Electron-Diffraction Study on an Amorphous Al-V Alloy Produced by Electron Irradiation of Quasicrystalline Al-16 at-%V

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Quasicrystalline Al-16 at-%V was transformed to the amorphous state by low-temperature electron irradiation in a high-voltage electron microscope. Electron diffraction experiments were carried out in the amorphous state and in the crystalline state obtained after subsequent heat treatment. From the results the total structure factor was determined. The pair correlation function was calculated which yields the radii of the different coordination spheres and the total coordination number. The results are discussed in terms of current topological models of the structure of metallic glasses.

1. Introduction

In an alloy which is in the quasicrystalline state the atoms are arranged in a quasiperiodic lattice which exhibits no translational symmetry but is characterized by long-range orientational order with icosahedral symmetry. Icosahedral atom correlations play a key role in a number of current models of metallic glasses [1–6]. These models are constructed on the basis of steric and topological arguments neglecting a possible influence of chemical effects on the structure.

Shortly after the discovery of Shechtman et al. [7] of the quasicrystalline state a discussion started on the consequences this might have for the models of metallic glasses. Unfortunately, with the exception of Pd-U-Si [8], non of the systems exhibiting a quasicrystalline phase can, by conventional quenching techniques, be produced in the amorphous state. This means that it is in general not possible to compare scattering data on the two different states which were obtained from one and the same system.

Recently Urban et al. [9] observed that, by high-energy electron irradiation, quasicrystalline Al-14 at-%Mn can be rendered amorphous. This transition is reversible, i.e. the quasicrystalline state can be recovered during a proper heat treatment. The amorphous to quasicrystalline transition has also been observed by other authors [8, 10, 11]. During the transition the system "avoids" the crystalline state. This can be interpreted as indication for a special structural relationship between the quasicrystalline and the amorphous phase.

In order to elucidate this question further we have studied Al-16 at-%V which is another alloy, which, from the quasicrystalline state, can be transformed to the amorphous state by electron irradiation [12, 13]. The Al-V material obtained by melt spinning is not homogeneously quasicrystalline. There are regions where the quasicrystals are very small, their diameters reaching some nanometers only. In other areas quasicrystal diameters are in the range of some micrometers. In between there are regions were the material is of different structure and composition. This makes conventional X-ray scattering techniques unreliable for the determination of structure factors and radial distribution functions. Therefore we have chosen to carry out a quantitative electron diffraction study in an electron microscope. This permits to select for the diffraction experiments a small specimen area of uniform composition and structure.

2. Theoretical Fundamentals

In the evaluation of diffraction patterns obtained from melts or amorphous materials it is necessary to

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correct the measured intensities for inelastic as well as for elastic multiple electron scattering. Subsequently the corrected intensities have, by normalization, to be transformed to absolute intensities. For neutron and X-ray diffraction the theory is well developed and the treatment of the primary data provides no fundamental difficulties [14, 15].

However, for electron diffraction corresponding tools are not available. Therefore in this work we use a method in which the background intensity is eliminated from the measured intensity by combining results obtained from the amorphous and the crystallized specimen [16, 17].

The method starts from the assumption that the angular dependence of the background intensity is the same for the crystalline and the amorphous specimen. This means that

$$I_{ba}(Q) = b\, I_{bc}(Q) + c\,, \tag{1}$$

where I_{ba} and I_{bc} denote the background intensity of the amorphous and the crystalline material, respectively. b is a proportionality factor and c is a constant determined by the angle-independent part of the background intensity and by experimental parameters (speed of film used for diffraction pattern recording, etc.). Q is the scattering parameter given by

$$Q = (4\pi/\lambda)\sin\theta\,, \tag{2}$$

where λ is the electron wavelength and 2θ is the angle between the incident and a scattered beam.

