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MOLECULAR DYNAMICS SIMULATION OF STRUCTURAL CHANGES OF LIQUID Fe_3Al ①

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ABSTRACT The relationship between the pair distributed function $g(r)$ of Fe_3Al and the temperature under rapid solidification was studied by the molecular dynamic method, it was found that the second peak of $g(r)$ of Fe_3Al has been split and an amorphism appeared in the liquid. The changes of clusters was investigated by the pair analysis technology, the changes of the orientational order parameters and bond pairs demonstrate that during transforming the liquid to the amorphism, the group of structure has been changed indeed, amorphism is not the freezing liquid.

Key words tight binding potential rapid solidification molecular dynamics

1 INTRODUCTION

Liquid metal is a very difficult and attractive branch of condensed state physics. As far as the metal crystal is concerned, we can use 14 kinds of lattice to describe it; as for gas, the Van Derwalls equation can be used to deal with it. However, it is difficult for us to understand the liquid because of its high temperature and undefinitive distance. But the liquid is the initial state of the metal materials, for example, in cast and metallurgy. With the deep research to the relationship between the solid and liquid, it is demonstrated that the structure and defect of crystal is related to the liquid structure and dynamics of phase. The distinct and overall microstructure is very helpful to discover the heredity nature of liquid and solid, so, it is very important to study the liquid metal. Foresight physist, materials scientist and chemist are all devoted to study the liquid[1, 2].

Now, the semiempirical and empirical multi-body potential was developed, the tight binding potential developed by Cleri was used to a lot of material regions[3- 8], but the research of liquid has not been reported and the tight binding potential has not been used to the amorphism and rapid solidification. It is not easy to detect the structure of disordered system, so, the computer simulation becomes an important method to learn some information about liquid. The paper report that the structure and the change of structure of melt Fe_3Al by use of tight binding potential and molecular dynamics simulation under the conditions of rapid solidification.

2 POTENTIAL OF ATOMS

According to the tight binding potential, the total of single atom in system is divided into two parts, one is the attractive part, its cohesive energy

$$E_B^i = \{\sum_j \xi_{\alpha\beta}^2 e^{-2q_{\alpha\beta}(\gamma_{ij}/r_0^{\alpha\beta} - 1)}\}^{0.5} \qquad (1)$$

the other is repulsive part its cohensive energy,

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$$E_{\mathrm{R}}^{i} = \sum_{j} A_{\alpha\beta} e^{p_{\alpha\beta}(\gamma_{ij}/r_{0}^{\alpha\beta} - 1)} \qquad (2)$$

where r_{ij} is the distance between atom i and atom j; $\gamma_0^{\alpha\beta}$ is the neighbour distance; A, p, ξ, q is the model parameters, respectively. The total cohesive energy of system is

$$E_{\mathrm{C}} = \sum_{i} (E_{\mathrm{R}}^{i} + E_{\mathrm{B}}^{i})$$

the force applied on atom i is written as $F_i = -\forall E_{\mathrm{C}}$, here $\forall$ is laplace character. Knowledge in detail about tight binding potential can be found in Ref. [3].

3 BRIEF PROCESS OF MOLECULAR DYNAMICS SIMULATION

As far as normal molecular dynamics simulation, 100~ 1 000 particles, 500 atoms and periodic conditions were used. The time step is less than vibration period of atom i. e. chose 5 fs, and the decline force method was used to decrease the temperature. At first, we run 10^5 time steps from initial configurations on temperature 2 000 K to get balance liquid, then the liquid was cooled to 300 K at the rate of 4×10^{13} K/s and the configurations were recorded every 200 K. The structure of liquid was defined by using the 20 average configurations.

4 METHOD OF STRUCTURE ANALYSIS

4.1 Pair distributed function[9]

The pair distributed function $g(r)$ can be gotten by Fourier transformation of X-ray diffractor structure factor $s(q)$, so, the $g(r)$ is the standard of the experimental data and theory. It is also the effective method to describe the liquid and amorphism. As far as the AB alloys, there would be three partial pair distributed functions, namely $g_{\mathrm{A-A}}(r)$, $g_{\mathrm{A-B}}(r)$ and $g_{\mathrm{B-B}}(r)$, and one total pair distributed function, $g_{\mathrm{Tol}}(r)$.

4.2 Bond orientational order parameters[10]

The bond orientational order is very useful to describe the local properties of system. Our analysis starts by associating a set of spherical harmonics with every bond joining an atom to its near neighbours. In order to eliminate the influence of the direction, we adopt the W_6 and Q_6 of the spherical harmonics. As for some structure, the W_6 and Q_6 would have suitable distribution. The icosahedra play an important role in the rapid solidification. If ideal icosahedra exists, the W_6 would be -0.169754.

