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ELECTRON DIFFRACTION STUDY ON THE STRUCTURE OF Ni–Fe FILMS PREPARED BY LOW TEMPERATURE CONDENSATION*

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Transmission electron diffraction of 80% Ni–20% Fe thin films prepared by low temperature condensation has been investigated. A radial distribution analysis of the obtained diffuse halo patterns has also been carried out. The radial distribution function RDF(r) and the Menke probability function $W(r)$ have been compared with RDF(r) and W(r) calculated from the proposed model.

1. Introduction

Several studies [1–6] on the structure of films prepared by low temperature condensation were previously carried out by means of the radial distribution analysis of diffraction patterns. Patterns of many of films prepared at low temperatures showed diffuse halos which are characteristic of a highly disordered structure. The radial distribution function (RDF) determined from such patterns suggest that films are certainly in an amorphous state.

It is the purpose of the present study to determine the radial distribution function RDF(r) and the Menke probability function $W(r)$ for 80% Ni–20%Fe films condensed on substrates at liquid nitrogen temperature (LNT) by carring out the radial distribution analysis of electron diffraction patterns and to compare them with functions calculated from a model.

2. *Experimental procedure*

The 80%Ni–20%Fe films produced by low temperature condensation are generally metastable and crystallize before reaching room temperature [7]. Therefore the experimental work must be performed in situ. For the diffraction studies reported here a low-temperature thin film deposition unit was constructed for the TESLA BS 413 electron microscope.

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Ni—Fe was evaporated at the evaporation rate of about 20 Å/s from a V-shape tungsten filament wire and was condensed on a substrate of carbon film about 200 Å thick at LNT in a vacuum of 10^{-5} torr. The thickness of films were roughly estimated from the amount of evaporated material to be about 100 — 600 Å, respectively. The optimum film thickness for diffraction experiments lies between 100 — 200 Å and in this range there was no detectable change in structure with thickness.

The observed diffraction patterns were recorded on photographic plates and the blacking procedure related to the diffraction intensity throught microdensitometer scans of the plates [8]. Diffraction rings of a polycrystalline gold film were used as a standard s-value ($s = 4\pi \sin\theta/\lambda$, where 2θ is the scattering angle and λ the electron wavelenght).

3. Radial distribution analysis

3.1. Theory

As is well known the radial distribution function RDF(r) is experimentally obtained by the Fourier transformation of the structure sensitive intensity $I_m(s)$

$$\mathrm{RDF} = 4\pi r^2 \varrho(r) = 4\pi r^2 \varrho_0 + \frac{2r}{\pi} \int_0^\infty s I_m(s) \sin(sr) ds, \tag{1}$$

where $\varrho(r)$ is the atomic density at the distance r from an atom, ϱ_0 the mean atomic density. In the present case ϱ_0 could not be determined experimentally and therefore the assumption was adopted that the density ϱ_0 in amorphous alloys is the same as that of the crystalline state. The method of deriving $Im(s)$ from the diffuse halo pattern is described in other papers [2, 4].

The range of s, where $I_m(s)$ is experimentally obtained, is limited because of very strong intensity near the direct beam and a great decrease of the observed intensity for the large s value. Therefore, a damping facor, $\exp(-Bs^2)$, called the artificial temperature factor is introduced to reduce the termination effect. The constant B is usually determined so that the damping factor equals 0.1 for the largest available s value ($s_{\max}$). Thus, equation (1) becomes:

$$\begin{aligned} \mathrm{RDF} &= 4\pi r^2 \varrho^*(r) \\ &= 4\pi r^2 \varrho_0 + \frac{2r}{\pi} \int_0^{s_{\max}} s I_m(s) \exp(-Bs^2) \sin(sr) ds, \end{aligned} \tag{2}$$

where $\varrho(r)$ is converted to $\varrho^*(r)$.

The atomic arrangement in the film should be discussed from the experimental Menke probability function

$$W(r) = \frac{\mathrm{RDF}(r)}{4\pi r^2 \varrho_0} = \frac{\varrho^*(r)}{\varrho_0}. \tag{3}$$

The integration of equation (2) and (3) are carried out numerically using ODRA 1304 electronic computer.

