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ULTRASONIC VELOCITY NEAR THE MARTENSITIC TRANSFORMATION TEMPERATURE


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Abstract.—Single crystal specimens of γ1'-martensite and γ1-austenite of Cu-Al-Ni alloy are prepared. The temperature dependence of the shear elastic constants of both phases is measured and found to be in good agreement with the prediction from a simple free energy model for the martensitic transformation.

Introduction.—In the martensitic transformation in such alloys as In-Tl, Nb3Sn or other A-15 compounds, the instability of the austenitic phase has been clearly revealed by the drastic decrease of the shear elastic constant $C' = (C_{11} - C_{12})/2$ as the temperature approaches the martensitic transformation temperature $M_s$. On the other hand, in most of the typical martensitic alloys such as Cu-Al-Ni or Cu-Zn-Al, the decrease of the shear elastic constant $C'$ is very small relative to its absolute value at $M_s$. The decrease of the elastic constant $C'$ in the austenitic phase alone cannot give any explicit evidence for the instability of the austenitic phase. Guenin, Rios Java, Murakami, Delaey and Gobin have succeeded, for the first time, in producing a single crystal of martensitic phase of Cu-Zn-Al alloy and in measuring the temperature dependence of the shear elastic constant in both the austenitic phase and the martensitic phase (1). They have found a decrease of the shear elastic constant in the martensitic phase with increasing temperature. However, the shear elastic constant of martensitic phase that Guenin et al. measured does not correspond to the deformation in the basal plane of the martensitic phase. The present authors have measured the temperature dependence of the shear elastic constant corresponding to the shear deformation in the basal slip plane of both the martensitic and the austenitic phases.

Specimen.—Single crystals in the B1 matrix phase were made by the modified Bridgman method. One of the B1-phase single crystals was transformed into γ1'-phase single crystal by the stress-inducement technique previously described (2). A rectangular parallelepiped martensitic phase specimen surrounded by (100), (010), and (001)γ1 faces was cut by the spark cutting machine after the crystal orientation was determined by the back reflection Laue method. A B1-phase specimen has one pair of (110)γ1 parallel planes. The compositions and transformation temperatures of the single crystal specimens are shown in Table 1. The crystal structure of the γ1'-martensite has the 2H structure which has ABABAB... stacking sequence as shown in Fig.1. In contrast to B1 (Austenite) phase which has a cubic crystal structure, the γ1'-martensite has an orthorhombic crystal structure. Hence, its second order elastic constant tensor has 9 independent non-zero components. Figure 2 shows the lattice structure relationship between B1-phase and γ1'-phase. The shuffle plane indicated in...
Method of measurement.- The measurement of ultrasonic velocity was carried out by the Pulse-Echo-Overlap (PEO) method (3) with 3MHz-X and Y cut quartz transducers. Elastic constants $C_{ij}$ were determined from $C_{ij} = \rho V^2$, where $\rho$ is the density of specimen and $V$ the sound velocity. Both $\beta_1$ and $\gamma_1'$-phase specimens were of about 5mm cube, which were suitable for measurement of transverse wave velocity, but were too short for longitudinal wave velocity with the PEO method. Therefore, the measurement of longitudinal wave velocity was carried out by measuring the echo time intervals directly on the oscilloscope without using the PEO method. Shear elastic constants $C'$ and $C_{44}$ in the $\beta_1$-phase were determined from the velocity of the transverse waves propagating along [110] and polarized to [110] and [001], respectively. On the other hand, diagonal components of $\gamma_1'$ elastic constant tensor were determined from the velocity of the one longitudinal and two transverse waves propagating in each of the following directions: [100], [010] and [001]. Although we carried out 9 measurements for 3 propagation and 3 polarization directions, we could not determine all of the 9 components of the tensor because of the duplicate elastic constants obtained. The measurement of off-diagonal 3 components has not been carried out at the time of the preparation of this paper. The temperature dependences of elastic constants $C_{44}$, $C'$ in the $\beta_1$-phase and those of $C_{55}$, $C_{44}$ in the $\gamma_1'$-phase were measured. The elastic constants $C'$ and $C_{55}$ correspond to the shear deformation parallel to the basal plane shown in Fig.1. On the other hand, the elastic constant $C_{44}$ corresponds to the shear

