

TK 155.206

KFKI-1980-84

GY. FAIGEL
W.H. DE VRIES
H.J.F. JANSEN
M. TEGZE
I. VINCZE

QUASI-CRYSTALLINE MODELLING
OF AMORPHOUS ALLOYS

Hungarian Academy of Sciences

CENTRAL
RESEARCH
INSTITUTE FOR
PHYSICS

BUDAPEST

2017

QUASI-CRYSTALLINE MODELLING
OF AMORPHOUS ALLOYS

Gy. Faigel, W.H. de Vries*, H.J.F. Jansen*, M. Tegze, I. Vincze**

Central Research Institute for Physics
H-1525 Budapest 114, P.O.B. 49, Hungary

*Solid State Physics Laboratory, University of Groningen,
The Netherlands

*To appear in the Proceedings of the
Conference on Metallic Glasses:
Science and Technology, Budapest,
Hungary, June 30 - July 4, 1980;
Paper S-05*

HU ISSN 0368 5330
ISBN 963 371 730 2

⁺On leave from the Central Research Institute for Physics, Budapest

АННОТАЦИЯ

Сравниваются функции парной корреляции и интерференции простых неупорядоченных систем, построенных с определенным локальным порядком, с соответствующими функциями, полученными из квазикристаллической модели. В этой упрощенной модели было предположено расширение по \sqrt{r} атомных позиций. На основе этой некорреляционной модели ожидается характерное изменение функций радиального распределения металлических стекол Fe_3B и Ni_3B , которое будет отражать отличие между ближними химическими порядками соответствующих кристаллических фаз.

KIVONAT

Meghatározott lokális renddel felépített egyszerű rendezetlen szerkezetek párkorrelációs- és interferencia-függvényeit hasonlítjuk össze a kvázikristályos modellből nyert megfelelő függvényekkel. Ebben a leegyszerűsített modellben az atomi pozíciók \sqrt{r} szerinti kiszélesedését tettük fel. Ennek alapján egy jellegzetes változást várhatunk a Fe_3B és Ni_3B üvegek radiális eloszlás függvénye között, melyben a megfelelő kristályos fázisok kémiai rövidtávu rendje közti különbségek tükröződnek.

ABSTRACT

The pair correlation and interference functions of simple disordered structures with a given local geometry are compared to those of an oversimplified, uncorrelated model using only \sqrt{r} broadening of the atomic positions. On the base of the uncorrelated model a characteristic change is predicted in the radial distribution function of Fe_3B and Ni_3B metallic glasses reflecting the change in the chemical short-range order of the crystalline counterparts.

An increasing number of experiments suggests that the local structure of transition metal-metalloid glasses is substantially better ordered than predicted by dense random packing models [1]. A reasonable first approach to the amorphous structural unit involves the distorted local atomic arrangement of the metastable crystalline phase appearing in the course of crystallization [2]. The use of large building units results in complicated calculations. In the case of dense random packing of spheres the seed of the structure is a cluster which contains three atoms and the newly added fourth atom has to touch the already existing cluster. Since no chemical or topological short-range order is incorporated in the structure (apart from excluding metalloid-metalloid nearest neighbours) the procedure does not take into account the relative positions of the three old atoms. Thus the four particle correlation function describing the probability of a certain configuration of three old atoms and one new atom is approximated by a product of the three appropriate two particle correlation func-

tions. No computer procedure has been described yet to simulate the separate, well-determined coordinations characteristic of chemical short-range order. In the only model [3] which is based on a random packing of trigonal prismatic units of cementite, Fe_3C (isostructural to Pd_3Si) the calculation starts from a physical, hand-built model which seriously limits the number of atoms in the structure. Thus it is of interest to investigate simple models which can simulate the differences in the chemical short-range order of different metallic glasses.