4.3 Pair analysis[11]

Pair analysis refers to use parameter *ijkl* to describe the relation of neighbour atoms. If $i = 1$, it means two atoms become a bond; j is the number of another atoms, of which all become bonds with pair atoms; k is the number of atoms, of which the neighbour atoms become bonds each other. Here, Bonds of course don't mean chemical bonds, but rather lines resulting from convenient assignation of near neighbour. l is another parameter to define the cluster. As for different structure, the *ijkl* would have a special distribution, for example, 1551 is related to the icosahedra, all kinds of polyhedra can be calculated by pair analysis.

5 RESULTS AND DISCUSSION

The relationships between the $g(r)$ and the temperature are shown in Fig. 1. The temperature is 2000, 1 800, 1 600, 1 400, 1 200, 1 000, 800, 600, 400, 300 K in turns from the bottom to the top. It is obvious that there is a maximum of $g_{\mathrm{Fe-Al}}(r)$, which demonstrates that there is much stronger potential between the atom Fe and atom Al; The maximum of first peak of $g_{\mathrm{Al-Al}}(r)$ is less than $g_{\mathrm{Fe-Fe}}(r)$ and $g_{\mathrm{Fe-Al}}(r)$, which means the probability of Fe atoms locating the position of Al atoms' surrounding is much more than that of Al atoms locating the position of Al atoms' surrounding. And with increase of packing density, the phenomenon would become more and more obvious. In the end, the second peak of $g_{\mathrm{Al-Al}}(r)$ is beyond the first peak of $g_{\mathrm{Al-Al}}(r)$.

The fact that the first peak would become bigger and bigger with decreasing temperature indicates that, during the cooling process, the ordering degree of liquid is strengthened and the disordering degree decreased. There is a strong

