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Theoretical and experimental investigations on elastic interactions between γ'-precipitates in a Ni–Al alloy

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Elastic interaction energies between two identical inhomogeneous plates in an anisotropic matrix were calculated with respect to particle separation distance and alignment orientation on the basis of the microscopic theory of elasticity developed by Yamauchi and de Fontaine. Four types of plate-arrangement along the cube directions; two plates whose faces were mutually parallel, two plates which were arranged on a cube plane as a raft, two plates which were perpendicularly arranged with face–edge configuration and two plates which were perpendicularly aligned with edge–edge configuration were energetically stable, while other arrangements were not stable. Compared with experimental observations on the local arrangement of γ' plates in a Ni–Al single crystal, the calculated results were confirmed to be qualitatively correct.

1. Introduction

Understanding of the elastic interaction energy between precipitates in an anisotropic medium has progressed rapidly. This is of great interest with regard to the regular arrangement of precipitates in a matrix. In most of the previous works on this subject the homogeneous system, wherein the elastic constants of the precipitate were assumed to be equal with those of the matrix, have chiefly been derived [1–4]. However, the regular arrangement of particles due to elastic interaction is usually observed in the inhomogeneous system wherein the elastic constants of the precipitate and matrix are different. On the basis of the continuum theory, Johnson and Lee [5] have theoretically investigated the interaction energy of the inhomogeneous sytem containing two identical precipitates, but their calculations have been limited to the case of spherical particles. Since a general expression of the stress field outside an ellipsoid has recently been developed by Mura and Cheng [6], quantitative estimations of the elastic interaction energy between two ellipsoidal homogeneous particles may be carried out. Nevertheless, the computation has been conducted only for spherical particles as far as authors know, probably because of the difficulties in calculating the stress field outside a plate or rod-shaped inclusion. The precipitates which are regularly aligned by elastic interaction are usually not spherical in shape but parallelepiped or plate-like. Therefore, in order to understand the cause of the regular arrangement of precipitates the interaction energies of plate or parallelepiped precipitates should be calculated. The elastic interaction energy in an isotropic medium has been estimated by Perovic *et al.* [7] for plate-shaped precipitates based upon the elastic strain field associated with dislocation loops. However, the interaction energies obtained by them seem to be partly incompatible with the actual arrangement of precipitates, which will be shown in Section 4 of this paper.

On the basis of the Fourier transform technique in the linear continuum elasticity formulation, Yamauchi and de Fontaine [8] have recently produced a general expression for the elastic interaction energy between small homogeneous clusters. In their treatment, although the cluster shape was assumed to be spherical for simplicity of calculation, tetragonality of the strain field has been

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introduced. Therefore, the calculation of the anisotropic strain field associated with a generally shaped inclusion can be carried out approximately.

In the present paper, the calculation of the elastic interaction energies of two inhomogeneous ellipsoids contained in an anisotropic medium will be shown on the basis of the method of Yamauchi and de Fontaine [8] as functions of particle separation distance and alignment direction, and then the experimental results on the microstructural configuration of plate shaped γ'-precipitates in a Ni–15at%Al single crystal will be shown. The Ni–Al alloy was chosen because the elastic constants of conjugating phases have been determined by many investigators.

2. Theoretical basis for calculation of elastic interaction energies

According to Yamauchi and de Fontaine [8], the elastic interaction energy between two homogeneous clusters (α and β) $E^{\alpha\beta}$ contained in an elastically anisotropic continuum of total volume V is given by Equation 1

$$E^{\alpha\beta} = (1/2V) \sum F^{\alpha\beta}(\mathbf{n}) S^{\alpha}(\mathbf{q}) S^{\beta}(\mathbf{q}) \exp(i\mathbf{qR}) \tag{1}$$

where, **R** shows separation of the two clusters and **n** is the unit vector along the Fourier wave vector **q**. $F^{\alpha\beta}(\mathbf{n})$, the elastic energy coefficient in Fourier space, is defined by Equation 2