The most simple approximation is based on the idea that the short-range order of glasses at certain compositions ("stoichiometric" glasses) can be reasonably well described [4] by the short-range order of the crystalline counterparts (quasi-crystalline (QC) models). It is worth to emphasize that always a careful investigation (preferably with nuclear methods, NMR, ME) is necessary to check whether this approximation is valid - the identical composition of the amorphous and single phase crystalline alloys is a necessary but not satisfactory condition. Our aim is to calculate the pair correlation function (PCF) within the QC model without having actual atomic coordinates on the base of the following assumptions: i, the first neighbours of an atom have a Gaussian probability distribution around the perfect crystal lattice sites, ii, the crystalline correlations are extra-

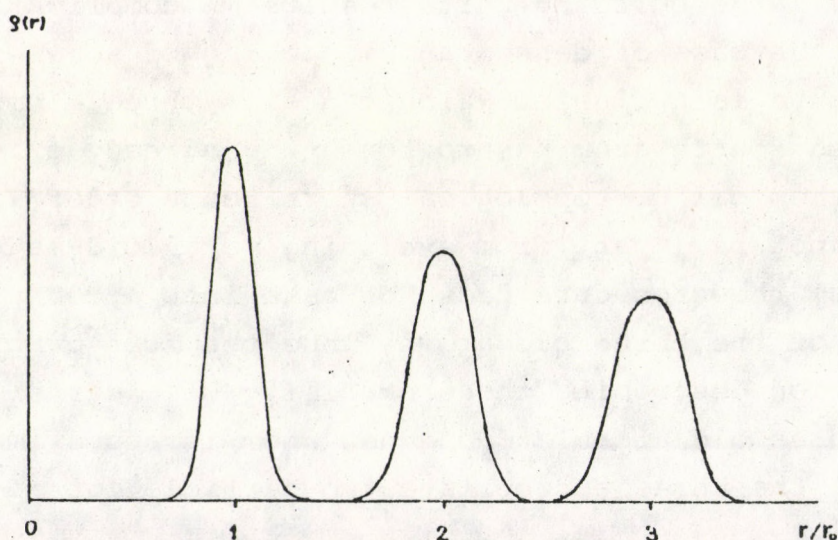


Fig. 1. Pair correlation function of a linear chain

polated to large distances. In one dimension the calculation of PCF is a simple convolution problem and can be treated analytically [5]. The result is a sum of broadening Gaussian distributions (Fig. 1) where the average deviation from the ideal position at \bar{r} , $\sigma(\bar{r})$ is proportional to $\sqrt{\bar{r}}$ ($\sigma^2(\bar{r}) \approx \bar{r}$). This result is exact because a linear chain of atoms can never be topologically disordered and no defects occur in the structure. In higher dimensions a similar calculation cannot be carried out because defects will occur due to the topological disorder and the exact distance and direction dependence of $\sigma(\bar{r})$ is not known. Therefore as a first approximation we will use the linear relationship valid in one dimension for $\sigma^2(r)$ in the expression for the PCF:

$$G(r) = (2\pi)^{-3/2} (\rho_0 r)^{-1} \sum_i (\sigma(r_i) r_i)^{-1} \exp\left(-\frac{r_i^2 + r^2}{2\sigma^2(r_i)}\right) \sinh\left(\frac{r r_i}{\sigma^2(r_i)}\right) \quad (1)$$

where the sum stands for all atomic positions (\bar{r}_i) of the corresponding crystal structure.