<table>
<thead>
<tr>
<th>Nominal Composition</th>
<th>Ms</th>
<th>Mc</th>
<th>Af</th>
<th>As</th>
</tr>
</thead>
<tbody>
<tr>
<td>Austenite phase</td>
<td>Cu-14.1wt%Al-3.0wt%Ni</td>
<td>-18.0</td>
<td>-46.0</td>
<td>-12.0</td>
</tr>
<tr>
<td>Martensite phase</td>
<td>Cu-13.8wt%Al-4.0wt%Ni</td>
<td>15.5</td>
<td>15.5</td>
<td>50.0</td>
</tr>
</tbody>
</table>

Table 1: Composition in the ingots and transformation temperature of the specimens.
deformation on the same basal plane but with the direction at right angle to the deformation corresponding to C' or C55 on the basal plane. The thermal expansions in the specimens, in the quartz transducer and in the adhesive between the specimen and the transducer, are estimated to be negligible in the small range of temperature in the present experiment. The Q-value of the quartz transducer bonded to the specimen by the adhesive is also checked to be practically independent of the temperature.

Experimental Results. 6 diagonal components of the elastic constant tensor of γ1 martensite were determined as shown in Table 2. The reason why results of C11, C22, and C33 have only two significant figures is that the measurement of the longitudinal wave velocity is carried out without the PEO method. It is to be noticed that C55 is much smaller than the other shear elastic constants C44 and C66. However, neither of the ratios C44/C55 nor C66/C55 in the γ1-phase are as large as the anisotropy ratio C44/C' in the β1-phase. The temperature dependences of C55, C44 in the γ1-phase and of C', C44 in the β1-phase near the As and Ms are shown in Fig.4. Figure 5 compares the temperature dependence of C' with that of C55. It is remarkable that there exists a gap between the corresponding shear elastic constants of the β1-phase and the γ1-phase at the transformation temperature. While the temperature dependence of C55 is negative, that of C' is positive. Although the temperature dependence of C' is positive, its value looks too small to indicate the presence of the lattice instability in the β1-phase. However, it is found from the analysis in the following

<table>
<thead>
<tr>
<th>C_{11}</th>
<th>C_{22}</th>
<th>C_{33}</th>
<th>C_{44}</th>
<th>C_{55}</th>
<th>C_{66}</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.9x10^2</td>
<td>1.4x10^2</td>
<td>2.2x10^2</td>
<td>55.9</td>
<td>21.2</td>
<td>62.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>57.5</td>
<td>19.4</td>
<td>58.7</td>
</tr>
</tbody>
</table>

(x10^10 dyne/cm²)

Table 2: Diagonal components of the elastic constant tensor of the orthorhombic γ1 martensite as determined from the 9 ultrasonic measurement.
Fig. 4: Temperature dependence of shear elastic constants in the martensite (A) and in the austenite (B).
Fig. 5: Gap and slope of the shear elastic constants ($C_{55}$ and $C'$) across the transformation temperature.

section that this small positive temperature dependence is indeed indispensable for the martensitic transformation.

**Free Energy Model.**—We try to interpret the behavior of the shear elastic constant corresponding to the shear deformation in the basal planes of both the martensitic and the austenitic phases in terms of simple free energy models. First, we try the simplest expression for free energy as a function of the relative displacement $\epsilon$.