$$\begin{aligned} F^{\alpha\beta}(\mathbf{n}) &= \Omega^{\alpha\beta} - \Phi_j^{\alpha}(G^{-1})_{jk}\Phi_k^{\beta} \\ \Phi_j^{\alpha}(\mathbf{n}) &= C_{jklm} n_k \eta_{lm}^{\alpha} \\ \Omega^{\alpha\beta} &= C_{jklm}\eta_{jk}^{\alpha}\eta_{lm}^{\beta} \\ G_{jk}(\mathbf{n}) &= C_{jklm} n_l n_m, \end{aligned} \tag{2}$$

in which C_{jklm} is the elastic stiffness constant and η_{jk} is the distortion tensor which depends on the type of cluster considered.

$S(\mathbf{q})$, the Fourier space shape function, is given by Equation 3, using a shape function $\Theta(\mathbf{q})$.

$$S^{\alpha}(\mathbf{q}) = \Theta^{\alpha}(\mathbf{q})[1 - \delta(\mathbf{q})], \tag{3}$$

where $\delta(\mathbf{q})$, the delta function, is unity at the origin of Fourier space and zero elsewhere. When an α-cluster is assumed to be spherical in shape, the shape function $\Theta^{\alpha}(\mathbf{q})$ is given by Equation 4.

$$\Theta^{\alpha}(\mathbf{q}) = 3V^{\alpha}[\sin(\mathbf{q}r_s) - (\mathbf{q}r_s\cos(\mathbf{q}r_s)]/(\mathbf{q}r_s)^3, \tag{4}$$

where **q** is the wave vector defined by the relation: $(4\pi/3)\mathbf{q}^3 = 32(\pi/a)^3$, where a is a lattice constant and r_s and V^{α} are the radius and volume of the cluster, respectively. Equation 1 was used in the present work as a basic equation to calculate the elastic interaction energy.

The η tensor in $F^{\alpha\beta}$ has been introduced as the following diagonal forms [8]

$$\eta^{\alpha} = \eta^{\beta} = \eta \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & t \end{pmatrix} \text{ for the parallel case}$$

and

$$\eta^{\alpha} = \eta \begin{pmatrix} 1 & 0 & 0 \\ 0 & t & 0 \\ 0 & 0 & 1 \end{pmatrix}$$

and

$$\eta^{\beta} = \eta \begin{pmatrix} t & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \text{ for the perpendicular case.}$$

The parameter t in these diagonal forms indicates the tetragonality of strain field and the scalar η shows the amount of strain. In the parallel case the tetragonality axes of the strain field of α and β clusters are parallel to the [001] direction, while in the perpendicular case the axes of the α and β clusters are along the [010] and [100] directions, respectively.

It has been known that the spherical inhomogeneity whose eigen strain is of pure dilatation produces an isotropic strain field, while plate-like inhomogeneity produces an anisotropic field [9]. Therefore, an evaluation of the tetragonality in the anisotropic strain field produced by a plate-like ellipsoid is conducted first, and then the interaction energy between them is calculated. The tetragonality of the anisotropic field can be determined from eigen strains (stress free strain) e_{ij}^{T} of "an equivalent ellipsoid" which forms a strain field equivalent to that of an ellipsoidal inhomogeneity. The equivalent eigen strain e_{ij}^{T} can be obtained from the simultaneous Equations 5 and 6 if the eigen strain of the inhomogeneous ellipsoid $e_{ij}^{\mathrm{T}*}$ is experimentally given [9, 10].

$$\sigma_{ij}^{\mathrm{I}} = C_{ijkl}(e_{kl}^{\mathrm{c}} - e_{kl}^{\mathrm{T}}) = C_{ijkl}^{*}(e_{kl}^{\mathrm{c}} - e_{kl}^{\mathrm{T}*}) \tag{5}$$

$$e_{ij}^{\mathrm{c}} = (1/4\pi)C_{mnkl}\bar{G}_{ijmn}e_{kl}^{\mathrm{T}}, \tag{6}$$

TABLE I Elastic constants of conjugating phases and eigen strain used in calculation

