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Authors
Sinclair, R.
Schneider, K.
Thomas, G.

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R. Sinclair, K. Schneider, and G. Thomas

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ANALYSIS OF ORDERING IN Cu₃Au UTILIZING LATTICE IMAGING TECHNIQUES

R. Sinclair, K. Schneider* 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

ABSTRACT

The resolution of lattice fringes in ordered and partially ordered Cu₃Au has been calculated using the dynamical theory of electron diffraction, taking into consideration the effects of such variables as foil thickness and orientation, objective lens defocus, degree of order and, for partially ordered alloys, the position of small ordered domains in the foil. The experimental observations were found to be in excellent agreement with these calculations. Lattice defects in ordered Cu₃Au have been studied using this technique. Thus, the domain wall thickness at translational antiphase domain boundaries has been shown to be one or two atomic diameters and the antiphase boundary separating the pair of fundamental lattice dislocations which constitute a superdislocation has been revealed. A clear distinction exists between the lattice image contrast of predominantly ordered and disordered regions and this has been utilized to study partial long-range order and short-range order. Short-range order in this system is characterized by a dispersion of small highly ordered domains, ~15Å in diameter, in a predominantly disordered matrix.

* Brown, Boveri and Cie A.G., D-6800 Mannheim 1, W. Germany
INTRODUCTION

Significant contributions to the understanding of the mechanisms of ordering reactions in alloys have come principally from x-ray diffraction and transmission electron microscopy techniques. At Berkeley we have been concerned with characterisation of short-range order and its relationship to the initial stages of the transformation by utilising electron diffraction and microscopy (for review, see Das and Thomas). X-ray analysis is a powerful investigative method but one which is generally limited to systems with a large difference in scattering factors of the atomic species, for which the diffracted intensity due to the ordered structure (viz. superlattice reflections) is relatively high, and is limited to averaging information from millions of unit cells. In recent x-ray diffraction studies, detailed quantitative diffraction data have been analysed to yield the most probable atomic arrangements in the alloy and, in the case of CoPt, such a treatment has been substantiated by direct observation of the distribution of atoms by field ion microscopy.

Electron microscopy is potentially capable of more direct and wider application. The qualitative crystallographic information, which may be obtained rapidly in the diffraction mode compared with x-ray data, is supplemented with direct observation of the microstructure in the imaging mode at resolutions down to ~10Å (point to point) or 2Å or better (fourier). Dark field micrographs taken with the objective aperture positioned to allow only a superlattice reflection to form the image are particularly important in elucidating
the shape, size and distribution of ordered regions during the disorder to order transformation \(^{(3)}\). Whilst such conventional modes are extremely valuable in studying the overall transformation they are still subject to several disadvantages for the study of the atomic mechanisms in the reaction:

(i) interpretation of diffraction patterns is not unambiguous (e.g. 3, 5)

(ii) dark field microscopy does not yield quantitative information concerning the local atomic environment

(iii) the direct interpretation of small bright areas on a superlattice dark field micrograph as indicative of regions of high degree of order compared to surrounding material is not necessarily valid \(^{(6)}\).

Unique data on short range order and the initial stages of ordering are lacking for many ordering reactions. In the present study, advantage has been taken of the increased resolution (~2Å) of modern electron microscopes to establish how direct lattice imaging may be able to provide the additional information not available from conventional transmission electron microscopy. The interpretation of experimental images has been facilitated by comparison with images simulated from multibeam dynamical electron diffraction calculations and the application of the technique is illustrated for the well-known alloy Cu\(_3\)Au. Specific attempts have also been made to characterise short-range order in this material.
II. COMPUTATION OF LATTICE IMAGES

There are several methods of calculating the change of amplitude and phase of an electron passing through a crystal. The electron wave function $\psi(r)$ at the bottom surface may thus be found and used to predict the appearance of the image obtained by transmission electron microscopy. In oxide systems (7) and in biological materials (8) the "phase grating" approximation is often used, sometimes extended to a multi-slice calculation (9, 10). In simpler systems where fewer diffracted beams have to be included in the calculation the dynamical theory of electron diffraction is preferred with Bloch waves describing the diffracted electron waves (11). This latter approach was used in the present study.

