Premartensitic and martensitic phase transitions in ferromagnetic Ni_2MnGa

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We present an experimental study of the premartensitic and martensitic phase transitions in a Ni_2MnGa single crystal by using ultrasonic techniques. The effect of applied magnetic field and uniaxial compressive stress has been investigated. It has been found that they substantially modify the elastic and magnetic behavior of the alloy. These experimental findings are a consequence of magnetoelastic effects. The measured magnetic and vibrational behavior agrees with the predictions of a recently proposed Landau-type model [A. Planes *et al.*, Phys. Rev. Lett. **79**, 3926 (1997)] that incorporates a magnetoelastic coupling as a key ingredient. [S0163-1829(99)01834-2]

I. INTRODUCTION

An interesting feature of martensitic transitions in shape-memory alloys is the existence of precursor phenomena. They are a consequence of weak restoring forces in specific crystallographic directions that announce the possibility of a dynamical instability. Commonly, these systems have a low-lying transverse TA_2 phonon branch together with a low value of the corresponding elastic constant C'; both the whole branch and the elastic constant soften with decreasing temperature. Other pretransitional effects are diffuse elastic scattering and phonon anomalies on the low-lying branch at certain wave vectors that are close to the reciprocal lattice vector corresponding to the modulation of the low-temperature martensitic phase. The prototypical example where these anomalies have been extensively studied is the Ni-Al alloy.[1]

Precursor phenomena are expected in second-order phase transitions and are not observed in strongly first-order transitions. The martensitic transition is a first-order transition taking place before the complete softening of a response function; that is, before the system becomes harmonically unstable. It has been proposed[2,3] that this is possible due to the anharmonic coupling between a phonon on the transverse TA_2 branch and the long-wavelength shear mode related to C'. This picture has been shown to be suitable for qualitatively describing the martensitic transition in Cu-based alloys.[4]

Among the systems undergoing martensitic transitions, shape-memory alloys are highly attractive. Recently, there has been increased interest in the study of magnetic alloys exhibiting shape-memory properties.[5,6] The coupling between structural and magnetic degrees of freedom opens the possibility of a magnetic control of the shape-memory effect associated with the martensitic structure, which confers to these alloys a potential technological interest. In this paper we study the Ni-Mn-Ga alloy close to the Heusler Ni_2MnGa stoichiometric composition, which undergoes a martensitic transition in the ferromagnetic state. Recently, large magnetic-field-induced strains have been obtained in this system.[7,8]

From a fundamental point of view, peculiar pretransitional phenomena have been reported in Ni-Mn-Ga. Remarkably interesting is the fact that, in a certain composition range, this alloy exhibits a pronounced temperature softening of the $(\frac{1}{3}\frac{1}{3}0)$ phonon on the transverse TA_2 branch, which condensates at a temperature T_I, leading to the appearance of a micromodulated structure preceding the martensitic transition.[9] Such a microstructure has been studied by high-resolution transmission electron microscopy and has been shown to be the reason of the extra spots observed on the corresponding electron-diffraction pattern.[10] The wave vector associated with this modulation is different from that corresponding to the five- or seven-layer modulation, characteristic of the martensitic phases in this alloy system. The premartensitic (or intermediate) phase transition has been shown to be a first-order transition originated by the magnetoelastic coupling between the magnetization and the anomalous TA_2 phonon.[11,12]

In this paper, we present a detailed ultrasonic investigation of a Ni-Mn-Ga single crystal with composition very close to the stoichiometric one. We focus on the magnetoelastic properties of this alloy system.

II. EXPERIMENTAL RESULTS

The sample investigated was a single crystal with composition $Ni_{49.5}Mn_{25.4}Ga_{25.1}$ grown by the Bridgman technique. The single crystal was obtained by melting appropriate amounts of single crystals labeled 3 and 6 in Ref. 13; the estimated error in the composition is ± 0.5 at. %. From the original rod, a parallepipedical specimen $(6.75\times 11.45$

