Partial Atomic Distribution Functions of Liquid $Fe_{75}B_{25}$

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Partial structure factors and atomic distribution functions for liquid $Fe_{75}B_{25}$ were determined for the Fe–Fe and Fe–B pairs by means of X-ray and neutron diffraction. The experimental results show a strong chemical short-range order in the melt which is similar to that of amorphous $Fe_{75}B_{25}$.

Key words: liquid Fe–B – X-ray, neutron diffraction on liquid Fe–B – partial structure factors of liquid Fe–B – partial atomic distribution functions of liquid Fe–B – short-range order in liquid Fe–B.

1. Introduction

Until now only total atomic distribution functions of liquid Fe–B alloys were obtained by means of X-ray [1–3] and neutron diffraction [4]. This paper reports on the determination of partial structure factors of a Fe–B melt by combining X-ray and neutron diffraction. From the experimental results information is got concerning the chemical short-range order in liquid $Fe_{75}B_{25}$. On the other hand, the amorphous $Fe_{75}B_{25}$ alloy can be prepared by the rapid quenching technique directly from the melt. The determination of partial radial distribution functions of amorphous $Fe_{75}B_{25}$ [5] and $Fe_{80}B_{20}$ [6] shows a strong chemical short-range order in the Fe–B glass. If the amorphous state is assumed to be a frozen in liquid it should be interesting to compare not only the total (see [3]) but also the partial pair correlation functions of liquid and amorphous $Fe_{75}B_{25}$.

2. Theoretical Background

From the diffraction experiment the total structure factor $S(Q)$ is extracted ($Q = 4\pi \sin \vartheta/\lambda$). According to the Faber-Zimann definition [7], $S(Q)$ is related to the coherent elastic scattering per atom I_{coh} by

$$S(Q) = (I_{\text{coh}} - \langle b^2\rangle + \langle b\rangle^2)/\langle b\rangle^2 . \qquad (1)$$

For a two-component alloy the mean values are given by $\langle b\rangle = c_A b_A + c_B b_B$ and $\langle b^2\rangle = c_A b_A^2 + c_B b_B^2$, where c_i is the atomic concentration and b_i the coherent scattering length in the case of neutron diffraction or the atomic scattering factor $f_i(Q)$ in the case of X-ray diffraction. In the latter case the scattering lengths are Q-dependent. The total structure factor $S(Q)$ represents a weighted sum of the partial structure factors $S_{ij}(Q)$ which is given by the following equation for a binary alloy:

$$\begin{aligned} S(Q) &= \sum_i \sum_j W_{ij} S_{ij}(Q) \\ &= \frac{1}{\langle b\rangle^2} [c_A^2 b_A^2 S_{AA}(Q) + 2 c_A c_B b_A b_B S_{AB}(Q) \\ &\qquad + c_B^2 b_B^2 S_{BB}(Q)] . \end{aligned} \qquad (2)$$

In order to evaluate $S_{ij}(Q)$, three independent diffraction measurements are required with considerable differences in the weighting factors W_{ij}. From the partial structure factors $S_{ij}(Q)$ the partial atomic distribution functions $G_{ij}(r)$ can be evaluated by the Fourier transform:

$$G_{ij} = \frac{2}{\pi} \int_0^{Q_{\max}} Q\,[S_{ij}(Q) - 1] \cdot \sin(Q r)\, \mathrm{d}Q . \qquad (3)$$

From the partial atomic distribution functions partial coordination numbers N_{ij} (number of j atoms in the coordination shell around an i atom) can be calculated by

$$N_{ij} = c_j \int_{r_1}^{r_u} [r\, G_{ij}(r) + 4\pi r^2 \varrho_0]\, \mathrm{d}r . \qquad (4)$$

In this equation ϱ_0 represents the mean atomic number density.

3. Experimental

The neutron diffraction experiments on liquid $Fe_{75}B_{25}$ were performed at a temperature of 1600 K.