The development of dynamical electron diffraction theory is described in full elsewhere (12-14). However, in order to establish the optimum conditions for observing lattice images and to account fully for their appearance, the effect of several experimental variables on $\psi(r)$ must be considered. In the electron microscope phase shifts are introduced into different diffracted beams by aberrations of the objective lens and by defects of focus (15). These can be included into the standard theory by considering the situation at the back focal plane of the objective lens (i.e. the diffraction pattern). For simplification, the magnification is assumed to be unity and there is assumed to be only one image and object plane with no extension in the direction of the electron beam ($z$). In the back focal plane the ideal wave function $\psi_{id}$ is given by the Fourier transform of the wave function $\psi_{id}$ leaving the crystal.
\[ \Psi_{id}(\mathbf{g}') = \frac{1}{2\pi} \int \Psi_{id}(r, \mathbf{g}') \exp \left( 2\pi i \mathbf{g}' \cdot \mathbf{r} \right) d^3 r \]  \hfill (1)

where \( \mathbf{g}' \) is a continuous variable in reciprocal space (\( \mathbf{g} \), the standard reciprocal lattice vector, may assume only discrete values). Lens aberrations are included by assuming they impose a phase shift between different directions in reciprocal space, i.e. \( \Psi_{id}(\mathbf{g}') \) is modified by a phase factor \( \exp \left( i \chi(\mathbf{g}') \right) \) giving the real wavefunction

\[ \Psi_{re}(\mathbf{g}') = \Psi_{id}(\mathbf{g}') \exp \left( i \chi(\mathbf{g}') \right) \]  \hfill (2)

with \( \chi \) given by (16) the expression

\[ \chi(\mathbf{g}') = -\frac{\pi}{2} C_s \lambda^3 g' - 2\pi C_c \frac{\Delta K}{K} \lambda g'^2 + \pi R \lambda g'^2 \]  \hfill (3)

The first part of this expression is the phase delay at off-axis beams (\( \mathbf{g}' \neq 0 \)) caused by spherical aberration, the second part is that due to chromatic aberration (with a spread \( \Delta K \) of the wavevector \( K \)) and the third term is the influence of defect of focus, \( R \). (Under-focus, weakening of the objective lens, corresponds to a positive value for \( R \).) Lens astigmatism may be neglected since it may be corrected by the microscope operator and in practice the chromatic aberration is small compared to the other two terms.

The real wavefunction \( \Psi_{re} \) in the image plane may now be obtained by Fourier transforming again:

\[ \Psi_{re}(r) = \int \Psi_{re}(\mathbf{g}') \exp(-2\pi i \mathbf{g}' \cdot \mathbf{r}) d^3 \mathbf{g}' \]  \hfill (4)

and the intensity distribution of the resulting wave interference pattern is given by:

\[ I = \left| \psi_{re}^* \psi_{re} \right| \]  \hfill (5)
Disordered f.c.c. alloys near the composition Cu₃Au form the Ll₂ structure (Fig. 1) on ordering below the critical temperature. The superlattice reflections are the non-f.c.c. reflections of the diffraction pattern and the periodicity of superlattice planes is twice that of the fundamental f.c.c. lattice. {100} planes have the largest spacing (3.8Å) and were the ones studied in the present investigation.

The imaging modes that were employed are shown in Fig. 2. The illumination was tilted so that the outermost reflections of the group were symmetrical to the optical axis of the microscope and the specimen orientation chosen so that only the desired [100] systematic row was strongly excited. The amplitudes of the chosen electron beams are very dependent on the influence of other reflections cut off by the objective aperture and the calculations have to take this into account. Seven beams along the systematic row were therefore considered (unless otherwise stated) corresponding to a distance of ±3g from the centre of the reciprocal lattice section (in Cu₃Au this distance is 0.75Å⁻¹). It is believed that for 100 kV electrons this approximation is adequate since higher order beams are too remote from the Ewald sphere to have a significant influence on the amplitudes of the (100) and (200) diffracted waves.