Elastic constant ($MN\,m^{-2}$)		
C_{11}	11.24×10^4	Elastic constants
C_{12}	6.27×10^4	of matrix at
C_{44}	5.69×10^4	1023 K
C_{11}^*	16.66×10^4	Elastic constants
C_{12}^*	10.65×10^4	of γ' at
C_{44}^*	9.92×10^4	1023 K
e^{T*}	5.63×10^{-3}	Eigen strain (Stress free strain)

where e_{ij}^c is the constrained strain, and C_{ijkl} and C_{ijkl}^* are the elastic stiffness constants of matrix and inclusion, respectively. $\bar{G}_{ijkl}$ has been concretely given as a function of the aspect ratio of an ellipsoid by Lin and Mura [11]. Thus η and t in the η tensor are now represented by e_{11}^T and e_{33}^T/e_{11}^T, respectively.

This procedure inevitably implies an assumption that the elastic strain field caused by the ellipsoidal cluster ($c/a \neq 1$) is identical to that of the spherical one, provided both particles have the same eigen strain e_{ij}^T, elastic constant C_{ijkl} and volume V. This assumption is not generally correct. Nevertheless, the approximate calculation mentioned above was conducted in the present work because the elastic strain energies of the sphere and ellipsoid are approximately equal unless the aspect ratio of the ellipsoid c/a deviates too far from unity, which will be shown in Section 3.

3. Numerical calculations on the γ'-precipitates in Ni–Al alloys

Numerical values used for the calculation are summarized in Table I, where the elastic stiffness constants at an ageing temperature of 1023 K for γ'-precipitates and matrix have been represented in our previous work [12]. The eigen strain (stress free strain) e^{T*} of γ'-precipitates at 1023 K was evaluated from the lattice constants of a Ni solid solution containing 13.5 at% Al, which had been measured by X-ray diffraction in the previous work [12], and from the lattice constants of the Ni_3Al single phase [13].

The results of the calculations are now presented. Table II shows the change of the equivalent eigen strain e_{ij}^T with aspect ratio of homogeneous ellipsoid. It is obvious that the tetragonality t ($= e_{33}^T/e_{11}^T$) gradually deviates from unity with a decrease in aspect ratio.

Here, the numerical errors caused by using the spherical shape function $\Theta(\mathbf{q})$ in the calculation of the anisotropic elastic strain field associated with the ellipsoid are examined. $E^{\alpha\beta}$ in Equation 1 gives the elastic strain energy, E_{self}, of a single particle (α or β) when the distance between the centres of the two particles R is put as zero [8]. The E_{self} of ellipsoids having various aspect ratios are represented in Table II, which may imply numerical errors arising from $\Theta(\mathbf{q})$. An elastic strain energy E'_{self} can be also obtained from the familiar Equation 7 developed by Eshelby [10] as well as Equation 1. This gives accurate strain energies of ellipsoids.

$$E'_{self} = -(1/2)\sigma_{ij}^I e_{ij}^{T*}. \quad (7)$$

The E_{self} should essentially coincide with E'_{self} for all aspect ratios, if the calculation of E_{self} is performed with the ellipsoidal shape function. The numerical values of E'_{self} for various aspect ratios of ellipsoids are also shown in Table II in the form of a ratio to E_{self}. It is obvious from the table that E_{self} nearly coincides with E'_{self} unless the aspect ratio deviates too far from unity. Thus it seems that, even if the spherical shape function $\Theta^\alpha(\mathbf{q})$ is used as a substitution of that of ellipsoid, Equation 1 is quantitatively available to evaluate the elastic interaction energy between two oblate spheroids.