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The experiments were described in detail in [4]. The neutron diffraction patterns were recorded from $Q = 2.5\ \mathrm{nm}^{-1}$ up to $Q = 89\ \mathrm{nm}^{-1}$.

The X-ray diffraction measurement was performed at a temperature of 1573 K on a liquid sample with the same nominal composition. The structure factor was determined from this experiment in the Q-range $7\ \mathrm{nm}^{-1} \leqq Q \leqq 125\ \mathrm{nm}^{-1}$. For other experimental details cf. [3].

Although there was a small difference in the temperatures of the melts for neutron and X-ray diffraction it seems possible to use the data for the estimation of the partial structure factors. As shown by the X-ray and neutron diffraction experiments on liquid $Fe_{75}B_{25}$ [3, 4] there is no strong temperature dependence of the structure factor in this temperature region.

The density of liquid $Fe_{75}B_{25}$ was estimated to be $\varrho_0 = 90.4\ \mathrm{atoms/nm}^3$ for a temperature of 1600 K [4].

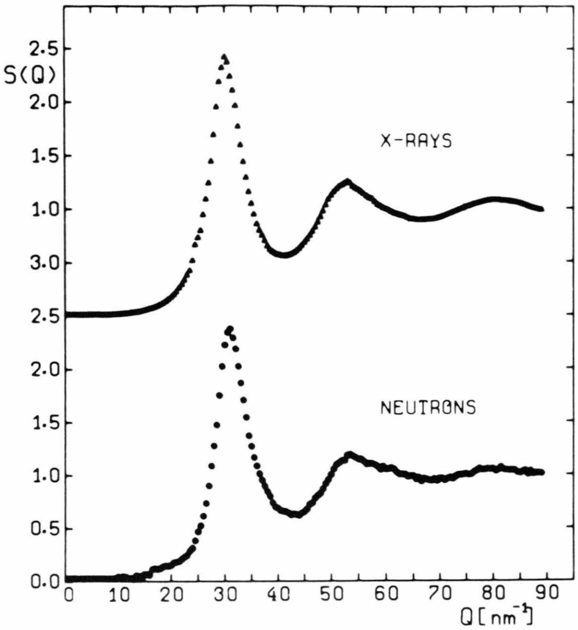

Fig. 1. Total structure factors from the neutron and the X-ray diffraction experiments on liquid $Fe_{75}B_{25}$.

4. Results and Discussion

The obtained total structure factors $S(Q)$ for the two independent experiments are shown in Figure 1. The X-ray data are limited here to the Q range of the neutron diffraction experiments.

From the experimentally determined total structure factors the partial structure factors $S_{ij}(Q)$ can be estimated. According to (2) we get for our experiments:

$$S_N(Q) = 0.660\, S_{FeFe}(Q) + 0.305\, S_{FeB}(Q) + 0.035\, S_{BB}(Q),$$

$$S_X(Q) = 0.883\, S_{FeFe}(Q) + 0.113\, S_{FeB}(Q) + 0.004\, S_{BB}(Q). \quad (5)$$

The coefficients W_{ij} for the X-ray experiment are given here for $Q = 0$. The very small weighting factor of $S_{BB}(Q)$ indicates that this contribution may be neglected permitting errors in the same order of magnitude as the experimental ones. So the partial structure factors $S_{FeFe}(Q)$ and $S_{FeB}(Q)$ were evaluated from the modified Eq. (5) with Q-dependent W_{ij} values for the X-ray equation. The results are shown in Figure 2. The general behaviour of the partial structure factors in the melt is similar to that of the amorphous state according to [5] and [6]. But the amplitudes of the modulations are smaller, as it was expected from the higher mobility of atoms in the liquid state. The iron-iron partial structure factor is very similar to the total