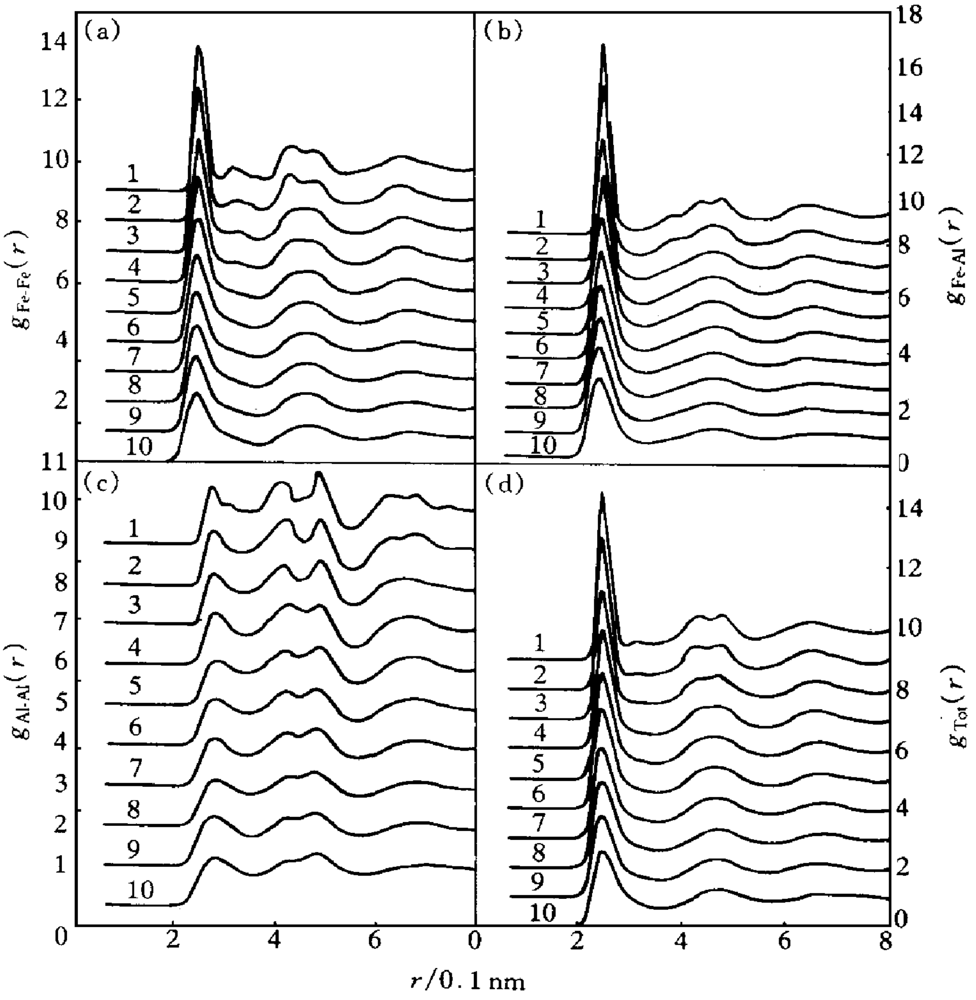

Fig. 1 Distributed functions $g(r)$ at different temperatures
(a) $-g_{Fe-Fe}(r)$; (b) $-g_{Fe-Al}(r)$; (c) $-g_{Al-Al}(r)$; (d) $-g_{Tol}(r)$
1 —300 K; 2 —400 K; 3 —600 K; 4 —800 K; 5 —1 000 K;
6 —1 200 K; 7 —1 400 K; 8 —1 600 K; 9 —1 800 K; 10 —2 000 K

chemical order in liquid metal but not random as thought. The structure of liquid is different at different temperature. It would be in accordance with the thermodynamic rule.

The four kinds of pair distributed functions $g(r)$ tell us that the second peaks of $g_{Fe-Fe}(r)$, $g_{Fe-Al}(r)$ and $g_{Tol}(r)$ have been split, which is the typical characteristic of amorphism formation. The phenomenon indicates some amorphism has formed in melt. The little peak between the first peak and second peak becomes clear with the temperature decreasing. This result means a part of crystal has formed accompanying with the formation of amorphism.

The bond orientational order parameters, which are sensitive to the icosahedra in different temperature during the rapid solidification are shown in Fig. 2. The results of simulation show that the Q_6 and W_6 which are sensitive to the icosahedra become bigger with decrease temperature (Tot means total, Def means Defect, Icos means icosahedra), which means the icoshedral ordering of liquid Fe_3Al becomes strong during the rapid solidification.

When the temperature is 300 K, the Tot Q_6 and Tot W_6 are suddenly equal to zero, which means the crystal appears in melt. The relation between the temperature and 1551, which is sensitive to icosahedra, is the same as the Tot Q_6 and Tot W_6. (see Fig. 3(a)).

The various kinds of bond pairs in different temperatures are shown in Fig. 3. It is obvious

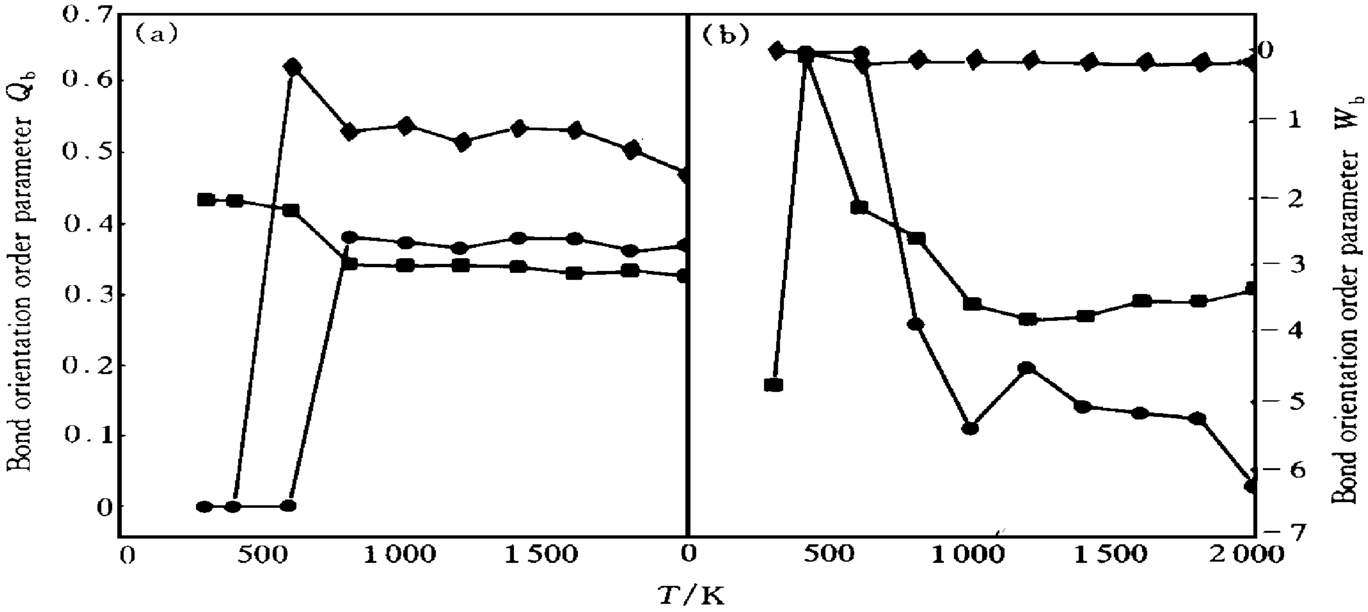

Fig. 2 Bond orientational order parameters at different temperatures
(a) — Q_6; (b) — W_6; ■—Tot; ●—Def; ◆—Icos