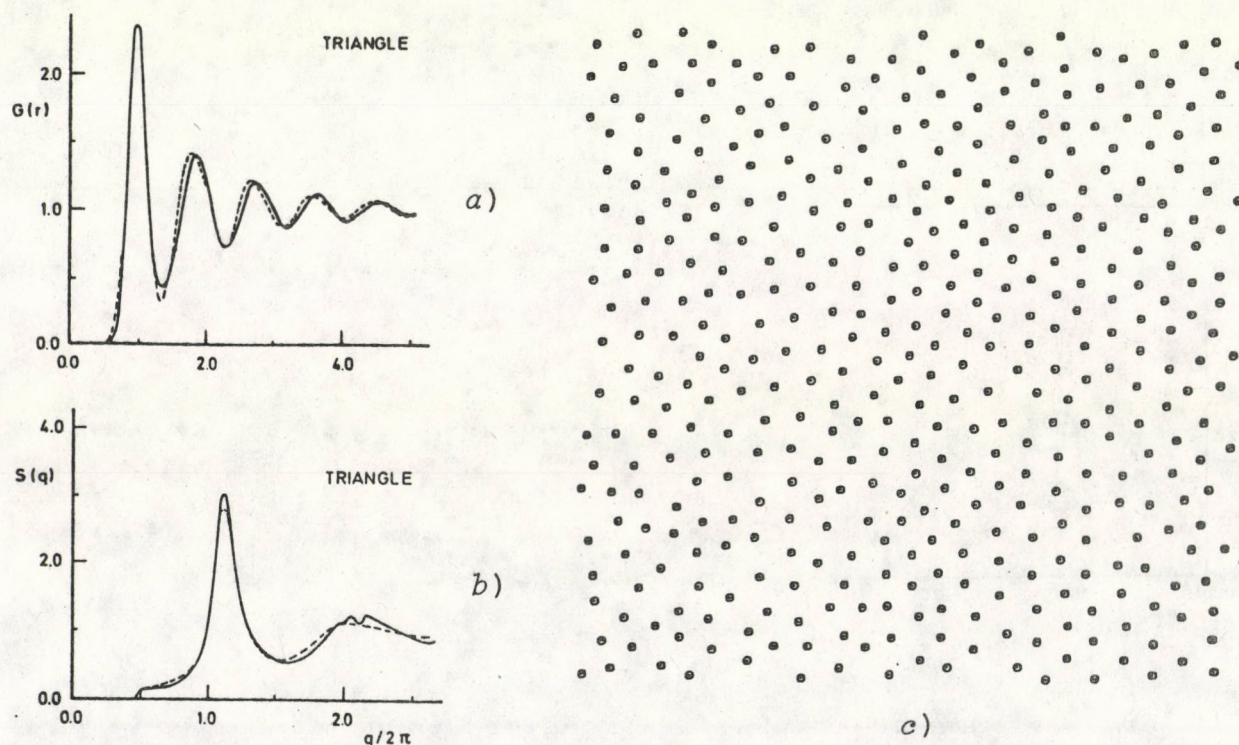


Fig. 2. Pair correlation and interference functions (a and b) calculated for two dimensional distorted triangular units. A typical set of the actual atomic arrangement is also shown (c). The dotted line corresponds to a QC calculation as explained in the text.

In the following we will compare the pair correlation and the interference functions calculated in the above mentioned, oversimplified, uncorrelated model (QC) with those of realistic physical models generated in two dimensions with a given local geometry (triangular, square). In the construction of the atomic arrangements only the first assumption has been used and defects (holes and extra atoms) were introduced when the distance between the generated new atomic positions became too small or too big. The agreement between the two types of calculations is quite good for the triangular unit cells (Fig. 2) while the medium-range order is strongly overestimated in the QC model for the square unit cells (Fig. 3). We have to be very careful in the extrapolation of these results for three dimensions but it is very tempting to believe that the agreement between actual physical models and QC calculations will be satisfactory for dense packed systems.