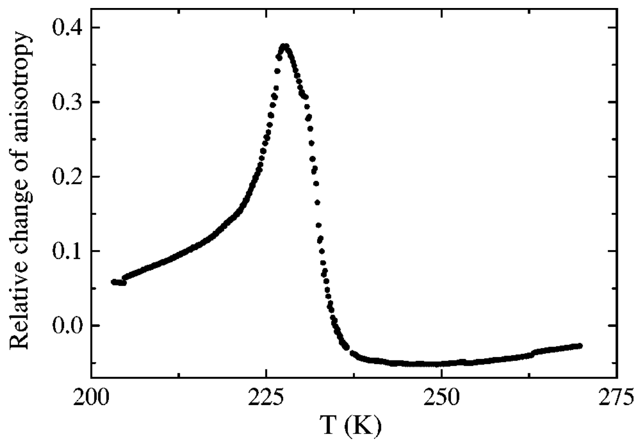

FIG. 1. Relative change of the elastic anisotropy ($A = C_{44}/C'$) as a function of temperature.

$\times 4.8$ mm^3) with faces parallel to the (001), (110), and ($1\bar{1}0$) planes was cut. In addition, two platelike small samples ($3.2\times1.4\times1.0$ mm^3) and ($2.8\times1.6\times0.7$ mm^3) were cut, with the longer direction along the [001] and [110] crystallographic axis, respectively, which were used for magnetization measurements.

The crystal exhibits an ordered $L2_1$ Heusler structure (space group $Fm3m$) at room temperature. For the investigated composition, the Curie point is at $T_c = 381$ K; the intermediate transition takes place at $T_I = 230$ K, and the martensitic start temperature is $T_M = 175$ K.

The velocity of ultrasonic waves was determined by the pulse-echo method, using the phase-sensitive technique. *X*-cut and *Y*-cut quartz transducers with resonant frequencies of 10 MHz were acoustically coupled to the surface of the sample by means of Dow resin 276-V9 in the temperature range 270–320 K, and by Nonaq stopcock grease in the temperature range 200–270 K. The room temperature values found for C_L (=229 GPa) and C_{44} (=102 GPa) are close to those reported for a Ni-Mn-Ga crystal with a slightly different composition.[14] The value found for C' (=22 GPa) is larger in our sample. It is important to point out that ultrasonic waves propagating along the [110] direction with [$1\bar{1}0$] polarization are affected by strong attenuation arising from magnetic domain scattering of ultrasonic waves. As a consequence, determination of the actual velocity for these waves is rather difficult. Such a difficulty is reflected by the difference in the values found from neutron data ($v = 1000$ m/s) (Ref. 15) and ultrasonic measurements ($v = 740$ m/s) (Ref. 14) performed on exactly the same sample by different authors. The value found for our crystal ($v = 1600$ m/s) is slightly larger, and the difference is likely to be due to the different composition between the two samples.

The two shear elastic constants C_{44} and C' show significant softening at the intermediate phase transition.[11,14,16] The softening of C' is more pronounced than that of C_{44}, thus resulting in a relative increase of the elastic anisotropy at the intermediate phase transition, as illustrated in Fig. 1. Below the intermediate phase transition C' increases on cooling. This relative increase in C' is larger than that of C_{44} and the elastic anisotropy decreases as the sample approaches the martensitic transformation (see Fig. 1). Such a behavior is contrary to the one exhibited by other shape-memory alloys.[17]

With the purpose of investigating the interdependence between elastic and magnetic properties, we have measured the magnetic-field dependence of the elastic constants. For these measurements, the sample was placed between the poles of an electromagnet, and magnetic fields up to 10 kOe were applied along the [110] and [001] directions. Prior to each magnetic-field scan, the sample was annealed for 45 min at 520 K (well above the Curie temperature). Such a heat treatment ensured that the measured dependence of each elastic constant corresponded to the first magnetization process. The results obtained are shown in Fig. 2, where solid and open symbols stand for increasing and decreasing magnetic fields, respectively. All elastic constants increase up to a saturation value with increasing magnetic field. C' is the constant that exhibits the largest relative change. The saturation values found for fields applied along [001] and [110] are, within the experimental error, coincident. Notice that the value achieved by the elastic constants when the field is removed is slightly larger than the initial one. This irreversible effect is a consequence of the magnetic remanence.

