

## The analysis of macrotwins in NiAl martensite

J.M. Ball and D. Schryvers<sup>1</sup>

*Mathematical Institute, University of Oxford, 24-29 St. Giles', Oxford OX1 3LB, U.K.*  
<sup>1</sup>*Electron Microscopy for Materials Research (EMAT), Department of Physics,  
University of Antwerp, RUCA, Groenenborgerlaan 171, 2020 Antwerp, Belgium*

**Abstract.** We present a theoretical study of macrotwins arising in cubic to tetragonal martensitic transformations. The results help to explain some features of such macrotwins observed in Ni<sub>65</sub>Al<sub>35</sub>.

### 1. INTRODUCTION

In their high resolution electron microscopy studies of bulk and splat-cooled Ni<sub>65</sub>Al<sub>35</sub> polycrystals, Boullay & Schryvers [6] made detailed observations of macrotwin interfaces separating different martensitic plates. This alloy undergoes a cubic (bcc B2 phase) to tetragonal (bct) martensitic transformation. Fig. 1 shows two macrotwin interfaces separating plates involving the same two martensitic variants but with different families of microtwins, the microtwin planes in adjacent plates being nearly orthogonal.

These macrotwin interfaces are of 'crossing type', as opposed to 'step type' interfaces that are also observed (see [14, 7, 15, 6] for a description). The step type interfaces seem to occur more frequently in splat-cooled samples, which have a small grain size of the order of 1 μm. Our discussion applies most directly to crossing type macrotwins in bulk-cooled samples, whose typical grain size is large (of the order of 1mm) compared to that of the macrotwins. In fact we treat the sample as if it were a single crystal, ignoring constraints arising from neighbouring grains. In terms of the calculations made, this is justified provided the microstructures have close to zero energy (the zero of energy being taken to be that of a pure variant of martensite or of undistorted austenite, these energies being equal at the transformation temperature  $\theta_c$ ).

The formation of martensitic microstructure is a dynamic process that should properly be modelled by appropriate dynamical equations. However, it is not clear exactly what equations should be used, and the prediction of martensitic morphology on the basis of any such equations seems to be currently out of reach. Furthermore, since the transformation is so fast, for the samples used there is no direct observational evidence of how the final morphology arises. However, by comparing details of the macrotwins with predictions based on the nonlinear elasticity model of martensitic transformations, we obtain indirect evidence confirming the natural scenario that macrotwins are formed through coalescence of martensitic plates nucleated at different points of the sample, and propose an explanation of why other types of macrotwins were not observed. We restrict attention throughout to macrotwins involving only two tetragonal variants.

### 2. NONLINEAR ELASTICITY MODEL

According to this model (see Ball & James [1], [2] and for corresponding linearized models [8], [9], [10], [12], [13], [5]) the total free-energy of a single crystal at temperature  $\theta$  is given in terms of the free-energy density  $\varphi = \varphi(A, \theta)$  by  $I_\theta(y) = \int_\Omega \varphi(\nabla y(x), \theta) dx$ , where  $y(x) = (y_1(x), y_2(x), y_3(x))$  denotes the deformed position of the material point of the crystal that has position  $x = (x_1, x_2, x_3)$  in the region  $\Omega$  occupied by the crystal in a reference configuration, which we take to be undistorted austenite at the transformation temperature  $\theta_c$ , and where  $\nabla y(x)$  denotes the deformation gradient, namely the  $3 \times 3$  matrix  $(\partial y_i / \partial x_j)$ .

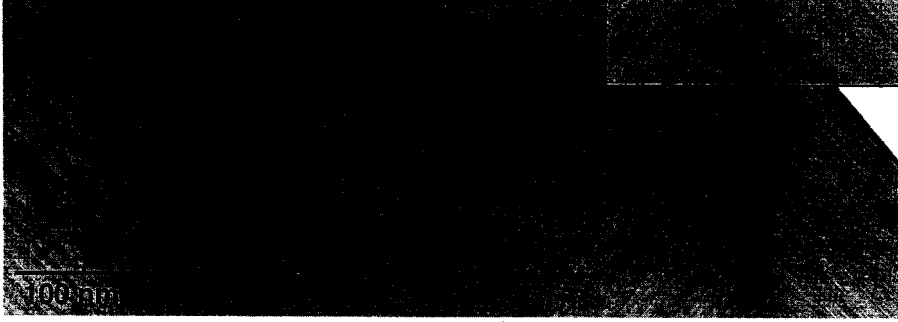


Figure 1: Low magnification image of crossing-type macrotwins. The insert shows details of the crossing at twice the magnification. Bands of different grey levels correspond to different variants  $U_1$  and  $U_2$

