Structural Study of a Decagonal $Al_{75}Fe_{15}Ni_{10}$ Alloy by Anomalous X-ray Scattering (AXS) Method

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A structural study of an as-quenched decagonal $Al_{75}Fe_{15}Ni_{10}$ alloy has been carried out by anomalous x-ray scattering (AXS) as well as ordinary x-ray diffraction. The environmental radial distribution functions (RDFs) for Fe and Ni determined by the AXS measurements turned out to resemble each other and to be similar to the ordinary RDF obtained by ordinary x-ray diffraction. These results clearly show that the Ni and Fe atoms are homogeneously distributed and occupy the same sites in the decagonal structure of $Al_{75}Fe_{15}Ni_{10}$.

Introduction

Decagonal phases are two-dimensional quasicrystals. Because of this, it has been suggested that the details of their structure may give an important clue for the understanding of three dimensional quasicrystals. Until quite recently, decagonal phases were found only as mixtures with other phases in a narrow composition range. This made an accurate characterization of the decagonal phase extremely difficult. Tsai et al. [1] produced a single decagonal phase in a wide composition range of Al-Ni-Fe and Al-Ni-Co ternary alloys by rapidly quenching. In the ternary Al-Co-Cu alloy, the stable decagonal phase can readily be produced by conventional solidification as well as by rapid quenching [2]. The discovery of such single decagonal alloys stimulates investigations of their fundamental physical and chemical properties including the atomic scale structures.

The quasicrystal has an infinite number of sites which are not exactly equivalent, which makes construction of a structural model for quasicrystals extremely complicate. One of successful approaches to understand its atomic structure is the "rigid geometry plus decoration" proposed by Henley [3]. Namely, the atomic structure of a quasicrystal can be described by placing atoms on the rigid geometrical frame with a certain decoration rule. Local probes, e.g. the EXAFS (extended x-ray absorption fine structure) technique, the anomalous x-ray scattering (AXS) technique and the neutron diffraction technique with isotopically substituted samples, are useful experimental methods to study the decoration rule in quasicrystals. In the present study, the AXS method was applied to the as-quenched $Al_{75}Fe_{15}Ni_{10}$ decagonal alloy.

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Experimental

An ingot of $Al_{75}Fe_{15}Ni_{10}$ was prepared by arch-melting from mixtures of high-purity (>99.7 wt%) metals of Al, Fe and Ni in argon atmosphere. From this master ingot, ribbons of about 0.02 mm thickness and 1 mm width were prepared by a single-roller melt-spinning technique. The details of the sample preparation are given in [1]. For the x-ray scattering measurements, powder samples were prepared by grinding these ribbons.

The AXS measurements were carried out with synchrotron radiation at the Photon Factory of the National Laboratory for High Energy Physics, Tsukuba, Japan. The details of the experimental setting and analysis are explained in [4]. MoKα radiation generated by a sealed Mo x-ray tube with a singly bent pyrolytic graphite monochromator in the diffracted beam was used for the ordinary x-ray measurement. The scattering intensity was observed with a scintillation detector combined with a pulse-height analyzer.

For convenience of the following discussion, only the fundamentals of the ordinary x-ray diffraction

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analysis as well as the AXS analysis are given below, while the details may be found, e.g. in [4–6]. The scattering intensity measured with MoKα radiation was used to estimate the ordinary radial distribution function (RDF), which describes the average structure of a sample. The measured intensity is corrected for absorption, polarization and Compton scattering, and converted to absolute units [6]. Fourier transformation of this resultant coherent intensity gives the ordinary RDF, i.e.

$$4\pi r^2 \varrho(r) = 4\pi r^2 \varrho_0 + \frac{2r}{\pi} \int_0^{Q_{max}} \frac{Q[I_{eu}^{coh}(Q) - \langle f^2 \rangle]}{\langle f \rangle^2} \sin(Qr)\, dQ, \quad (1)$$

where

$$\langle f \rangle = \sum_{j=1}^{N} c_j f_j, \quad \langle f^2 \rangle = \sum_{j=1}^{N} c_j f_j^2,$$

and $Q = 4\pi \sin\theta/\lambda$. c_j and f_j are the atomic fraction and the x-ray atomic scattering factor of the j-th element, 2θ is the scattering angle, μ the linear absorption coefficient, N the total number of constituent elements, Q_{max} the maximum value of Q used for the measurements, $\varrho(r)$ the average number density function and ϱ_0 the average number density.

