

# Short Range Order in Structural Models for the Amorphous $\text{Fe}_{80}\text{B}_{20}$ - and $\text{Ni}_{81}\text{B}_{19}$ -Alloys

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The short range order in several structural models for amorphous alloys is investigated with respect to pair correlation functions, bond angle distributions and other properties. Despite of the different principles of modelling, the topological short range order turned out to be almost identical. Therefore also the question for the triplet correlations to some extent is found solved. Remaining differences thus consist regarding more complex correlations among the atoms.

## 1. Introduction

During the last years the partial pair correlation functions of binary amorphous alloys have been determined with high accuracy from diffraction experiments making use of contrast variance like the isotopic substitution technique ( $\text{Fe}_{80}\text{B}_{20}$  [1],  $\text{Ni}_{81}\text{B}_{19}$  [2],  $\text{Ni}_{64}\text{B}_{36}$  [3],  $\text{Ni}_{80}\text{P}_{20}$  [4]). From these functions detailed information about the short range order could be derived in terms of atomic distances, coordination numbers, and short range order parameters. This in turn stimulated the development of computer models for the atomic structure of these materials where, however, quite different principles were used for their construction [5, 6].

The motivation for modelling is not only given by the aim of a better understanding of the atomic arrangement as such. The models can also be used to calculate solid state properties, for instance elastic constants, they can be useful for the interpretation of high resolution electron microscope images, spin-spin-interactions, electronic densities of states, simulation of X-ray absorption coefficients in the near edge region (XANES) (see forthcoming publication [7]) and in the extended edge region (EXAFS) [8].

At the present stage a comparison between the different models, using selected criteria as well as comparison with experimental data, seems to be useful. With respect to diffraction experiments, however, it should be noted that pair correlation functions contain only a limited amount of information about the real structure because they represent a one-dimen-

sional as well as averaged description. For a better understanding of the three-dimensional structure of an amorphous system the necessity for information about higher order correlation functions has been pointed out frequently [9]. One simple manifestation of those functions are bond angle distributions, which are part of triplet correlations [10–12]. There are modern experimental techniques which promise access to bond angle distributions like field ion microscopy [13] and X-ray near edge spectroscopy (XANES) [14]. For the application of XANES to investigate the structure of real amorphous alloys and of clusters we refer to a forthcoming paper [7].

In the present study five available structural models for the amorphous alloys  $\text{Fe}_{80}\text{B}_{20}$  and  $\text{Ni}_{81}\text{B}_{19}$  are compared with each other and with experimental data. The short range order of the models is analyzed with respect to bond angles, coordination numbers, and certain local asymmetries.

## 2. Models for Amorphous $\text{Fe}_{80}\text{B}_{20}$ and $\text{Ni}_{81}\text{B}_{19}$ ; Pair Correlations

Table 1 gives a compilation of models presented up to now to describe the partial pair correlation functions of amorphous  $\text{Fe}_{80}\text{B}_{20}$  and  $\text{Ni}_{81}\text{B}_{19}$ , respectively.

For details of the models see the corresponding references. It should be noted that in most of the modelling principles there is a common finishing procedure: After the construction of an initial atomic configuration according to the philosophy of the respective authors a relaxation procedure is applied to the atomic arrangement, which leads to an increase of

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Table 1. Amorphous  $\text{Fe}_{80}\text{B}_{20}$  and  $\text{Ni}_{81}\text{B}_{19}$ . Models. DRPHS = dense random packing of hard spheres, SRO = short range order, MD = molecular dynamics. \* Models used in the present paper. Always only the first author of the first paper is mentioned in the second column.