Each imaging mode should give information concerning ordering, but the mode shown in Fig. 2(c) (termed 0-g-2g) was considered to be the most important since it simultaneously yields information not only about the superlattice (as in (0-g) and (g-0-g) imaging) but also about the fundamental lattice (0-2g) imaging). The results of the calculations in this condition are presented in more detail.
Rather than presenting examples of each individual profile, a convenient way of condensing information about the details of the fringe pattern is to define the visibility \( V \) of the lattice fringes (Fig. 3)

\[
V = \frac{I_p - I_B}{I_p + I_B}
\]

(6)

When the fringe pattern is simple, with a clear maximum and minimum, \( I_B \) was taken as the minimum intensity and \( I_p \) as the peak. If a subsidiary weaker peak existed between the main superlattice peaks, \( I_B \) was taken as the height of the subsidiary peak for the purpose of defining the visibility of superlattice fringes. The profiles were also smoothed from the raw calculated data by assuming an ultimate spatial resolution of \( 1\lambda \), which appeared to be a realistic estimate of the experimental resolution limit judging from lattice images obtained of the (200) planes of pure gold (2.04Å spacing).

The fringe patterns were found, as in previous studies, to depend on several factors e.g. the magnitude of defocus, \( R^{(9)} \); the thickness of the foil, \( z^{(17)} \); and the deviation from the Bragg condition, \( s \), \( (7) \). An example of individual profiles illustrating the effect of defocus on the fringe pattern is shown in Fig. 4 for a fully ordered Cu₃Au foil of thickness 250Å. It can be seen that the profile depends strongly on \( R \).

The corresponding variation of visibility on \( R \) is shown in Fig. 5. The minima have a separation of \( d^{2}_{100}/\lambda \), which follows from equation 3, and the position of the first maximum is given by the magnitude of the coefficient of spherical aberration, \( C_s \). For the Philips EM 301, the maximum superlattice fringe visibility is predicted to occur at 300Å underfocus.
The effect of specimen thickness on fringe visibility was also found to be periodic (Fig. 6). In the (0-g-2g) case a distinction has to be made between visibility of (100) fringes (heavy line) and the appearance of any fringes at all. For instance at a thickness of 350Å the visibility of (100) fringes is poor but the contrast of fundamental lattice (200) fringes (dashed line) is quite strong. A minimum of $V_{100}$ such as this arises when the amplitude of the (100) beam is very small. A change of defocus $R$ yields a change in the relative phases of the (100) and other beams but does not alter their amplitudes. Thus, it is not possible to compensate for a non-ideal specimen thickness by focus changes using the objective lens. The optimum specimen thickness for observing superlattice fringes can be seen to be ~250Å for this imaging mode.

The importance of nearly perfect diffraction conditions for intense superlattice fringes must also be stressed. The fringe visibility rapidly decreases with deviation from the exact Bragg condition and for larger tilts the character of the fringes may also alter, from superlattice to fundamental (e.g. Fig. 7).

In a completely disordered alloy, the superlattice beam intensity is zero and the fringe spacing is that of the fundamental lattice (1.9Å) independent of the experimental variables considered above.

Having established that, under appropriate experimental conditions, a distinction may be made between the lattice image of ordered and disordered Cu$_3$Au it is important to examine how the nature of the image changes in partially ordered samples. For a homogeneous specimen in which the degree of order is identical throughout, the variation
of visibility with degree of long range order \( (S) \) may be calculated by multiplying by \( S \) those coefficients of the lattice potential that have values of \( g \) corresponding to superlattice reflections. Fig. 8 shows the result of these calculations for foil thicknesses of 250\( \AA \) and 300\( \AA \). The former is the specimen thickness for optimum superlattice fringe visibility and it can be seen that a marked increase of \( V_{100} \) is predicted from complete disorder (in which fundamental lattice fringes of equal intensity exist) up to a value of \( S = 0.5 \). Fringe visibility hardly increases thereafter up to the fully ordered condition of \( S = 1 \). For constant experimental conditions it would in principle be possible to obtain a value of the local degree of long-range order by measuring fringe visibility, although in practice such constant conditions would be extremely difficult to obtain.

