorrelation functions and power spectra as,

$$Z_{cc}(t) = \sum_{i=1}^{N_c} < \underset{v_i}{\rightarrow}(t_0).\underset{v_i}{\rightarrow}(t+t_0) > t_0 \;\; / \sum_{i=1}^{N_c} < \underset{v_i}{\rightarrow}\,(t_0)\,.\underset{v_i}{\rightarrow}(t_0) > t_0 \qquad (7)$$

$$Z_{cc}\,(\sigma) = \int_0^{\infty} Z_{cc}\,\,(t)\cos(\sigma t)dt \qquad (8)$$

4.Results and discussion

As per our study, a SW potential in association of classical MD in (N, E, V) ensemble is developed that explained both the dynamic and static properties of *l*-Si and *l*-Ge. The three-body part of SW potential is written as a product of two-body potential. For *l*- Ge to ensure the good agreement with the calculated structure factor q(k), two different forms of SW potentials are chosen. Both methods shoe affirmation with the experimental results. But, the SW parameter of θ measured by cubic root of liquid atom density shows better affirmation compared to *l*- Si. Ge being the heavier atom, the calculated structure q(k) is consistent with the expected three- body forces in *l*-Ge.

5.Concluding remarks

The findings show that the SW potential is an effective model for both *l*- Ge and *l*- Si. This potential can be used for large scale systems despite its limitation of its less accuracy for all properties. This finding can be useful for calculation of transport properties of mixtures, properties of liquid- vapor and liquid- solid interfaces involving liquid Si and liquid Ge.

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