Year	Author	Alloy	Ref.	Construction principle
1972	Polk	$\text{Fe}_{80}\text{B}_{20}$	[15]	DRPHS
1981	Boudreaux	$\text{Fe}_{80}\text{B}_{20}$	[16]	prescribed SRO
1981	Mattern	$\text{Fe}_{80}\text{B}_{20}$	[17, 18]	prescribed SRO; two phases
1982	Fujiwara *	$\text{Fe}_{80}\text{B}_{20}$	[19]	DRPHS
1982	Blétry	$\text{Ni}_{81}\text{B}_{19}$	[2]	DRPHS
1983	Beyer *	$\text{Ni}_{81}\text{B}_{19}$	[20]	MD simulation
1984	Lewis	$\text{Fe}_{80}\text{B}_{20}$	[21]	DRPHS
1984	Sadoc	$\text{Fe}_{80}\text{B}_{20}$	[22]	prescribed SRO
1985	Hamada	$\text{Fe}_{80}\text{B}_{20}$	[23]	prescribed SRO; two phases
1985	Belashchenko	$\text{Fe}_{80}\text{B}_{20}$	[24]	MD simulation
1985	Dubois *	$\text{Ni}_{81}\text{B}_{19}$	[25]	prescribed SRO
1987	Zweck *	$\text{Fe}_{80}\text{B}_{20}$	[26, 27]	simplified MD simulation
1986	Jing-Han	$\text{Fe}_{80}\text{B}_{20}$	[28]	prescribed SRO; two phases
1987	Brandt *	$\text{Fe}_{80}\text{B}_{20}$	[29]	MD simulation

the density of the structure and to a reduction of the model's free energy.

For the relaxation algorithm pair potentials have to be introduced for the metal-metal-, metal-boron-, and boron-boron-interactions. For the analytical form of these potentials different approaches are in use: Lenard-Jones- and Morse-potentials as well as parabolas of higher order. The isotropic character of the pair potentials may truly be criticized, however at present no viable anisotropic potential approach is available. Even the modern embedded-atom-method [30] is of isotropic nature. Non-central potentials to describe directed chemical bonding for metalloids in amorphous alloys have been applied for a two-dimensional [31], but not yet for a three-dimensional model.

A few comments on some of the models have to be added:

i) Dubois model: Among the different versions of that model, the one which was used for the present study corresponds to the partial pair correlation functions which are shown in Fig. 11 a of [25].

ii) Zweck model: In addition to the final model also a second arrangement was used, which was obtained from the initial configuration, but after just 1/40 of the necessary relaxation steps. In the following it is named "test model". Of course this structure agrees worse with diffraction data, and it may seem useless to dis-

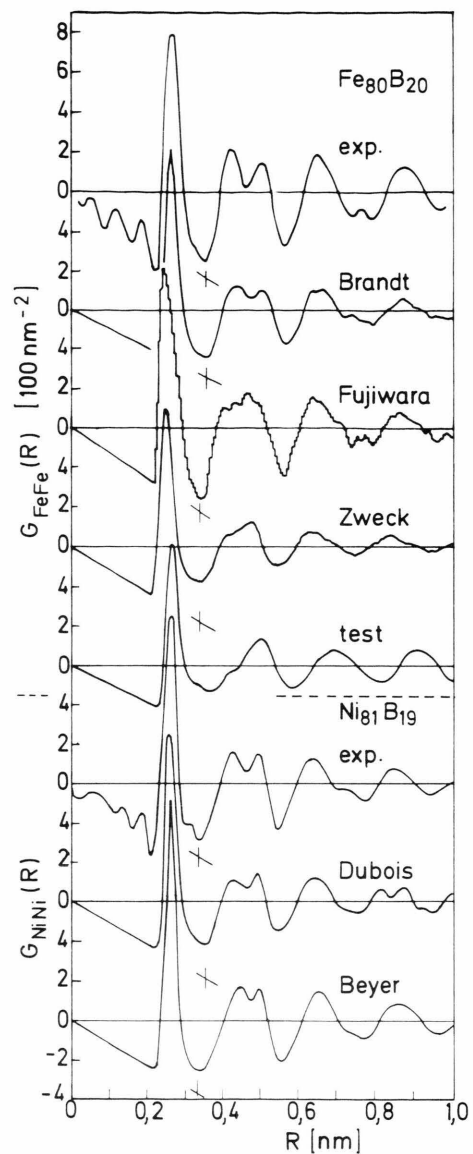


Fig. 1. Amorphous  $\text{Fe}_{80}\text{B}_{20}$  and  $\text{Ni}_{81}\text{B}_{19}$ . Experimental and calculated Faber-Ziman partial pair correlation functions  $G_{\text{FeFe}}(R)$  and  $G_{\text{NiNi}}(R)$ .

cuss it at all. However, for the XANES calculations in [7] this model yielded an interesting insight into the performance of XANES.

iii) Brandt model: The original model of Brandt was set up with full MD simulation which means starting from a configuration where the atoms have a non-zero velocity (i.e. temperature). The relaxation procedure goes parallel to a slowing down of the velocities (i.e.

