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# Anomalous temperature behaviour of the elastic constants of antiferromagnetic Mn–Invar alloys

U Kawald†, O Mitze†, H Bach†, J Pelzl† and G A Saunders‡

† Institut für Experimentalphysik, Arbeitsgruppe Festkörperspektroskopie, Ruhr-Universität Bochum, D-44780 Bochum, Germany

‡ School of Physics, University of Bath, Claverton Down, Bath BA2 7AY, UK

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**Abstract.** The temperature dependences of the elastic constants of the antiferromagnetic Invar alloys $Fe_{60}Mn_{40}$ and $Co_{46}Mn_{54}$ have been investigated between 4.2 K and 800 K. Anomalies in the temperature behaviour of the sound velocities have been found in the vicinity of the magnetic phase transition indicating a strong magnetoelastic coupling in these antiferromagnetic Invar systems. The Landau theory has been adapted to the particularities of the systems under study for a phenomenological interpretation of the experimental data. Comparing our results with data obtained on ferromagnetic Invar alloys, we conclude that our phenomenological model is able to describe consistently the elastic anomalies in ferromagnetic and antiferromagnetic Invar systems.

## 1. Introduction

Since its discovery in 1897 [1] the Invar effect has been used in numerous applications. For example the anomalous behaviour of the thermal expansion coefficient of Invar systems in the vicinity of their magnetic phase transition has been very useful for achieving dimensional stability in precision instruments [2]. However, the physical origin of the large magnetovolume effects in the Invar alloys has only recently been explained in terms of the electronic band structure of 3d transition metals and alloys [3, 4, 5]. A quantitative comparison of the theory with experimental data is nevertheless extremely difficult due to the impracticability of including in a microscopic theory all the relevant parameters influencing the actual physical properties.

Strong magnetovolume effects responsible for the Invar anomalies are observed in ferromagnetic as well as in antiferromagnetic 3d transition metal alloys when the electron/atom ratio reaches certain critical values, i.e. $e/a = 8.6$ and $e/a = 7.6$ [6]. Whereas the temperature behaviours of the elastic constants of the ferromagnetic Invar alloys have been studied quite extensively [7, 8, 9, 10, 11, 12], their antiferromagnetic counterparts have received much less attention. Both Lenkkeri [13] and Renaud [9] investigated an Fe–Mn alloy with a composition of approximately $Fe_{60}Mn_{40}$, and they both found a positive magnetic contribution to the longitudinal elastic constants. This is in contrast to experimental results for ferromagnetic Invar alloys where a substantial softening of all elastic constants below the normal Debye behaviour has been observed.

At first sight these results on Invar effects in ferromagnets and antiferromagnets seemed to defy unified explanation. Recently, however, the range of elastic measurements has been extended to higher and lower temperatures in a sample of similar composition, $Fe_{60}Mn_{40}$

  

[14], and it has been found that the discrepancy was due to a previous invalid extrapolation of the high-temperature data.

Invar anomalies of the thermal expansion also occur in the Co–Mn alloy system. For a composition between about 30 and 55 at.% Mn these alloys crystallize in a disordered face-centred cubic structure [15]. The magnetic structure changes from ferromagnetic to antiferromagnetic with increasing manganese concentration at approximately 35 at.% Mn. Close to this critical concentration mixed magnetic behaviour is observed [15]. Both ferromagnetic and antiferromagnetic CoMn alloys show magnetovolume anomalies, although these are smaller than in the classical ferromagnetic Invar substance $Fe_{65}Ni_{35}$. We have started an investigation of the elastic behaviour of CoMn alloys because of the possibility of studying the effects on the elastic constants of the transition from ferromagnetic to antiferromagnetic order in a binary alloy system.

In this work we have investigated the elastic behaviour of an antiferromagnetic single crystal of $Co_{46}Mn_{54}$ between 4.2 K and 800 K. The experimental results are compared with measurements of the elastic constants of antiferromagnetic $Fe_{60}Mn_{40}$ which have been published previously [14].

