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STRUCTURE CHANGES IN $In_{20.49}Ga_{66.96}Sn_{12.55}$ EUTECTIC MELT UPON ALLOYING WITH Ni

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Abstract. The structure of an $In_{20.49}Ga_{66.96}Sn_{12.55}$ molten eutectic alloy and $In_{20.49}Ga_{66.96}Sn_{12.55}$+5:10 at.% Ni liquid mixtures have been studied by means of the X-ray diffraction method at different temperatures. The structure factors and pair correlation functions are obtained and analyzed. It is shown that an addition of Ni to a molten eutectic alloy promotes the chemical ordering of the structure.

1. INTRODUCTION

Ternary eutectic alloys are less studied in comparison with binary ones. However, interest in these alloys has increased during the last two decades due to their application in nanocomposite systems, particularly in ferrocolloidal suspensions. Ferrocolloidal suspensions attract attention of researches due to their application as magnetic liquids [1]. As these materials belong to nanocomposite systems they are interesting also from a fundamental viewpoint [2].

Nowadays various types of materials are used as matrices of such nanocomposites, however most of them are non-metallic materials. At the same time, in many cases, it is necessary to have a metallic matrix. Certainly, the low melting temperature is one of the most important characteristics of a metallic matrix. Thus, ternary eutectic alloys on the base of Ga are most suitable for this purpose. On the other hand, improving eutectic alloys properties by an addition of other elements has not been studied in a liquid state. This process is known as a modification of eutectics and it is related to the structure transformation before solidification.

Due to above mentioned facts the influence of Ni additions on the structure of an $In_{20.49}Ga_{66.96}Sn_{12.55}$ eutectic melt has been studied in this work by means of the X-ray diffraction method.

2. EXPERIMENTAL

The samples were prepared in an arc melting furnace filled with pure argon. The purity of the initial metals was 99.999%. The diffraction studies were carried out using a high-temperature diffractometer with a special attachment that allows solid and liquid samples to be investigated at different temperatures up to 1800K. Cu-K_α radiation monochromatized by means of a LiF single crystal as a monochromator and Bragg-Brentano focusing geometry was used. The scattered intensities as a function of the scattering angle were recorded within the range of 1 $Å^{-1} < k < 7Å^{-1}$, with a different angular step which was equal to 0.05° within the principal peak region and 0.5° at the rest values of wave vectors. The measuring of scattered intensity was done with an accuracy better than 2%. The scan time was equal to 100 s in order to obtain more accurate scattered intensities. The diffracted intensity was recorded

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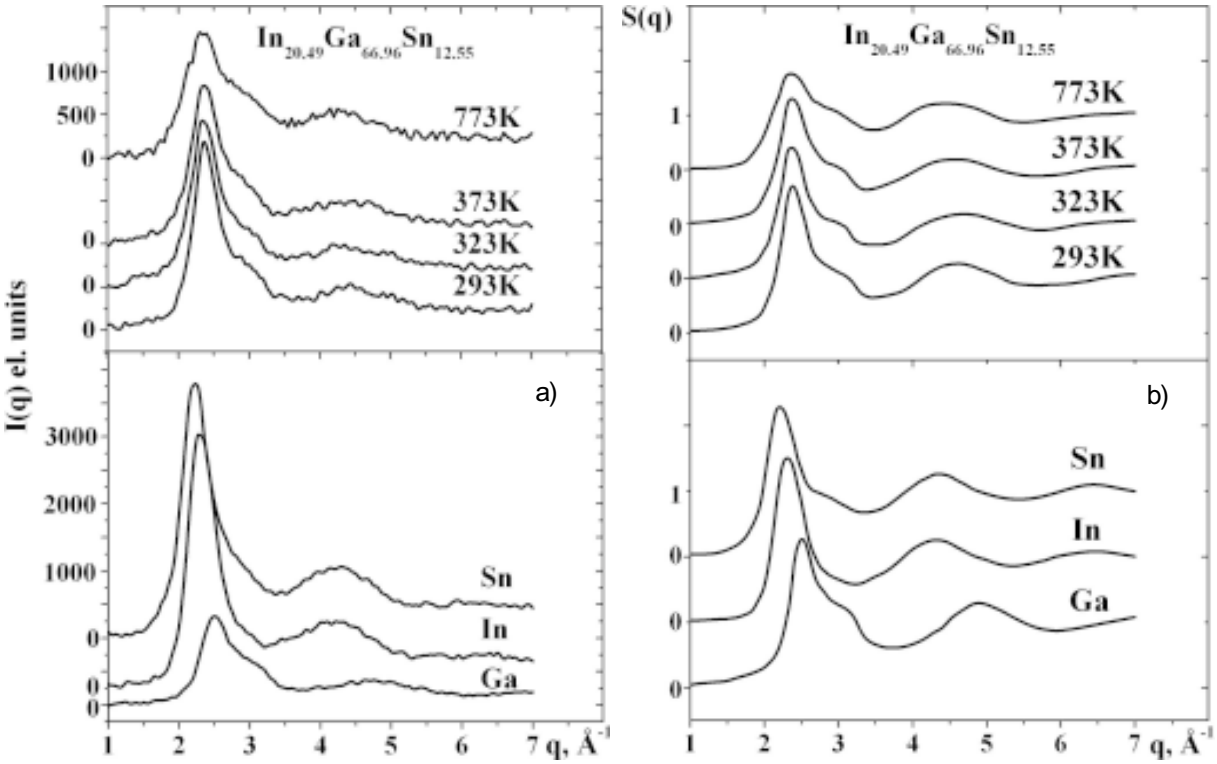