It is also instructive to look at the magnetic-field dependence of the ultrasonic attenuation. It is clear from Fig. 2 that, for all acoustic modes, the ultrasonic attenuation decreases with increasing magnetic field.[18] The attenuation of ultrasonic waves in ferromagnetic materials is mostly due to the scattering produced by magnetic domains. Present results show that scattering of ultrasonic waves is smaller when magnetic domains are aligned along the same direction. For the modes corresponding to C_L and C_{44}, the relative decrease is similar for magnetic fields along [001] and [110] directions. For C', the relative decrease for the magnetic field in [110] direction is larger than for the other modes.

Finally, we have also investigated the dependence of elastic constants upon uniaxial compressive stress. For these measurements, the sample was placed inside a universal tensile machine equipped with compression grips. The machine was equipped with a cryofurnace, which enabled us to conduct measurements at different temperatures. As an example, in Fig. 3 we present the room-temperature dependence found for the three independent elastic constants for uniaxial compressive stresses applied along the [001] direction. All elastic constants increase with increasing stress, indicating an overall stiffening of the crystal. The increase is clearly nonlinear and seems to reach a saturation value. The relative change in the elastic constants is similar for all of them. The amount of change in C' is of the order of that measured in other bcc alloys; however, the change in C_L and C_{44} is about one or two orders of magnitude larger than the typical changes reported for other bcc alloys.[19]

The combined temperature and stress dependence of ultrasonic waves provides a convenient way of investigating the stress dependence of the premartensitic transition. With this aim, we have measured the temperature dependence of the shear elastic constant C_{44} at different levels of applied uniaxial stress along the [001] and [$1\bar{1}0$] directions. We have used this shear elastic constant instead of C' because of the poor quality of the ultrasonic echoes for the waves asso-