In order to obtain a phenomenological interpretation of the experimental data, the Landau theory of second-order phase transitions has been applied to the systems under study. The symmetry of these systems leads to some conclusions concerning the size of magnetoelastic effects, which are confirmed by experimental observations in Invar samples. The parameters of this Landau approach may serve as a basis for a more quantitative understanding of the microscopic basis of the Invar effect.

In the next section the experimental set-up is described and the results obtained for $Co_{46}Mn_{54}$ are shown. They are compared to the temperature dependence of the elastic constants of $Fe_{60}Mn_{40}$. In section 3 Landau's theory of second-order phase transitions is adapted to the case of Invar systems. In section 4 the theory is compared to the experimental data on ferromagnetic and antiferromagnetic Invar alloys.

## 2. Experimental and results

Single crystals of the samples investigated have been grown from high-purity elements by a modified Bridgman–Stockbarger technique. The crystals were of a cylindrical shape with a diameter of 8 mm and a length of 7 mm. These dimensions are large enough for the determination of sound velocity from the transit time of ultrasonic pulses across the sample.

The characteristics of the samples are compared in table 1 to those of some ferromagnetic Invar alloys which were investigated previously [11, 12]. The compositions were determined by microprobe analysis to be $Fe_{60}Mn_{40}$ and $Co_{46}Mn_{54}$, within an experimental accuracy of 1 at.%. The lattice parameter was measured by x-ray diffraction. The Néel temperatures of the samples were determined from temperature-dependent measurements of the electrical resistivity $R$ of a small piece cut from the sample; they were identified as the respective temperature of an anomaly of the derivative $(1/R_0)(dR/dT)$. The Debye temperature $\theta_D$ was determined from the measured elastic constants after [16].

The crystals were oriented on a three-arc goniometer to $\pm 0.5°$ using x-ray Laue back-reflection photography. Samples for the ultrasonic experiment were cut and polished with two faces normal to the [110] direction. Both faces were flat to surface irregularities of about 3 $\mu$m and parallel to better than $10^{-3}$ rad.

Ultrasonic wave velocity measurements were made on the three independent acoustic modes propagating along the [110] direction. 10 MHz ultrasonic pulses were generated and detected by X-cut and AC-cut quartz transducers bonded to the samples using Apiezon N or

**Table 1.** Room temperature values of characteristic data for the two antiferromagnetic alloys $Fe_{60}Mn_{40}$ and $Co_{46}Mn_{54}$ investigated in this work compared to those of the ferromagnetic Invar alloys $Fe_{75}Pt_{25}$ and $Fe_{65}Ni_{35}$ taken from [27].

| | $Fe_{60}Mn_{40}$ | $Co_{46}Mn_{54}$ | $Fe_{75}Pt_{25}$ | $Fe_{65}Ni_{35}$ |
|---|---|---|---|---|
| Density (kg $m^{-3}$) | 7820 | 7870 | 11 620 | 8121 |
| Lattice constant (nm) | 0.3614 | 0.3626 | 0.3719 | 0.3593 |
| Elastic constants (GPa) : | | | | |
| $C_{11}$ | 170 | 158 | 115 | 136 |
| $C_{12}$ | 98 | 81 | 63 | 92 |
| $C_{44}$ | 141 | 130 | 94 | 98 |
| $C' = \frac{1}{2}(C_{11} - C_{12})$ | 36 | 38 | 26 | 22 |
| $B^S = \frac{1}{3}(C_{11} + 2C_{12})$ | 123 | 107 | 80 | 107 |
| $T_N$ (K) | 467 | 425 | — | — |
| $T_C$ (K) | — | — | 363 | 500 |
| Debye temperature $\theta_D$ (K) | 514 | 487 | 348 | 430 |

UHU-Plus for the low-temperature measurements and Krautkrämer ZGM high-temperature coupling paste above room temperature. The transit times of the ultrasonic pulses were determined using the pulse–echo overlap technique [17], which is capable of a resolution of velocity changes to 1 part in $10^5$. The absolute accuracy of the measured elastic constants is approximately 3%.

The temperature dependence of the velocities of the three independent sound waves along the [110] direction was measured between 4.2 K and a maximum temperature of 800 K at which the bonding between sample and transducer started to dissolve. The maximum temperature was well above the respective Néel temperatures of both samples, so an extrapolation of the non-magnetic background behaviour of the sound velocities was possible. Temperature-induced changes of sample dimension and density were taken into account using thermal expansion data on alloys of similar concentration taken from [18, 19].