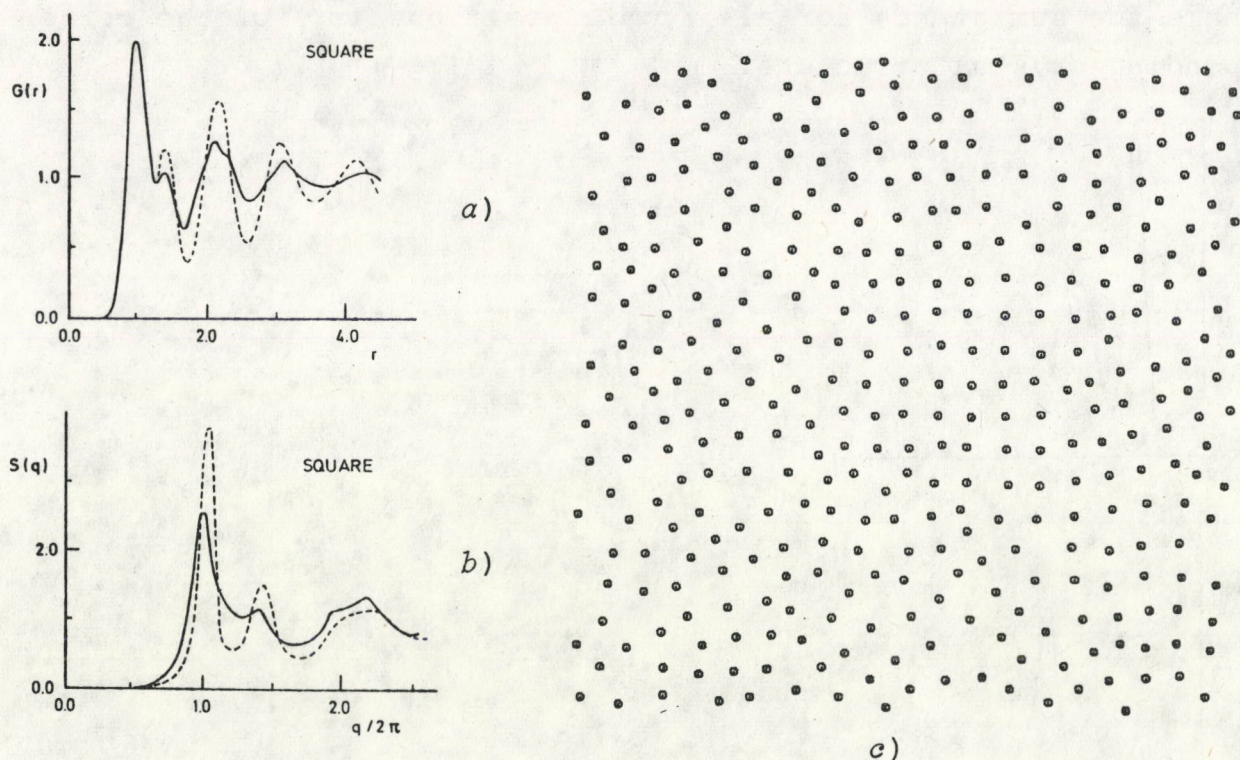
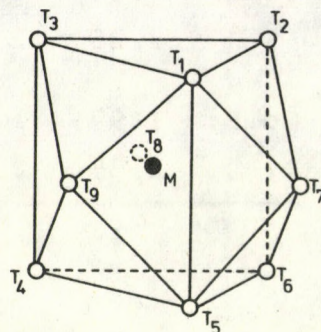


Fig. 3. Same as Fig. 2 for two dimensional distorted square units

In the following it will be shown that QC calculations can be used as simple guides to predict what kind of changes are to be accepted in the PCF due to different types of chemical short

range order. Fe_3B and Ni_3B (or Co_3B) have tetragonal and orthorhombic crystal structures [6], respectively, with the metalloids surrounded with 9 TM atoms (Fig. 4). The different topology

Fig. 4. Typical transition metal (T) surrounding of the metalloid (M) in the tetragonal Fe_3B and orthorhombic Co_3B , Ni_3B , respectively