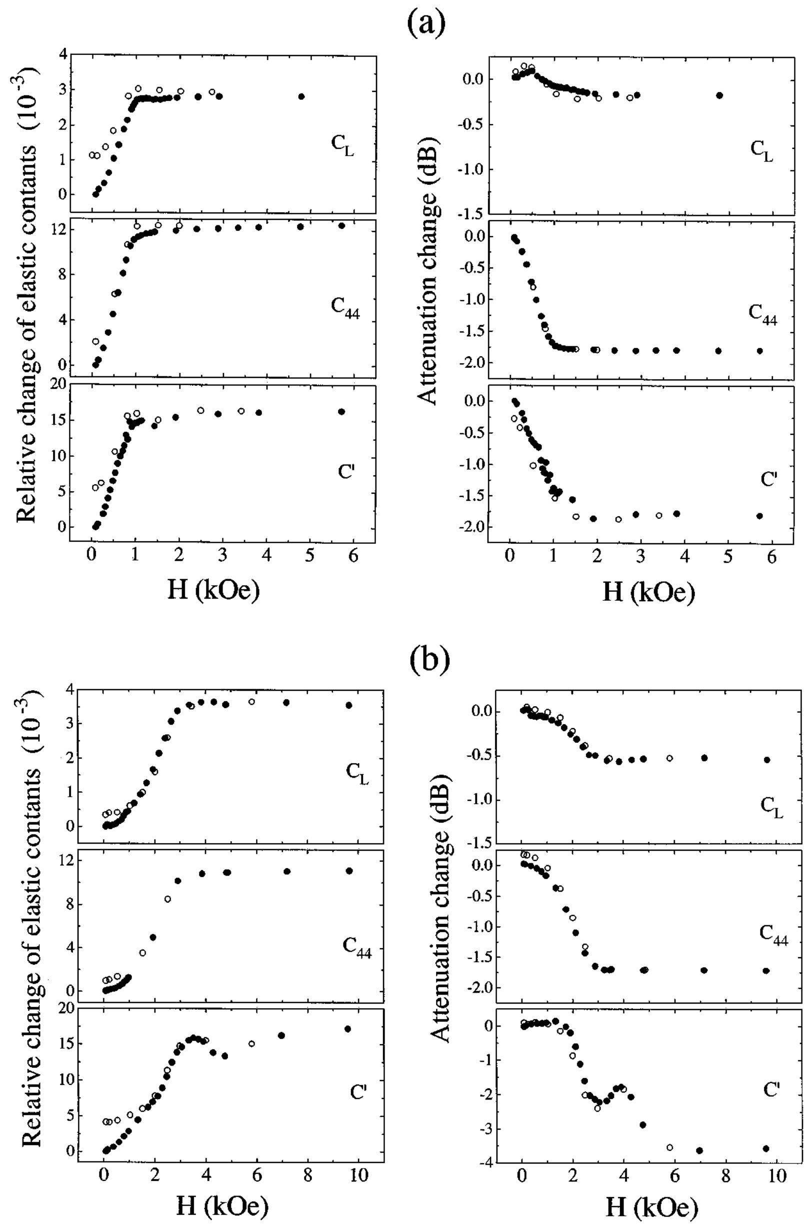

FIG. 2. Relative change in the elastic constants and change in the corresponding relative ultrasonic attenuation, for dc magnetic fields applied along the [001] (a), and [110] (b) directions. Solid symbols correspond to increasing magnetic field and open symbols, to decreasing magnetic field.

ciated with this elastic constant, which in some cases made it difficult to clearly define the position of the minimum in the C' vs T curve. In Fig. 4 we show an example of the results found during heating runs at stresses of 0 MPa (circles), 1 MPa (squares), and 4.5 MPa (triangles), applied along the $[1\bar{1}0]$ direction. The temperature T_I of the minimum of the C_{44} vs T curves for different stress levels along the [001] and $[1\bar{1}0]$ directions is plotted in Fig. 5. It is clear that the application of a compressive stress decreases the temperature of the forward transition and increases that of the reverse one. Therefore, the premartensitic transition under applied stress occurs with thermal hysteresis. The change in the forward and reverse intermediate transition temperatures seems to reach a saturation value for stresses above 3–4 MPa (in the case of a stress along the [001] direction the saturation is not observed for the forward transition because at higher stress levels there was a significant distortion of ultrasonic echoes).

III. DISCUSSION

It has been customary to compare the lattice dynamical behavior of Ni_2MnGa to that exhibited by the Ni-Al shape-memory alloy. In both alloys pronounced temperature softening of TA_2 phonons [close to $(\frac{1}{3}\frac{1}{3}0)$ for Ni_2MnGa and to $(\frac{1}{6}\frac{1}{6}0)$ for Ni-Al (Ref. 20)], accompanied by a ''central peak'' has been reported.[1,9] In spite of these similarities, the premartensitic behavior of Ni_2MnGa turns out to be quite different. The important differences are (i) the phonon softening in Ni_2MnGa is more pronounced; and (ii) for Ni-Al, all elastic constants stiffen with reducing temperature, with the exception of C', which decreases monotonously down to the martensitic transition temperature.[21] As a result, in Ni-Al the elastic anisotropy increases monotonously with reducing temperature (approaching the martensitic transition). This long-wavelength acoustic-mode behavior is completely different from that exhibited by Ni_2MnGa (see Fig. 1). The origin of this unique vibrational behavior of Ni_2MnGa lies on

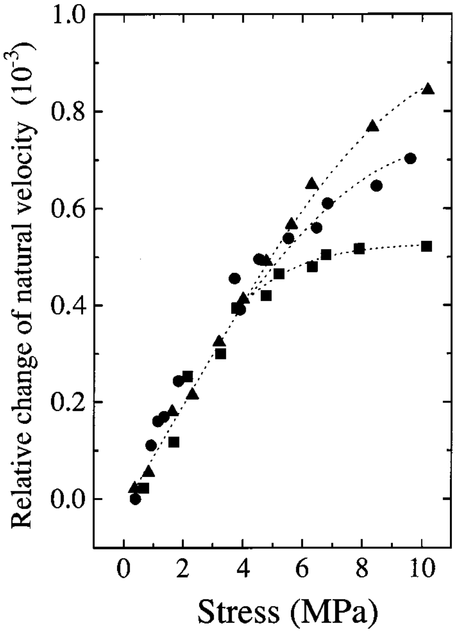

FIG. 3. Relative change in the natural velocity of ultrasonic waves associated with the elastic constants C_L (triangles), C_{44} (squares), and C' (circles), for uniaxial stresses applied along the [001] direction.