Many materials, including Cu\(_3\)Au, do not order homogeneously but rather by a mechanism of growth of highly ordered domains consuming predominantly disordered material (e.g. Ref. 2). In this situation it is necessary to establish the contrast of such a highly ordered domain in a disordered matrix. Our calculations show that the appearance of the lattice fringes depended critically on the position of the domain within the foil. Thus, Fig. 9 shows the predicted fringe profile for a disordered 300\( \AA \) Cu\(_3\)Au foil in which a fully ordered 50\( \AA \) domain (\( S = 1 \)) exists. If the domain existed at the top or bottom of the foil, the profile (Fig. 9a) would be hardly distinguishable from that of the disordered fcc matrix which has equal intensity (200) fringes. However, when the domain exists near the centre of the foil, the intensity of alternate fringes is markedly
different (Fig. 9b). The relative difference of intensity between the fringes decreases with the degree of order of the domain and with distance of the domain from the foil centre. If the position of the domain in the foil were precisely known then the profile may be used to estimate the degree of order in the ordered region.

Absorption has not been taken into account in this treatment, but since the specimen thicknesses used in the present study were very small, absorption will not have a significant effect. For example, an estimate of its effect found by adding an imaginary part of $iU_0/10$ to the lattice potential (13) showed that whereas the absolute value of intensity drops to one half due to an average absorption term $iU_0/10$ the change in intensities of the individual beams is only about 10%.

III. EXPERIMENTAL PROCEDURE

Lattice fringe images were taken in a Philips EM 301 microscope fitted with a high resolution stage. A magnification at the final screen of 450,000x was generally employed with the second condenser lens slightly defocussed by an amount to double the exposure time from the fully focussed condition (a fully focussed condenser did not generally yield lattice images). Exposure times were typically 16 or 32 seconds and a through-focus series was necessary to capture the details of 2Å fringe spacings.

Specimens were prepared in (001) orientation to the desired thickness by thin film evaporation techniques. The experimental conditions were similar to those described by Das et al. (18), with direct evaporation of appropriate amounts of Cu and Au on to a heated
rocksalt substrate. This technique is known not to maintain the original stoichiometry, but only small deviations were encountered when the composition was checked by lattice parameter measurements. It was not thought this would affect interpretation of the structural data obtained in this study. At the optimum thickness for lattice imaging of these specimens (250-300Å), the thin foils were found to be in the "channel and hole" stage\(^{(19)}\). Although this made the foils rather fragile, the holes were found to be extremely useful in aiding correction of astigmatism. Heat treatments were generally performed prior to removal of the foil from the rocksalt.

The objective lens defocus for optimum visibility of the fringes was found experimentally by observing the coincidence at the edge of the specimen of the transmitted beam image (usually dark) and the diffracted beam image (usually light). For the tilted (0-g-2g) case, this occurs at approximately 4000 Å overfocus from the disappearance of the Fresnel fringe at the edge of the specimen (the usual criterion for judging best focus). The absolute value of defocus cannot be determined in this tilted condition. Although weak fringes were obtained periodically throughout a through-focus series, the optimum condition was always very well defined, with sharp, high visibility fringes. The range of objective lens settings over which these infocus images were obtained was about ±70Å. Thus the subsidiary visibility maxima have lower visibilities than those predicted in Fig. 4. This disadvantage is considered in more detail later.
IV. RESULTS

A. Fully Ordered Cu₃Au

1. Fringe Profile

(100) fringe images were obtained in ordered specimens with thickness in the range 250-300Å by the three imaging modes shown in Fig. 2. Since alternate (200) planes of the L₁₂ structure contain either all copper atoms or half copper-half gold atoms the image may be likened to the imaging of one set of alternate (200) planes. This is an important result when the (000)+(100)+(200) beams are used to form the image and confirms the prediction of our calculations. The relative intensity of individual fringes was not as even as expected, and no small half-spacing peaks were indicated either visually or by microdensitometer traces perpendicular to the fringes (Fig. 10).

