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Authors

Sinclair, R.
Thomas, G.

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ANTIPHASE DOMAINS AND SUPERLATTICE SPOT SPLITTING
IN Cu_3Au I

R. Sinclair and G. Thomas

Department of Materials Science and Engineering, College of Engineering,
and Inorganic Materials Research Division, Lawrence Berkeley Laboratory,
University of California, Berkeley, California 94720.

ABSTRACT

Lattice imaging by high resolution electron microscopy has been utilised in the study of antiphase boundaries and antiphase domains in Cu_3AuI . A displacement of half the (100) fringe spacing characterises the antiphase boundary and, by following the trace of (100) fringes from one ordered domain to another, the relative phase of adjacent domains may be established. The technique has shown that the discrete ordered domains formed in the early stages of ordering in Cu_3AuI are in small groups in which adjacent domains are in antiphase, with good correlation between the antiphase domain separation and the splitting of superlattice reflections in the corresponding diffraction patterns. It has also provided direct evidence that small regions of relatively high local degree of order (microdomains) exist in the disordered alloy.

I. INTRODUCTION

One of the principal features of diffraction patterns taken of Cu_3AuI alloys during the early stages of ordering is the splitting of the superlattice reflections. The character of the splitting is similar to, but not as marked as, that observed in $\text{Cu}_3\text{Au II}$, the long period superlattice form of the alloy in which antiphase boundaries (A.P.B.'s) are present periodically throughout the lattice. The splitting has previously been interpreted in terms of a distribution of APB's in the material⁽¹⁻³⁾ but direct evidence for this suggestion was lacking. Dark field transmission electron microscopy using a superlattice reflection revealed that in these early stages the alloy is composed of small discrete domains of order within a disordered matrix⁽³⁾. The ordered domains are not in contact with one another. It is impossible to determine from such images whether adjacent domains are in phase or antiphase since a disordered matrix, not an antiphase boundary, exists between them. Consequently characteristic APB contrast cannot be achieved⁽⁴⁾. This difficulty had been ignored in previous studies, but has been overcome in the present work utilising the high resolution lattice imaging electron microscopy technique.

II. EXPERIMENTAL PROCEDURE

Thin foil specimens of Cu_3Au were prepared in the [001] orientation by vacuum deposition methods. Although exact stoichiometry is difficult to achieve, it was not thought that this would affect the conclusions drawn from structural observations. Lattice images were taken, in a Philips EM 301 electron microscope, of specimens oriented in a strong [100] systematic row with the (200) reflection satisfying the Bragg

condition. The transmitted, (100) superlattice and (200) fundamental reflections were selected to form the image with an appropriate objective aperture (40μ in diameter) and with the illumination tilted so that the (100) beam was parallel to the optical axis of the microscope. These are the optimum conditions for imaging the (100) superlattice planes in Cu_3Au ⁽⁵⁾.

III. RESULTS

A. Antiphase Boundaries in Fully Ordered Cu_3Au .

Previous electron microscope observations⁽⁶⁾ have demonstrated that translational antiphase boundaries in Cu_3Au lie principally on {100} planes with the antiphase vector, $\frac{a}{2}\langle 011 \rangle$, lying in the boundary plane. This brings the all-copper (200) atomic planes of the L1_2 structure into coincidence with the mixed copper-gold planes on the other side of the boundary. The effect on the lattice image of (010) planes perpendicular to the boundary plane, (100), would be to shift the fringes by an amount $\frac{a}{2}[010]$ ⁽⁷⁾. Thus, the superlattice (010) planes should exhibit a discontinuity of half a fringe spacing at the APB. This effect is illustrated in Fig. 1 for an APB lying predominantly parallel to (010). The discontinuity is quite clear with a dark fringe on one side of the boundary meeting a white fringe on the other side.