In the crystalline specimen I_{bc} can be obtained from the measured intensity curve by connecting the minima thus eliminating the Bragg reflections. The corrected intensity of the amorphous specimen I_{ca} in arbitrary units can then be calculated from the experimental intensity distribution I_{ea} using (1) and the relation

$$I_{ca}(Q) = I_{ea}(Q) - I_{ba}(Q)\,. \tag{3}$$

In order to normalize the corrected intensity the Krogh-Moe method is applied [18]. Accordingly the intensity I_a of electrons coherently scattered in the specimen is related to I_{ca} introducing the normalization coefficient β, i.e.

$$I_a = \beta\, I_{ca}\,. \tag{4}$$

For the determination of β not only $I_{ca}(Q)$ at large values of Q is used but also the product $Q^2 I_{ea}(Q)$ along the whole Q-range. Thus

$$\beta = \frac{\int_0^{Q_M} q^2[\langle f^2(Q)\rangle + b\, I_{ba} + c]\,\mathrm{d}Q - 2\pi^2\varrho_0[\langle f(Q)\rangle^2/\langle f^2(Q)\rangle]}{\int_0^{Q_M} q^2\, I_{ea}\,\mathrm{d}Q}\,. \tag{5}$$

with

$$\langle f^2(Q)\rangle = C_A f_A^2(Q) + C_B f_B^2(Q)\,, \tag{6}$$

where C_A and C_B are the atomic concentrations of the two alloy components A and B ($C_A + C_B = 1$). f_A and f_B denote the scattering factors of A and B atoms, respectively. In addition we have used $q = Q/\langle f(Q)\rangle$, and ϱ_0 is the mean atomic number density.

Equation (5) contains two unknowns, b and c. In a first approximation c is neglected and the so-called high-angle method [19] is used which finally yields [17]

$$b = \frac{\int_{Q_M-\varepsilon}^{Q_M} I_{ea}(Q)\,\mathrm{d}Q - (1/\beta)\int_{Q_M-\varepsilon}^{Q_M}\langle f^2(Q)\rangle\,\mathrm{d}Q}{\int_{Q_M-\varepsilon}^{Q_M} I_{bc}(Q)\,\mathrm{d}Q}\,. \tag{7}$$

For Q_M we take the maximum value of Q for which we have experimental data.

The integration is carried out in the high-Q region between the limits $Q_M - \varepsilon$ and Q_M in which the intensity shows only oscillations with very small amplitude.

According to Ashcroft and Langreth [20] the total structure factor $S(Q)$ is given by

$$S(Q) = I_a(Q)/\langle f^2(Q)\rangle\,. \tag{8}$$

The parameter c in (1) is adjusted in such a way that finally a "correct" shape of the $S(Q)$ curve is obtained. This means that $S(Q)$ oscillates around the value 1.

From the structure factor we obtain the pair correlation function $G(R)$ using the relation

$$G(R) = 4\pi R(\varrho(R) - \varrho_0) \tag{9}$$
$$= (2R/\pi) \int_0^\infty Q^2[S(Q) - 1][\sin(QR)/QR]\,\mathrm{d}Q\,,$$

where R is the coordinate in real space and $\varrho(R)$ is the local atomic number density.

In amorphous specimens $G(R)$ exhibits in general the following typical features: (i) A pronounced first maximum whose position, R_1, is essentially determined by the radius of the first coordination shell around an atom. Further maxima exist which are, however, much less pronounced. In general the second maximum is split up into two submaxima. (ii) For $0 \leq R \leq R_1$ theory yields

$$G(R) = -4\pi R\varrho_0\,. \tag{10}$$

In practice, the upper integration limit in (9) cannot be infinity but an experimentally determined value, Q_M. This leads to the so called termination effect, i.e. the occurrence of spurious maxima in $G(R)$ which have no physical meaning. They can be recognized by their strong Q_M-dependence.