The results of the temperature-dependent measurements on both the $Fe_{60}Mn_{40}$ and the $Co_{46}Mn_{54}$ alloy are shown in figures 1 to 3. Each figure depicts one of the elastic constants $C_{IJ}$ determined from the sound velocity of an independent mode propagating along the [110] direction determined as $C_{IJ} = \rho v^2$. Then $C_L = \frac{1}{2}(C_{11} + C_{12} + 2C_{44})$ corresponds to the longitudinal mode, and the transverse modes polarized in the [001] and $[1\bar{1}0]$ directions supply two other combinations— $C_{44}$ and $C'$ $(= \frac{1}{2}(C_{11} - C_{12}))$, respectively. The Néel points of the samples are indicated by the arrows.

In the paramagnetic range all the elastic stiffnesses decrease approximately linearly with increasing temperature. This is the normal temperature behaviour observed in many different classes of substances which can be successfully described in the quasiharmonic approximation of lattice dynamics using the Debye model [20]. The magnetic contribution to the elastic constants is obtained by subtracting the measured data from the normal contribution deduced from an extrapolation of the high-temperature behaviour. The function describing the lattice contribution in the Debye model is given by

$$C(T) = C(T = 0) - 3K\theta_D F\left(\frac{T}{\theta_D}\right) \tag{1}$$

with

$$F\left(\frac{T}{\theta_D}\right) = \frac{1}{8} - \left(\frac{T}{\theta_D}\right)^4 \int_0^{\theta_D/T} \frac{x^3\,dx}{e^x - 1}.$$

The Debye temperature $\theta_D$ has been estimated by linear extrapolation of the high-temperature behaviour of the elastic constants to 0 K and using these values to compute $\theta_D$ according to [16]. Using this value of $\theta_D$ the function given above has been fitted to the high-temperature part of the data ($T > 600$ K). The results of this procedure are displayed as continuous lines in the figures. The magnetic contribution to the elastic stiffnesses is the difference between the continuous line and the points obtained experimentally.

In the vicinity of the Néel temperature all elastic constants are influenced by the onset of magnetic order. $Fe_{60}Mn_{40}$ and $Co_{46}Mn_{54}$ behave in a similar way showing a reduction of $C_L$ and $C'$ and a change of slope to a higher value for $C_{44}$. In accordance with the thermal expansion data which reveal a smaller Invar effect for antiferromagnetic CoMn alloys than for FeMn [3], the magnetic influence on the elastic behaviour is smaller in the Co alloy.

The results are also in accord with the previous measurements on alloys of composition $Fe_{60}Mn_{40}$ [9, 13]. However, these investigations were conducted only from room temperature up to 600 K where the antiferromagnetic transition has already affected the elastic constants. Extrapolation of normal Debye behaviour from this region led to a smaller background elastic constant, so those authors reported a positive magnetic contribution to the longitudinal mode. Inspection of figure 1 clearly indicates that a more reliable estimate of the background behaviour requires measurements at higher temperatures. Extrapolation of the data taken at temperatures above $T_N + 150$ K shows that the magnetic contribution to the longitudinal elastic constant $C_L$ is always negative in accord with observations made on ferromagnetic Invar alloys [12].