In thicker specimens, the visibility of (100) fringes was poor, but fundamental lattice fringes were not observed. Furthermore, the result of defocus from the optimum condition was to cause blurring and loss of the fringe pattern rather than introduction of subsidiary lattice fringes as predicted by Fig. 4(20).

2. Observation of Lattice Defects

Translational antiphase domain boundaries in Cu₃Au lie predominantly on {100} planes and have a misfit vector \( \frac{a}{2} \langle 011 \rangle \). This brings the all copper atomic plane of one domain in contact with the mixed copper-gold plane in the adjacent domain. In the (100) lattice image a shift of half a fringe spacing is produced on (100) fringes perpendicular to the boundary. This phenomenon is best illustrated by viewing the print at an oblique angle (~15°), as is shown in Fig. 11a. The fringe discontinuity, as expected, runs
approximately parallel to the (010) plane, although atomic roughness ~5-10Å is evident (Fig. 11b). At certain places on the boundary, fringe contrast is lost completely. This is likely to be caused by abuttal of one domain into the other at some depth in the specimen, so that the electron beam travels through both domains in this region. Since the domains are in antiphase the lattice fringe images then mutually cancel. It can also be appreciated that the lattice image demonstrates the width of the boundary to be of the order of one or two atomic diameters although conventional bright field images have a width as great as 50Å (22). However, it is not possible to assess whether the degree of order falls off as the boundary is approached as the superlattice fringe visibility is rather insensitive in the range $S = 0.5 - 1.0$, although the absence of fundamental lattice fringes indicates that $S$ does not decrease below 0.5.

Fig. 12 illustrates an example of the effect of a superdislocation on the lattice image of ordered Cu$_3$Au. In the regions indicated as C, normal fringe continuity is present. At B the fringes become discontinuous, with the amount of shift increasing towards the centre of the disturbance. At A the discontinuity is equal to half a fringe spacing, and thus APB contrast is demonstrated. A Burger's circuit around the disturbance indicates an extra (100) fringe in the bottom half of the crystal with respect to the top. This contrast is consistent with a pair of $\frac{a}{2}[110]$ fundamental lattice dislocations separated by an antiphase boundary, which are well-known as constituting a superlattice dislocation in Cu$_3$Au (23). An approximate value for the partial separation may be derived by counting lattice fringes between the centre of the fringe disturbances at B. The value obtained, 130Å, is in very good agreement with the original determination of
partial separation for fully ordered Cu₃Au by conventional transmission electron microscopy (23). It is also interesting to note that the APB again lies on the lowest energy (100) plane. During slip the partial dislocations travel in pairs on the same (111) slip plane to destroy the APB created by the leading dislocation. However, in this case the partial dislocations must be present on different (111) slip planes in order to lower the total energy of the superlattice dislocation, as previously found for Cu₃Au (24) and isostructural Ni₃Al (25).

It must be emphasized that lattice fringe distortions near a dislocation, the line of which lies at an angle to the imaging plane, are known to be rather complex (11) and the interpretation of the fringe pattern in terms of a one to one correspondence of fringes and lattice planes in the specimen is generally not possible in such structures. This is the case for the regions indicated at B where the fringe pattern is changing. However, the rather narrow width of these regions, the relatively larger area over which APB contrast is observed and the agreement between conventional and lattice image values for the partial dislocation separation all indicate that in this special circumstance the above interpretation may be valid. If this is the case, the total information concerning the dislocation obtained from the lattice image is superior to conventional bright field microscopy and to the higher resolution dark field weak beam and high order bright field techniques (26, 27).