The structure factor $S(Q)$ and the pair correlation function $G(R)$ can be written in terms of the corresponding partial functions, S_{ij} and G_{ij}, respectively. They represent the individual contributions of atom pairs of type ij $(i,j = \mathrm{A}, \mathrm{B})$. According to [20],

$$S(Q) = W_{AA}S_{AA}(Q) + W_{BB}S_{BB}(Q) + 2W_{AB}S_{AB}(Q)\,, \tag{11}$$

$$G(R) = C_A^2 W_{AA}G_{AA}(R) + C_B^2 W_{BB}G_{BB}(R) + 2C_A C_B W_{AB}G_{AB}(R)\,, \tag{12}$$

where $W_{ij} = f_i f_j/\langle f^2\rangle$.

On the other hand, according to Bhatia and Thornton [21], $S(Q)$ can also be expressed in terms of the contributions of correlations between fluctuations in atom number density and concentration, i.e.

$$S(Q) = [\langle f\rangle^2 S_{NN}(Q) + C_A C_B(f_A - f_B)^2 \cdot S_{CC}(Q) + 2\langle f\rangle(f_A - f_B)S_{NC}(Q)]/\langle f^2\rangle\,. \tag{13}$$

Here S_{NN} denotes the contribution of correlations between density fluctuations, S_{CC} is the contribution of correlations between concentration fluctuations, and S_{NC} represents the contribution of correlations between density and concentration fluctuations.

The area under the first maximum of the pair correlation function yields the nearest-neighbour coordination number N. In terms of the partial coordination numbers N_{ij} this can be written as [20]

$$N = C_A W_{AA}N_{AA} + C_B W_{BB}N_{BB} + 2C_A W_{AB}N_{AB}\,. \tag{14}$$

N_{AB} denotes the number of B-atoms around an A-atom.

3. Experimental Details

Melt-spun Al-16 at-%V alloy was obtained from Bell Laboratories. The material consisted of thin flakes about 30 μm thick, about 1 mm wide and some millimeters long. The flakes contained holes. Along the hole edges the material was, without any further preparation, thin enough for investigation in the AEI EM7 transmission electron microscope operated at 1 MV.

Thin specimen areas (less than 50 nm in thickness) were selected. There the material was, due to the very high cooling rate during melt spinning, in a microquasicrystalline state [13] with quasicrystal diameters between 5 and 20 nm. The irradiation was carried out at 30 K in a helium-cooled specimen holder. The electron dose rate was $10^{24}\,\mathrm{e\,s^{-1}\,m^{-2}}$. After 5 min of irradiation the Debye-Scherrer pattern of the microquasicrystalline state was replaced by the ring pattern typical for the amorphous state. The irradiation was continued up to 15 min in order to ensure that the quasicrystal structure was destroyed completely. The diffraction pattern was recorded on photographic film after heating up to room temperature. Subsequently the specimen was transferred into a hot stage and heated up, inside the microscope, to 700 K, where the amorphous area crystallized. The corresponding diffration pattern was again recorded at room temperature.

The photographic density of the developed films was measured in a microdensitometer. By means of experimentally determined calibration curves the density values were corrected for the nonlinear behaviour of the film material.

4. Results

4.1. Electron Diffraction Pattern

Figure 1 shows diffraction patterns before and after irradiation. There is a direct correspondence between the position of the most intense Debye-Scherrer rings of the quasicrystalline state and the rings of intensity in the amorphous state. Using the notation of [22] the radial distance of the innermost and most intense ring in the amorphous material coincides with the position of the (100000) and (110000) Debye-Scherrer rings. The two maxima of the second ring coincide with the (101000) and the (110010) ring, respectively. Such a correlation was also found in the case of Al-14 at-%Mn [9, 12].