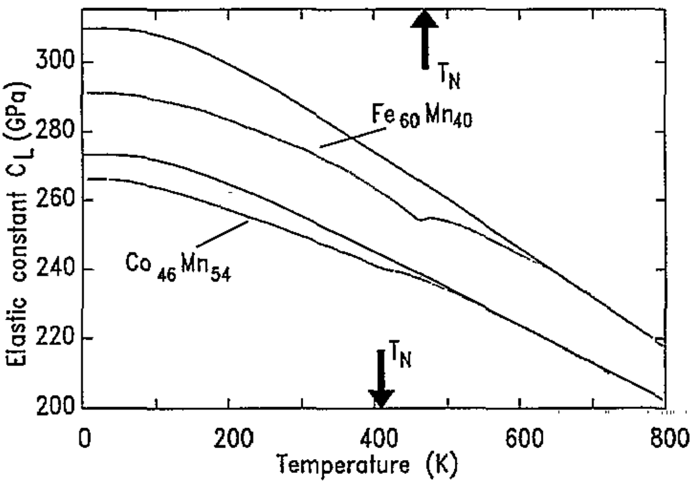


**Figure 1.** Temperature dependence of the elastic constant $C_L = \frac{1}{2}(C_{11} + C_{12} + 2C_{44})$ corresponding to the longitudinal mode propagating along [110] for $Fe_{60}Mn_{40}$ and $Co_{46}Mn_{54}$. The arrows mark the respective Néel temperatures. The continuous lines represent the normal quasiharmonic behaviour as deduced from the high temperature limit of $C_L$.

The transverse elastic constants $C'$ and $C_{44}$ show a behaviour markedly different to that of ferromagnetic Invar alloys. In the ferromagnetic crystals large anomalies are observed for $C'$ as well as for $C_{44}$ starting at the Curie point $T_C$ and showing a similar temperature

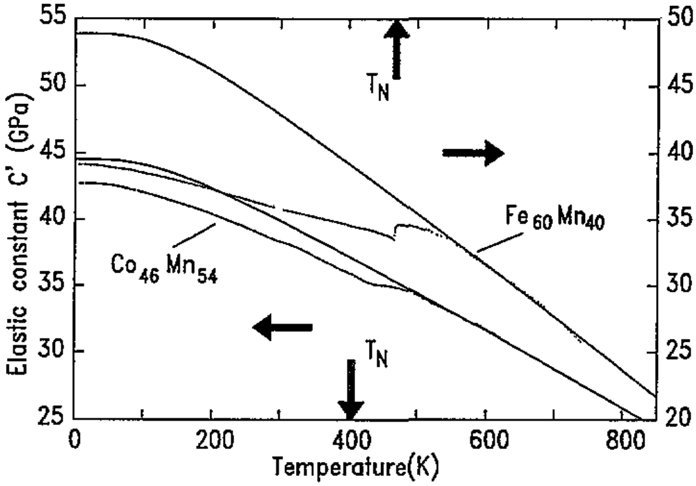


**Figure 2.** Temperature dependence of the elastic constant $C' = \frac{1}{2}(C_{11} - C_{12})$ corresponding to the $[1\bar{1}0]$-polarized transverse mode propagating along [110] for $Fe_{60}Mn_{40}$ and $Co_{46}Mn_{54}$. The arrows mark the respective Néel temperatures. The continuous lines represent the normal quasiharmonic behaviour as deduced from the high-temperature limit of $C'$.

dependence. The magnetic contribution to the elastic constants is negative for both modes and increases nearly linearly with decreasing temperature. In the antiferromagnetic samples there are only small elastic anomalies at $T_N$. The magnetic contribution to $C_{44}$ is positive (see figure 3) and only results in a change of slope at $T_N$. The magnetic part of $C'$ is negative, and in $Fe_{60}Mn_{40}$ there is even a small downward jump at $T_N$. The significance of these differences between ferromagnetic and antiferromagnetic Invar alloys will be discussed in section 4.

## 3. Theory

Recent theories of the Invar effect explain the magnetovolume anomalies in terms of an instabilitity of the magnetic moment of 3d transition metals [3]. The conclusions are based on band-structure calculations using the fixed-spin-moment procedure [4, 5]. Up to now, however, it was not possible to perform these calculations for systems which were studied experimentally taking into account the marked effects of alloy disorder.

In order to obtain a theoretical interpretation of our experiments in spite of these difficulties, we have adapted the phenomenological Landau theory of phase transitions to the alloys under study. The parameters of this expansion determined from experiment could serve as a suitable starting point for comparing experimental data to more refined microscopic theories.