3.2. Results

A Ni—Fe film about 200 Å in thickness which was prepared by condensation at LNT gave a diffuse halo pattern as shown in Fig. 1(a). The pattern rapidly changed to a ring pattern as showns in Fig. 1(b) at about 120 K on heating.

A curve in Fig. 2 showns the structure sensitive intensity $Im(s)$ derived from the halo pattern of the 80%Ni—20%Fe at LNT. Values of $I_m(s)$ were obtained in the range from $s = 1.5$ to $10 Å^{-1}$. Those in the range $s = 0$ to $1.5 Å^{-1}$ were reasonably extrapolated to

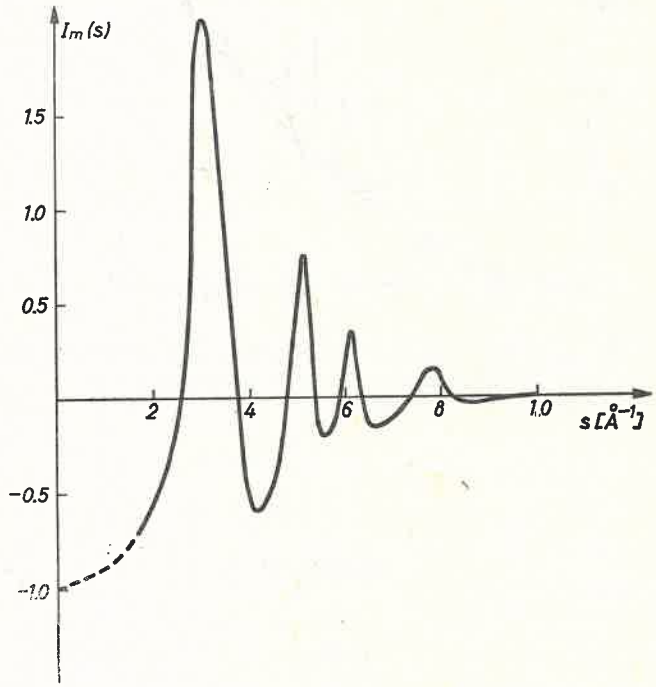


Fig. 2. $I_m(s)$ of a Ni–Fe film condensed at LNT

-1.0 at $s = 0$, because an appreciably diffracted intensity in the neighborhood of $s = 0$ due to long-ranged density fluctuation in the film was not found by visible observation. The maxima in the curve are observed at $s = 3.0$, 5.1, 6.1, 7.9, and $9.2 Å^{-1}$.

The RDF in Fig. 3 was determined by the Fourier transformation of $I_m(s)$ with $\varrho_0 = 8.33 \cdot 10^{-2}$ atoms/Å^3 and $B = 0.02 Å^2$ as the damping factor. Seven peaks are observed at 2.6, 4.28, 5.20, 6.53, 7.57, 8.76 and 10.4 Å. Table I is a comparison of peak positions obtained experimentally and by model building. It can be seen that there is very good agreement between the experimental results described here and the theoretical results of the Ichikawa model [10]. Interatomic distances in fcc 80%Ni−20%Fe are shown by bars placed on Fig. 3 whose length is proportional to the number of neighbors having the respective distance. The co-ordination number (CN) was determined by the area under the first peak of a RDF curve. The CN of Ni—Fe film is 11 which is larger than that of the liquid metal.