Fig. 1. Experimental X-ray scattering intensity functions (a) and structure factors (b) for liquid In, Ga, Sn, and $In_{20.49}Ga_{66.96}Sn_{12.55}$.

using a NaI(Tl) scintillator detector in conjunction with an amplification system. The sample was placed in a rounded cup, 20 mm in diameter. The intensity curves were corrected on polarization, absorption and incoherent scattering [3]. After this procedure, they were normalized to electron units by the Krogh-Moe method [4]. The obtained intensity curves were used to calculate the structure factors (SF), and then, the Pair Correlation Function (PCF). The main structure parameters, obtained from the SF and PCF, were analyzed.

3. RESULTS AND DISCUSSION

A liquid $In_{20.49}Ga_{66.96}Sn_{12.55}$ eutectic alloy has been studied by the X-ray diffraction method and viscosity measurements [5,6]. It is shown that its structure is inhomogeneous and there are no structural units where all kinds of atoms are randomly distributed. In case of liquid eutectic + ferromagnetic powder of a nanoscale size it can be supposed that an active interaction between them occurs which is due to two reasons: a high chemical activity of powder surface atoms and an In(Ga, Sn)-ferromagnetic element difference. For that reason, it is of large importance to study the influence of 3d-elements on the structure of this eutectic melt.

The structure of $(In_{0.2049}Ga_{0.6696}Sn_{0.1255})_{1-x}Ni_x$ molten alloys has been studied in this work. As is shown in Fig. 1, the scattering intensities and SF of a molten eutectic alloy at different temperatures are different from those for each element (In, Ga, Sn). The shoulder on the right hand side of the principal peak is similar to that of liquid Sn and Ga. On the other hand, the principal peak position is shifted significantly to low *q*-values (Table 1.) although the content of Ga in the eutectic melt prevails. If the In-atoms are diluted in Ga forming structural units with a Ga-like atomic arrangement, the principal peak position should be the same as in liquid Ga. In fact, a shift to low wave vector values can be observed.

The most probable interatomic distances r_1 and r_2 are larger than the corresponding values for Ga and significantly smaller that those for liquid In and Sn. The number of neighbours Z is lower in comparison with those in liquid components (In, Ga, Sn).

The specific atomic distribution existing at temperatures near the melting point temperature persists within some temperature range. As can be seen from Fig. 1 and Table 1, the main structure parameters show no significant changes within the 293-773K range except for the SF principal peak