On the lower energy side of the edge of a specific element A, the detected variation in intensity is attributed only to the change of the real part of the anomalous dispersion term f' of A. The environmental RDF for A was determined by Fourier transformation of the difference between the scattering intensities measured at two energies E_1 and E_2 ($E_1 < E_2$).

$$4\pi r^2 \varrho_A(r) = 4\pi r^2 \varrho_0 + \frac{2r}{\pi} \int_0^{Q_{max}} \frac{Q\, \Delta I_A(Q) \sin(Qr)}{c_A(f_A'(E_1) - f_A'(E_2))\, W(Q)} dQ, \quad (2)$$

$$\Delta I_A(Q) = \{I_{eu}^{coh}(Q, E_1) - \langle f(Q, E_1)^2 \rangle\} - \{I_{eu}^{coh}(Q, E_2) - \langle f(Q, E_2)^2 \rangle\}, \quad (3)$$

$$W(Q) = \sum_{j=1}^{N} c_j \,\mathrm{Re}[f_j(Q, E_1) + f_j(Q, E_2)], \quad (4)$$

where $\varrho_A(r)$ is the number density function around A. "Re" indicates the real part of the function in the braces. For the x-ray atomic scattering factors, the values tabulated in the International Tables for X-ray Crystallography, Vol. IV [7] were used and the theoretical values [5] computed by Cromer and Liberman's method [8] were used for the anomalous dispersion terms.

By comparing the environmental RDF around A in (2) with the ordinary RDF in (1), the merit of the AXS method is easily recognized. Six partial RDFs overlapping in the ordinary RDF are reduced to only three partial RDFs for pairs including the element A in the environmental RDF.

Results and Discussion

Scattering intensities of the as-quenched decagonal $Al_{75}Fe_{15}Ni_{10}$ alloy measured at 7.086 and 6.811 keV below the Fe K-absorption edge (7.111 keV) and their differential profile are shown in Figure 1. The peaks are indexed by the indexing scheme of the decagonal reflections proposed by Takeuchi and Kimura [9]. Similarly, the results obtained at 8.306 and 8.031 keV below the Ni K-absorption edge (8.332 keV) and their difference are shown in Figure 2. These differential intensity profiles at the Fe and Ni K-absorption edges have some fundamental features in common: They resemble the original intensity profile, which implies that both of Fe and Ni atoms are homogeneously distributed and probably occupy similar atomic sites in the decagonal structure. This might be a reason why a single decagonal phase can be formed in a quite wide composition range, e.g. 9 to 16% Ni and 9 to 21% Fe in the ternary Al-Fe-Ni alloy [1].

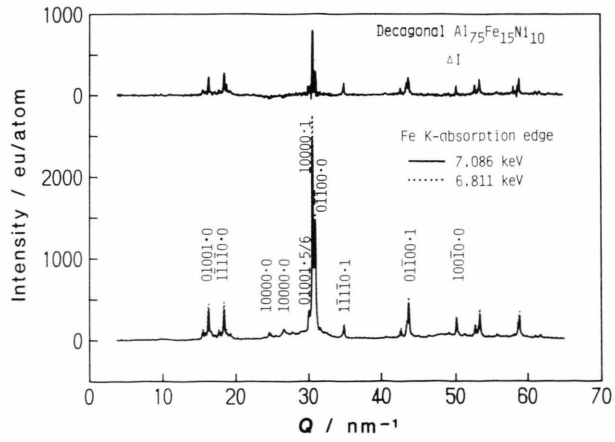

Fig. 1. Differential intensity profile of as-quenched decagonal $Al_{75}Fe_{15}Ni_{10}$ alloy (top) computed from intensities (bottom) measured at 7.086 and 6.811 keV below the Fe K-absorption edge (7.111 keV).

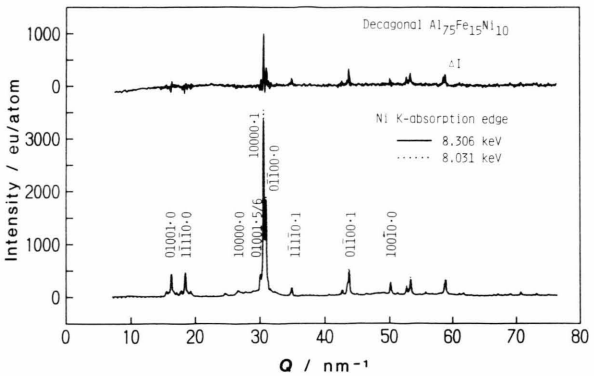

Fig. 2. Differential intensity profile of as-quenched decagonal $Al_{75}Fe_{15}Ni_{10}$ alloy (top) computed from intensities (bottom) measured at 8.306 and 8.031 keV below the Ni K-absorption edge (8.332 keV).