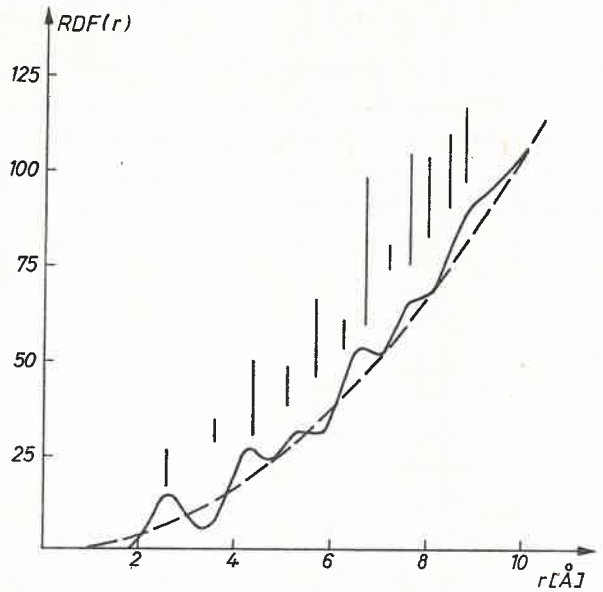


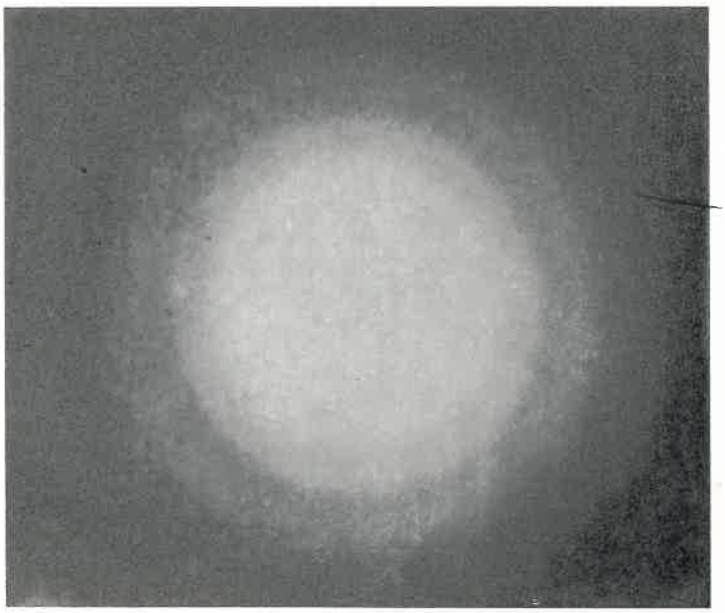
Fig. 3. RDF(r) curve for Ni–Fe film condensed at LNT. Bars show interatomic distances in fcc Ni–Fe alloy. Dotted curve shows $4\pi r^2 \varrho_0$

TABLE I

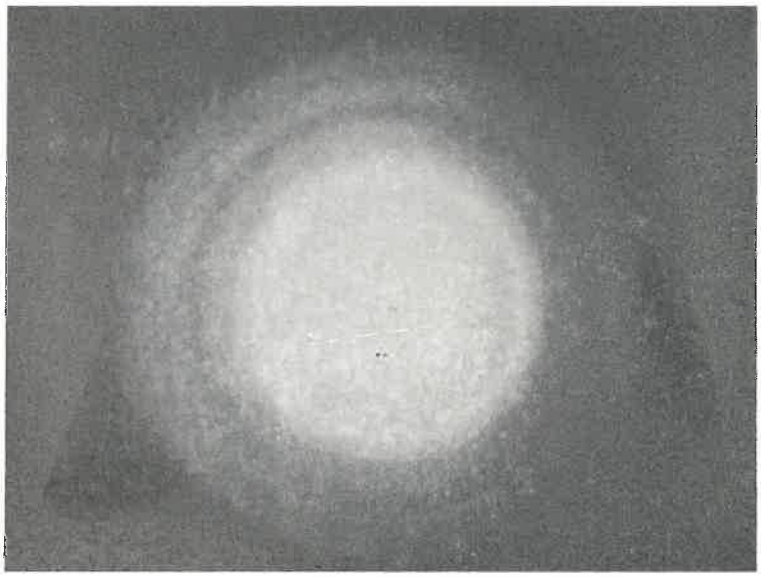
Element	r_2/r_1	r_3/r_1	r_4/r_1	r_5/r_1	r_6/r_1	Reference
			Experimental			
80% Ni–20% Fe	1.65	2.00	2.51	2.91	3.36	Present work
50% Ni–50% Fe	1.67	—	2.50	—	3.29	Fujime [3]
Ni	1.71	1.92	2.56	—	3.38	Ichikawa [5]
Fe	1.67	1.96	2.51	—	3.38	Ichikawa [5]
			Calculated			
	1.66	1.99	2.52	2.96	3.34	Ichikawa [10]
	1.67	2.00	2.52	—	3.34	Leung and Wright [11]
	1.68	2.00	—	—	—	Bennett [12]
	1.73	2.00	2.68	—	3.53 (a)	Finney [13]
	1.73	1.99	2.68	—	3.53 (b)	
	1.65	2.00	—	—	—	Sadoc et al. [14]