Half spacing fringe shifts due to electron optical effects are known to occur for fringes perpendicular to a foil edge at specimen thicknesses of half an extinction distance and multiples thereof⁽⁸⁾. However, in the present case no confusion with this phenomenon was possible since the AP boundary lay approximately perpendicular to the edge of the foil and also since specimens prepared by the evaporation method

are approximately parallel-sided.

B. Partially Ordered Cu_3Au

Partially ordered specimens were obtained by slowly cooling the thin foils through the critical ordering temperature. The electron diffraction patterns of such specimens exhibited the superlattice reflection splitting which causes the $\{hko\}$ reflections ($h \neq k$) to appear spindle-shaped, and the $\{hho\}$ reflections to appear cross-shaped. (e.g. Fig. 2a). The individual maxima were generally recognizable in the $\{100\}$ reflection, and these were used in measurements of the spot splitting, s . Dark field images using the superlattice reflection clearly indicated a dispersion of discrete ordered domains ($\approx 25\text{\AA}$ in diameter) in a disordered matrix (Fig. 2b). In a previous electron microscope correlation of s with the domain distribution⁽³⁾, it had been assumed that alternate domains were in antiphase. This assumption strongly affects the measured separation of antiphase domains and clearly cannot be verified by the dark field technique. However, in the lattice image it is possible to determine the relative phase of ordered domains by following the line of superlattice fringes from one domain to another.

Lattice images taken with $(000) + (100) + (200)$ reflections allow simultaneous imaging to be achieved of the disordered matrix (i.e. $(000) + (200)$ beams yielding (200) spacing fringes) and the ordered domains (i.e. $(000) + (100) + (200)$ beams with the (100) fringe periodicity of the superlattice). Discrete regions with alternately stronger and weaker fringe visibility were evident (these give the appearance of alternately wider and narrower white fringes in normal prints, as shown in Fig. 3). These regions thus

possess the fringe periodicity of the superlattice, which is twice the periodicity of the fundamental lattice in this case. This result is interpreted as imaging of regions in which the local degree of order is fairly high. These regions also correlated with the positions of bright areas in the corresponding conventional superlattice dark field micrographs.

The ordered domains are not always very distinctive in this simultaneous image but their visibility can be enhanced by three methods⁽⁹⁾:

- (i) by viewing the print at an oblique angle;
- (ii) by making a positive print of the original photographic plate;
- (iii) by defocus of the objective lens.

In this latter method, small changes of focus were made to cause blurring of the image sufficiently to lose resolution of the (200) fringes of the disordered matrix but to leave (100) fringes visible in the ordered regions.

Examination of images of this type showed that many of the domains existed in groups of three to four in which adjacent domains were in antiphase with one another (e.g. Fig. 4). The average separation of these antiphase domains was found to be $26 \pm 3 \text{ \AA}$. Raether⁽¹⁾ proposed that the superlattice reflection splitting was related to the separation of antiphase domains (M) by an expression

$$s = \frac{\text{constant}}{M}$$

where the constant varied from 0.37, if a group of only two domains were in antiphase, to 0.5 when five or more domains existed in the group. Using a value of 0.45 for the constant, corresponding to a group of between three and four, the value of M calculated from the observed splitting was found

to be $29 \pm 3\text{\AA}$. The agreement between experimental and calculated values is good. Our observations confirm Raether's suggestion that domains may exist in small groups in antiphase with one another.

C. Disordered Cu_3Au

The diffraction pattern of foils quenched from above the critical temperature is known to exhibit weak diffuse scattering at the superlattice positions with shapes similar to that obtained in the early stages of ordering⁽¹⁰⁾. Two models have been proposed in explanation of this phenomenon:

- (i) the disordered alloy contains small ordered domains which have some antiphase relation as in the early stages of ordering^(1,11);
- (ii) the diffuse scattering intensity, being related to the Fourier transform of the atomic pair interaction parameter, is reflective of the shape of the Fermi surface of the alloy⁽¹²⁾.