Using e_{ij}^T shown in Table II, the interaction energy $E^{\alpha\beta}$ is now represented. Fig. 1 shows the change of $E^{\alpha\beta}$ with interecentre distance between two precipitates for the case of $c/a = 1$ (sphere). The ordinate and abscissa are normalized by E_{self} and the particle diameter, respectively. From the figure the most energetically favourable position

TABLE II Dependencies of e_{ij}^T, tetragonality of strain t, E_{self} and E'_{self}/E_{self} with aspect ratio of ellipsoid

Aspect ratio c/a	e_{33}^T	e_{11}^T	Tetragonality of strain, t	E_{self} ($J\,m^{-3}$)	E'_{self}/E_{self}
1.0	0.651×10^{-2}	0.651×10^{-2}	1.00	0.522×10^7	1.00
0.7	0.673×10^{-2}	0.604×10^{-2}	0.90	0.519×10^7	1.00
0.5	0.694×10^{-2}	0.558×10^{-2}	0.80	0.517×10^7	0.99
0.3	0.721×10^{-2}	0.496×10^{-2}	0.69	0.513×10^7	0.96
0.1	0.756×10^{-2}	0.409×10^{-2}	0.54	0.508×10^7	0.91

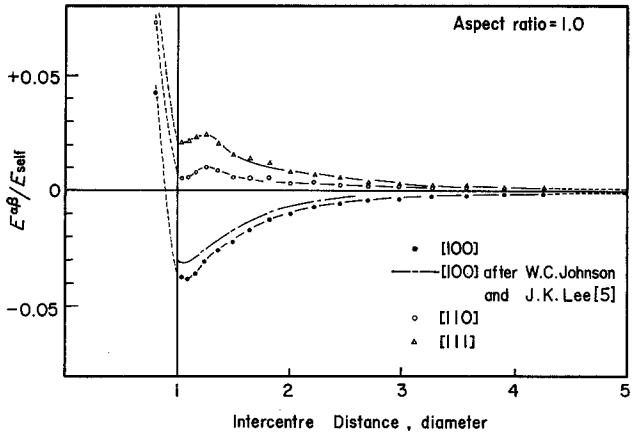

Figure 1 Elastic interaction energies between two spherical particles as functions of particle separation and alignment direction.

of a neighbouring particle is found to be slightly away from the original sphere along the ⟨100⟩ cube directions. A chain line in Fig. 1 is the ⟨100⟩ interaction energy which has been calculated by Johnson and Lee [5], based upon the modified Eshelby method, for the case of spherical γ'-particles in Ni–Al alloys. A little incompatibility between the lines can be seen. However, this results from the difference in the value of the elastic constants used in the computations, not from the calculation method since both lines were confirmed to be nearly equal when the same value of stiffness was used.

Results for the case where plate-shaped γ'-precipitates ($c/a = 0.5$) are aligned in parallel are summarized in Fig. 2. Relative configurations of plate-arrangement are schematically drawn in the figure. It is clear from Fig. 2 that two plates whose faces are mutually parallel (face–face configuration) along the orthogonal ⟨100⟩ directions [(e) of Fig. 2] interact most attractively, and then the case of two plates which are arranged on a plane as a raft along the ⟨100⟩ directions [(d) in Fig. 2] is next in order of attractive interaction. Whereas other arrangements [(a), (b) and (c) in Fig. 2] are repulsive.

Fig. 3 shows the calculated results for perpendicular cases. The arrangements (d) and (e) show energetically favourable positions, while arrangements (a), (b) and (c) are not stable.

On the basis of the stress field of dislocation loops, Perovic *et al.* [7] have evaluated the elastic interactions between two identical plates to be repulsive for the mutually parallel case [(e) of Fig. 2], and attractive for the mutually perpendicular arrangement [(e) of Fig. 3]. This is opposite to our result for the mutually parallel case. Electron microscopic observations on the actual configuration of γ'-precipitates will be shown in the next section.

4. Comparison with some experimental observations

The results presented in Figs 2 and 3 are discussed below. Fig. 4 shows an example of dark-field images of γ'-(Ni_3Al) precipitates, taken from a foil

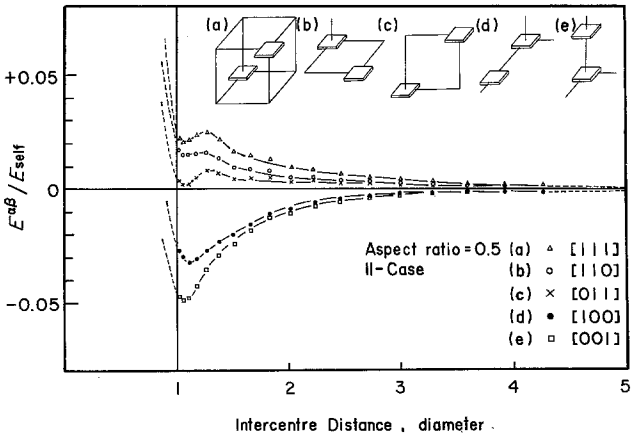

Figure 2 Elastic interaction energies for a parallel arrangement.