In the case of the models constructed by Finney (a) refers to the local model and (b) the global model.

To quantitatively describe this structure the cluster model have been proposed. This model starts with atomic shell distances of the atoms and assumes a Gaussian distribution function for each shell in order to account for disordering displacement around the



a



b

Fig. 1. Diffraction pattern of a 80% Ni–20% Fe film 200 Å thick; a) immediately after condensation at LNT, b) after crystallization

center of the shell [9, 15]. The RDF(r) and $W(r)$ were calculated by the following formulas:

$$\mathrm{RDF}(r) = 4\pi r^2 \varrho_0 + S_z \left\{ \sum \frac{N_i \exp\left[-(r-r_i)^2/(4\sigma_i+4B)\right]}{\sqrt{4\pi(\sigma_i+B)}} \right\}, \tag{4}$$

$$W(r) = \frac{\mathrm{RDF}(r)}{4\pi r^2 \varrho_0}$$

$$= 1 + \frac{S_z}{4\pi r^2 \varrho_0} \left\{ \sum \frac{N_i \exp\left[-(r-r_i)^2/(4\sigma_i+4B)\right]}{\sqrt{4\pi(\sigma_i+B)}} \right\}, \tag{5}$$

where N_i is the number of atoms in i'th radial shell, S_z — a damping factor determined from the size of the cluster, r_i the interatomic distance to the i'th neighbor, B the damping factor used in (2), and σ_i a mean square deviation of the i'th neighbor distance.

For a spherical cluster of atoms S_z my be expressed by the following formula [15]:

$$S_z(r, r_c) = 1 - \tfrac{3}{4}\frac{r}{r_c} + \tfrac{1}{16}\left(\frac{r}{r_c}\right)^3 \quad \text{for } r \leqslant 2r_c$$

$$S_z(r, r_c) = 0 \quad \text{for } r > 2r_c, \tag{6}$$

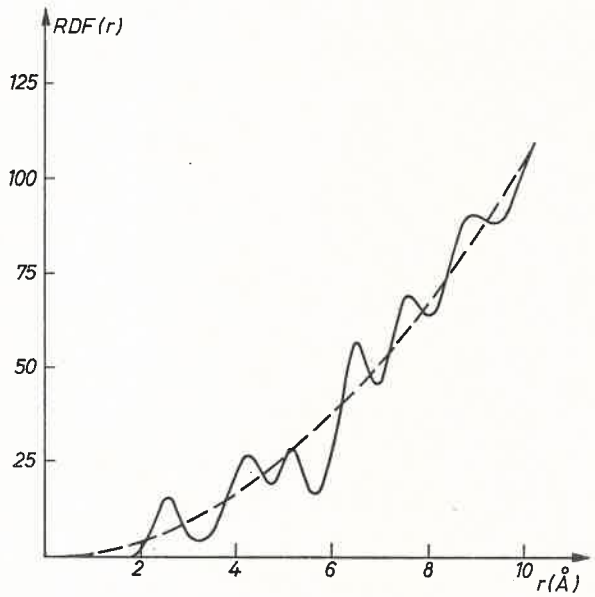


Fig. 4. The theoretical RDF(r) curve calculated from the proposed model

were r_c is the radius of a cluster. The r_c, thus chosen for this model was 10 Å. Atoms of the i'th shell in the amorphous state after crystallization belong to these crystallographic planes whose distance from the central atom are in the RDF(r) peak region. In this state