that the proportion of 1422 which is related to hcp and the proportion of 1421 which is related to fcc become small with decreasing temperature. But when the temperature is 750 K or so, the proportion of 1422 and 1421 all become big suddenly. The hcp pairs become more quickly than fcc pairs, which means the dynamic conditions are very important in the liquid structure.

It is interesting that the structural energy of hcp is not much more different than that of fcc, but the proportion of hcp increases more quickly than that of fcc when the temperature is over 750 K. It is possible that hcp structure is more stable than fcc one during rapid solidification. The proportion of 1441 and 1661 bond pairs which are related to bcc structure increases with decreasing temperature. But the proportions of 1441 and 1661 are suddenly equal to zero when the temperature is 750 K or so. Some normal structure pairs, for example, 1431, 1321, 1311 in disordered system decrease with increasing temperature. The proportion of 1551 pair which is related to the icosahedra decreases with decreasing temperature, which means the ordered degree of icosahedra becomes strong during the rapid solidification.

The results of structure analysis show that the version of structure is continous when the

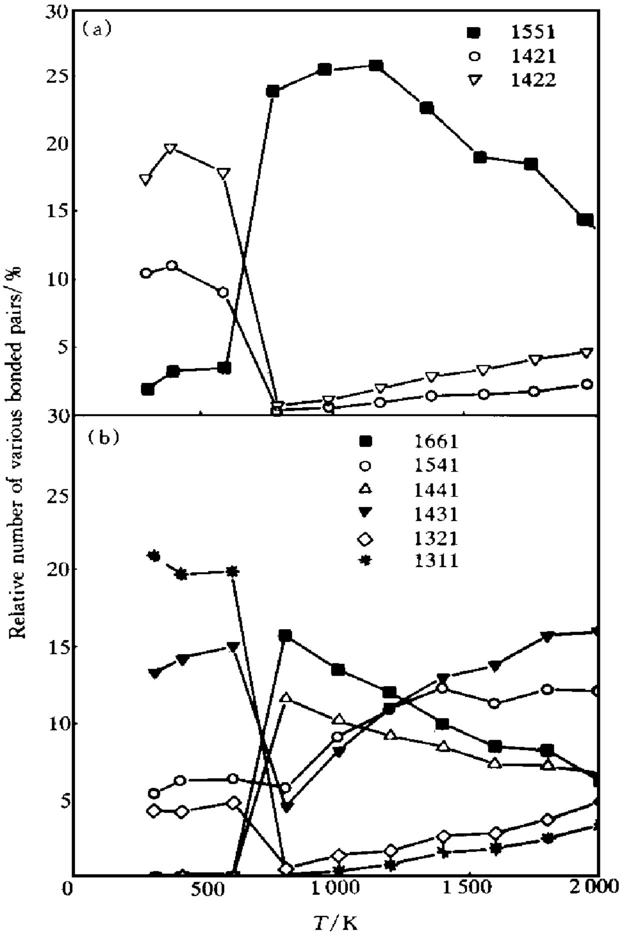

Fig. 3 Number of bond pairs at different temperatures
(a) —1551, 1421, 1422;
(b) —1661, 1541, 1441, 1431, 1301, 1311

temperature is over 750 K, although the high cooling rate has been adopted. There is a sudden change of liquid structure when the temperature is 750 K. Some amorphism has been formed in system. All results show that there is a difference between liquid and amorphism, amorphism is not freezing liquid, but special structure.

6 CONCLUSION

The tight binding potential can be used to predict the liquid structure and version of amorphism effectively. The results of structure analysis demonstrate that during rapid solidification, the icosahedra order is strengthened and inclination of the formation of hcp structure is much more than that of fcc, which is influenced by dynamic factors. The version of structure is continous when the temperature is over 750 K. But when the temperature is 750 K, the structure has a sudden version, which means the formation of amorphism. The ordered degree of amorphism is much higher than that of liquid and amorphism is not cooled liquid.

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