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INTERNAL FRICTION AND ELASTIC CONSTANT ANOMALIES OF ANTIFERROMAGNETIC Mn-Ni ALLOYS

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Résumé - Les variations du module élastique et du coefficient de frottement interne avec la température ont été mesurées sur des alliages poly- et mono-cristallins de Mn-Ni dans le domaine de concentration 14 à 30 %. Les alliages riches en Mn montrent de grandes anomalies à la température de Néel et au-dessous, accompagnées d'un frottement interne élevé avec $Q^{-1} = 10^{-2}$. Ces anomalies sont dues aux transformations magnétiques et/ou structurales se produisant dans ces alliages.

Abstract - The temperature variation of the elastic moduli, as well as of the internal friction of poly-and single crystalline Mn-Ni alloys in the concentration range 14 - 30% Ni is reported. Mn-rich alloys exhibit large elastic anomalies at and below the Néel temperature and a high internal friction with $Q^{-1} = 10^{-2}$. These anomalies are due to the magnetic and/or structural transformations occurring in these alloys.

Introduction - The elastic moduli of fcc Mn-Ni alloys show large anomalies at and below their Néel temperature $T_N$ due to either antiferromagnetic ordering and/or to a structural transformation which occurs concurrently with magnetic ordering [1 - 6]. The latter transformation is of a displacive type and is often called martensitic [6]. Single crystal elastic measurements have indicated that the $C' = (C_{11} - C_{12})/2$ mode is highly anomalous whereas the $C_L = (C_{11} + C_{12} - 2C_{44})/2$ and the $C_{44}$ modes are less strongly affected by the antiferromagnetic ordering and the associated structural transformation [5 - 6].

These alloys exhibit also pronounced internal friction anomalies at and below the Néel temperature [1 - 5, 7 - 9] similar to Mn-Cu alloys where the occurrence of high damping [1, 7 - 9] is both promoted and complicated by the decomposition into a coherent two-phase structure according to a metastable miscibility gap [10]. In the Mn-Ni system a corresponding miscibility gap does not exist. In both the Mn-Cu and Mn-Ni systems the tetragonal or orthorhombic distortion of the fcc lattice concurrent with the antiferromagnetic transition has been shown to give rise to a high internal friction peak ($Q^{-1} = 10^{-2}$) near 0°C. It is concluded that this peak arises if the lattice distortion leads to the formation of a high number of twin domain boundaries [7 - 10] which are arranged in a characteristic lamellar microstructure.

Experimental - Polycrystalline Mn-Ni alloys were prepared containing 14.3, 18.3, 20.6, 22.4 and 31.7 at.% Ni. Single crystalline specimens were grown by the Bridgeman technique with compositions 15 and 18.5 at.% Ni respectively. All alloys were quenched into water after
a homogenizing treatment at 950°C. The elastic and anelastic properties of the polycrystalline specimens, E, G and Q\(^{-1}\), were determined using a Tectanel apparatus using specimens with dimensions 50 x 5 x 1 mm\(^3\) or a Förster Elastomat using samples in the form of rods. In the former equipment an electrostatic driving and detection system is used while in the latter equipment an electromagnetic system is employed.

In the case of single crystals, the sound velocities were measured on specimens oriented in a [110] direction by the pulse overlap technique using MATEC equipment. The sound velocities, v, are related to the elastic constants, C\(_{ij}\), as follows:

\[
\begin{align*}
C_L &= v^2_l = (C_{11} + C_{12} + 2C_{44})/2 \\
C' &= v^2_t = (C_{11} - C_{12})/2 \\
C_{44} &= v^2_t = C_{44}
\end{align*}
\]

where \(\rho\) is the density; \(\rho = 7.33\) for Mn-18.5 at.% Ni and \(\rho = 7.285\) for Mn-15 at.% Ni was used to convert sound velocities into elastic constants.

Results and Discussion — The temperature dependence of Young's modulus E, of the shear modulus G and of the internal friction Q\(^{-1}\) of polycrystalline Mn-Ni alloys containing 14.3, 18.3, 20.6, 22.4 and 31.7% Ni is shown in Figs 1 and 2 respectively. Some of the results for Mn-14.3% Ni and Mn-18.3% Ni have been reported earlier [5]; they are included here for completeness.

Around \(T_N\), which was determined by differential scanning calorimetry, a strong decrease occurs with decreasing temperature for both E and G, while the internal friction Q\(^{-1}\) reveals peaks at \(T_N\) and at \(\approx 0°C\), respectively. All effects are the larger the higher the Mn content of the alloy. The temperature variation of E and G for Mn-Ni alloys with 14.3, 18.3 and 20.6 at.% Ni is non-monotonic below \(T_N\); these alloys exhibit modulus minima at about 0°C, 100°C and -100°C, respectively.

A large depression in E and G is commonly observed at the paramagnetic antiferromagnetic transition irrespective of whether magnetic ordering leads to a structural distortion of the parent phase or not. It is interesting to note that E and G show practically the same anomalous behaviour. From the relation

\[
\frac{\Delta E}{E} = \frac{8}{9} \frac{\Delta G}{G} + \frac{1}{9} \frac{\Delta B}{B}
\]

one can conclude that - for Mn-Ni alloys - shear modes dominate in being responsible for the anomalous elastic behaviour, while volume effects as described by the bulk modulus B are less important. This is quite different from the results obtained by Tsunoda et al [11] who suggested - in the case of Mn-Cu alloys - that the effects associated with the bulk modulus are most important; i.e., Mn-Ni alloys are quite different from Mn-Cu alloys as far as their magnetoelastic properties are concerned.

