Convergent Beam Dark Field Imaging Technique with Its Application to Observation of Multiphase Materials

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A new technique for the observation of multiphase materials has been proposed, which enables us to distinguish easily different phases by their different contrasts. The technique is based on the broadening of diffraction spots and the overlapping of dark field images due to beam convergence. Since contrasts are adjustable by changing the beam convergence, the contrast of a weak diffraction phase can be made brighter than a strong diffraction phase. An observation of the precipitates in a Ti-10V-2Fe-3Al alloy by this technique showed that a stable α phase precipitates nucleating at the metastable ω phase in the supersaturated β matrix.

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I. Introduction

There are many cases in which more than one phase in a specimen need to be identified by transmission electron microscopy (TEM). However, such identification is difficult for TEM, because overlapping of images of different phases is intrinsic in TEM observations. Stereo microscopy provides three-dimensional pictures, however, it is not useful for a specimen containing many particles. To distinguish the overlapping one from the direct contact between particles of different phases, it is necessary to observe many particles in order to see all the particles connected with another phase. Superimposition of dark field images (DFIs) taken with diffraction spots of each phase can be applied in such case, however, it is difficult to locate the same positions in both images when particles are small. High resolution electron microscopy (HREM) is capable of observing several phases at the same time⁽¹⁾. However, a small number of precipitates is observable due to the high magnification in HREM method. Moreover HREM images strongly depend on such conditions as focussing, thickness of a specimen, orientations, etc. Using two or three diffraction spots within a large objective aperture, one can obtain a DFI containing particles of different phases in a single recording. However, this does not give sharp images. The "2 1/2 D technique"⁽²⁾ is useful to distinguish phases, however images are defocused and positions of particles are shifted in principle.

The aim of the present study is to describe a new technique based on DFI for obtaining images of several kinds of phases simultaneously, using a small objective aperture. The technique is applied to clarify a nucleation site of precipitates in a near- β Ti alloy.

II. Experimental Procedures

A Ti-10V-2Fe-3Al (in mass%) alloy studied previously⁽³⁾ was used. The chemical composition of the alloy is shown in **Table 1**. Specimens of about $7 \times 7 \times 3$ (mm³) in size were heat-treated at 1033 K for 7.2 ks in argon atmosphere, and then quenched into ice water, followed by aging at 573 K for 26 Ms. The specimens were mechanically polished and then electropolished by a twin-jet method using a mixture of 50 ml methanol, 3 ml n-butyl alcohol and 30 ml perchloric acid. TEM observations were carried out using a JEM-200CX operating at an accelerating voltage of 200 kV.

III. Results

The alloy is composed of a bcc (β) matrix and primary hcp (α) grains in the as-quenched condition⁽³⁾⁽⁴⁾. It is generally recognized that the metastable ω phase precipitates preceding the precipitation of the stable α phase in supersaturated β grains during aging at relatively low temperatures⁽³⁾⁻⁽⁶⁾. Examples of dark field images of the specimen are shown in **Fig. 1** along with the corresponding diffraction pattern. The beam direction is parallel to $[110]_{\beta}$. Since the precise lattice constants of α and β phases in the present alloy were not obtained, they were estimated based on the data reviewed by Pearson⁽⁷⁾⁽⁸⁾ as a Ti-10 mass%V alloy and Ti-2 mass%Al alloys, respectively, that is, $a_{\beta}=0.325$ nm and $a_{\alpha}=0.295$, $c_{\alpha}=0.472$ nm, respectively. The lattice parameters of ω phase were calculated to be $a_{\omega}=\sqrt{2}a_{\beta}=0.459$ nm and

Table 1 Chemical composition of the alloy (mass%).

V	Fe	Al	O	Ti
10.13	2.04	3.03	0.099	bal.

Fig. 1 Conventional dark field images of the Ti-10V-2Fe-3Al aged at 573 K for 26 Ms. (a) Selected area diffraction pattern, (b) dark field image of ω_1 precipitates, (c) dark field image of α_1 and α_2 precipitates.