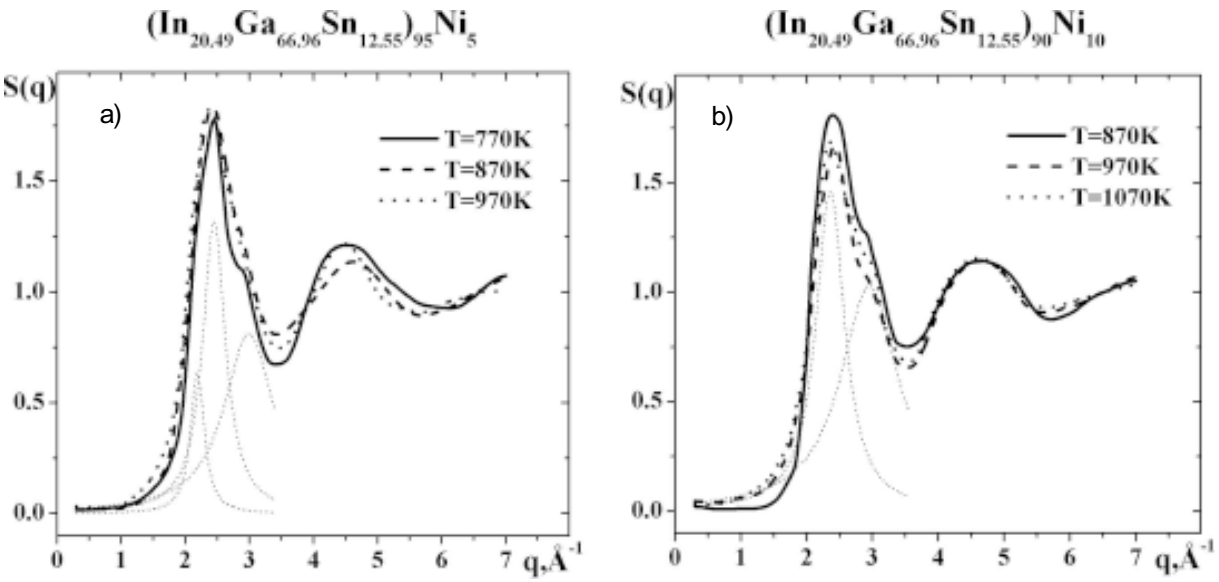

Fig. 2. Structure factors change upon addition of Ni: (a) 5 at.%; (b) 10 at.%.

Table 1. Main structure parameters for an $In_{20.49}Ga_{66.96}Sn_{12.55}$ liquid eutectic alloy.

T,K	q_1	q_2	$S(q_1)$	r_1	r_2	Z
293	2.37	4.61	2.70	3.00	5.80	9.5
323	2.37	4.70	2.39	3.02	5.89	9.8
373	2.36	4.59	2.30	3.04	5.82	10.1
773	2.35	4.43	1.75	3.08	5.82	10.0
Ga	2.52	4.9	2.29	2.82	5.6	10.4
In	2.30	4.32	2.51	3.23	6.10	11.6
Sn	2.21	4.33	2.29	3.23	6.3	10.9

Table 2. Main structure parameters for an $In_{20.49}Ga_{66.96}Sn_{12.55}$ liquid eutectic alloy with Ni.

At.%Ni	T,K	s_1, Å^{-1}	s_2, Å^{-1}	$a(s_1)$	r_1, Å	r_2, Å^{-1}	Z
0	293	2.37	4.61	2.70	3.00	5.80	9.5
5	770	2.45	4.51	1.78	2.99	6.14	9.3
5	870	2.42	4.52	1.83	3.01	6.37	9.8
5	970	2.38	4.47	1,82	3.03	6.47	10.6
10	870	2.39	4.64	1.81	2.99	6.11	10.1
10	970	2.40	4.63	1.67	3.02	6.23	10.1
10	1070	2.36	4,61	1.69	3.03	6.51	10.4

height $S(q_1)$ which drastically decreases at T=773K. Such a decrease is commonly attributed to the formation of a low packing density structure. At the same time a slight increase of Z can be seen which indicates some growth of the atomic density in the first coordination sphere. Therefore, in order to agree these experimental facts we may suppose that structural units decrease their size with temperature revealing thermal disordering inside. Using the model interpretation methods of structure parameters we find that better agreement with the experimental data is in the assumption that Ga and Sn form random solution units whereas In-atoms are self-associated. This fact is in accordance with the results obtained in [7,8] for binary Ga-Sn and Ga-In alloys. Some increase in the number of neighbours

