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The Structure of Thin Films of CuAu at Room and Elevated Temperatures

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An experimental study of thin single crystal films of CuAu has resulted in electron diffraction effects which can be interpreted as follows:

(*a*) The low temperature CuAu I ordered structure is made up of domains wherein the *c*-axes of the tetragonal unit cells are parallel with each other but perpendicular to the *c*-axes in adjacent domains.

(*b*) The AuCu II ordered structure has an orthorhombic unit cell as proposed by Johannssen & Linde (1936).

(*c*) The disordered lattice has a face-centered-cubic structure. Since the diffraction effects are analogous in every respect to those found by X-rays, it is concluded that CuAu, when in the form of a single crystal film about 300 Å thick, behaves crystallographically as it does when in massive form.

Introduction

The purpose of this study was to determine the crystalline characteristics of a CuAu alloy in the form of a very thin film and compare them with known crystalline characteristics of the alloy when in massive form. The present report describes electron diffraction effects from thin single crystal films of CuAu and their interpretation. The composition CuAu was of interest because an orthorhombic lattice, corresponding to the non-equilibrium configuration observed in Cu_3Au is known to be stable in a narrow temperature range just below the ordus. The results of this study are the same as those obtained by Ogawa and Watanabe (1954), however, the method of obtaining them is somewhat different.

Procedure

A master alloy of CuAu was prepared by melting OFHC copper and 'fine' gold in an evacuated quartz tube. The composition of the homogenized (1000 °C. in vacuum for 14 hours) ingot was 50 atom percent Au as determined by the lattice parameter (a_0 = 3·874 Å) of the disordered phase.

A single crystal film of CuAu was made by evaporating a measured amount of the master alloy from a hot tungsten basket onto a freshly cleaved (100) face of rock salt held at 410 °C. After the evaporation, the film and substrate were slowly cooled to room temperature in order to allow the alloy to come to a high degree of order. Care was taken to evaporate all of the basket charge. The composition of the deposited film was then assumed to be the same as that of the master alloy. The thickness of the film was adjusted to approximately 300 Å by controlling the charge weight and the geometry of the evaporation system. There was no experimental check of film thickness. The temperature of the substrate and film was measured by means of a 0·005 in. chromel-alumel thermocouple placed in a small hole in the rock salt.

When the specimen was cool, the substrate and film were separated by dissolving the salt in water. The freed film was then washed several times in distilled water, and finally picked up on a fine-mesh nickel screen for subsequent study in the electron diffraction camera. Transmission photographs were made at room temperature and elevated temperatures, using a special furnace which is described in detail elsewhere (Alessandrini, 1949). An accelerating potential of 50 kV. was used for all photographs.

Results

The diffraction pattern given by the slowly cooled film is shown in Fig. 1. The same film held at approximately 400 °C. during the exposure gave the pattern shown in Fig. 2. After the film had been held 10 minutes at 400 °C. and quenched (400 °C. to 100 °C. in about 10 seconds), it gave the pattern in Fig. 3. The pattern given by the disordered structure was obtained when the film was held at 450 °C. during the exposure. This is shown in Fig. 4. Thus, Figs. 1–4 demonstrate the transition of the crystal film from the ordered tetragonal CuAu I structure to the orthorhombic CuAu II to the disordered face-centered cubic forms.

As the ordered film was heated to 400 °C., some of the super-lattice reflections became diffuse. After the film had been held at 400 °C., crosses could be seen at the normal superstructure reflection positions. These crosses finally resolved into four new diffuse spots (Fig. 2). After the film was quenched from 400 °C. (Fig. 3) the new spots were sharp and intense. They disappeared completely when the film was heated above 425 °C.

Fig. 1. Pattern from AuCu film (500 Å in thickness) as evaporated showing tetragonal AuCu I.

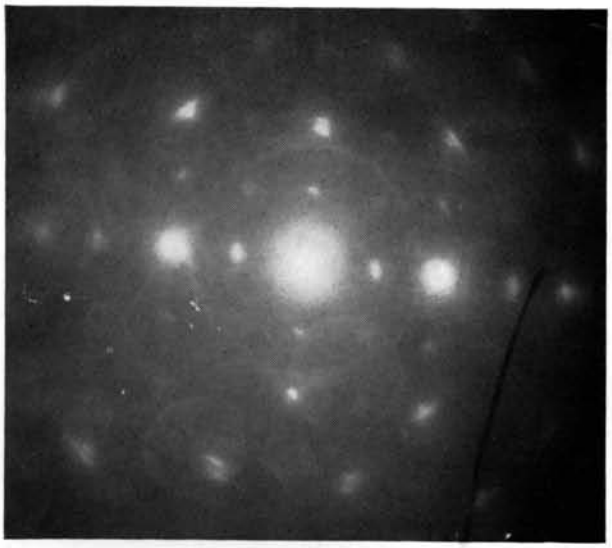

Fig. 2. Pattern from same single crystal film at 400 °C.