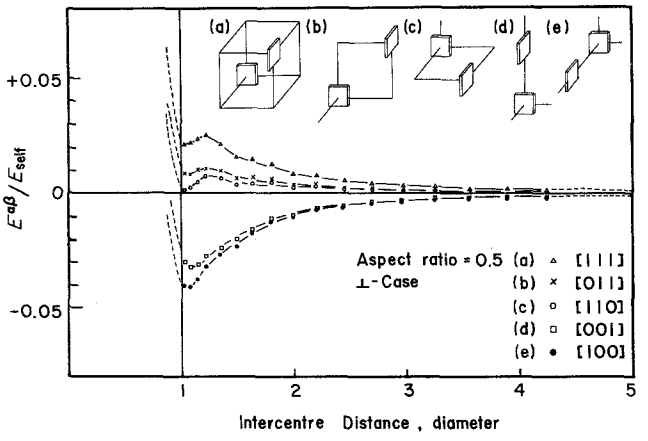

Figure 3 Elastic interaction energies for a perpendicular arrangement.

which was prepared from Ni–15at%Al single crystal aged at 1073 K for 1800 min. The foil was cut parallel to the (100) plane. Many plate-shaped γ'-precipitates, about 0.4 in aspect ratio, are regularly aligned along the orthogonal ⟨100⟩ directions. Particle-arrangement can be classified into four types which are represented in Fig. 5a to d. In Fig. 5a the plates are seen to be arranged in the (100) plane along the [010] direction. This type of arrangement is clearly seen from Fig. 2 to be stable. Fig. 5b shows a mutually perpendicular configuration (face-edge), where one particle is in the (100) and the other in the (010) plane, which corresponds to (e) of Fig. 3. A practical example of the arrangement of (d) of Fig. 3 is found in Fig. 5c. Fig. 5d shows an example of the mutually parallel case (face–face configuration), where the two plates are seen to be arranged in the (010) plane. In this case it may be considered that, being a fairly large distance away, each plate is outside the elastic strain field produced by the other particle. However, the centre to centre distance of the particles is nearly equal to those in Fig. 5a to c. Hence, it is understood that the two particles in Fig. 5d are located in their energetically favourable positions indicated in Fig. 2. This face–face configuration was reported by Perovic *et al.* [7] to be repulsive, but this is obviously incompatible with Fig. 5d. Further experimental evidence clearly showing that the face–face configuration of particles is energetically stable is given. Fig. 6a and b are bright-field micrographs of γ'-particles taken from a thin foil of Ni–40at%Cu–5.0at%Si alloy aged at 773 K for 1000 min. Structural changes with ageing in this alloy system have been previously reported [14], wher the γ'-particles (Ni_3Si) are nucleated at a later stage of ageing. In Fig. 6a the γ'-particles are sparsely dispersed because of a small amount of Si in the alloy. Fig. 6b shows enlarged precipitates. The morphology of the γ'-precipitates should be noted. It can be seen that all precipitates, of which the aspect ratio is about 0.6, are separated into two mutually parallel plates [in this photograph the crevices parallel to the [01 $\bar{1}$] direction are only observable because of the foil plane (011)]. Such pairing of γ'-particles was experimentally verified to take place when ageing was performed inside but near the miscibility gap of phase transformation of this alloy system. Each pair of γ'-plates is almost iso-