B. Partially Ordered Cu₃Au

The microstructure of partially ordered Cu₃Au, prepared by slowly cooling the evaporated foils through T_c, was determined by conventional
superlattice dark field microscopy to be one of small ordered domains (-25Å in diameter) in a predominantly disordered matrix \(^{(28)}\). Fig. 13a shows the lattice image of this material, using the \((0-g-2g)\) imaging mode, in which it is evident that two types of region may be distinguished:

(i) the continuous matrix possesses uniform intensity fringes with the spacing of the fundamental lattice (1.9Å).

(ii) isolated regions in which alternate white fringes are wider and narrower. These possess the periodicity of the superlattice and thus are interpreted as ordered domains existing near the centre of the foil.

On the original plate the fringe pattern in these domains was composed of alternately stronger and weaker lines and this is illustrated more clearly in a positive print of the plate \(^{(29)}\). A microdensitometer trace across the domains also clearly reveals the alternately stronger and weaker fringes (Fig. 13b).

This result is in excellent agreement with the computed profile of an ordered domain in a disordered matrix (Fig. 9). Although precise information about the position of the domain within the foil is necessary before a local degree of order can be found for the domain, it is evident from the fringes that the matrix is predominantly disordered, with \(S < 0.4\), and that the local order in the small domain is significantly higher.

The effect of small changes in objective lens current (i.e. focus) has a marked effect on the image. With a slightly underfocussed setting, even intensity fringes of the fundamental lattice are
obtained (29) with no indication of the presence of the ordered domains. At slight overfocus from the above "simultaneous image" the (200) fringes of the disordered matrix become blurred and are no longer resolved, whilst in the ordered domains only (100) fringes are seen (29). The domains now stand out more clearly against a blurred background. This contrast may be utilized to indicate the presence of regions in the material with a significantly higher degree of order than the surrounding material. In this way it has been shown that small "microdomains" of high local order (~15 Å) are present in quenched Cu₃Au, accounting for the weak diffuse scattering at superlattice positions in the diffraction patterns (28).

It may also be noted that the relative phase of adjacent domains remains unaltered by changes of focus. Thus the distribution of domains in antiphase with their neighbours may be investigated by this technique. It has been shown that a distribution of antiphase domain groups can account for the observed superlattice spot splitting found in diffraction patterns of Cu₃Au in the early stages of ordering (28).

The lattice imaging technique also demonstrated that even in the early stages of ordering some of the small ordered domains already possess an anti-phase boundary, as illustrated in Fig. 13a. Although it is possible that such contrast arises from a fortuitous overlapping of domains, the good fit and similar fringe visibility found in such cases between the two portions of the domain indicate that they do in fact belong together. Since it is likely that these are produced by coalescence of adjacent domains which were formed initially in antiphase, this result again
emphasises the role of cooperative antiphase domain phenomena in the early stages of ordering in this alloy system.

A comparison of the conventional superlattice dark field micrograph and the corresponding lattice image is shown in Fig. 14. The number of ordered domains indicated in the lattice image is considerably less than the number of bright areas on the dark field micrograph, as expected by the calculations shown in Fig. 9. A direct correlation of the two images in terms of shape and positioning of the domains is not always found. For instance at positions marked A in Fig. 14, the domains revealed by the lattice image correlate well with bright regions in the superlattice dark field image. At B, the domains in the lattice image show weak contrast compared with the dark field, whilst at C the lattice image contrast is predominantly that of fundamental fringes although bright regions are quite clear on the dark field. Since domains located at various depths in the foil and having varying degrees of order all contribute to the dark field image it is consistent with our calculations that some ordered domains indicated in the dark field are missing from the lattice image. Furthermore, spurious bright spot images as described by Cowley(6) will also be part of the contribution to the dark field image and will not be revealed as ordered domains by lattice imaging.

V. DISCUSSION

The experimental lattice images of ordered and disordered Cu₃Au have been found to correlate well with the profiles calculated from dynamical electron diffraction theory. Thus, in the ordered alloy, superlattice fringes are obtained under appropriate conditions using
(0-g-2g) imaging beams, with a variation of fringe visibility with degree of order, and in partially ordered alloys the profile of predominantly ordered or disordered material can be distinguished. These have been utilized in studying lattice defects and ordering in Cu₃Au and the interpretation of experimental results has been fully justified by comparison with the theoretical calculations.