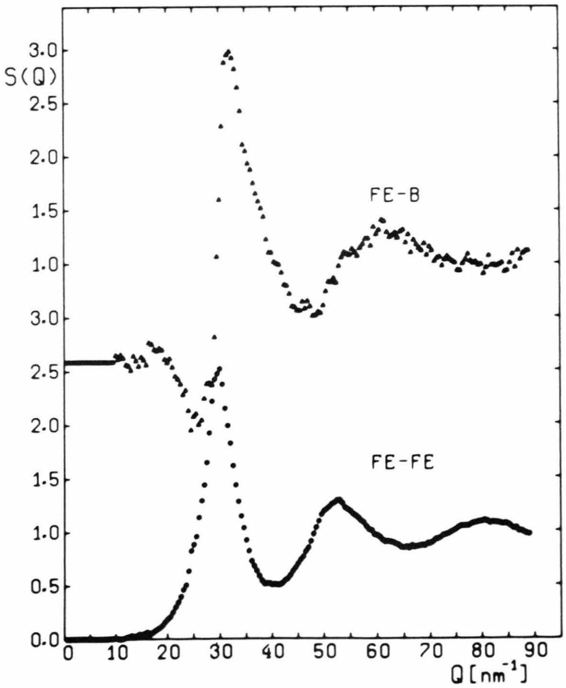

Fig. 2. Partial structure factors for the Fe-Fe and Fe-B pairs of liquid $Fe_{75}B_{25}$.

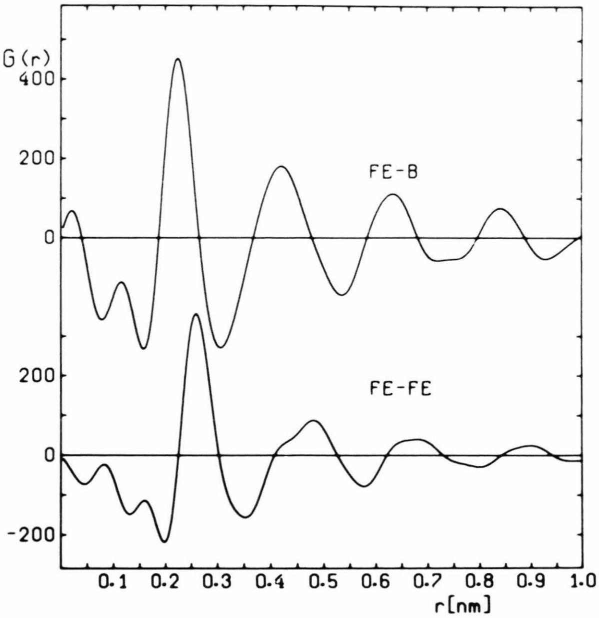

Fig. 3. Partial atomic distribution functions $G_{ij}^{(r)}$ for liquid $Fe_{75}B_{25}$.

Table 1. Partial nearest neighbour distances and coordination numbers of $Fe_{75}B_{25}$.

	r^1_{FeFe}/nm	N^1_{FeFe}	r^1_{FeB}/nm	N^1_{FeB}
Liquid $Fe_{75}B_{25}$ (this paper)	0.260	11.2 ± 0.3	0.225	2.8 ± 0.3
amorphous $Fe_{75}B_{25}$ [5]	0.260	11.4 ± 0.6	0.218	2.7 ± 0.2
tetr.b.c. Fe_3B	0.261	10.7	0.222	3.0
DRP-model $Fe_{76}B_{24}$ [12]		9.7		2.0

structure factor from the X-ray experiments, since its intensity is dominated by the scattering from iron. Compared to the amorphous state, the main maximum is shifted to lower Q-values ($Q_1 = 30\ \mathrm{nm}^{-1}$) and the second maximum exhibits no shoulder at the high Q side; only a slight asymmetry is visible. The main maximum of the iron-boron partial structure factor is shifted to higher Q-values in comparison with $S_{FeFe}(Q)$, and a broad second maximum around $65\ \mathrm{nm}^{-1}$ occurs. A prepeak with a dublette structure between 10 and $25\ \mathrm{nm}^{-1}$ like that in the amorphous state was not observed for $S_{FeB}(Q)$. From the partial structure factors the partial atomic distribution functions $G_{ij}(r)$ were calculated according to (3). The resulting curves are shown in Figure 3. The position of the first maximum in $G_{ij}(r)$ and the first neighbour coordination number N_1^{ij} are given in Table 1. The partial atomic correlation function of iron-boron pairs is different from that of iron-iron pairs and indicates a high degree of chemical ordering in liquid $Fe_{75}B_{25}$. Boron atoms have about 9 iron nearest neighbours at smaller interatomic distance than in case of the iron-iron first neighbours. The correlation for iron-boron pairs at higher distances is relatively strong.