●:matrix (β), □: ω_1 , ■: ω_2 , ○: α_1 , ●: α_2

Fig. 2 Key diagram for Fig. 1(a).

$c_\omega = \sqrt{3} a_\beta / 2 = 0.281 \text{ nm}^{(6)(9)}$. A key diagram of the diffraction pattern shown in Fig. 1 is indexed in Fig. 2. The orientation relationships between β and ω , and that between β and α are

$$(110)_\beta // (\bar{2} \ 110)_\omega, \quad [\bar{1} \ 11]_\beta // [0001]_\omega$$

and

$$(110)_\beta // (0001)_\alpha, \quad [\bar{1} \ 12]_\beta // [10 \ \bar{1} \ 0]_\alpha,$$

respectively, which coincide with those reported in the literatures⁽³⁾⁽⁵⁾⁽⁶⁾⁽¹⁰⁾⁻⁽¹²⁾. There are four variants of the ω phase and twelve variants of the α phase. The key diagram (Fig. 2) shows two variants for both of the α (α_1 , α_2) and ω (ω_1 , ω_2) phases. Preliminary calculations of the diffraction pattern show that such spots indicated in Fig. 1(a) do not overlap with other variants. Figure 1(b) shows a dark field image taken with ω_1 . Although Fig. 1(c) is taken with ($\alpha_1 + \alpha_2$), the precipitates α_1 and α_2

Fig. 3 Conventional dark field image taken with spots of ω_1 , α_1 and α_2 phases.

can be distinguished from each other as indicated in (c), because the spots from α_1 and α_2 are elongated in different directions due to the shape effect of precipitates. Superimposition of DFIs ((b) and (c)) each other did not reveal whether the ω_1 precipitates coincide with α_1 or α_2 precipitates.

A dark field image taken with ($\omega_1 + \alpha_1 + \alpha_2$) using a large aperture is shown in Fig. 3. Because the intensity of ω_1 is stronger than those of α_1 and α_2 (see Fig. 1(a)), images of α precipitates are difficult to recognize. Besides, contrasts like planer defects are seen in ω precipitates, which may be confused with a precipitates.

Thus, a new technique was attempted, that is, spots of $0\bar{1} \ 10_{\alpha_1}$ and $0\bar{1} \ 10_{\alpha_2}$ (see Fig. 2) were taken into a small objective aperture, then an electron beam was gradually converged and DFIs were taken. The resultant images are shown in Fig. 4. Diffraction spots used for observations are indicated in each DFI with the objective aperture. When the specimen is illuminated by a nearly parallel beam (a), only α_1 and α_2 precipitates are observed though weak in contrast. With increasing convergence of the beam, images of α_1 and α_2 become clear ((b), (c)). In (d), an additional faint contrast of precipitates appears,

Fig. 4 Series of dark field images. Beam convergence increases in the order of (a) to (i). Inserted pictures show diffraction spots in the objective aperture.

which is due to $01\bar{1} \ 1_{\omega 1}$ spot. The center of this spot is out of the objective aperture as schematically illustrated in (d), but a diffusely broadened portion of the spot contributes to the image because of beam convergence. The intensities of $\alpha 1$ and $\alpha 2$ spots are higher than the diffuse portion of the ω spot, therefore the images of α are brighter than those of ω . Since brightness of $\omega 1$ precipitates increases with beam convergence, which makes it difficult to distinguish between α and ω ((g), (h), (i)). The condition of convergence shown in (e) or (f) is suitable for observation. This image is named the “convergent beam dark field image (CBDFI)”. Changes in intensity distribution of the diffraction spots are schematically shown in Fig. 5. When the specimen is illuminated by a parallel beam, diffraction spots g_a and g_b due to the phases a and b, respectively, are apart from each other at the distance of d_s , and only the spot g_a can be selected by an objective aperture with radius r_o , (a). With increasing

beam convergence, the intensities of g_a and g_b increase and the diffusely broadened portion of the spot g_b enters the objective apertures, (b) to (d). At the maximum convergence, the diffraction spots change into discs with radius r_c which depends on the size of a condenser aperture, (e). Conditions of taking the images of Fig. 4(a), (f) and (i) correspond to those of Figs. 5(a), (c) and (e), respectively.