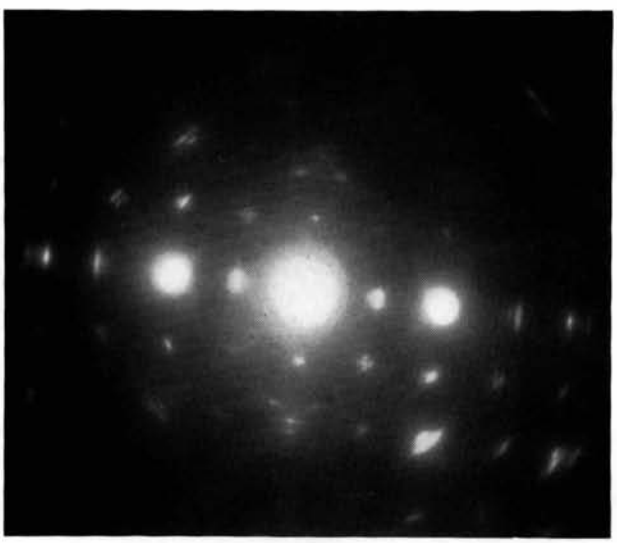

Fig. 3. Pattern from same single crystal film after heating for 10 min. at 400 °C. and quenching. AuCu II temperature range.

Fig. 4. Pattern from same single crystal film at 450 °C. showing intensities in reciprocal space corresponding to f.c.c. lattice.

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Discussion

The patterns in Figs. 1–3 may be considered to be a composite of three superimposed nets each corresponding to the CuAu I tetragonal structure, in the three orientations drawn in Fig. 5. The three nets

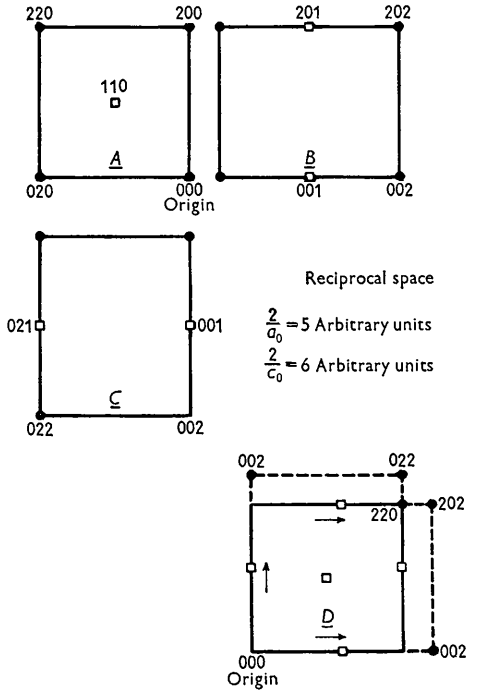

Fig. 5. Four figures in reciprocal space. In A the C axis is $\perp$ to the paper and in B and C the axis is lying in opposite directions in this plane. The diagram D shows how, by superimposing these planes, the three spots can be accounted for in the pattern.

superimposed in this way account for the three spots observed at the {200} positions. The diffuse character of the diffraction spots in Figs. 1 and 2 can be interpreted respectively as a consequence of small domain size (Fig. 1) and imperfect order within the domains (Fig. 2).

The splitting of the superstructure reflections (Figs. 2 and 3) into four symmetrically arranged intensity spots may be interpreted as arising from an elongated unit cell made up of five tetragonal cells in line with five more cells displaced $\frac{1}{2}a_0+\frac{1}{2}c_0$. This constitutes a multiple unit cell made up of ten ordered tetragonal cells bisected by an 'out-of-step' boundry and is identical with the cell proposed by Johannssen & Linde (1936) see Fig. 6. These effects are similar to

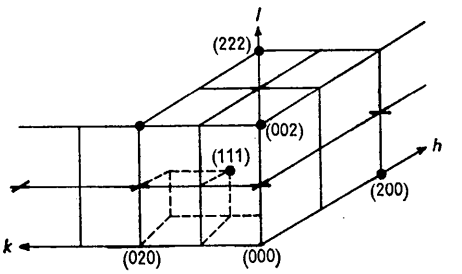

Fig. 6. Reciprocal lattice of AuCu II position of crosses as they appear in reciprocal space.

diffuse X-ray effects given by Cu_3Au. Their interpretation is the same as that given by Guinier & Griffoul (1948).

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X-ray Scattering from CCl_4

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By using a recent development for the calculation of the X-ray scattering from a gas of non-spherical molecules the scattering factor for CCl_4 has been computed. In this way it was possible to establish some comparison with experiment regarding the 'valence' electron distribution in such a molecule when described by a Thomas–Fermi method. Further, a test was provided of the range of applicability of the general treatment used for evaluating the scattering factor.

1. Introduction

In a recent paper by Coulson & March (1956) a modified Thomas–Fermi (T.F.) treatment was developed for tetrahedral and octahedral molecules with heavy atoms in the outer positions. Whilst retaining within their approximation the simplicity of a central-