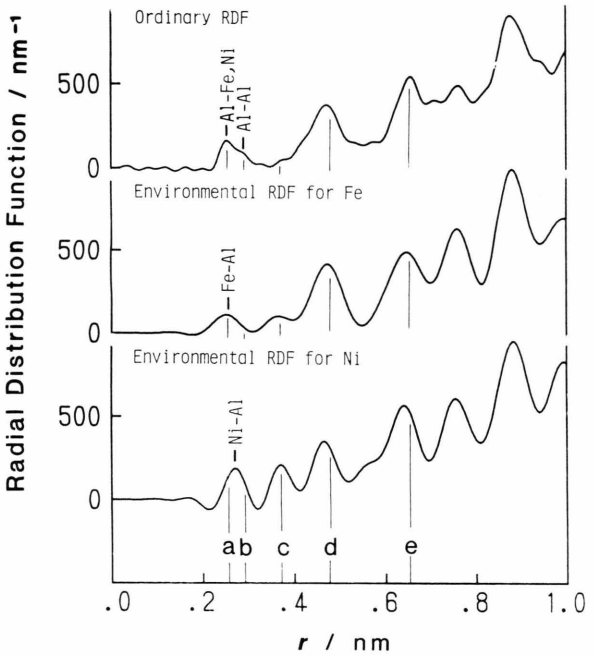

Fig. 3. Ordinary radial distribution function (RDF) and environmental RDFs for Fe and Ni of as-quenched decagonal $Al_{75}Fe_{15}Ni_{10}$ alloy (density = 3.740 g/cm^3).

By observing the similar basic profiles in the three RDFs in Fig. 3, the same conclusion is deduced. For convenience of discussion, the first 5 major peaks are labeled "a" to "e" in the figure. The peak "a" in the ordinary RDF has a shoulder labeled "b" while the first peak in the environmental RDFs for Fe and Ni has no shoulder. Taking account of the definition of the environmental RDF in (2) and the atomic concentration of each constituent element, this first peak at "a" is attributed to pairs of Al and the transition metals, and its shoulder at "b" to Al and Al pairs. The coordination numbers and atomic distances calculated from the first peak of the three RDFs are summarized in Table 1. Since Fe-Al and Ni-Al pairs are located at the maximum of the first peak in the ordinary RDF, the tabulated coordination number was estimated as an average coordination number of Al atoms around Fe and Ni atoms. All of the coordination numbers computed from these three RDFs conform with each other. It is also worth noting that the atomic distances of Al-(Fe, Ni) and Al-Al pairs estimated from the ordinary RDF in Table 1 are very close to the nearest neighboring distances of Al-Fe and Al-Al pairs in $Al_{13}Fe_4$ [10], and their coordination numbers are roughly equal to the values of $Al_{13}Fe_4$. Henley [11] demonstrated that the layer structure of this crys-