In Landau's theory the free-energy density of the system is expanded in terms of the

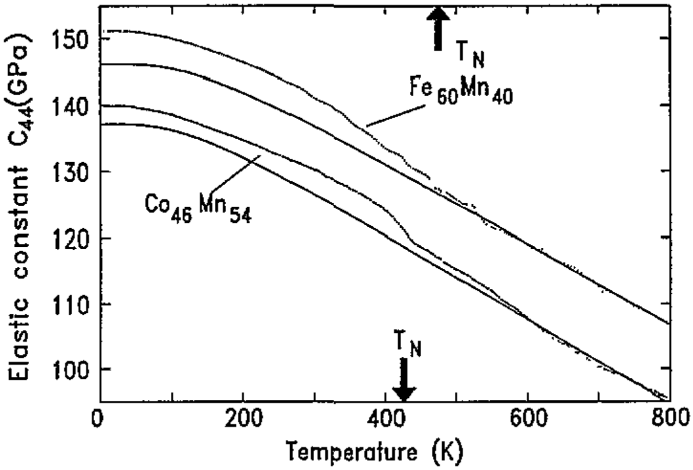


**Figure 3.** Temperature dependence of the elastic constant $C_{44}$ corresponding to the [001]-polarized transverse mode propagating along [110] for $Fe_{60}Mn_{40}$ and $Co_{46}Mn_{54}$. The arrows mark the respective Néel temperatures. The continuous lines represent the normal quasiharmonic behaviour as deduced from the high-temperature limit of $C_{44}$.

three-component order parameter $\boldsymbol{Q}$

$$f = \frac{1}{2}aQ^2 + \frac{1}{4}b_1Q^4 + \frac{1}{2}b_2\sum_{i<j=1}^{3} Q_i^2Q_j^2 + \frac{1}{2}\sum_{\Gamma} C_\Gamma\varepsilon_\Gamma^2 + \sum_{\Gamma} g_\Gamma\varepsilon_\Gamma\, Q_\Gamma^2 \qquad (2)$$

where $Q_\Gamma^2$ is the symmetry-adapted square of the order parameter components and the $\varepsilon_\Gamma$ are the symmetry-adapted components of the strain tensor corresponding to an irreducible representation $\Gamma$ of the cubic point group $m3m$:

$$Q_A^2 = Q_1^2 + Q_2^2 + Q_3^2$$
$$Q_E^2 = \frac{1}{\sqrt{3}}\begin{pmatrix} 2Q_3^2 - Q_1^2 - Q_2^2 \\ \sqrt{3}(Q_1^2 - Q_2^2) \end{pmatrix}$$
$$Q_T^2 = \begin{pmatrix} Q_2Q_3 \\ Q_1Q_3 \\ Q_1Q_2 \end{pmatrix}.$$

The symbols A, E, and T characterize the irreducible representations $A_{1g}$, $E_g$, and $T_{2g}$. In ferromagnetic systems the order parameter $\boldsymbol{Q}$ can be identified with the magnetization vector $\boldsymbol{M}$. In antiferromagnetic systems $\boldsymbol{Q}$ corresponds to the antiferromagnetic vector $\boldsymbol{L}$ which is a linear combination of the components of the sublattice magnetization vectors [21].

The first three terms on the right-hand side of (2) describe the magnetic part of the free-energy density, the next term is the elastic energy density and the last term represents the lowest-order magnetoelastic coupling responsible for the Invar anomalies. A bilinear coupling of the strain and order parameter is forbidden by symmetry.

Following the usual procedure of Landau's theory the free-energy density is minimized with respect to strain and order parameter and the following conclusions can be drawn [21].

(i) At the phase transition a spontaneous strain develops according to

$$\varepsilon_\Gamma^{\mathrm{sp}} = \frac{g_\Gamma}{C_\Gamma} Q_\Gamma^2. \tag{3}$$

For $\Gamma = \mathrm{A_{1g}}$ the spontaneous strain corresponds to the spontaneous volume magnetostriction. The large value of this quantity observed in Invar alloys points towards a large value of the coupling parameter $g_\mathrm{A}$. Depending on the symmetry of the order parameter a non-zero value of one of the other coupling constants would lead to a symmetry-breaking strain at the phase transition which has never been observed in Invar alloys. Therefore we assume that there is no significant contribution of this coupling mechanism to the transverse elastic constants. To explain the observed anomalies of the transverse elastic constants a higher-order coupling between order parameter and strain has to be taken into account [22] which gives no contribution to a spontaneous strain. The magnitude of this higher-order coupling may be different for different Invar systems without affecting the Invar behaviour. This could account for the differences in the behaviour of the transverse modes between ferromagnetic and antiferromagnetic Invar systems.