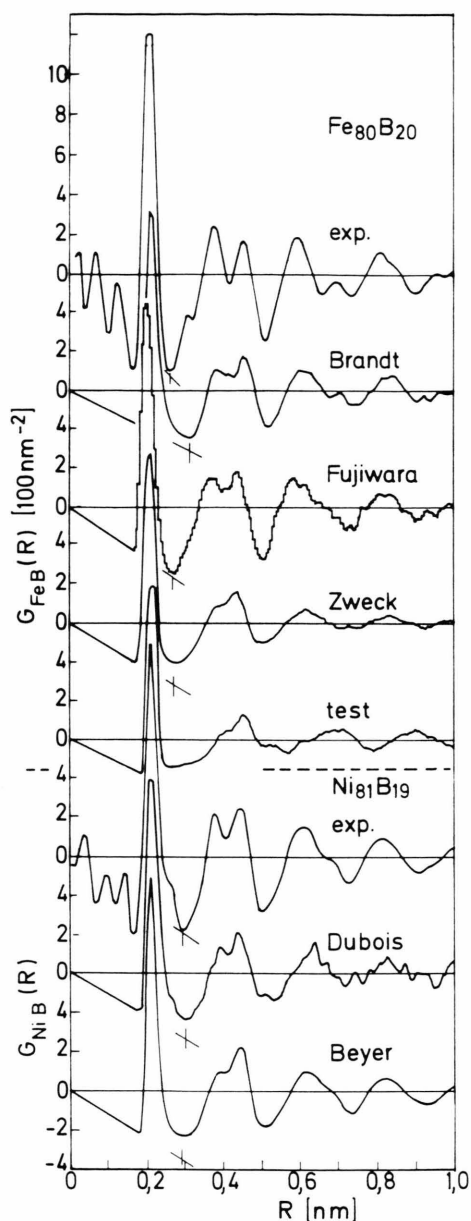


Fig. 2. Amorphous  $\text{Fe}_{80}\text{B}_{20}$  and  $\text{Ni}_{81}\text{B}_{19}$ . Experimental and calculated Faber-Ziman partial pair correlation functions  $G_{\text{FeB}}(R)$  and  $G_{\text{NiB}}(R)$ .

cooling down) of the atoms. The model which was used in the present study was set up with a simplified version of the mentioned computer code. Hereby the atoms have no velocity, which means that the calculation treats a “static” relaxation at zero temperature. Indeed this simplified algorithm leads to a model with

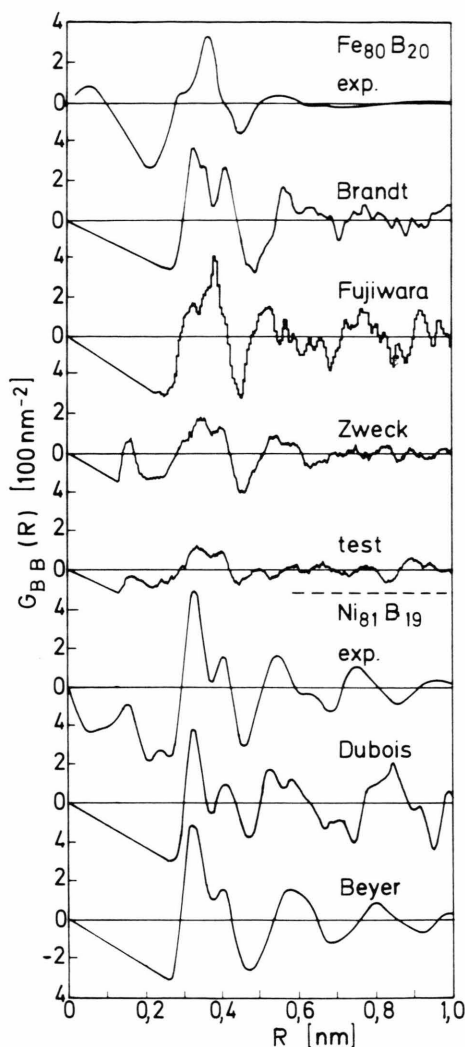


Fig. 3. Amorphous  $\text{Fe}_{80}\text{B}_{20}$  and  $\text{Ni}_{81}\text{B}_{19}$ . Experimental and calculated Faber-Ziman partial pair correlation functions  $G_{\text{BB}}(R)$ .

the same partial pair correlation functions as for the more sophisticated calculation with “real” MD simulation.