of these structures can be easily illustrated with the M-TM distances (Table I): in Fe_3B all 9 Fe atoms are quite homogeneously distributed around the average distance $\bar{R} = 2.21 \text{ \AA}$ ($\Delta R = R_{\text{max}} - R_{\text{min}} = 0.14 \text{ \AA}$), which is valid only for the 6 inner TM atoms in the orthorhombic structure ($\bar{R} = 2.01 \text{ \AA}$ $\Delta R = 0.09 \text{ \AA}$ for the 6 inner TM atoms while the remaining 3 TM atoms are pushed out for larger distances ($\bar{R} = 2.17$, $\Delta R = 0.81 \text{ \AA}$ for the 9 TM atoms). With other words, the packing of the orthorhombic structure (Co_3B , Ni_3B) is more dense than that of the tetragonal units, e.g. the density of Ni_3B is ~10% larger than that of Fe_3B and only half of it can be explained with atomic weight differences. It has been shown [7] that the arrangement of near neighbours in $(\text{Fe}_{1-x}\text{Ni}_x)_3\text{B}$ glasses closely follows that of the crystalline counterparts indicating that the local symmetry in the amorphous and crystalline structure at this special composition is similar and changing with the Ni substitution. This change in chemical short range order is reflected in the PCF of amorphous Fe_3B and Ni_3B calculated in the QC model using the corresponding crystal structures with the single adjusted parameter, $\sigma(r_0)/r_0 = 8.5\%$ (Fig. 5). The PCF of amorphous Fe_3B in Fig. 5 agrees reasonably well with recent X-ray measurement [8]. We may suggest from Fig. 5 that the main change expected for the substitution of Fe by Ni in the radial distribution function of amorphous Fe_3B is the increase in the shoulder intensity of the second split peak reflecting the above discussed atomic rearrangement. Preliminary EDXD measurements indicate [9] the presence of this suggested change.

Table I

Metalloid-transition metal distances [6] (in Å) in tetragonal Fe_3B and orthorhombic Co_3B

Fe_3B :	2.13,	2.14,	2.20,	2.21,	2.23,	2.23,	2.25,	2.25,	2.27
Co_3B :	1.97,	1.97,	1.99,	2.01,	2.06,	2.06,	2.34,	2.35,	2.78