(ii) The spontaneous magnetization developing below $T_\mathrm{C}$ is modified by the magnetoelastic coupling. Assuming $g_\mathrm{E}, g_\mathrm{T} \ll g_\mathrm{A}$ we have:

$$Q^2 = -\frac{a - 2g_\mathrm{A}\varepsilon_\mathrm{A}}{f(b_1, b_2)} \tag{4}$$

where $f$ is a linear combination of the magnetic parameters $b_1$ and $b_2$ and depends on the symmetry of the order parameter. $\varepsilon_\mathrm{A}$ corresponds to the volume strain $\Delta V/V$. The change of the temperature dependence of $Q^2$ induced by a non-zero value of $g_\mathrm{A}$ may be absorbed in the transition temperature because $T_\mathrm{C}$ is defined as the temperature where $Q^2$ goes to zero. Thus one obtains a volume-dependent, and therefore also pressure-dependent, phase transition temperature $T_\mathrm{C}$:

$$T_\mathrm{C}(\varepsilon_\mathrm{A}) = T_\mathrm{C}(0) + \frac{2g_\mathrm{A}\varepsilon_\mathrm{A}}{\alpha}$$

and therefore

$$\frac{\mathrm{d}T_\mathrm{C}}{\mathrm{d}p} = -\frac{2g_\mathrm{A}}{\alpha B^S}. \tag{5}$$

$\alpha$ is defined on the usual assumption of Landau's theory that $a = \alpha(T - T_\mathrm{C})$.

(ii) The magnetoelastic coupling leads to a jump of the corresponding elastic constant at the transition point. For a coupling to the volume strain one obtains for the bulk modulus

$$B^S(T < T_\mathrm{C}) = B^S(T > T_\mathrm{C}) - \frac{2g_\mathrm{A}^2}{f(b_1, b_2)} \tag{6}$$

where $f$ is the same combination as in (4). A large value of the coupling constant $g_\mathrm{A}$ typical of Invar alloys (see above) leads to a large jump of the bulk modulus. For the shear constants $C_\mathrm{E} = C'$ and $C_{44}$ no jump should be observed due to the small values of $g_\mathrm{E}$ and $g_\mathrm{T}$ required to conserve symmetry. The small jump of $C'$ observed in $\mathrm{Fe_{60}Mn_{40}}$ does not contradict this argument because the symmetry of the order parameter in this system corresponds to the irreducible representation $\mathrm{T_{2g}}$. In this case the spontaneous strain induced by a non-zero value of $g_\mathrm{E}$ is zero because the symmetry-adapted square of the order parameter components

$Q_E^2$ is zero for an order parameter of symmetry $T_{2g}$. Combining equations (4), (5), and (6) we obtain

$$g_A = \frac{\Delta B^S/B^S}{(\mathrm{d}Q^2/\mathrm{d}T)(\mathrm{d}T_C/\mathrm{d}p)}. \tag{7}$$

Accordingly the coupling parameter $g_A$ may be computed from the experimental values of the temperature dependence of the order parameter, the pressure dependence of the transition temperature, and the jump of the bulk modulus.

From the parameters of Landau's theory determined from experiment using equations (4), (5), and (6) the spontaneous strain responsible for the Invar anomaly of thermal expansion may also be computed using (3). If the experimental temperature dependence of the spontaneous strain below $T_C$ is known, this procedure provides a consistency check of the experimental data in the framework of Landau's theory.

## 4. Discussion

A comparison of our experimental results on antiferromagnetic Invar alloys with already published results on ferromagnetic Invars [12] leads to different conclusions about the behaviours of the longitudinal and transverse modes.

The effects of temperature on the longitudinal elastic constants and of the bulk modulus are similar in both cases. Starting at temperatures high above the temperature of the magnetic transition the elastic constants begin to drop below the extrapolated normal Debye behaviour already 100–200 K above $T_N$. In all cases this softening resembles a jump to lower values which is broadened by fluctuations. The magnitude of this jump is much larger in the ferromagnetic alloys in accord with the larger Invar anomaly of the thermal expansion observed in these systems.

