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ON THE EXPERIMENTAL DETERMINATION OF LOW ORDER STRUCTURE FACTORS IN TiAl BY ENERGY FILTERED CONVERGENT BEAM ELECTRON DIFFRACTION.

S. Swaminathan, I. P. Jones*, N. J. Zaluzec**, D. M. Maher*** and H. L. Fraser.

Dept. of Materials Science and Engineering, The Ohio State University, Columbus, OH 43210

*School of Metallurgy and Materials, University of Birmingham, Birmingham, UK

**Center for Electron Microscopy, Argonne National Laboratory†, Argonne, IL

***Dept. of Materials Science and Engineering, North Carolina State University, Raleigh, NC

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It has been claimed that the effective Peierls stresses and mobilities of certain dislocations in TiAl are influenced by the anisotropy of bonding charge densities [1, 2]. This claim is based on the angular variation of electron charge density calculated by theory [e.g. 3,4]. It is important to verify the results of these calculations experimentally, and the present paper describes a series of such experiments. A description of the bonding charge density distribution in materials can be obtained by utilizing the charge deformation density ($\Delta\rho(\mathbf{r})$) defined by (1)

$$\Delta\rho(\mathbf{r}) = \frac{1}{V} \sum (\mathbf{F}_{\text{obs}} - \mathbf{F}_{\text{calc}}) \exp(-2\pi i \mathbf{H} \cdot \mathbf{r}) \quad (1)$$

where V is the volume of the unit cell, $\mathbf{F}_{\text{obs}}$ is the experimentally determined low order structure factor and $\mathbf{F}_{\text{calc}}$ is the structure factor calculated using the Hartree-Fock neutral atom model [5]. To determine the experimental low order structure factors, a technique involving a combination of convergent beam electron diffraction (CBED) and electron energy loss spectroscopy (EELS) [6] has been used. Apart from the intrinsic accuracy of this technique for low order structure factors compared to measurements made using x-ray diffraction, it is our experience that measurements made using x-rays are also subject to error because of the large linear absorption coefficient ($\approx 64 \text{ cm}^{-1}$) and heavy defect population introduced during specimen preparation. In contrast, the CBED technique permits the diffracted intensities to be obtained on a small spatial scale from a defect free region of a crystal.

The experimental technique involved recording CBED patterns obtained from a number of reflections, with the sample oriented in each case at the Bragg condition. A Philips EM420 transmission/scanning transmission electron microscope (TEM/STEM) interfaced to a combination of commercial and custom built ancillary equipment in the Electron Microscopy Center of Argonne National Laboratory was used. Digital scans were produced using customized software and hardware with the data acquisition computer interface which allowed variable size and orientation of two dimensional rasters to be directed to the post-specimen double deflection coils of the TEM. The electron intensity was directed through the electron spectrometer to a photomultiplier tube (PMT) where upon it was processed using analog and digital signal processing equipment. The energy resolution limit of the spectrometer employed during these measurements was less than 0.75 eV. The angular resolution at a camera length of 600 mm and the 200 μm size entrance aperture was $\approx 0.3 \text{ mRad}$.

A thin foil of Ti52at%Al, prepared by electrochemical techniques, was used in the experiments. An example of a two dimensional energy filtered CBED pattern containing only the (200) dark field disk in TiAl is shown in Fig 1a. A rocking curve shown in Fig 1b, has been obtained by plotting the intensity profile through the center of the disk along line AB. The crystal orientation was chosen such that there is minimal interaction from higher order laue zone (HOLZ) lines on the bright-field and the dark-field disks. In the cases where there was a HOLZ line in the intensity profile AB, that part of the rocking curve affected by the HOLZ line was replaced by data unaffected by HOLZ lines taken from above or below the line. Removing these HOLZ effects permitted simulations to be performed while neglecting non-systematic reflections, which resulted in a considerable reduction of computation time. To establish confidence in this experimental technique, including the adjustment described above, it has been applied to the determination of the Si (220) structure factor at two different temperatures. The deduced structure

factor is in excellent agreement with those measured using other methods. A total of nine low order rocking curves have been determined in TiAl experimentally using the technique described.

These rocking curves have been matched using the three dimensional Bloch wave formulation of the dynamical theory; the thickness t , the extinction distance ξ_g and absorption coefficient of the extinction ξ_g , was fitted for each reflection. The quantity $\chi^2 = (I_{\text{calc}} - I_{\text{expt}})^2$ was minimized by a quasi-Newton method with a finite difference gradient using the subroutine BCONF from the IMSL mathematical library [7]. The fitted values of the extinction distance and absorption coefficient were verified by repeating the experiments and calculations for various crystal thicknesses. The χ^2 minimization was also performed for different normalizing data point so that the extinction distance ξ_g and absorption coefficient of the extinction ξ_g were returned within one percent accuracy. A planar section of the deformation electron charge density distribution taken perpendicular to [001] (through the origin of the TiAl unit cell) is shown in Fig. 2. This deformation map is in good agreement with that produced by theory [e.g. 3,4].

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Fig. 1a - (200) CBED dark field disk.

Fig. 1b - Rocking curve from profile AB with the theoretically calculated fit.

Fig. 2- Electron charge density deformation map for the (001) section in TiAl: Contour level = 0.025 electrons/ $\text{\AA}^3$

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