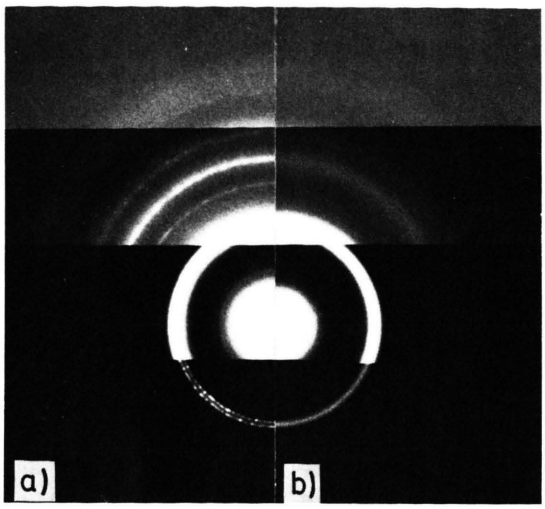

Fig. 1. Diffraction pattern of microquasicrystalline Al-16 at-%V alloy. (a) before and (b) after irradiation.

4.2. Structure Factor

The total structure factor $S(Q)$ is shown in Figure 2. For the evaluation of the primary data the atomic number density was approximated by the density which can be derived from that of the elementary crystalline materials, i.e. $\varrho_0 = 0.062\ \text{Å}^{-3}$. The curve exhibits the shape also observed in "normal" amorphous alloys. A pronounced first maximum occurs at $Q_1 = 2.75\ \text{Å}^{-1}$. The second maximum is split into two submaxima at $Q_{2a} = 4.67\ \text{Å}^{-1}$ and $Q_{2b} = 5.32\ \text{Å}^{-1}$. A third and also a fourth maximum are visible. For the ratios Q_{2a}/Q_1 and Q_{2b}/Q_1 we find the values 1.71 and 1.94, respectively.

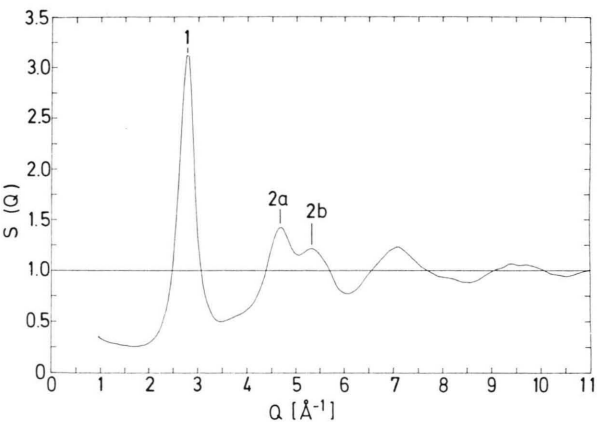

Fig. 2. Total structure factor $S(Q)$ of amorphous Al-16 at-%V alloy. Q is the scattering parameter defined in (2).

On the basis of experiments in which only electron-radiation is employed the partial structure factors cannot be determined. Nevertheless, the contribution of the different types of atom pairs can be estimated calculating the weighting factors W_{ij} in (11). Inserting the atomic scattering factors of aluminum and vanadium at Q_1 [23] we find

$$S(Q) = 0.81\ S_{\text{AlAl}}(Q) + 2.04\ S_{\text{VV}}(Q) + 2.56\ S_{\text{AlV}}(Q)\,. \tag{15}$$

We recognize that the weight at which V-V and Al-V atom pairs contribute to the total structure factor is much larger than that of the Al-Al pairs.

The weight at which the different kinds of fluctuations contribute to the total structure factor can be estimated calculating the composition and scattering-factor dependent coefficients in (13):

$$S(Q) = 0.96\ S_{\text{NN}}(Q) + 0.04\ S_{\text{CC}}(Q) - 1.05\ S_{\text{NC}}(Q)\,. \tag{16}$$

If the atomic radii of the two alloy constituents are similar, S_{NC} is very small [21, 24]. This is indeed the case in the Al-V system. The Goldschmidt radii are 1.40 Å for Al and 1.35 Å for V [25]. Since in addition the weighting factor of the second term is very small, the structure factor is essentially determined by S_{NN}, i.e. by atomic number density fluctuations.