The relative displacement $\epsilon$ is defined in terms of the displacements of the neighboring basal planes, $u_n$ and $u_{n-1}$, as

$$\epsilon = \kappa (u_n - u_{n-1}),$$  \hspace{1cm} (1)

where $\kappa$ is a geometrical constant. The simplest expression for the free energy, that has two minima with the same depth at the equilibrium temperature $T_0$, is given as follows:

$$F = a_2 ((1 + \delta \cdot (T-T_0)) \frac{\epsilon^2}{2} - \epsilon^3 + \frac{\epsilon^4}{2}),$$  \hspace{1cm} (2)

where $a_2$ is the second order shear elastic constant of the austenite at $T=T_0$ and $\delta$ is a constant. The equilibrium condition $\delta F/\delta \epsilon = 0$ has two solutions for minima, one at $\epsilon = 0$ corresponding to the austenite and another at $\epsilon = \epsilon_M$. The latter moves from 1 as $T$ moves from $T_0$. The second order shear elastic constants of the austenite and the martensite are proportional to the second derivatives of the free energy given by Eq. (2) at the respective minima. They are

$$\frac{\partial^2 F}{\partial \epsilon^2} \epsilon = 0 = a_2 (1 + \delta \cdot (T-T_0))$$  \hspace{1cm} (3)

and

$$\frac{\partial^2 F}{\partial \epsilon^2} \epsilon = \epsilon_M = a_2 (1 - 5 \delta \cdot (T-T_0)).$$  \hspace{1cm} (4)
The free energy model given by Eq.(2) requires that the ratio of the shear elastic constant of the martensite to that of the austenite at the equilibrium temperature $T_0$ is equal to 1 and the ratio of the temperature derivative of the shear elastic constant of the martensite to that of the austenite is equal to $-5$. However, Fig.5 shows that the ratio of $C_{55}$ to $C'$ at $T_0$ is estimated to be approximately 3 and the ratio of $dC_{55}/dT$ to $dC'/dT$ is approximately $-11.3$. Thus, it is seen that the experimental results cannot be interpreted in terms of the free energy model $F_3$ given by Eq.(2).

Second, we try the next simplest expression for the free energy $F_4$, which has now one minimum (at $\varepsilon=0$) corresponding to the austenite and another two minima (at $\varepsilon=\varepsilon_M$) corresponding to the martensite. The latter moves from $T_1$ as $T$ moves from $T_0$.

The expression for the free energy $F_4$, which is an even function of $\varepsilon$, is given by

$$F_4 = a_2 (1 + 6 \cdot (T-T_0)) \varepsilon^2 - \varepsilon^4 + \varepsilon^6 / 2.$$  \hspace{1cm} (5)

The free energy model Eq.(5) is very similar to the one used by Devonshire (4) to study the ferroelectric transition and also to the one proposed by Falk (5) as a model for the shape memory alloy. However, a subtle difference in the interpretation of the expansion parameter of the free energy exists between the present paper and the paper by Falk. Whereas the expansion parameter used in the paper by Falk represents the macroscopic shear strain, the expansion parameter in the present paper represents a relative displacement of the neighboring basal slip planes. The second order shear elastic constants of the austenite and the martensite are proportional to the second derivatives of the free energy given by Eq.(5) estimated at $\varepsilon=0$ and $\varepsilon=\varepsilon_M$. They are

$$\left( \frac{\partial^2 F_4}{\partial \varepsilon^2} \right)_{\varepsilon=0} = a_2 (1 + 6 \cdot (T-T_0))$$  \hspace{1cm} (6)

and

$$\left( \frac{\partial^2 F_4}{\partial \varepsilon^2} \right)_{\varepsilon=\varepsilon_M} = a_2 (4 - 8 \cdot 6 \cdot (T-T_0)).$$  \hspace{1cm} (7)

The free energy model Eq.(5) requires that the ratio of the shear elastic constant of the martensite to that of austenite at $T_0$ is equal to $4$ and the ratio of the temperature dependence of the shear elastic constant of the martensite to that of the austenite is equal to $-8$. Hence, it is concluded that the free energy model $F_4$, which is an even function of $\varepsilon$, gives a definitely better agreement with the experimental results shown in Fig.5 than the free energy model $F_3$. It is noticed that while the free energy model $F_4$ can reflect the fact that the $DO_{3}$ basal slip plane can be shear deformed in either way ($\varepsilon=\varepsilon_M$) to obtain the 2H-type stacking, the free energy model $F_3$ cannot.

References.
(2) OTSUWA.K., SAKAMOTO.H., SHIMIZU.K., Scripta Met. 10 (1976) 981.