In Figs. 1 to 3 we compare the experimental partial pair correlation functions  $G_{\text{MM}}(R)$  ( $M = \text{Fe}, \text{Ni}$ ),  $G_{\text{MB}}(R)$ , and  $G_{\text{BB}}(R)$  with the calculated ones. The following items are of importance:

i) Between calculated and experimental pair correlation functions we observe no drastic differences and thus we can state a more or less satisfactory coincidence.

ii) The slope of the functions at the origin should be  $-4\pi q_0$  with  $q_0 = \text{mean atomic number density}$ . The smaller slopes in the calculated curves show that all models yield too small mean densities. This also is visible from the smaller amplitudes in the  $G(R)$ -functions and in Table 2 from the tendency to smaller coordination numbers.

Table 2 shows the partial coordination numbers and atomic distances.

Since the different models are obviously able to describe the experimental partial pair correlations more or less with the same accuracy, the question arises: Are structural differences, which might in fact exist between the three-dimensional atomic arrangements of the models, washed out by the calculation of the  $G(R)$ 's? Note that this procedure involves *averaging* over the atomic sites as well as one-dimensional *projection*. To examine this question, in the following a more detailed comparative analysis of the models has been carried out where the *distributions* of structural features as well as aspects of *triplet correlations* are considered. This is important with respect to experimental techniques which, in contrast to diffraction methods, promise to be sensitive to those features (XANES [14], FIM [13], etc.).

### 3. Detailed Analysis of the TSRO in the Models

Small angle scattering methods as performed with amorphous  $\text{Fe}_{80}\text{B}_{20}$  [32, 33] and  $\text{Ni}_{81}\text{B}_{19}$  [34], for example, have revealed the existence of medium range fluctuations of density or stoichiometrical composition. Thus we have to assume the existence of two "phases". However, we are surely not too wrong if in the present paper we merge the two "phases" with their very similar or almost identical topological short range order (TSRO) theoretically into one single phase, unless further experiments show the failure of this approximation.

#### 3.1. Distribution in Partial Coordination Numbers

In Fig. 4 we show for six models of amorphous  $\text{Fe}_{80}\text{B}_{20}$  and  $\text{Ni}_{81}\text{B}_{19}$  the probability of occurrence of the partial coordination number  $N_{\text{MM}}$  in the first coordination sphere. We recognize that the individual partial coordination number per site can vary from 6 to 14 atoms and that the different models exhibit different shapes of the distribution curves. Corresponding

Table 2. Amorphous  $\text{Fe}_{80}\text{B}_{20}$  and  $\text{Ni}_{81}\text{B}_{19}$ . Partial coordination numbers and atomic distances.

Alloy	Ref.	Partial coordination numbers				Distances [nm]		
		$N_{\text{MM}}$	$N_{\text{MB}}$	$N_{\text{BM}}$	$N_{\text{BB}}$	$R_{\text{MM}}$	$R_{\text{MB}}$	$R_{\text{BB}}$
Experiments								
Fe <sub>80</sub> B <sub>20</sub>	[1]	12.4	2.16	8.64	6.5	0.257	0.214	0.357
Ni <sub>81</sub> B <sub>19</sub>	[2]	10.8	2.2	9.3	3.6	0.252	0.211	0.329
					3.7			0.402
Models								
Fe <sub>80</sub> B <sub>20</sub>								
Brandt		11.2	2.2	7.7	7.5	agreement with experiment: see Figures 1 to 3		
Fujiwara		11.9	1.9	7.7	6.5			
Zweck		10.9	1.9	6.8	6.5			
“Test”		10.8	1.0	6.3	6.7			
Models								
Ni <sub>81</sub> B <sub>19</sub>								
Dubois		9.7	1.9	7.6	6.5			
Beyer		10.6	2.0	8.6	7.3			