Although the CBDFI taken under the maximum convergence is not always suitable for observations (see Fig. 4(i)), r_c is useful for expressing the necessary condition to take a CBDFI as follows:

$$r_c \geq d_s + r_o \quad (1)$$

or

$$d_s + r_o > r_c > d_s - r_o \quad (2)$$

In mathematical meaning, eqs. (1) and (2) are simply de-

Fig. 5 Schematic illustration of change in distribution of intensity in diffracted beams and position of an objective aperture. Beam convergence increases in the order of (a) to (e). g_a and g_b : diffraction spots. d_s : distance between diffraction spots. r_o , r_c : radii of objective and condenser apertures on the back focal plane of the objective lens.

Fig. 6 Schematic illustration showing the condition for CBDFI. d_s , r_c , r_o : same as Fig. 5. Objective aperture is fully; (a) or partly; (b) covered by the overlapped region of diffraction discs.

scribed as $r_c > d_s - r_o$. However, as schematically shown in Fig. 6, eq. (1) is for the condition in which the objective aperture is fully covered by the overlapped region of diffraction discs. On the other hand, eq. (2) is for the condition in which the objective aperture is partly covered by the overlapped region, that is, a part of a micrograph taken at the condition of eq. (2) lacks an image of the "b" phase. In the present study, condenser and objective apertures 300 and 20 μm in diameter were used. This corresponds to $r_c = 1.1$ and $r_o = 0.6 \text{ nm}^{-1}$, respectively, in a reciprocal lattice space, and the distance d_s between the discs of $0\bar{1}10_{\alpha 1}$ and $01\bar{1}1_{\omega 1}$ were about 0.7 nm^{-1} , which satisfies eq. (2).

The distance between $01\bar{1}1_{\omega 1}$ and $01\bar{1}1_{\omega 2}$ is about 2.5 nm^{-1} , which does not satisfy eq. (1) nor (2). Therefore, in order to observe $\omega 2$ precipitates together with $\alpha 1$ and $\alpha 2$, it is necessary to take another image by putting $\bar{1}010_{\alpha 1}$ and $\bar{1}010_{\alpha 2}$ spots into the objective aperture and using the diffuse part of $01\bar{1}1_{\omega 2}$ (see Fig. 2). Figures 7(a), (b) and (c) show the CBDFIs taken with $(0\bar{1}10_{\alpha 1} + 0\bar{1}10_{\alpha 2} + \text{diffuse part of } 01\bar{1}1_{\omega 1})$, $(\bar{1}010_{\alpha 1} + \bar{1}010_{\alpha 2} + \text{diffuse part of } 01\bar{1}1_{\omega 2})$, and an image printed by superimposing the films for (a) and (b), respectively. The same precipitates of $\alpha 1$, $\alpha 2$, $\omega 1$ and $\omega 2$ are indicated in Fig. 5. Most of the $\alpha 1$ precipitates are observed correlating with the $\omega 1$ precipitates in (a). A part of the $\alpha 2$ precipitates is

observed at $\omega 1$, as indicated by arrow "a" in (a), however, it is correlated with $\omega 2$ as marked by arrow "b" in (b). That is, $\omega 1$ and $\omega 2$ are overlapped in such a case. It is clearly shown from (c) that the $\alpha 1$ and $\alpha 2$ precipitates are formed at the $\omega 1$ and $\omega 2$ precipitates, respectively. The orientation relationship between ω and α is as follows:

$$\begin{aligned} (\bar{2}110)_{\omega} // (0001)_{\alpha}, \\ [0001]_{\omega} // [\bar{1}2\bar{1}0]_{\alpha} \end{aligned}$$

in agreement with that reported by Takemoto *et al.*⁽¹³⁾ based on an HREM image of an aged Ti-Mo binary alloy. However, as mentioned in Section I, HREM has an ambiguity in the interpretation of images, and also observable precipitates are few.