Alloys in the concentration range 15 - 20% Ni show non-monotonic variations below \(T_N\) which agrees very well with the previous measurements by Honda et al [4]. This is the concentration range where magnetic ordering induces a distortion of the lattice (tetragonal with c/a \(\approx 1\) or orthorhombic, depending on concentration [4]). The magnetically induced transformation of the lattice from fcc to fct (or fco) leads to the formation of a substructure in which - because of internal stress relief - the direction of tetragonality alternates. The boundaries (see Fig 5) between adjacent regions can be described as being microtwin boundaries as well as antiferromagnetic domain
Fig 1 - Temperature dependence of Young's modulus $E$ and of the shear modulus $G$ of Mn-Ni alloys. The Néel temperature is indicated by an arrow. Boundaries. These coherent low-energy boundaries move easily under the application of stress and give rise - similar to the case of ferromagnetic materials - to a nonintrinsic $\Delta E$ effect, as well as to high damping. The anomalous behaviour of $E$ and $G$ below $T_N$, as well as the internal friction peak occurring at $\approx 0^\circ\text{C}$, is attributed to this mechanism. For Mn-14.3% Ni and Mn-18% Ni, this internal friction peak is very large, $Q^{-1}$ max $\approx 10^{-2}$ for $\epsilon = 10^{-6}$ where $\epsilon$ is the strain amplitude. Evaluating the activation energy, $H$, for the $0^\circ\text{C}$ peak using the peak shift method ($H_s$) as well as the half-width method ($H_\Delta$) we obtain similar results as those obtained by Sugimoto et al [1]:

$$H_s = 6.5 \times 10^4 \text{ J mole}^{-1} \quad (\text{Mn-14.3} \% \text{ Ni})$$
$$= 6.0 \times 10^4 \text{ J mole}^{-1} \quad (\text{Mn-18.3} \% \text{ Ni})$$

$$H_\Delta = 2.4 \times 10^4 \text{ J mole}^{-1} \quad (\text{Mn-14.3 at.} \% \text{ Ni})$$
$$= 2.3 \times 10^4 \text{ J mole}^{-1} \quad (\text{Mn-18.3 at.} \% \text{ Ni})$$

The large difference between $H_s$ and $H_\Delta$ is indicative of a distribution of relaxation times.

The elastic behaviour of the single crystalline Mn-Ni alloys (15 and 18.5% Ni) is shown in Figs 3 and 4. $C_L$ shows a drop of about 3% at $T_N$ which correlates well with the peak in the specific heat. Such a discontinuous behaviour is absent in the case of $C_{44}$. Unfortunately, no results could be obtained for $C'$, neither for the 15% Ni, nor for the
Pig 3 - Temperature variation of the elastic constant
\[ C_L = \frac{(C_{11} + C_{12} + 2C_{44})}{2}. \]
\( C_p \) is the specific heat measured by differential scanning calorimetry.

18.5\% Ni specimen, possibly due to a high damping of this mode. After completion of the measurement, the sample length was reduced to 5 mm, without any improvement, however.

Comparing Fig 1 with Figs 3 and 4, it appears that a large difference exists between the elastic behaviour found for polycrystalline and single crystalline Mn-Ni alloys. Such a behaviour we have found also in the case of Fe-Mn alloys [12,13].

From \( C = C_{44} \) and \( C' \), the shear modulus \( G \) can be calculated by means of an appropriate averaging scheme. By using the Voigt-Reuss-Hill average, we obtain

\[ G = \frac{1}{10} \frac{38C' + 6C'' + 6C^2}{2C + 3C'} \]

Since we could not measure \( C' \), we assume the largest possible anomaly, i.e., \( C' = 0 \). This assumption does not introduce a serious error in the calculation since \( C' \ll C \) for highly anisotropic materials. In this case, \( G \) is simply given by

\[ G \approx 0.3 \] C_{44} \]
The behaviour of $C_{44}$ (Fig 4) is totally different from that found for $G$ (Fig 1). We have no rigorous explanation for this discrepancy. Possibly the measurement done for polycrystals contains a large non-intrinsic $\Delta E$ effect caused by the generation of a large anelastic strain due to the presence of a domain structure (Fig 5). At ultrasonic frequencies, on the other hand, the domain boundaries may not follow the stress wave and the $\Delta E$ effect does not occur.

Fig 4 shows that $C_{44}$ is increased by antiferromagnetic ordering, at least near $T_N$, which is different from our previous analysis [5], i.e., $\Delta C^M_{44}$, where $\Delta C^M_{44}$ is the exchange contribution [14], is positive. $C_L$ also exhibits a positive exchange contribution (Fig 3).

Single crystalline elastic constant data were reported by Lowde et al [6] for MnNiC alloys. These authors were able to measure $C'$ in these alloys which showed a marked softening of $C'$ on lowering the temperature. Moreover, a peculiar behaviour of $C_{11}$ was found by Lowde et al [6] in that $C_{11}$ became soft on cooling and in that $C_{11}$ had a lower value than $C_{44}$. In this respect it should be noted that not only $C_{11}$ and $C'$ behave anomalously but also $C_{44}$. When comparing $C_{44}$ with other materials, it appears that $C_{44}$ of Mn-Ni alloys is unusually high.

Fig 5 shows the microstructure of the Mn-15% Ni single crystal exhibiting a banded microstructure. The characteristic variations in band width are due to both sectioning effects and true differences in the average density of twin domains. These differences arise from local variations in the magnitude and direction of transformation strains which increase along with the tetragonal (orthorhombic) magnetostrictive distortions with decreasing temperature. Consequently, the mobility of the boundaries (and their node lines) is expected to be a strong function of temperature in agreement with the evidence that can be derived from the internal friction behaviour.

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