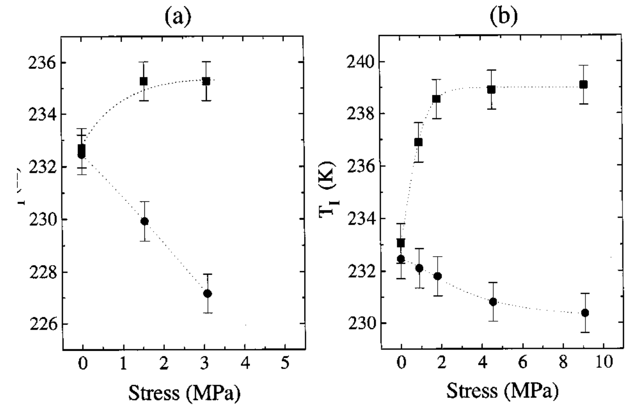

FIG. 5. Change in the intermediate transition temperatures during cooling (circles) and heating (squares) runs, for stresses applied along the [001] (a), and $[1\bar{1}0]$ (b) directions. Dashed lines are guides to the eye.

the ferromagnetic order exhibited by this alloy.[11] Such a possibility was first ruled out by Zheludev *et al.*[9,15] These authors based their assertion on the fact that they observed a wiggle on the TA_2 phonon branch at $\xi=\frac{1}{3}$ at a temperature slightly above the Curie point. However, very recently, Stuhr *et al.*[22] have performed neutron scattering experiments over a broad temperature range covering the ferromagnetic and paramagnetic phases of a $Ni_{51.5}Mn_{23.6}Ga_{24.9}$ crystal (this crystal did not exhibit any transition to the intermediate phase). They have found that the temperature dependence of the energy of the soft phonon changes significantly at the Curie point. Such a change indicates that the phonon softening depends on the magnetic ordering in the sample. Interestingly, the phonon softening in the paramagnetic phase

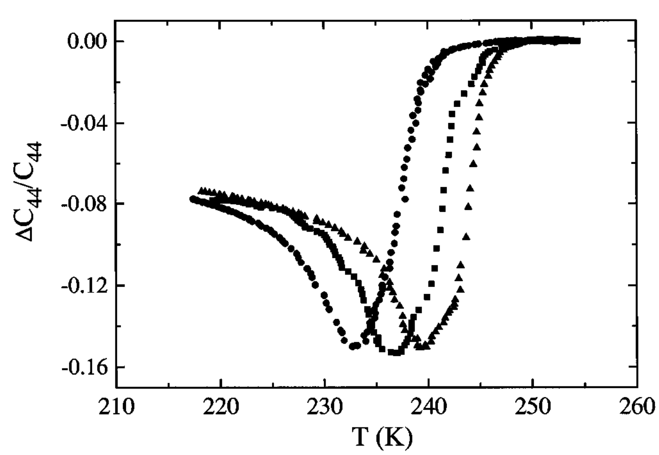

FIG. 4. Relative change of the shear elastic constant C_{44} with temperature (heating run), for applied uniaxial stresses of 0 (circles), 1 MPa (squares), and 4.5 MPa (triangles), applied along the $[1\bar{1}0]$ direction.

($\simeq 0.019\ \mathrm{meV^2/K}$) is similar to that of Ni-Al ($\simeq 0.016\ \mathrm{meV^2/K}$).[1] This result shows that when the sample becomes magnetically ordered, the softening is enhanced as a consequence of the interaction between the magnetization and the phonon energy.

For Ni-Mn-Ga alloys close to the stoichiometric composition, the soft phonon can freeze at a given temperature (T_I). This freezing gives rise to the development through a first-order phase transition of a micromodulated structure that can easily be detected by the narrowing in the peak width and a remarkable increase in the integrated intensity of the diffraction peaks at $(\frac{1}{3}\frac{1}{3}0)$. Below T_I, the energy of the $(\frac{1}{3}\frac{1}{3}0)$ TA_2 phonon increases with further decreasing temperature.[9] A recent Landau-type model has shown[12] that the occurrence of this first-order phase transition must be ascribed to the existence of a magnetoelastic coupling. Notwithstanding, the transition from the $L2_1$ structure towards the micromodulated structure has not been observed in all Ni-Mn-Ga samples investigated by different authors. In order to clarify this point it is interesting to collect data for the different transition temperatures from the literature for different samples with compositions around the stoichiometric one. We have observed that in the range of compositions close to the stoichiometric (hatched region in the inset of Fig. 6), these data can be compiled in a compact representation by plotting the different transition temperatures as functions of a parameter (α) obtained as a weighted composition ($\alpha = x$ at. % Ga$+y$ at. % Mn composition, with $x+y=1$; it can easily be related to the electron per atom ratio[23]). We have found (Fig. 6) that the x,y values that give the best representation are $x=0.6$ and $y=0.4$. The diagram shown in Fig. 6 delimits the four principal phases: $L2_1$ paramagnetic, paramagnetic martensite, $L2_1$ ferromagnetic, and ferromagnetic martensite. The intermediate phase has only been observed in alloy systems in the ferromagnetic $L2_1$ phase, for which the martensitic transformation temperature is far enough from the Curie point (dashed line in the diagram). This finding is probably an indication that the magnetoelastic interaction is sufficiently strong to drive the system through the transition towards the intermediate phase. Actually, this would be in agreement with the model presented in Ref. 12,