Concerning the transverse modes, there are distinct differences between the ferromagnetically and the antiferromagnetically ordered alloys. In the ferromagnetic Invars the transverse elastic constants $C'$ and $C_{44}$ which are independent of each other behave in a similar way. Both start softening at the Curie point and the deviation from the normal temperature dependence increases linearly with decreasing temperature. This behaviour is in good accord with predictions of Landau's theory for a second-order magnetoelastic coupling [22]. In the antiferromagnets a jump to a lower value is observed at $T_N$ for $C'$ whereas $C_{44}$ exhibits no such discontinuity at $T_N$ but its value increases in the magnetically ordered phase (figures 2 and 3). This kind of temperature dependence has also been observed in MnNi [23], another antiferromagnetic Invar system.

The differences observed for the transverse modes in the ferromagnetic and antiferromagnetic Invar alloys prove that the magnetic anomalies of the transverse elastic constants are not connected directly with the Invar effect. They are the results of higher-order magnetoelastic coupling and their characteristics depend on the specific magnetic structure of the sample.

By contrast the temperature behaviours of the bulk moduli in the ferro- and antiferromagnetic Invar alloys are very similar. The softening in the two kinds of system is governed by the coupling described in (2) which is also responsible for the Invar anomalies of the thermal expansion. From the experimental data the coupling parameter $g_A$ has been determined by the procedure described at the end of the last section. The results are displayed in table 2 where the parameters obtained for our antiferromagnetic sample $Fe_{60}Mn_{40}$ are compared to those of ferromagnetic Invars. For our $Co_{46}Mn_{54}$ sample no experimental data are available yet on the pressure dependence of the Néel temperature and on the temperature dependence of the order parameter.

**Table 2.** Magnetic parameters of ferromagnetic and antiferromagnetic Invar alloys as determined from experiment. The magnetoelastic coupling constant $g_A$ was calculated according to (7) using the given values of the pressure dependence of the transition temperature $dT_C/dp$, the temperature dependence of the order parameter $dQ^2/dT$ and the jump of the bulk modulus at the magnetic transition $\Delta B^S$.

| | $Fe_{60}Mn_{40}$ | $Fe_{75}Pt_{25}$ | $Fe_{65}Ni_{35}$ |
|---|---|---|---|
| $dQ^2/dT$ ((A m$^2$ kg$^{-1}$)$^2$ K$^{-1}$) | 49.4 [28] | 57.4 [24] | 74.5 [29] |
| $dT_C/dp$ (K GPa$^{-1}$) | −25.0 [30] | −57.0 [3] | −40.0 [3] |
| $\Delta B^S$ (GPa) | −8.4 | −110 [22] | −54 [9] |
| $g_A$ (MPa kg$^2$ (A$^2$ m$^4$)$^{-1}$) | 0.055 | 0.185 | 0.114 |

It is obvious that all the coupling parameters have the same order of magnitude but the $g_A$-values of the ferromagnetic alloys are larger that of $Fe_{60}Mn_{40}$. This is in accord with the observation that the thermal expansion anomalies are also larger in ferromagnetic Invars. From the parameters given in table 2 the spontaneous strain below the transition temperature was computed using (3) and compared to experimental data for $Fe_{75}Pt_{25}$ [24] and $Fe_{65}Ni_{35}$ [25]. The satisfactory agreement between the data indicates that the observed behaviours of various magnetic and elastic properties can be described consistently in the framework of Landau's theory.

The value of $g_A$ which describes the magnetoelastic coupling might be a suitable parameter to use in comparisions with to more refined theories of the Invar effect, especially as there have already been attempts to link band-structure calculations to a Landau expansion of the free energy [26].