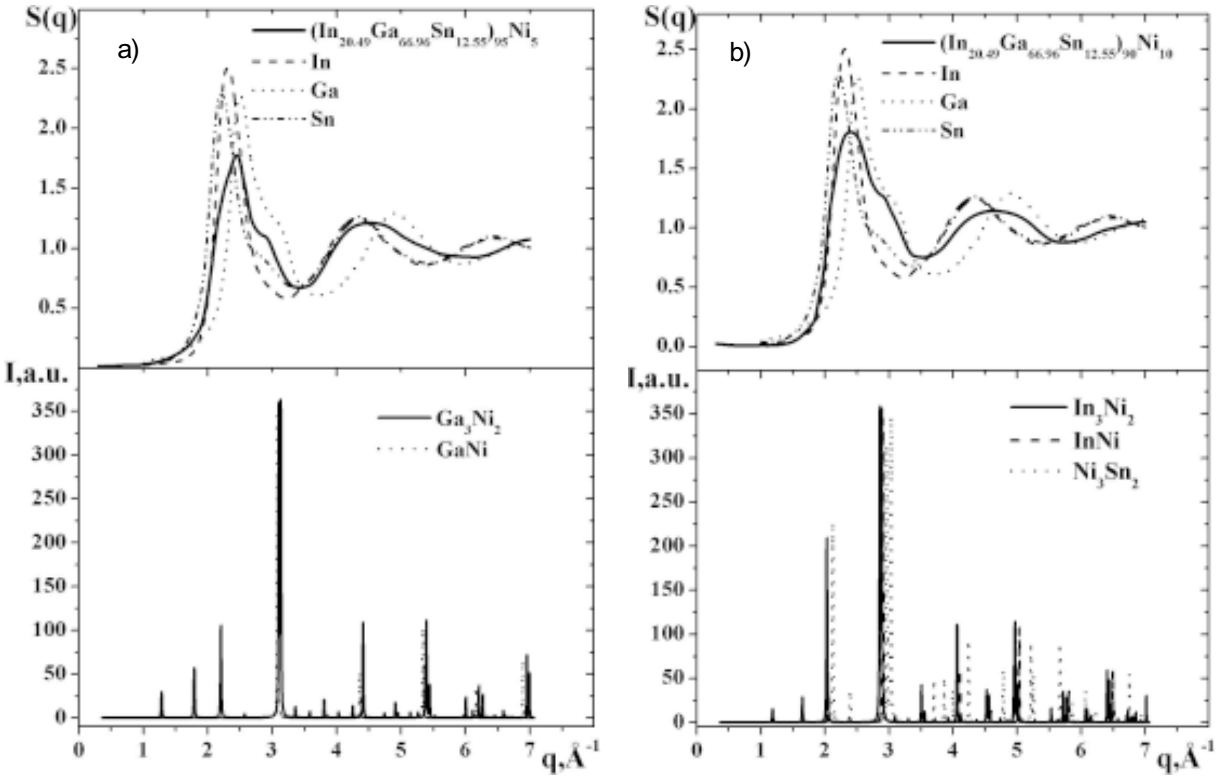

Fig. 3. Comparison of structure factors for $(In_{20.49}Ga_{66.96}Sn_{12.55})_{1-x}Ni_x$ with diffraction patterns for intermetallics: (a) 5 at.%; (b) 10 at.%.

is considered to result from the first coordination sphere broadening.

The changes in the SF profile with an addition of Ni are shown in Fig. 2. The SF changes significantly with an addition of 5 at.% of Ni. Namely, the principal peak shifts to large *q*-values and its height becomes smaller. A shoulder on right hand side of the structure factor principal peak is also observed. The most probable interatomic distance is in fact the same as well as the number of neighbours (Table 2). An increase in the Ni content up to 10 at.% is accompanied by a further shift of the principal peak to large *q*-values and its broadening.

It is important to compare the temperature dependences of the structure parameters for these two molten alloys with Ni and compare them with those for liquid $In_{20.49}Ga_{66.96}Sn_{12.55}$. It can be seen (Fig. 2.) that in case of melts with Ni the shoulder disappears with heating more quickly than in case of an eutectic melt.

In order to explain the behaviour of the shoulder and its relation to the atomic distribution, the principal peak has been interpreted as a sum of Gaussian-like maxima. In case of $In_{20.49}Ga_{66.96}Sn_{12.55}$+5 at.% Ni the main maximum of the SF is a sum of three subpeaks whose positions correspond to three kinds of structural units. It is possible to analyse these positions and compare them with similar parameters for various phases which exist in the structure of melts. Such analysis follows from the equilibrium phase diagram. We suppose that the first of these subpeaks corresponds to In-In clusters. The most intensive maximum describes the random atomic distribution of Ga and Sn atoms. The last of the subpeaks is supposed to be attributed to chemically ordered clusters with Ga_3Ni_2, GaNi, InNi, and Ni_3Sn_2-like structures. This is confirmed by comparing the structure factors for $(In_{20.49}Ga_{66.96}Sn_{12.55})_{1-x}Ni_x$ with diffraction patterns for intermetallics: (a) 5 at.%; (b) 10 at.%. (Fig. 3). It is difficult to say on the basis of the diffraction data only which of these intermetallics dominate in the formation of chemical ordering in the atomic arrangement.

It should be also noted that the width of subpeaks is different resulting in a different size of structural units (clusters). The size of chemically ordered structural units on the base of intermetallics is significantly smaller than the size of In-In and Ga-Sn units. The subpeak corresponding to In-In self-associated atomic groups disappears in a molten alloy, con-

taining 10 at.% Ni. Thus, it can be supposed that the tendency to forming chemically ordered structural units increases in case of a larger Ni content.

4. CONCLUSIONS.

The structure of a liquid $In_{20.49}Ga_{66.96}Sn_{12.55}$ eutectic melt shows an inhomogeneous structure in a wide temperature range above the melting point.

Our data suggest that an addition of Ni-atoms to molten eutectics results in an increase in the chemical ordering of the structure and in formation of chemically ordered structural units with an intermetallics-like atomic arrangement. The size of these atomic groups is significantly smaller than the size of In-In and Ga-Sn clusters, existing also in the structure of melts.

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