Apart from the assumptions involved in the dynamical theory, two further factors have not been taken into account in the present study. Firstly, it has been assumed throughout that only one amplitude \( \psi(g) \) exists at the reciprocal coordinate \( g' = g \). However, for the diffuse intensity which exists at superlattice positions in disordered and partially ordered specimens, a function \( \psi(g') \) would be more appropriate, altering the influence of the superlattice beams on the resulting image.

Secondly, the incident electron beam has been assumed to be a plane wave. In practice, a nearly fully focussed second condenser lens was used to minimize the exposure time. The resultant beam divergence introduces a spatial incoherence into the beam which can be accommodated by using wave vectors \( K \) with different directions, but the same magnitude (i.e. a spherical wave). Thus, the microscope aberrations considered in the theory are not sufficient and this could explain some experimental observations on the optimum conditions for fringe visibility:

(i) fringe images were not successfully obtained at maximum beam divergence (i.e. fully focussed second condenser lens);

(ii) the fringe visibility at subsidiary defocus maxima (Fig. 4) was considerably lower than at the optimum defocus condition.
However, these disadvantages affect only minor points concerning comparison of experiments and theory and do not invalidate the major results that have been found.

The experimental lattice images of ordered Cu₃Au have illustrated that the fully ordered lattice is continuous to within one or two atomic diameters of the boundary. However, the boundary itself is not perfectly smooth but small protrusions from a (100) plane are present. The separation of paired dislocations has also been found to be in excellent agreement with that previously determined and the lattice image has further demonstrated that the partials are separated by an antiphase boundary on a (100) plane.

In partially ordered alloys, the characteristics of the lattice image depend on the objective lens setting, the degree of order and, for an ordered domain existing in a predominantly disordered matrix, the position of the domain in the foil. Since the optimum conditions may be obtained from a through-focal series and the position by stereo analysis of the superlattice dark field, the local degree of order in a small region may be obtained. Thus the development of order whether by growth of imperfectly ordered or highly ordered domains may be studied directly by this technique. It is capable, therefore, of providing valuable information on an extremely fine scale in a relatively straightforward manner and in a form readily accessible to interpretation.

Results presented elsewhere (28) have demonstrated how the technique has been utilized to yield information on the role of antiphase domain groups in the early stages of ordering of Cu₃Au, with the first direct
evidence that the splitting of superlattice reflections in partially ordered Cu₃Au arises from domains existing in antiphase with one another. The surprising result that APBs exist in the very early stages of ordering has further been confirmed in the present paper and it is possible that they occur in order to accommodate extra wrong-site atoms in non-stoichiometric alloys as indicated by the x-ray diffraction work of Schwartz and Cohen (30). Experiments on bulk alloy specimens of known composition are in progress to clarify this suggestion.

Finally, it has also been demonstrated elsewhere (28) that the bright regions which appear in superlattice dark field micrographs of disordered Cu₃Au correlate well in size and distribution with ordered domains revealed by the lattice image technique. Since the interpretation of such high-resolution dark field micrographs has recently been shown to be more complex than originally thought (6), it is apparent that high resolution lattice imaging is necessary for a complete structural characterisation of short-range order and partial long-range order in alloys. Furthermore, these new results are in excellent agreement with the computer simulated model of short range order in Cu₃Au derived from diffuse x-ray scattering data (31).

VI. CONCLUSIONS

1. Experimental and computed lattice images of ordered and partially ordered Cu₃Au have been shown to be in excellent agreement.
2. Information concerning the mechanism of ordering and details of lattice defects in ordered alloys may be obtained by the lattice imaging technique.
3. Antiphase boundaries in Cu₃Au on (100) planes contain small 5–10Å protrusions.
4. The equilibrium configuration of superdislocations in ordered Cu₃Au is indicated to be two \( \frac{a}{2} [110] \) fundamental lattice dislocations separated by a \{100\} antiphase boundary. Information concerning the dislocations may, in certain circumstances, be superior to conventional imaging techniques.