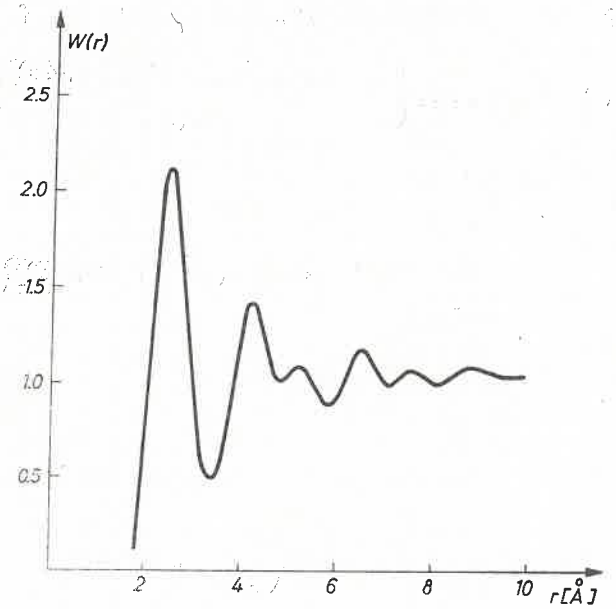


Fig. 5. $W(r)$ curve for Ni–Fe film condensed at LNT

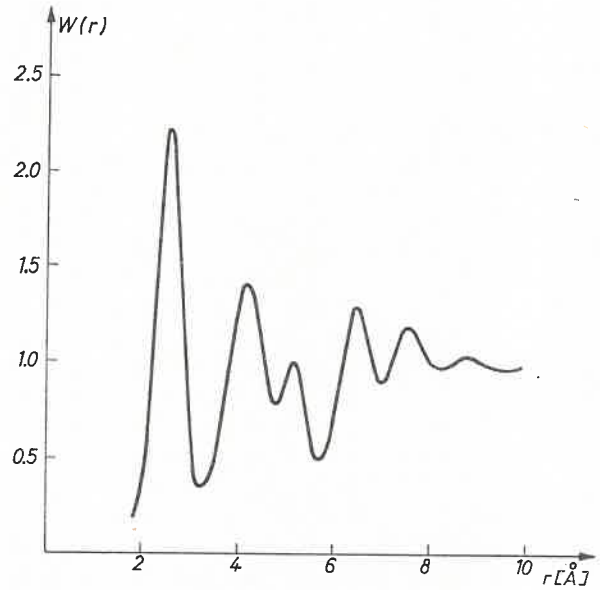


Fig. 6. The theoretical $W(r)$ curve calculated from the proposed model

there are many defects and impurities and thus the number of atoms in each shell is smaller than the number of atoms that belong to suitable crystal planes.

The values of r_i, σ_i and N_i have been obtained from RDF(r) fitted with the Gaussian functions and were 0.013 and 11 for the ones at 2.6 Å, 0.05 and 29 for the ones at 4.28 Å,

0.05 and 32 for the ones at 5.2 Å, 0.03 and 50 for the ones at 6.53 Å, 0.05 and 66 for the ones at 7.57 Å, 0.1 and 120 for the ones at 8.76 Å and 0.2 and 180 for the ones at 10.4 Å, respectively. The agreement of the experimental RDF(r) (Fig. 3) and $W(r)$ (Fig. 5) curves with the calculated functions (Fig. 4 and 6) was satisfactory.

4. Conclusion

Film of 80%Ni–20%Fe prepared by LNT condensation showed the following properties characteristic of highly disordered structure:

— Electron diffraction patterns obtained immediately after condensation consist of diffuse halos.

— On heating, diffraction patterns changed from halos to rings at temperature 120 — 150 K depending upon film thickness. At the same time electric resistance decreased rapidly [7].

— The radial distribution analysis suggested that the films are not in a "liquid-like" structure.

It is concluded from the above results that 80%Ni–20%Fe films prepared by LNT are in an amorphous state.

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