Cargill and Speapen [8] introduced a parameter, η_{AB}, to describe the degree of chemical order in binary amorphous materials based on the partial coordination numbers N_{ij}. For statistical distribution, $\eta_{AB} = 0$, for compound-forming tendency, $\eta_{AB} > 0$, and for phase separation tendency $\eta_{AB} < 0$. According to [8] the short-range order parameter for liquid $Fe_{75}B_{25}$ can be calculated as follows

$$\eta_{FeB} = \frac{N^1_{FeB}\langle N\rangle}{c_B N_{Fe} N_B} - 1 \qquad (6)$$

with $\langle N\rangle = c_{Fe} N_{Fe} + c_B N_B$, $N_{Fe} = N^1_{FeFe} + N^1_{FeB}$ and $N_B = N^1_{BFe} + N^1_{BB}$. With the determined values given in Table 1 we obtain $\eta_{FeB} = 0.200$ ($N^1_{BB} = 0$). This means that in liquid $Fe_{75}B_{25}$ there is a preferred compound-forming tendency. For the normalized short-range order parameter $\eta^0_{FeB} = \eta_{FeB}/\eta^{max}_{FeB}$, which is normalized to the maximum degree of order $\eta^{max}_{FeB} = c_B N_B/c_{Fe} N_{Fe}$, the calculation yields $\eta^0_{FeB} = 1.0$. This points to maximum chemical short-range order in liquid $Fe_{75}B_{25}$.

The parameters of the first neighbourhood of liquid $Fe_{75}B_{25}$ are compared in Table 1 with those of amorphous $Fe_{75}B_{25}$ given in [5]. From Table 1 it is visible that the typical data for the first neighbourhood of liquid and amorphous $Fe_{75}B_{25}$ are very similar. Differences in the G_{ij} curves exist between the liquid and the amorphous state in the shape of the second peaks. In the melt the second peak of G_{FeB} is not split and that of G_{FeFe} shows a reversed shape with a shoulder on the left hand (Figure 3). The comparison of the total structure factors and the total atomic distribution functions of liquid and amorphous Fe-B alloys in [3] indicates that the amorphous state is not a simple frozen liquid. Structural changes arise during rapid quenching of the melt which result in a changed size and connection of structural units.

At present two important conceptual structure moduls for the local atomic order exist for transitionmetal-metalloid (T-M) glasses: (i) dense random packed (DRP) models for binary alloys and (ii) stereochemically defined models. Gaskell [9] reviewed the properties of these models in comparison with the essential experimental results of structural investigations. In spite of the apparent success in reproducing experimental total pair distribution functions, structure models of binary alloys which are based on central force atomic interactions [10–12] fail to reproduce the specific kind of chemical order present in T-M glasses [13]. For instance the number N^1_{FeB} of boron atoms within the first coordination shell around iron atoms predicted by the DRP models [10–12] for amorphous Fe-B alloys is too low. In Table 1 the coordination numbers are given for the DRP model of Takacs [12] which are considerably smaller than the experimental values of amorphous and liquid $Fe_{75}B_{25}$. The stereochemically defined models are based on the assumption of the existence of the same local structural units in the amorphous and crystalline state of the alloy. Amorphous $Fe_{75}B_{25}$ crystallizes to metastable Fe_3B. The short-range order parameters of the Fe_3B phase given in Table 1 show good agreement with the experimental results within the errors. From the comparison of the data we can conclude that in liquid $Fe_{75}B_{25}$ the atomic arrangement within the first neighbourhood is similar to that of metastable Fe_3B.

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