5. Antiphase boundaries exist in the very early stages of ordering in Cu₃Au.

6. Local heterogeneities in degree of order may be studied by lattice imaging. Thus, the presence of highly ordered domains in the predominantly disordered matrix during ordering of Cu₃Au has been confirmed by the technique and the existence of small microdomains has been indicated in the short range ordered state quenched from above the critical temperature.

**VII. ACKNOWLEDGMENTS**

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Fig. 1. The Ll₂ structure of ordered Cu₃Au.

Fig. 2. The imaging modes employed to study superlattice fringes in Cu₃Au. The deviation from the exact Bragg condition, s, is indicated for the imaging beams.

Fig. 3. Definition of fringe visibility, V.

Fig. 4. The fringe profile of a fully ordered Cu₃Au foil of thickness 250Å using the (0-g-2g) imaging mode: (a) with objective lens overfocus 300Å; (b) with objective lens overfocus 500Å. The nature of the fringe pattern changes from superlattice in (a) to fundamental in (b).

Fig. 5. The dependence of the visibility of (100) superlattice fringes with defocus for an ordered Cu₃Au foil of thickness 250Å.

Fig. 6. The effect of specimen thickness on (100) fringe visibility of ordered Cu₃Au, at the optimum defocus condition for the three imaging modes shown in Figure 2. The dashed line illustrates the overall fringe visibility for the (0-g-2g) imaging mode.

Fig. 7. The effect of large deviation from the exact Bragg condition (1° tilt) on the optimum superlattice fringes (shown in (a)) obtained in the (0-g-2g) imaging mode. The new orientation (b) is near the exact Bragg condition for the (600) reflection.

Fig. 8. The variation of superlattice fringe visibility, (0-g-2g) imaging, with Bragg-Williams long-range order parameter (S) at specimen thicknesses 250Å and 300 Å.

Fig. 9. The fringe profile of a 50Å ordered domain (S=1) in a disordered matrix (S=0), (0-g-2g) imaging at optimum focussing conditions and foil thickness 300Å: (a) when the domain is at the top or bottom of the foil; (b) when the domain is at the centre of the foil.
Fig. 10. Microdensitometer trace (i.e. fringe profile) of the (100) superlattice fringes of ordered Cu₃Au obtained by the (0-g-2g) imaging mode.

Fig. 11. The lattice image of a translational antiphase domain boundary (APB) in ordered Cu₃Au: (a) viewed at an oblique angle; (b) detailed, normal view near the APB showing small protrusions from a (100) plane.

Fig. 12. The lattice image of a superdislocation in ordered Cu₃Au. The antiphase boundary (at C) separating the fundamental lattice dislocations (at B) is clearly revealed.

Fig. 13. (a) Lattice image of partially ordered Cu₃Au showing the alternately stronger and weaker fringes in a highly ordered domain in a disordered matrix. An antiphase boundary is indicated at the centre of the ordered domain. (b) Fringe profile across such an ordered domain in partially ordered Cu₃Au. The alternately stronger and weaker fringes in the domain are quite clear.

Fig. 14. A comparison of the lattice image (a) and superlattice dark field image (b) of partially ordered Cu₃Au. Bright regions in the dark field micrograph correlate well with ordered domains indicated in the lattice image at A, correlate partially at B and show poor correlation at C. The corresponding diffraction pattern is shown in (c).
Fig. 3
\[ R = 300 \, \text{Å} \]
\[ z = 250 \, \text{Å} \]
\[ V_{100} = 0.84 \]
Fig. 4b
Fig. 6
Fig. 7
Fig. 8
Fig. 10

INTENSITY
(arbitrary units)

DISTANCE (Å)

3.75 Å

XBL7411-7552
Fig. 11a
Fig. 13 a
Fig. 13b
Fig. 14
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