We may assume that  $\min_A \varphi(A, \theta_c) = 0$ . For a cubic-to-tetragonal transformation, the set  $K(\theta_c) = \{A : \varphi(A, \theta_c) = 0\}$  of minimizing gradients is assumed to have the form  $K(\theta_c) = SO(3) \cup \bigcup_{i=1}^3 SO(3)U_i$ , where  $U_1 = \text{diag}(\eta_3, \eta_1, \eta_1)$ ,  $U_2 = \text{diag}(\eta_1, \eta_3, \eta_1)$ ,  $U_3 = \text{diag}(\eta_1, \eta_1, \eta_3)$  are the transformation strains of the three martensitic variants, and  $\eta_1 > 0, \eta_3 > 0$  the deformation parameters.

We can identify zero-energy microstructures with sequences of deformations  $y^{(j)}$  with  $I_{\theta_c}(y^{(j)}) \rightarrow 0$  as  $j \rightarrow \infty$ , or with the *Young measures*  $(\nu_x)_{x \in \Omega}$  corresponding to the deformation gradients  $\nabla y^{(j)}$  of such sequences. However, for the purposes of this paper we just need the compatibility conditions that are satisfied by twins and martensitic plates. As is well known, twins correspond to rank-one connections  $A - B = a \otimes n$  between two of the martensitic energy wells  $SO(3)U_i$ ,  $A$  and  $B$  being the deformation gradients on opposite sides of the twin plane, which has normal  $n$ . Taking without loss of generality twins involving the first two variants and  $B = U_1$ , the two possible twins with  $A \in SO(3)U_2$  are given by  $a = \sqrt{2} \frac{\eta_3^2 - \eta_1^2}{\eta_1^2 + \eta_3^2} (-\eta_3, \kappa \eta_1, 0)$ ,  $n = \frac{1}{\sqrt{2}}(1, \kappa, 0)$ , where  $\kappa = \pm 1$ .

A martensitic plate with twins  $A, B$  in the volume fraction  $\lambda$  to  $1 - \lambda$  has corresponding macroscopic deformation gradient  $F = \lambda A + (1 - \lambda)B$ . In order for such a plate to be compatible with undistorted austenite across the habit plane  $\{x \cdot m = k\}$  the equation  $\lambda A + (1 - \lambda)B = \mathbf{1} + b \otimes m$  must hold for some vector  $b$ . The solutions  $A \in SO(3)U_i$ ,  $B \in SO(3)U_j$ ,  $\lambda \in [0, 1]$ ,  $b, m$  are given by the formulae of the crystallographic theory of martensite [16]; for details see [1]. Taking  $i = 1, j = 2$  we have that  $A = QU_1$ ,  $B = Q(U_1 + a \otimes n)$  with  $Q \in SO(3)$ , that  $\lambda = \lambda^*$  or  $1 - \lambda^*$ , where  $\lambda^* = \frac{1}{2} \left( 1 - \sqrt{\frac{2(\eta_3^2 - 1)(\eta_1^2 - 1)(\eta_1^2 + \eta_3^2)}{(\eta_3^2 - \eta_1^2)^2} + 1} \right)$ , and that

$$m = \left( \frac{1}{2} \chi (\delta + \nu \tau), \frac{1}{2} \chi \kappa (\nu \tau - \delta), 1 \right), \quad b = \left( \frac{1}{2} \chi \zeta (\delta + \nu \tau), \frac{1}{2} \chi \zeta \kappa (\nu \tau - \delta), \beta \right), \quad (1)$$

where  $\nu = 1$  for  $\lambda = \lambda^*$ ,  $\nu = -1$  for  $\lambda = 1 - \lambda^*$ . Here  $\delta = [(\eta_3^2 + \eta_1^2 - 2)(1 - \eta_1^2)^{-1}]^{\frac{1}{2}}$ ,  $\tau = [(2\eta_1^2\eta_3^2 - \eta_1^2 - \eta_3^2)(1 - \eta_1^2)^{-1}]^{\frac{1}{2}}$ ,  $\zeta = (1 - \eta_1^2)/(1 + \eta_3)$ ,  $\beta = \eta_3(\eta_1^2 - 1)/(1 + \eta_3)$ ,  $\chi = \pm 1$ . These solutions exist provided the inequalities  $\eta_1^2 + \eta_3^2 < 2$  if  $\eta_1 > 1$ ,  $\eta_1^{-2} + \eta_3^{-2} < 2$  if  $\eta_1 < 1$  hold. In the calculations below we assume the experimentally measured values  $\eta_1 = .93, \eta_3 = 1.15$ .