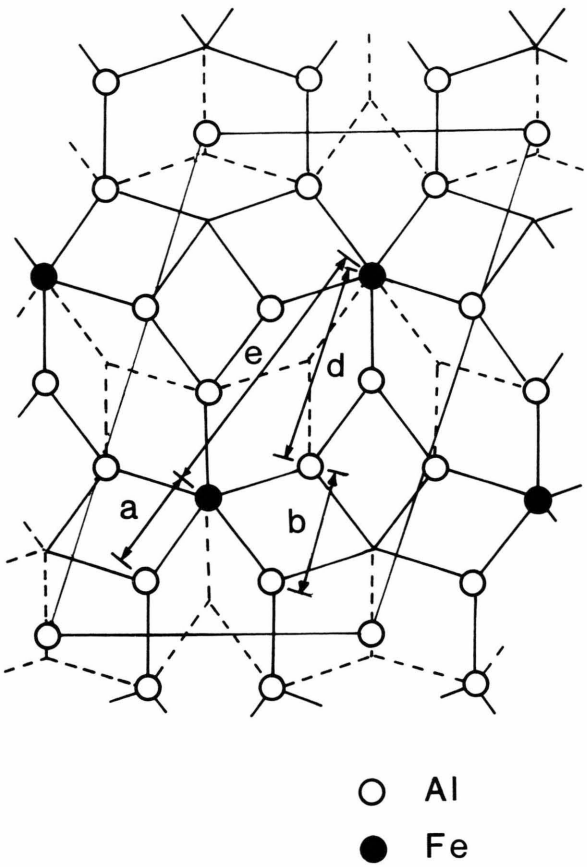

Fig. 4. The puckered layer of the $Al_{13}Fe_4$ structure with the decoration of the two-dimensional Penrose tiling proposed by Henley [11].

Table 1. Coordination numbers and distances in the as-quenched decagonal $Al_{75}Fe_{15}Ni_{10}$ alloy experimentally determined from the first peaks of the ordinary radial distribution function (RDF) and environmental RDFs for Fe and Ni with the values computed from the crystalline data of $Al_{13}Fe_4$ [10].

	r	N
Ordinary RDF		
(Fe, Ni)–Al	0.252 ± 0.002 nm	7.7 ± 0.1
Al–Al	0.284 ± 0.002 nm	8.6 ± 1.1
Environmental RDF for Fe		
Fe–Al	0.252 ± 0.002 nm	7.6 ± 0.2
Environmental RDF for Ni		
Ni–Al	0.270 ± 0.002 nm	8.5 ± 0.3
$Al_{13}Fe_4$		
Fe–Al	0.254 nm	9.7
Al–Al	0.279 nm	7.6

talline $Al_{13}Fe_4$ alloy can analogously be decomposed into the rhombic tiles of the two dimensional Penrose tiling. The puckered layer which is one of the two layers of the $Al_{13}Fe_4$ structure is shown in Fig. 4 [10] with a decoration as the rhombic tiles [11]. The distances labeled "a", "b", "d" and "e" in this figure, for example, correspond to the atomic distances indicated in Fig. 3. This suggests that the local atomic structure of the decagonal $Al_{75}Fe_{15}Ni_{10}$ alloy is very similar to the crystalline structure of the $Al_{13}Fe_4$ alloy. Since the atomic distances between Fe and Al atoms, or Fe and Fe atoms belonging to different layers along the c-axis in $Al_{13}Fe_4$ are approximately equal to the distance "c", the peak labeled "c" may be attributed to some atomic arrangements perpendicular to the decagonal plane.

Edagawa et al. [12] demonstrated that sometimes local atomic structures of icosahedral decagonal phases are successfully approximated by some crystalline structures with very large lattice constants. Our present results in the decagonal $Al_{75}Fe_{15}Ni_{10}$ alloy by the AXS method support their result, and the similarity of the local atomic structure between the decagonal $Al_{75}Fe_{15}Ni_{10}$ and crystalline $Al_{13}Fe_4$ alloys has experimentally been proved.

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[1] A. P. Tsai, A. Inoue, and T. Masumoto, Mater. Trans. Japan. Inst. Metals **30,** 150 (1989).
[2] A. P. Tsai, A. Inoue, and T. Masumoto, ibid. **30,** 300 (1989).
[3] C. L. Henley, Comments on Condensed Matter Physics **13,** 59 (1987).
[4] E. Matsubara, K. Harada, Y. Waseda, and M. Iwase, Z. Naturforsch. **43a,** 181 (1988).
[5] Y. Waseda, Novel Application of Anomalous X-ray Scattering for Structural Characterization of Disordered Materials, Springer-Verlag, New York 1984, p. 84.
[6] Y. Waseda, Structure of Non-Crystalline Materials, McGraw-Hill, New York 1980, p. 60.
[7] International Tables for X-ray Crystallography, Vol. IV, Kynoch, Birmingham 1974, p. 148.
[8] D. T. Cromer and D. Liberman, J. Chem. Phys. **53,** 1891 (1970).
[9] S. Takeuchi and K. Kimura, J. Phys. Soc. Japan **56,** 982 (1987).
[10] P. J. Black, Acta Cryst. **8,** 175 (1955).
[11] C. L. Henley, J. Non-Cryst. Solids **75,** 91 (1985).
[12] K. Edagawa, K. Suzuki, M. Ichihara, S. Takeuchi, and T. Shibuya, Phil. Mag. B, to be submitted.