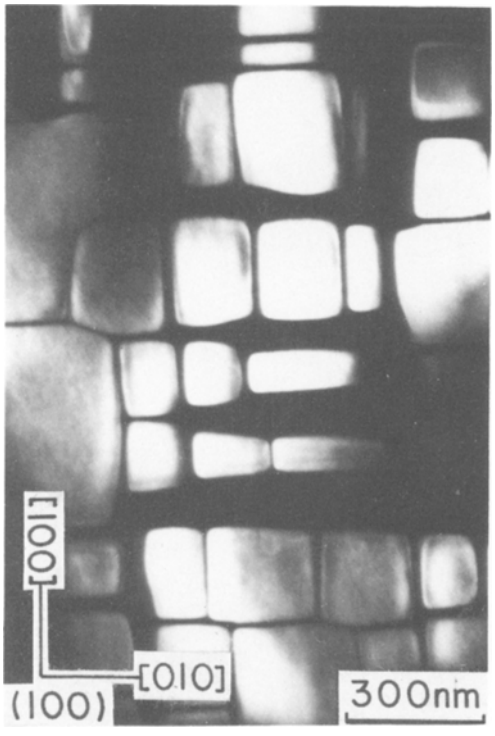

Figure 4 A dark-field image of γ'-plates in a Ni–15at%Al single crystal aged at 1073 K for 1800 min.

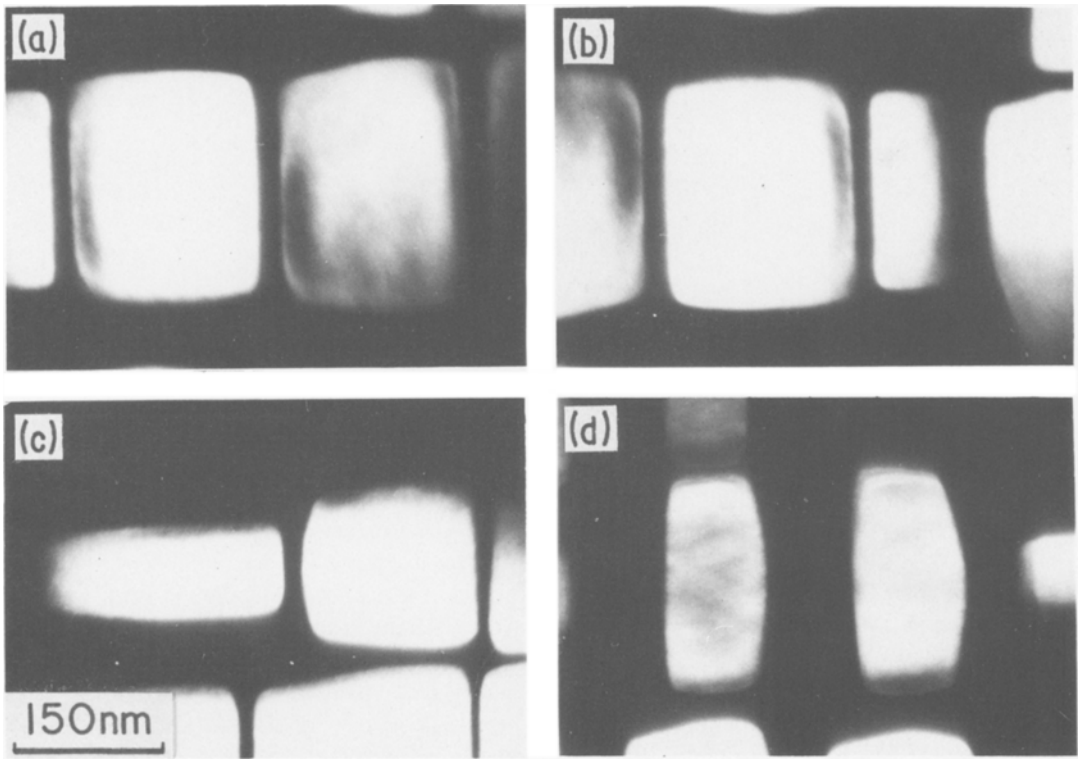

Figure 5 Four types of local arrangement of γ'-plates in a Ni–15 at%Al single crystal aged at 1073 K for 1800 min.