4.3. Pair Correlation Function

Figure 3 shows the total pair correlation function $G(R)$ as derived from $S(Q)$ in Figure 2. The oscil-

Table 1. The position of the maxima of the pair correlation function of amorphous Al-16 at-%V (upper line). For comparison values calculated on the basis of a tetrahedron model [3] are given in the second line.

	R_1 (nm)	R_{2a}/R_1	R_{2b}/R_1	R_3/R_1	R_4/R_1	R_5/R_1
Al-V	0.284	1.71	1.95	2.52	2.98	3.40
Theory [3]		1.65	1.99	2.49	2.97	3.38

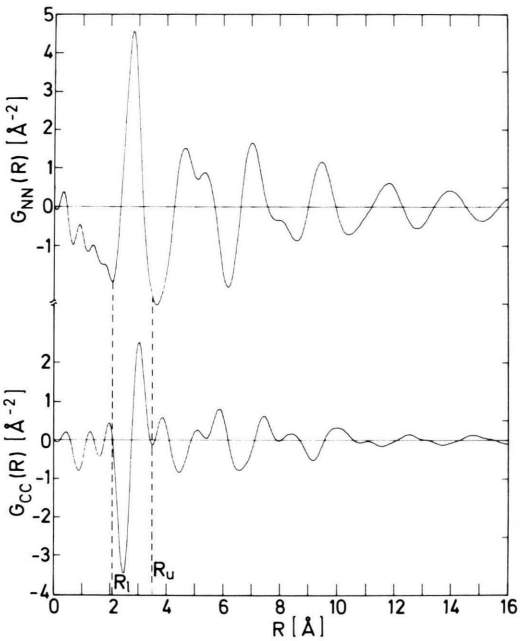

Fig. 3. Pair correlation function $G(R)$ of amorphous Al-16 at-%V alloy. R is the coordinate in real space.

lations at small R-values arise from the termination effect described above, but the main trend of the curve follows (10). The atomic distances which pertain to the maxima in Fig. 3 are compiled in Table 1. For convenience the radii of the higher maxima are normalized to the radius R_1 of the first maximum. For $r_{2a} = R_{2a}/R_1$ and for $r_{2b} = R_{2b}/R_1$ we find the values 1.71 and 1.95, respectively.

For the purpose of this paper it is important to compare our values for r_{2a} and r_{2b} to those found in "normal" amorphous materials. We find good agreement with the values for the single-atom systems Ni [26] and Co [27]. In the case of cobalt, $r_{2a} = 1.69$ and $r_{2b} = 1.97$. In the case of the binary or ternary alloys the respective values are somewhat smaller [28], i. e. typically $1.65 \leq r_{2a} \leq 1.70$ and $1.87 \leq r_{2b} \leq 1.91$. This can be attributed to the different sizes of the alloy atoms. In contrast, the atomic radii of Al and V are very similar, which explains the good agreement with the single-atom systems.

The relative contribution of the different types of atom pairs to the total correlation function can be estimated calculating the coefficients in (12), inserting the atomic scattering factors for Q_1 [23]:

$$G(R) = 0.56\, G_{\mathrm{AlAl}}(R) + 0.05\, G_{\mathrm{VV}}(R) + 0.34\, G_{\mathrm{AlV}}(R)\,. \tag{17}$$

The coefficients in this equation contain the squares of the concentrations and are therefore of different order of magnitude than those occurring in (15). The main maximum of $G(R)$ lies at 2.84 Å. According to (17) its position is essentially determined by the Al-Al and Al-V nearest-neighbour distances. The measured value fits well to the atomic diameters of Al and V given above.

The total coordination number N of the first coordination shell was calculated from the area below the first maximum of $G(R)$, taking as integration limits the position of the adjacent minima. The value is about 12. Such a value is also found in "normal" amorphous materials [29]. Calculating the coefficients of (14) we find that the weight of unequal neighbours is relatively large:

$$N = 0.67\, N_{\mathrm{AlAl}} + 0.33\, N_{\mathrm{VV}} + 2.15\, N_{\mathrm{AlV}}\,. \tag{18}$$

5. Discussion

Our results indicate that with respect to electron diffraction amorphous Al-16 at-%V behaves as other well known "normal" metallic glasses.

