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Medium-range order in Al-based liquid binary alloys

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ABSTRACT

A short-to-intermediate range order of liquid Al–Cu binary alloys has been studied by X-ray diffraction and the reverse Monte Carlo modeling. The comprehensive analysis of three-dimensional models of the liquid Al–TM (TM = Co, Ni, Cu) binary alloys has been performed by means of Voronoi–Delaunay method. The existence of a prepeak in the S(Q) function of the liquid alloys reached with Al is caused by a specific arrangement of 3d-transition metal atoms in dense packed polytetrahedral clusters. It has been found that the icosahedral short-range order in dense polytetrahedral clusters and chemical short-range order lead to the formation of the medium range order in the investigated melts.

1. Introduction

A comparison of a local atomic structure of liquid metallic alloys and corresponding quasicrystalline (QC) phases as well as the systematic study of structural prerequisites of the QC formation are the most important directions in developing a scientific base of QC materials preparation. A great number of the QC materials can be produced from the Al-based liquid binary Al–TM (TM – 3d-transition metal) and ternary Al–TM_1–TM_2 alloys [1,2]. Therefore, the study of interaction between a short and medium-range orders in liquids, quasicrystals and approximants is particularly relevant to quasicrystal-forming Al-based systems.

The crystalline state is characterized by the long-range order (LRO), i.e. by correlations between the positions of each two atoms located as far as possible from one another. Conversely, the non-crystalline state (like liquid or amorphous metallic alloys) is characterized by the short-range order (SRO), which is defined by the interatomic correlations in the first coordination sphere of an arbitrary atom. The intermediate range order or medium range order (MRO) are defined by the order extending to larger nearest interatomic distances in the non-crystalline state than those corresponding to SRO [3]. Usually the X-ray diffraction data give structural information only about short-range order (<3 Å). Sometimes there is a 'prepeak' before the main peak in the structure factor curves, which indicates the ordering beyond the first few nearest neighbor's distances.

The presence of prepeaks in the structure factors of the liquid Al-based alloys with transition metals was reported in [4–11]. Prepeaks observed in the structure factors of liquid alloys at small Q-values are generally attributed to the medium-range order in liquid alloys [8–10]. The nature of prepeaks in the SF's of liquid Al–Ni, Al–Cu, Al–Fe alloys was explained in [5,6] as the result of atomic clusters, whose composition corresponded to the stoichiometry of the solid intermetallide Al_3Ni, Al_2Cu, Al_5Fe_2, respectively. In contrast, Maret et al. [7] explained the existence of a prepeak for liquid $Al_{80}Ni_{20}$ alloy as attributed to Ni–Ni pairs and interpreted as belonging to a superstructure that originated from Al–Ni interactions. As it was shown by the molecular dynamics (MD) simulations of the liquid binary Al–Ni alloys [10], the prepeak manifested itself in the partial structure factors, $S_{NiNi}(Q)$, for the Al-rich compositions and in the $S_{AlAl}(Q)$ factors for the Ni-rich compositions. Holland–Moritz et al. [8] investigated undercooled $Al_{13}(Co,Fe)_4$ melts by neutron diffraction isomorphously substituting of Fe for Co. The Faber–Ziman structure factors $S_{AlAl}(Q)$, $S_{AlTM}(Q)$ and $S_{TMTM}(Q)$ were obtained from three neutron scattering examinations with different scattering contrast of the components. The prepeaks appeared in the $S_{TMTM}(Q)$ curves for low Q-values. The neutron scattering studies of the liquid $Al_{13}(Co_xFe_{1-x})_4$ (with $x = 1$, 0.75, 0.5, 0.25, 0) alloys [9] pointed to the prepeak existence in the partial structure factor $S_{TMTM}(Q)$.

Holland-Moritz et al. [8] and Schenk et al. [9] considered the prepeaks as the signature of a chemical order. The formation of Al–TM pairs as the nearest neighbours is preferred, while TM–TM pairs are avoided. The icosahedral short-range order is accompanied by a strong chemical order such that the first coordination sphere of a TM atom consists preferentially of Al atoms.

The results of X-ray diffraction studies and simulations of local atomic structure in the liquid Al–Co and Al–Ni binary alloys were

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