### 3. INCOMPATIBILITY OF MARTENSITIC PLATES

Consider two martensitic plates I and II comprising only the variants  $U_1$  and  $U_2$  and having distinct macroscopic gradients  $\mathbf{1} + b_1 \otimes m_1$  and  $\mathbf{1} + b_2 \otimes m_2$  with corresponding choices of parameters  $\kappa_1, \chi_1, \nu_1$  and  $\kappa_2, \chi_2, \nu_2$  respectively. By relabelling axes we can suppose that  $\kappa_1 = \chi_1 = \nu_1 = 1$  so that in particular Plate I corresponds to twinning on  $[110]_{B2}$  planes with volume fraction  $\lambda^*$ . We examine whether there is a rotation  $Q$  that brings Plate II into compatibility with Plate I across an interface with some normal  $N$ . Thus we have to solve the equation

$$\mathbf{1} + b_1 \otimes m_1 = Q(\mathbf{1} + b_2 \otimes m_2) + c \otimes N \quad (2)$$

Table 1: Rotations  $Q_1$  and  $Q_2$  that bring Plate II into compatibility with Plate I ( $\kappa_1 = \chi_1 = \nu_1 = 1$ ) and the corresponding macrotwin normals  $N_1$  and  $N_2$ . The direction of rotation is that of a right-handed screw in the direction of the given axis. For the case  $\kappa_2 = \nu_2 = 1, \chi_2 = -1$  see the text.

Parameter Values			$Q_1$			$Q_2$		
$\kappa_2$	$\chi_2$	$\nu_2$	Axis	Angle	$N_1$	Axis	Angle	$N_2$
-1	1	1	(.70,0,-.71)	1.64°	(0,1,0)	(.75,0,.66)	1.75°	(1,0,0)
-1	-1	1	(0,.99,.16)	7.99°	(1,0,0)	(0,.99,-.14)	7.99°	(0,1,0)
-1	1	-1	(.65,.48,-.59)	6.76°	(.59,-.81,0)	(.68,.50,.54)	6.91°	(-.81,-.59,0)
-1	-1	-1	(-.48,.65,.59)	6.76°	(-.81,-.59,0)	(-.50,.68,-.54)	6.91°	(.59,-.81,0)
1	1	-1	(-.54,.54,.64)	5.87°	$\frac{1}{\sqrt{2}}(1,1,0)$	(-.57,.57,-.59)	6.08°	$\frac{1}{\sqrt{2}}(1,-1,0)$
1	-1	-1	(.60,.60,-.52)	7.37°	$\frac{1}{\sqrt{2}}(1,-1,0)$	(.62,.62,.47)	7.47°	$\frac{1}{\sqrt{2}}(1,1,0)$

for  $Q \in SO(3)$  and  $c, N \in \mathbf{R}^3$  with  $|N| = 1$ : Note that (2) is equivalent to the equation

$$F = Q(\mathbf{1} + \bar{c} \otimes \bar{N}), \quad (3)$$

where  $F = (\mathbf{1} + b_1 \otimes m_1)(\mathbf{1} + b_2 \otimes m_2)^{-1}$ ,  $\bar{c} = Q^T c$  and  $\bar{N} = (\mathbf{1} + b_2 \otimes m_2)^{-T} N$ . The calculation of Bhattacharya [4] shows that it is not possible to solve (2) with  $Q = \mathbf{1}$ , that is *distinct undistorted plates are always incompatible*. This strongly suggests that the macrotwins are not zero-energy microstructures, but undergo distortions near the macrotwin interface to achieve compatibility there, with consequential nonzero stresses. In seeking solutions to (2) we take the view that the plates rotate as the macrotwin interface is approached and that in the vicinity of the interface reach the rotation necessary to make them compatible at near to zero energy. Note, however, that the distortion of the plates cannot be a pure rotation without stretching, on account of the theorem (see Reshetnyak [11]) that a deformation whose gradient is a rotation everywhere is a *constant* rotation.