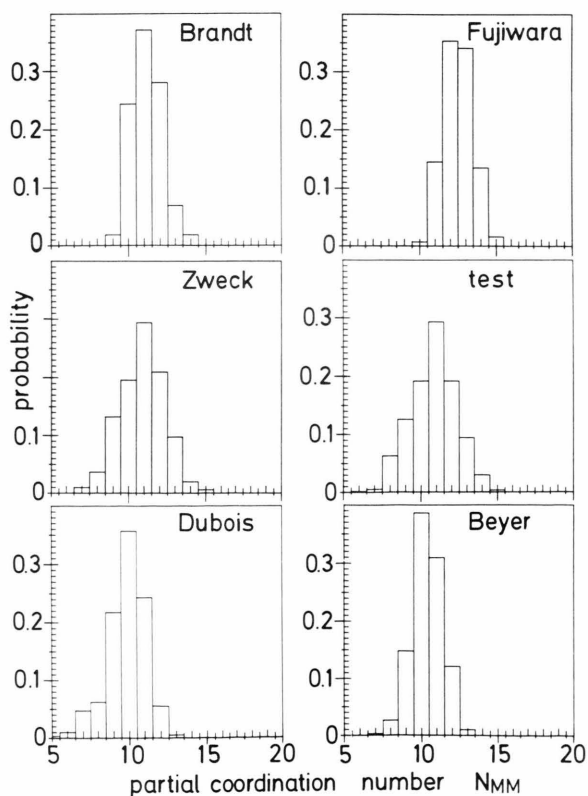


Fig. 4. Amorphous  $\text{Fe}_{80}\text{B}_{20}$  and  $\text{Ni}_{81}\text{B}_{19}$ . Probability of occurrence of the partial coordination number  $N_{\text{MM}}$ .

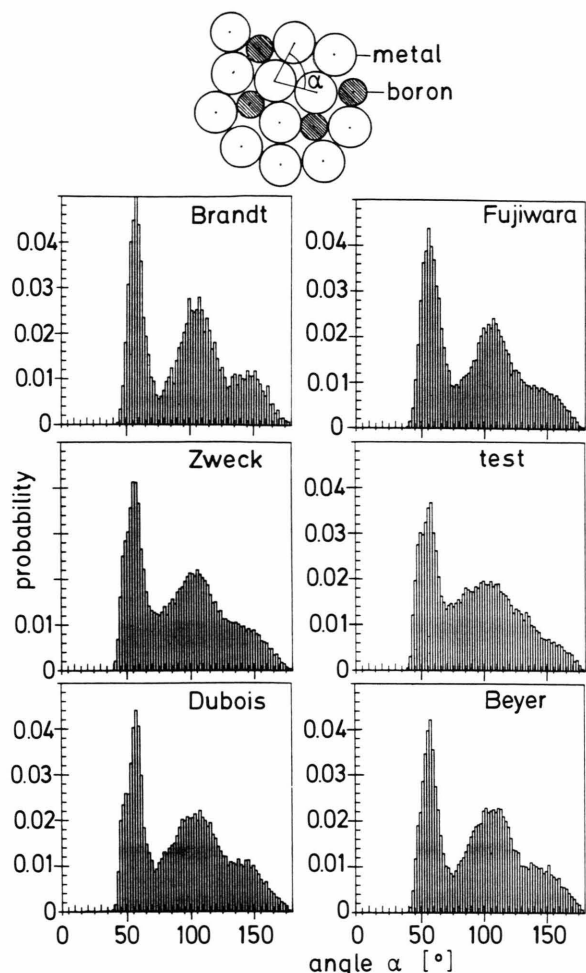


Fig. 5. Amorphous  $\text{Fe}_{80}\text{B}_{20}$  and  $\text{Ni}_{81}\text{B}_{19}$ . Probability of occurrence of the angle  $\alpha$ .

results are found also for  $N_{\text{BM}}$ ,  $N_{\text{MB}}$ , and  $N_{\text{BB}}$  [35]. At this point it is worth mentioning that also from Moessbauer spectroscopy such distributions of the coordination numbers have been derived [36, 37]. Since these distributions, however, are not characteristic for the one or the other principle of construction [35], the results of [36, 37] do not confirm or contradict any structure.

### 3.2. Distribution in Bond Angles

In Fig. 5 we show for six models of amorphous  $\text{Fe}_{80}\text{B}_{20}$  and  $\text{Ni}_{81}\text{B}_{19}$  the probability of occurrence of the angle  $\alpha$  defined in the upper sketch as the angle