In the latter model the structure of the alloy is essentially homogeneous whereas in the former heterogeneity in the local degree of order on the scale of $\approx 10 - 100\text{\AA}$ is suggested (i.e. short-range order)⁽¹³⁾.

Previous electron microscope studies have utilised dark field microscopy from the diffuse reflections to demonstrate the presence of small ordered domains (microdomains). However Cowley⁽¹⁴⁾ has recently shown that a direct correlation between small bright areas in such images and regions supposedly giving rise to the diffuse scattering maxima (i.e. microdomains) may not be valid. In the present study therefore lattice imaging has been used to provide extra information about the nature of short-range order in this system.

Fig. 5a shows the superlattice dark field image of a Cu_3Au specimen quenched from 600°C in which small equiaxed bright areas $\approx 15\text{\AA}$ in diameter are evident. The corresponding electron diffraction pattern is shown in Fig. 5b with weak diffuse maxima at superlattice positions. A defocussed lattice image taken in the above imaging mode is similar to those of the partially ordered alloy, with discrete regions showing the superlattice fringe spacing (Fig. 6). These regions are thus indicated to have a substantially higher local degree of order than the surrounding material with a size in the range three to eight fringe spacings (approximately $10 - 30\text{\AA}$). The number density, and size are lower than for domains in the partially ordered alloys consistent with the much weaker diffracted intensity at the superlattice positions. Examination of the images did not reveal definite antiphase groups as in the partially ordered alloy, although occasionally a pair of antiphase domains was found. The images thus give direct evidence in favor of a microdomain model for the short-range ordered state in Cu_3Au but a firm correlation of the shape of the diffraction spots with antiphase domain groups has not been found for the quenched alloy.

IV. CONCLUSIONS

Lattice fringe imaging has proved successful in revealing the out-of-step nature of translation APB's in Cu_3Au , in distinguishing small ordered domains in a disordered matrix both in partially ordered and in disordered alloys, and in the study of the relative phase of neighboring domains. In this study it has provided direct evidence that the concept that antiphase microdomains gives rise to superlattice spot splitting in the diffraction patterns of Cu_3AuI and that small regions, with local order substantially

higher than the overall matrix, exist in the short-range ordered state. The technique should find increasing importance in the investigation of ordering reactions in materials.

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FIGURE CAPTIONS

FIGURE 1. Lattice image of the (100) planes in fully ordered Cu_3Au .

An antiphase boundary (arrowed) perpendicular to the fringes is manifested by a half-spacing discontinuity in the fringes.

FIGURE 2. (a) [001] Selected area electron diffraction pattern of partially ordered Cu_3Au , showing characteristic splitting of the superlattice reflections. Arrows point to the [100] and $[\bar{1}00]$ reflections.

(b) Dark field electron micrograph taken with the superlattice (100) reflection showing the presence of discrete ordered domains (bright) in a disordered matrix.

FIGURE 3. A lattice image of partially ordered Cu_3Au in which the ordered domains (arrowed) are imaged simultaneously with the disordered matrix.

FIGURE 4. Lattice image of partially ordered Cu_3Au with slight objective lens defocus from the best image. A group of five ordered domains in antiphase is indicated at A and a group of three at B.

FIGURE 5. (a) Dark field electron micrograph of Cu_3Au quenched from above the critical temperature, using the diffuse intensity maximum at the (100) superlattice position to form the image. (b) [001] Electron diffraction pattern corresponding to (a) showing weak diffuse scattering at superlattice positions with shape similar to that in the partially ordered alloy.

FIGURE 6. Lattice image of disordered and quenched Cu_3Au (slightly defocussed as in Figure 4) showing the presence of microdomains with the fringe periodicity of the superlattice.

XBB 741-285

Fig. 1

b

a

XBB 744-2356

Fig. 2

XBB 741-286

Fig. 3

XBB 744-2352

Fig. 4

a

b

XBB 744-2355

Fig. 5

XBB 744-2354

Fig. 6

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