## 5. Conclusion

The temperature dependences of the elastic constants of the antiferromagnetic Invar alloys $Fe_{60}Mn_{40}$ and $Co_{46}Mn_{54}$ which have been analysed on the basis of the Landau theory show distinct similarities to the behaviour observed in the ferromagnetic Invar alloys. In both types of Invar alloy the bulk modulus and the elastic constants related to it show a decrease in the magnetically ordered state which has already started some hundreds of degrees above the ordering temperature. This softening of the bulk modulus is identified as a common feature of ferromagnetic and antiferromagnetic Invar alloys that can be explained consistently in the context of the Landau theory by the appearance of a spontaneous volume strain. In contrast, the shear elastic constants of $Fe_{60}Mn_{40}$ and $Co_{46}Mn_{54}$ display temperature dependences which can only be described assuming a non-linear coupling between strain and order parameter. As a consequence the shear elastic anomalies observed in ferromagnetic and antiferromagnetic Invar alloys are not a common property of the Invar state but are determined by the peculiarities of the exchange and magnetoelastic interactions in the magnetically ordered state of the material.

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## References


[1] Guillaume C E, 1897 *C. R. Acad. Sci., Paris* **125** 235
[2] Wassermann E F 1987 *Adv. Solid State Phys.* **27** 85
[3] Wassermann E F 1990 *Ferromagnetic Materials V* ed K H J Buschow and E P Wohlfarth (Amsterdam: Elsevier)
[4] Moruzzi V L 1990 *Phys. Rev.* B **41** 6939
[5] Podgorny M 1991 *Phys. Rev.* B **43** 11300
[6] Wassermann E F 1991 *Europhys. News* **22** 150
[7] Hausch G and Warlimont H 1973 *Acta Metall.* **21** 401
[8] Hausch G 1974 *J. Phys. Soc. Japan* **37** 819
[9] Renaud P 1988 *PhD Thesis* ETH, Lausanne
[10] Shiga M, Makita K, Uematsu K, Muraoka Y and Nakamura Y 1990 *J. Phys.: Condens. Matter* **2** 1239
[11] Mañosa Ll, Saunders G A, Rahdi H, Kawald U, Pelzl J and Bach H 1991 *J. Phys.: Condens. Matter* **3** 2773
[12] Mañosa Ll, Saunders G A, Rahdi H, Kawald U, Pelzl J and Bach H 1992 *Phys. Rev.* B **45** 2224
[13] Lenkkeri J T 1981 *J. Phys. F: Met. Phys.* **11** 1991
[14] Cankurtaran M, Saunders G A, Ray P, Wang Q, Kawald U, Pelzl J and Bach H 1993 *Phys. Rev.* B **47** 3161
[15] Men'shikov A Z, Takzeï G A, Dorofeev Y A, Kazantsev V A, Kostyshin A K and Sych I I 1985 *Sov. Phys.–JETP* **62** 734
[16] Alers G A 1965 *Physical Acoustics* vol 3B, ed W P Mason (New York: Academic)
[17] Papadakis E P 1967 *J. Acoust. Soc. Am.* **42** 1045
[18] Stamm W 1988 *PhD Thesis* Duisburg
[19] John C, Zähres H, Acet M, Stamm W, Wassermann E F and Pepperhoff W 1990 *J. Appl. Phys.* **67** 5268
[20] Ludwig W 1967 *Springer Tracts in Modern Physics* vol 43 (Berlin: Springer)
[21] Tolédano J C and Tolédano P 1987 *The Landau Theory of Phase Transitions* (Singapore: World Scientific)
[22] Kawald U, Schulenberg P, Bach H, Pelzl J, Eckold G and Saunders G A 1991 *J. Appl. Phys.* **70** 6537
[23] Hausch G, Schmolz A, Török E and Warlimont H 1983 *J. Physique Coll.* **12** C9 471
[24] Sumiyama K, Emoto Y, Shiga M and Nakamura Y 1981 *J. Phys. Soc. Japan* **50** 3296
[25] Graham G M 1972 *Phys. Lett.* **39A** 325
[26] Podgórny M, Thon M and Wagner D 1992 *J. Magn. Magn. Mater.* **104–107** 703
[27] Kawald U 1992 *PhD Thesis* Bochum
[28] Ishikawa Y and Endoh Y 1967 *J. Phys. Soc. Japan* **23** 205
[29] Yamada O, Ono F, Nakai I, Maruyama H, Arae F and Ohta K 1982 *Solid State Commun* **42** 473
[30] Fujimori H 1966 *J. Phys. Soc. Japan* **21** 1860