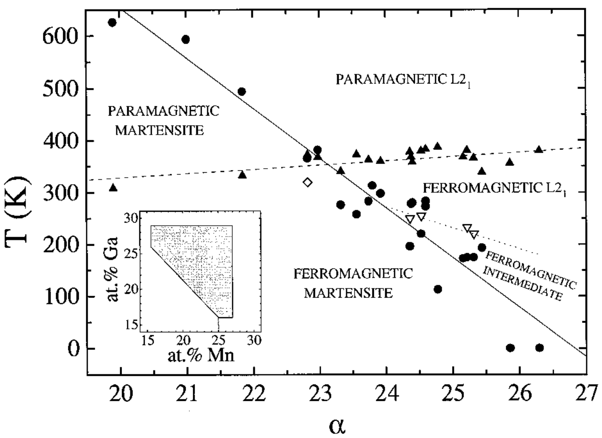

FIG. 6. Martensitic (solid circles), Curie (solid up triangles), and intermediate (open down triangles) transition temperatures as a function of a weighted composition parameter. Most data have been collected from Refs. 8 and 13. The open diamond corresponds to the temperature of the change in the modulation of the martensitic phase (Ref. 24). The hatched region in the inset shows the composition range from which data have been taken for this compact representation.

which shows the necessity of a large enough interaction in order to drive the system through the intermediate first-order phase transition. The temperatures of the premartensitic and martensitic transition become closer each to other with decreasing α. As a consequence, in a certain composition range, the martensitic transition can mask (or even inhibit) the existence of the intermediate phase. It is also interesting to notice, that changes in the modulation of the martensitic structure have been reported with decreasing temperature[24] in systems that do not exhibit a premartensitic transition (open diamond in Fig. 6). They could be a reminiscence of the intermediate transition in the $L2_1$ phase.

The set of experimental evidences undoubtfully state the existence of a magnetoelastic coupling in Ni_2MnGa alloys. All elastic constants increase with magnetic field. The relative change in the elastic constants correlates with the square of the magnetization.[25] This is consistent with the bilinear coupling between the magnetization and the homogeneous shear proposed in Ref. 12 and 26. It is worth remarking that such an M^2 dependence of the magnetoelastic energy has also been proposed by other authors[27] in order to account for their experimental observations.

It is acknowledged that the application of a uniaxial stress increases the martensitic transition temperature. For Ni_2MnGa, a dependence of $\sim$2.5 MPa/K for uniaxial stresses along the [001] crystallographic direction has been reported.[28] In our investigations, we have restricted our stress range in order to ensure that the temperature increase of the martensitic transition does not interfere with the intermediate transition. We have found that the application of uniaxial stresses modifies the characteristics of the intermediate phase transition: when the sample is subjected to a mechanical stress, the transition occurs with a certain thermal hysteresis. Dynamic mechanical tests[10] and neutron-scattering experiments under uniaxial stress[29] reported the existence of thermal hysteresis at the intermediate phase transition, although no detectable thermal hysteresis was observed by using other experimental techniques.[11] Present results confirm the fact the application of mechanical stresses results in a modification of the kinetic characteristics of the phase transition. Moreover, it has theoretically been shown[30] that the effect of applying a mechanical stress is an enhancement of the first-order character of the intermediate transition.

All elastic constants exhibit an anomalous stress dependence (Fig. 3): the measured increase is not linear, and the relative change is larger than that expected from purely vibrational anharmonic contributions. We argue that such an anomalous behavior could also be related to the magnetoelastic interaction. That is, the application of a uniaxial stress may induce rotation of magnetic domains, resulting in a change in the magnetization. This effect would lead to a modification of the values of elastic constants. This argument is consistent with the experimental finding that the relative change of elastic constants with hydrostatic pressure has been found[31] to be in the usual range for cubic alloys. Measurements of magnetization on samples subjected to controlled uniaxial stresses could provide experimental justification for this hypothesis.