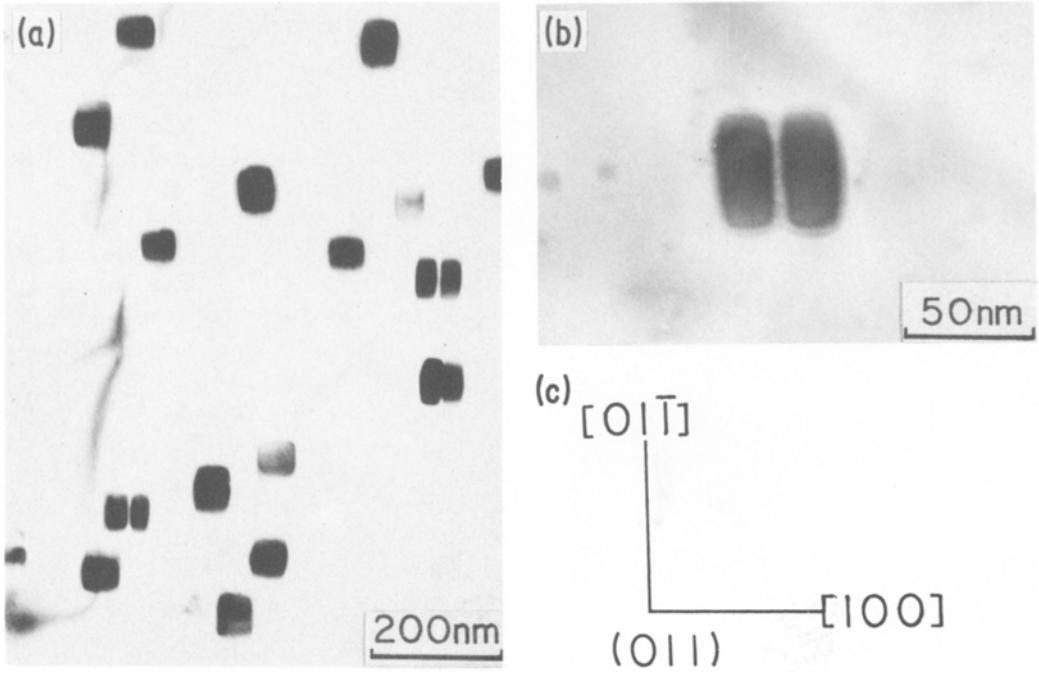

Figure 6 Dark-field micrographs of γ'-precipitates (Ni_3Si) in a Ni–40 at%Cu–5 at%Si alloy aged at 773 K for 1000 min, noting paired precipitates.

lated from other pairs, as can be seen in Fig. 6a, so that the two pairing plates seem to be elastically affected only by its partner's elastic strain field without any other elastic influence due to third or fourth particles. This is therefore a typical example of the "two-body problem" that the theoretical calculation dealt with. Since this alloy system has almost the same elastic constant and eigen strain values as those of the Ni–Al alloy system [12], the elastic interaction energies calculated for the γ'-particles of Ni–Al alloy ought to be qualitatively applicable to the γ'-particles in the Ni–Cu–Si alloy system. Consequently, the paired γ'-plates shown in Fig. 6 are sufficient evidence for the theory that mutually parallel plates along the cube directions are in energetically stable positions.

5. Conclusions

Numerical calculations on the elastic interaction energies between two identical inhomogeneous plates in an anisotropic medium were derived, based upon the microscopic elasticity theory as functions of particle separation distance and alignment orientation. Four types of arrangement along the orthogonal ⟨1 0 0⟩ directions are energetically favourable. These are (a) two plates with mutually parallel faces, (b) two plates arranged in a cube plane as a raft, (c) two plates which were perpendicularly aligned with face–edge configuration and (d) two plates which were perpendicularly arranged with edge–edge configuration. Other arrangements were not stable. Compared with experimental observations on the local arrangement of γ'-precipitates in a Ni–Al single crystal, the calculation results were confirmed to be qualitatively correct.

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