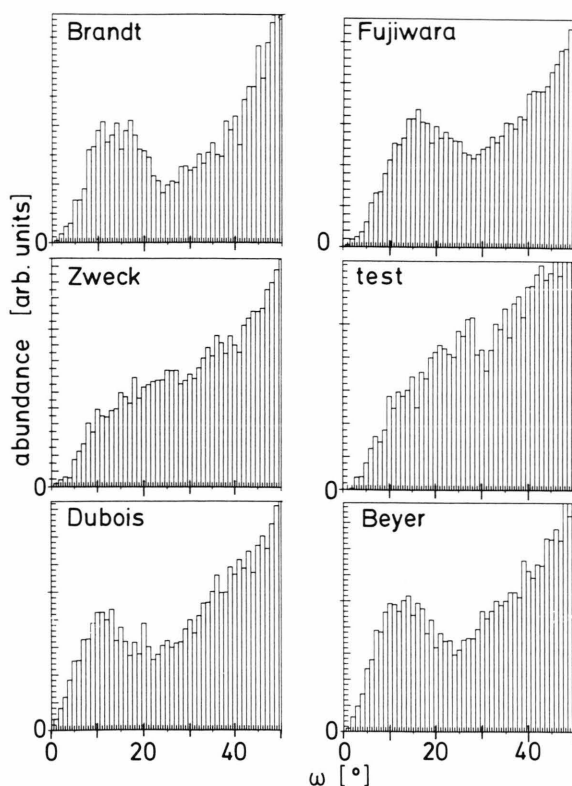


Fig. 6. Amorphous  $\text{Fe}_{80}\text{B}_{20}$  and  $\text{Ni}_{81}\text{B}_{19}$ . Abundance of  $\omega$  for tetrahedra with four metal atoms.

between two metal atoms of the first coordination sphere and the central metal site.

The six distributions are rather similar, each of them showing a pronounced peak for  $50^\circ \leq \alpha \leq 60^\circ$  and a smaller peak at about  $120^\circ$  followed by a hump at  $150^\circ$ . Peaks are most pronounced for the Brandt-model with its very pronounced  $G_{\text{FeFe}}(R)$ -curve (see Figure 1).

At this point it should be stressed that apparently the distributions of bond angles within the first coordination shell are not very suitable for distinguishing between the different models. With other words: It is very astonishing that five models which are based on rather different construction principles yield almost the same angular distributions.

Also the distributions of the following angles show close similarities for the five models [35]:

- i) Central site: metal. Two boron-atoms in the first sphere.
- ii) Central site: metal. One metal- and one boron-atom in the first sphere.



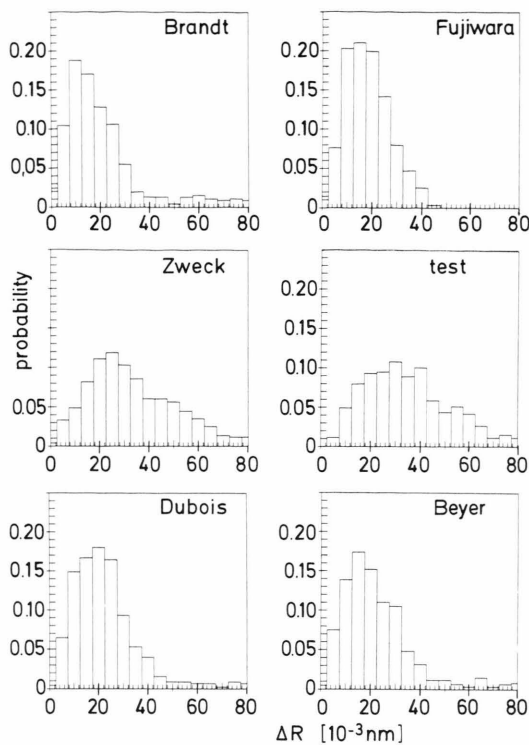


Fig. 7. Amorphous  $\text{Fe}_{80}\text{B}_{20}$  and  $\text{Ni}_{81}\text{B}_{19}$ . Deviation  $\Delta R$  of the central metal atom from the center of mass of the first coordination sphere.