The magnetic-field dependence of the structural transitions has been investigated by several authors.[7,12,27] For polycrystalline samples[27] no magnetic-field dependence has been found for the martensitic transition temperature, and the premartensitic transition temperature was not changed by fields less than 0.8 kOe but it was found to decrease for higher fields. We recently investigated more accurately the premartensitic transition at very low magnetic fields by means of an ac susceptometer for fields applied along the [001] direction.[12] These measurements have now been extended to magnetic fields along the [110] direction and the same behavior is obtained: a monotonous decrease of the intermediate transition temperature with increasing magnetic field. Since we have not found any significant dependence upon the direction of application of the magnetic field, a similar behavior is expected for polycrystalline samples. On the other hand, for the martensitic transition temperature, Ullakko *et al.*[7] reported a decrease of $\sim$2 K from strain vs temperature curves recorded at 0 and 10 kOe. This result is not consistent with the measurements by Zuo *et al.*[27] With the aim of making an estimation of the temperature change with magnetic field based upon thermodynamic data using the Clausius-Clapeyron equation, we have used recent values for the entropy change at this transition,[12] and have measured the temperature dependence of the saturated magnetization, shown in Fig. 7. An increase in M ($\Delta M \simeq 130$ emu/mol and $\Delta M \simeq 70$ emu/mol for fields along the [001] and [110] directions, respectively) is observed at the martensitic transition. These data render a maximum change in the martensitic transition temperature $dT/dH \sim 2\times10^{-2}$ K/kOe. This value is consistent with the results reported by Zuo *et al.*:[27] an increase of $\sim$1 K will fall within the experimental errors. Although the results by Ullakko *et al.*[7] are not consistent with the Clausius-Clapeyron predictions, it must be taken into account that nucleation effects may play a relevant role in determining the actual transition temperature of a given sample.

IV. SUMMARY AND CONCLUDING REMARKS

We have performed an experimental investigation of the premartensitic and martensitic transition in a Ni-Mn-Ga

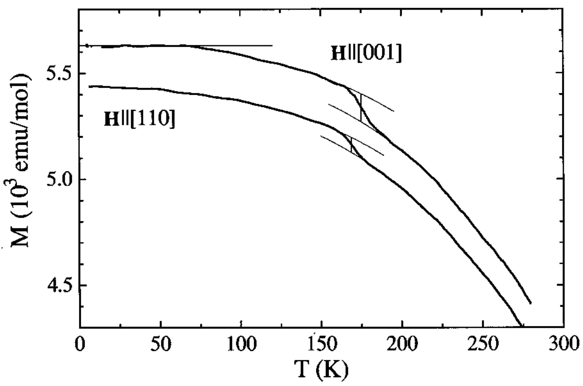

FIG. 7. Temperature dependence of the saturated magnetization measured under applied fields of 50 kOe along the [001] and [110] directions. Thin lines are guides to the eye to show the jump at the martensitic transition, and the horizontal behavior at low temperatures for the [001] direction.

single crystal. The main results outcoming from this investigation are as follows.

(1) All elastic constants and ultrasonic attenuation show a significant dependence upon the magnetic field.

(2) The elastic constants show an unusual dependence upon the applied stress which cannot be accounted for by purely anharmonic vibrational theories.

(3) The application of uniaxial stress results in a modification of the premartensitic transition: the transition takes place with thermal hysteresis.

(4) By making use of the measured values of ΔM and ΔS, it has been proven that the Clausius-Clapeyron equation predicts a change in the martensitic transition temperature with magnetic field around 2×10^{-2} K/kOe.

(5) The premartensitic transition temperature decreases with application of magnetic field *even* at low (0–20 Oe) magnetic fields. This behavior does not depend upon the direction of the applied field.

Present results undoubtfully state the existence of a magnetoelastic coupling in this alloy. Such a magnetoelastic coupling is responsible for the first-order phase transition from the $L2_1$ towards a micromodulated (intermediate) phase.

ACKNOWLEDGMENTS

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