The shape of α precipitates is plate-like, which caused the contrast like planar defects in Fig. 3. The α plates are parallel to $(10\bar{1}0)_{\alpha} // (01\bar{1}0)_{\omega}$ planes, which leads to twelve α variants as a result of three α variants for each ω in four variants.

As an optical micrograph, the image in which the precipitates are dark and the matrix is bright is useful for visually understanding a microstructure. Thus, superimposed CBDFI like as Fig. 7(c) was negatively printed and shown in Fig. 8, in which the dark precipitates are α and the halftone precipitates are ω . Many authors deduced the α nucleation at the ω from similarity in size and distribution of α precipitates to those of ω when the α formed after ω precipitation⁽⁶⁾⁽¹²⁾⁽¹⁴⁾⁽¹⁵⁾. The present results give direct evidence for their speculation.

IV. Discussion on the CBDFI Method

Reciprocal lattice points have Gaussian distribution in intensity with a half-peak width of about $1/w^{(16)}$, where w is a size of an object, under parallel beam illumination. When the beam is converged, the peak intensity is increased and the reciprocal lattice point becomes spread. The size of diffraction spot reaches to the maximum when the beam is focused on the specimen and a disc pattern appears. The diameter of the disc depends on the incident beam half-angle, $\alpha^{(17)}$. Overlapping of diffraction

Fig. 7 CBDFIs taken with (a) $(0\bar{1}10_{\alpha1} + 0\bar{1}10_{\alpha2} + \text{diffuse part of } 01\bar{1}1_{\omega1})$, (b) $(\bar{1}010_{\alpha1} + \bar{1}010_{\alpha2} + \text{diffuse part of } 01\bar{1}1_{\omega2})$, (c) an image printed by superimposing the films for (a) and (b).

Fig. 8 Negative print of superimposed CBDFIs.

spots or discs would occur when there are two or three phases in a specimen and they have similar lattice spacings each other. By placing a small objective aperture in the overlapping region, one can obtain the images of two or more phases depending on the number of overlapping spots.

The main advantages of CBDFI technique are as follows: (1) one can identify the different phases at the same time through the contrast change with beam convergence, and (2) one can make the contrast of a weak diffraction phase higher than that of a strong diffraction phase (see Fig. 5(c)). However, it should be noticed that there are limitations to use this technique. The key limitations are as follows: (1) fundamental analysis of a diffrac-

tion pattern is necessary to find an orientation that one of the diffracted spots of each phase to be close to each other, (2) the size of the particle should be small to allow the relatively uniform distribution of intensities within the disc when the disc pattern is used, (3) the smallest aperture preferably used to take a sharp image, and (4) series of images should be taken changing the beam convergence like as HREM images taken changing defocus values.

V. Summary

A new technique for observation of multiphase materials is proposed. The procedure of the technique is as fol-

lowings: (1) one of the diffraction spots belonging to one of the phases is centered in an objective aperture, (2) the electron beam is converged to broaden the diffraction spots using a condenser lens focus, (3) a part of the diffusely broadened spot belonging to another phase is taken into the aperture, and (4) a dark field image is taken. The contrast of each phase depends on beam convergence and can be adjusted to easily distinguish the different phases from each another. This technique enables one to observe a phase with weak diffraction intensity in brighter contrast than a phase with strong diffraction intensity.

Observations of precipitates in a Ti-10V-2Fe-3Al alloy by this technique showed that the stable α precipitate is nucleated at the metastable ω precipitate with the following orientation relationship:

$$(\bar{2} 110)_{\omega} // (0001)_{\alpha}, \text{ and } [0001]_{\omega} // [\bar{1} 2\bar{1} 0]_{\alpha}.$$

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