In the case  $\chi_2 = -1, \kappa_2 = \nu_2 = 1$ , we have that  $F$ , and hence  $FQ^{-1}$ , is a rotation, from which it follows that the only solution to (2) is  $c = 0, Q = F$ ,  $Q$  being a rotation about the axis (.15, .99, 0) of 8.00°. In each of the other cases it can be proved that there are exactly two rotations  $Q_1$  and  $Q_2$  which solve (2) for corresponding pairs of vectors  $c_1, N_1$  and  $c_2, N_2$ . The axes and angles of rotation and the corresponding normals are given in Table 1, the normals in the first two and last two lines being exact results.

#### 4. COMPARISON WITH EXPERIMENTS

Boullay & Schryvers only observed the case  $\kappa_2 = -1, \nu_2 = 1$  corresponding to opposite families of microtwins having equal volume fractions  $\lambda^*$ . Planar macrotwin interfaces were found to have normals  $(100)_{B2}$  and  $(010)_{B2}$  as given in Table 1, with the angles between the microtwin planes in each plate being greater than 90° for  $(100)_{B2}$  macrotwins, and less than 90° for  $(010)_{B2}$  macrotwins, as also predicted by the calculations. The calculated value  $\lambda^* = .35$  agrees with that observed. Furthermore, the microtwin planes are seen to bend slightly in the vicinity of the macrotwin interface. Away from the interface (at distances measured along the microtwin planes of the order of 500nm) measurement of the angles between the microtwin traces in each plate correlate with the hypothesis that the plates are undistorted there. These angles decrease slightly as the macrotwin interface is approached, consistent with the rotations in Table 1. In the vicinity of the macrotwin plane, high resolution images resolve the atoms and show that the atomic columns on either side of the macrotwin are very nearly parallel, as predicted by the compatibility calculations. In addition, the sense and magnitudes of the rotations of the variants as the macrotwin is approached are consistent with these calculations (see [3, 6] for details).

The angles of rotation in Table 1 give an indication of the degree of incompatibility of the plates and thus of the stored-energy of the macrotwin. The considerably larger angles for all parameter values but  $\kappa_2 = -1, \chi_2 = 1, \nu_2 = 1$  suggest that this case is preferred on grounds of lower energy and that pairs of plates corresponding to the other cases may find different, energetically preferred ways to coalesce. It might be expected, however, that the formation of a macrotwin is aided if the common line of the two habit planes lies in the macrotwin plane; this is the case for the macrotwin with normal  $N_1$  but not for those with normal  $N_2$ .

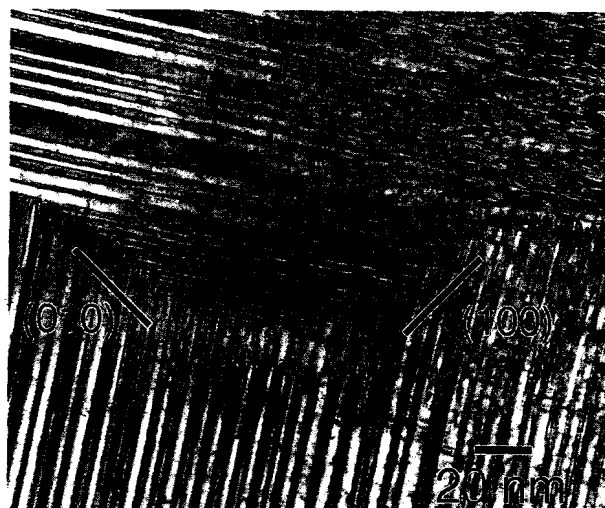


Figure 2: Two plates making contact across both  $(100)_{B2}$  and  $(010)_{B2}$  at either ends of a curved macro twin interface.

In the cases  $\kappa_2 = -1$ ,  $\chi_2 = \pm 1$ ,  $\nu_2 = 1$  the rotations  $Q_1$  and  $Q_2$  are quite close to each other, with the angle of rotation of  $Q_2^{-1}Q_1$  being  $2.33^\circ$ . In one case (see Fig. 2) two plates were observed to make contact across both  $(100)_{B2}$  and  $(010)_{B2}$  at either ends of a curved macro twin interface, with a corresponding small splaying of the microtwin planes. In fact it is to be expected that the overall stress and strain fields will change as one moves parallel to a macro twin due to the incompatibility of the plates in the far field, and perhaps this is responsible for the curving of the macro twin interface.

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