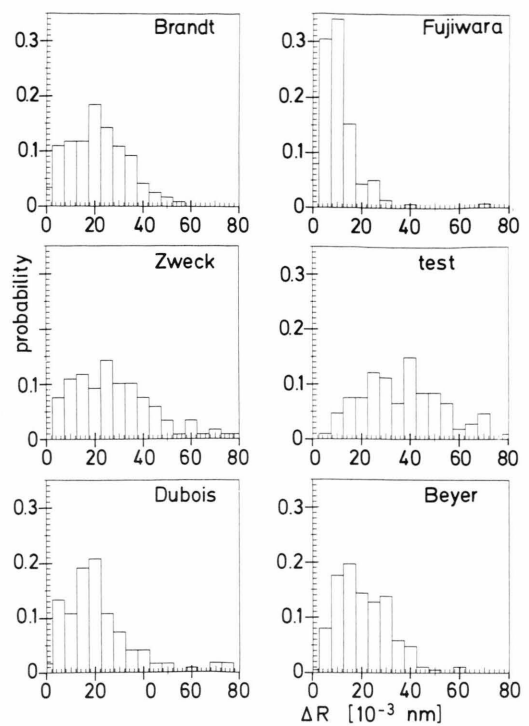


Fig. 8. Amorphous  $\text{Fe}_{80}\text{B}_{20}$  and  $\text{Ni}_{81}\text{B}_{19}$ . Deviation  $\Delta R$  of the central B-atom from the center of mass of the first coordination sphere.

- iii) Central site: boron. Two metal-atoms in the first sphere.
- iv) Central site: boron. Two boron-atoms in first sphere in  $G_{\text{BB}}(R)$ .

### 3.3. Search for Tetrahedra

An interesting task is the search for the occurrence of special polyhedra in the model of an amorphous alloy. A polyhedron in each case is distorted to a certain degree. Therefore, to allow a polyhedron to be recognized at all, tolerances concerning bond lengths and bond angles have to be allowed. These variable criteria make the search for polyhedra somewhat arbitrary [38, 39]. In some studies the search for polyhedra in structural models has been performed with special care and caution [16, 40, 41]. In the present paper tetrahedra will be treated, which is the only polyhedron which can be found without ambiguity. Thus a simplified question was applied to the models as follows: How much are the tetrahedra distorted? For

this purpose a tetrahedron is defined as follows: A central metal atom together with three neighbouring metal atoms are regarded together. There is no restriction concerning the distance except that the neighbouring atoms have to reside within the first coordination shell. The bond angles between the central atom and each two of the three neighbouring atoms are called  $\alpha$ ,  $\beta$ , and  $\gamma$ . If  $\alpha = \beta = \gamma = 60^\circ$ , we have a perfect tetrahedron in the sense of our definition, and we define an excess angle

$$\omega = |\alpha - 60^\circ| + |\beta - 60^\circ| + |\gamma - 60^\circ|. \quad (1)$$

The excess angle  $\omega$  is a measure for the distortion of the tetrahedron; in the case of an exact tetrahedron in the sense of our definition  $\omega = 0^\circ$ . Using this criterion, the models were examined for tetrahedra consisting of four metal atoms. Figure 6 shows the result for the six models. Apparently no exact tetrahedron with  $\omega = 0^\circ$  exists. But in four models there is a peak centered around  $\omega = 12^\circ$ . This means the existence of slightly distorted tetrahedra. In [35] also tetrahedra

with boron at one vertice and three metal atoms were treated in the same way with a result very similar to that of Figure 6.

### 3.4. Asymmetry of the Central Atom within the Near Neighbour Cage

In Fig. 7 we show the probability of the occurrence of deviations of the position of a central metal atom from the center of mass of the surrounding near neighbour cage for the six different models. We recognize from Fig. 7 that the probability has a maximum at  $\Delta R$ -values between  $10 \cdot 10^{-3}$  nm and  $30 \cdot 10^{-3}$  nm, depending on the model under consideration. Thus we state local asymmetries around the metal atoms in the different models.

According to Fig. 8 a similar result follows for first neighbour cages centered around a boron-atom. Local asymmetries in a structural model have already been discussed in terms of the distribution of internal stresses [42]. From Figs. 7 and 8 follows that such local asymmetries may vary from model to model.

## Conclusions

In the present study some of the most important structural models for amorphous M<sub>80</sub>B<sub>20</sub> alloys are compiled and investigated regarding their structural properties. It is found that despite the different principles of construction all of them are able to simulate

experimental partial pair correlation functions, which have been determined with high accuracy.

More detailed analysis shows striking similarities among the models concerning the angular distribution functions.

It is shown that within each model structure the kind of atomic neighbourhood varies appreciable from site to site. In general it can be concluded that for the simulation of the short range structure the relaxation under the influence of interatomic pair potentials is essential, whereas the principle of constructing the initial arrangement seems to be of smaller importance.

However, it shall not be excluded that concerning the medium range structure the principle of construction in fact may be important.

A challenge for the future is now to construct larger structural models which are capable for the description of the medium range order and the short range order simultaneously.

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