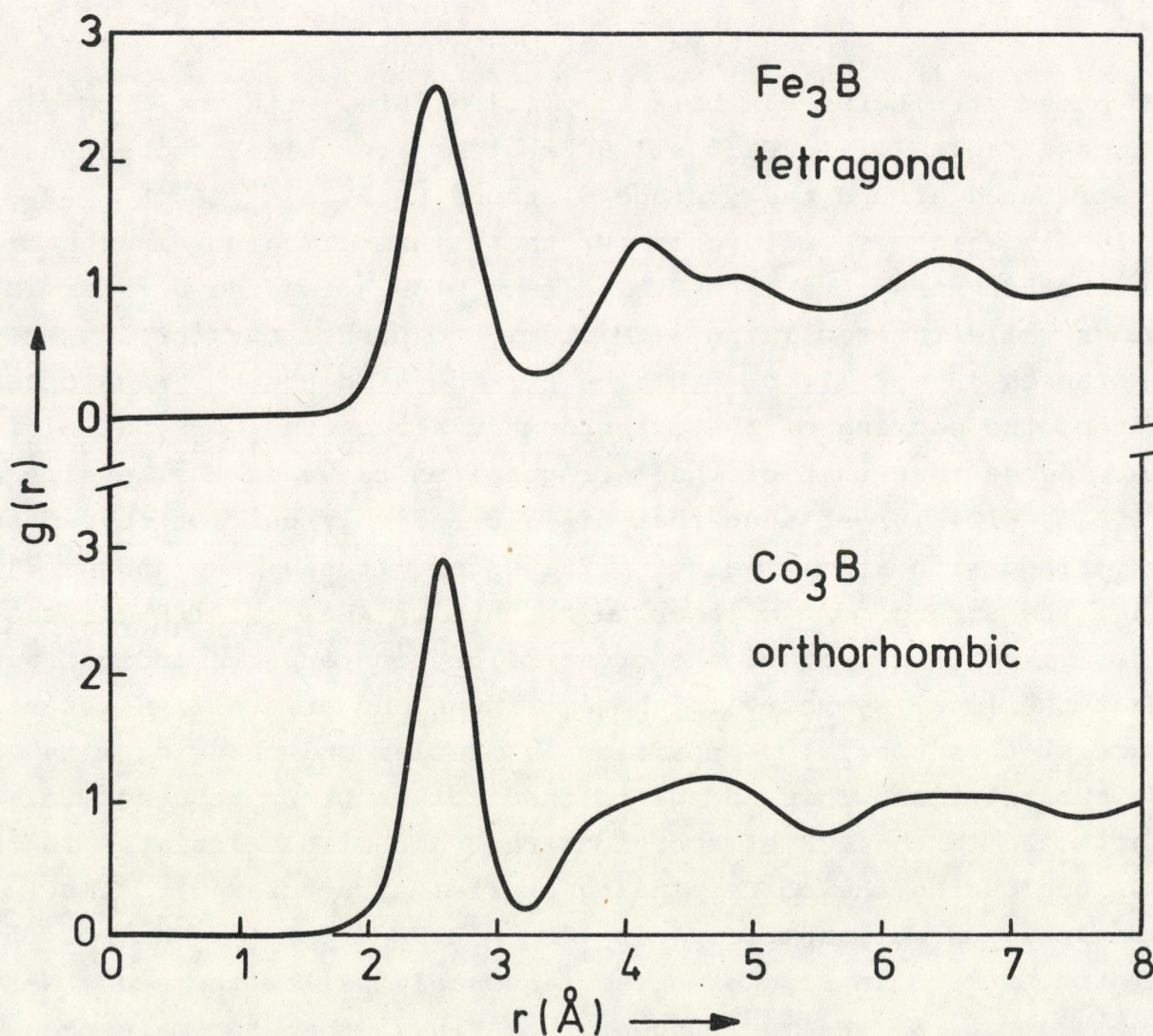


Fig. 5. Pair correlation functions for TM atoms calculated in QC model using tetragonal Fe_3B (upper) and orthorhombic Co_3B (lower) units

We wish to thank A.J. Dekker, T. Kemény, A.S. Schaafsma, C.N.J. Wagner and F. van der Woude for helpful discussions.

This work forms part of the research program of the Foundation for Fundamental Research on Matter (FOM) with financial support from the Netherlands Organization for the Advancement of Pure Research (ZWO).

REFERENCES

- [1] P.H. Gaskell, J. Phys. C: Solid State Phys. 12 (1979) 4337.
- [2] P.H. Gaskell, Phil. Mag. 32 (1975) 211; T. Kemény, I. Vincze, B. Fogarassy and S. Arajs, Phys. Rev. B20 (1979) 476.
- [3] P.H. Gaskell, J. Non-Cryst. Solids 32 (1979) 207.
- [4] I. Vincze, T. Kemény and S. Arajs, Phys. Rev. B21 (1980) 937.
- [5] J.A. Prins, Naturwissenschaften 19 (1931) 435.
- [6] U. Herold and U. Köster, Z. Metallk. 69 (1978) 326; R.W.C. Wyckoff, Crystal Structures, 2nd ed., Intersciences, New York (1964), Vol. 1.
- [7] I. Vincze, F. van der Woude, T. Kemény and A.S. Schaafsma, J. Magn. Magn. Mat. 15-18 (1980) 1336; I. Vincze, T. Kemény, A.S. Schaafsma, A. Lövas and F. van der Woude, this conference, paper S-19.
- [8] Y. Waseda and H.S. Chen, Phys. Stat. Sol. (a) 43 (1978) 387.
- [9] T. Egami, private communication.

63.060



Kiadja a Központi Fizikai Kutató Intézet
Felelős kiadó: Tompa Kálmán
Szakmai lektor: Hargitai Csaba
Nyelvi lektor: Hargitai Csaba
Gépelte: Végvári Istvánné
Példányszám: 220 Törzsszám: 80-624
Készült a KFKI sokszorosító üzemében
Felelős vezető: Nagy Károly
Budapest, 1980. október hó