Within the last decade a number of topological models have been developed for the structure of metallic glasses. In these models any influence of chemical effects on the structure is neglected. Takeuchi and Kobayashi [3], on purely geometrical grounds, stressed the importance of the four-atom

tetrahedron as structural unit. They concluded that these units are arranged in extended chains which interpenetrate each other giving rise to icosahedral atom arrangements. Their model predicts values for the peaks in the radial distribution function which deviate by less than 3.5% from our experimental values (cf. Table 1). A different way was followed by Briant and Burton [2], Hoare [1], and Steinhardt et al. [4]. They used a computer to relax a dense random packing of atoms in a Lennard-Jones interaction potential. While only a modest amount of orientational order appears in the hard-sphere model, very pronounced icosahedral atom correlations were found to occur in the "relaxed" structure. That icosahedral short-range order reduces the free energy of liquids has first been proposed by Frank [30]. In the computer studies the icosahedral atom correlations were found to increase with increasing undercooling [4]. From this it was inferred that during a sufficiently rapid quench this short-range order is preserved, which then determines the structure of the amorphous state.

The formation of a quasicrystal can be considered as another way to preserve, in the solid state, the icosahedral atom correlations postulated for the undercooled melt. In particular, it was suggested by Sachdev and Nelson [31] that the primary difference between the quasicrystalline and amorphous phases lies in the value of the correlation length of icosahedral orientational order. These authors compared the diffraction patterns of quasicrystalline Al-Mn to the structure factor $S(Q)$ of various metallic glasses. They found that the maxima of $S(Q)$ correspond to the values of the reciprocal lattice vectors of the most intense spots in the quasicrystal diffration patterns. This is directly confirmed by the work reported here (cf. Fig. 1) and in [9, 13], in which diffraction data on either state is obtained from one and the same alloy. In the framework of the density-wave model [32–34], intensity in the diffraction pattern at a particular value of the reciprocal-lattice vector indicates the presence of a density wave in the specimen of appropriate amplitude and wave vector. Thus the experimental results indicate a direct correspondence of the basic density waves in the quasicrystalline and the amorphous states.

A number of authors [5, 6, 35, 36] have argued that icosahedral order in glasses is related to an ideal, icosahedral crystal, the so called polytope $\{3, 3, 5\}$, which can be constructed on the three-dimensional surface of a fourdimensional sphere. In this model the regions of short-range $\{3, 3, 5\}$ order in the glass are broken up by a tangled array of wedge disclination lines required to compensate for the incompatibility of flat space with a spacefilling, perfectly icosahedral solid. On this basis Sachdev and Nelson [6] have employed Landau theory to calculate the density correlations in a metallic glass. They found that the maxima in $S(Q)$ correspond to particular reciprocal-lattice vectors of the polytope $\{3, 3, 5\}$. The theory predicts values for the ratios Q_{2a}/Q_1 and Q_{2b}/Q_1 of 1.7 and 2, respectively. The former value virtually coincides with the one measured during the present paper, while the latter is by about 3% too large.

Although our experimental results fit well to the predictions of the topological models described above we do not consider this as decisive evidence for the general validity of the theory. The total structure factor and pair correlation function are not sensitive enough to distinguish unambiguously between the various physical models. On the other hand, there is clear evidence that in a large family of metallic glasses chemical effects are important for the details of the structure, which deviates considerably from that expected on purely geometrical grounds [14, 15]. Although our results and in particular the phase-transformation studies reported in [9, 12, 13] suggest a special structural relationship between the quasicrystalline and the amorphous state final conclusions can only be drawn after the partial structure factors, pair correlation functions, and coordination numbers have been determined.

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