The magnetoelastic properties of a Ni_2MnGa single crystal

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The temperature dependence of the electromagneto-acoustic conversion efficiency, low-field susceptibility, and propagation velocities of transverse and longitudinal sound are investigated in a single crystal of the ferromagnetically ordered Heusler alloy Ni_2MnGa. Anomalies of the characteristics are discovered at the Curie point ($T_c = 374$ K) and in the vicinity of the cubic-to-tetragonal phase transition ($T_m = 270$–280 K). The Debye temperature and a complete set of elasticity moduli in the cubic phase of this compound are determined.

1. INTRODUCTION

The ternary intermetallic compound Ni_2MnGa is the only ferromagnetically ordered Heusler alloy that experiences a reversible martensite-type structural transformation upon cooling. The transition is from the $L2_1$-ordered bulk-centered cubic phase to the tetragonal phase with an axial ratio $c/a = 0.94$ (Ref. 1). The crystalline structure of the low-temperature phase can be described as a recurring five-layer motif formed by displacements of one of the (110) planes in the $[1\bar{1}0]$ direction.[2] The resistance of the lattice to such a transition is given by the shear elasticity modulus C'. Since a structural transformation in Ni_2MnGa occurs in the ferromagnetic matrix (Curie temperature $T_c = 360$ K) this material is convenient for determining the relationship between changes in the structural state of the alloy and its magnetic and elastic properties..

2. EXPERIMENT

The Ni_2MnGa single crystals were grown by the Bridgeman method from the melt; according to x-ray spectroscopic analysis data the alloy was nonstoichiometric due to the reduction by 1 at.% in the manganese concentration through nickel substitution. A wafer was cleaved from the slab by the electric spark method for the acoustical measurements; the normal to the wafer plane coincided with the [110] crystallographic axis.

Ordinary acoustic analyses of the elastic properties of compounds experiencing martensite transformations encounter fundamental limits associated with the formation of the large-scale surface relief of the crystal and the breakdown of the acoustic contact between the transducer and sample. This problem was solved in the present paper by using a noncontact sound generation and detection method based on direct electromagneto-acoustic conversion (EMAC) in the metal skin-layer.[3] This approach makes it possible not only to determine the elasticity moduli of Ni_2MnGa but also to obtain information on the magnetic and magnetoelastic properties of the crystal.

The resonance method was used to measure the EMAC efficiency and the propagation velocities of transverse and longitudinal sound over the temperature range 4–270 K. Two induction coils—an excitation coil and a detection coil—were wound in parallel around the sample. The first was connected to an RF oscillator and the signal from the second coil, after amplification, was presented to a phase difference instrument and a recorder. The sample was placed in the "thermal field" channel of a superconducting solenoid, which allowed measurements to be made in magnetic fields of $H \leqslant 8$ T. Resonant changes in the surface impedance of the wafer were observed at the frequencies where the half-integer number of acoustic wavelengths was equal to the thickness of the sample. Transverse elastic waves were excited in the constant magnetic field normal to the wafer, while longitudinal waves were excited in the constant magnetic field tangential to the wafer. The acoustic velocities were calculated by the frequencies of resonant phenomena, while the EMAC efficiency was calculated from their amplitudes.

The echo-pulse technique was used over the temperature range 260–360 K. In this case the acoustic oscillations were both generated and detected in the metal by the same coil wound around the sample. RF pulses 1 μs in duration at a 10 MHz carrier frequency and a voltage of approximately 1 kV were fed to this coil. The measurements were carried out in an electromagnetically generated magnetic field $H \leqslant 1$ T. Both constant and oscillatory magnetic fields in the metal skin layer were employed to generate elastic wave pulses traveling normal to the wafer surface and decaying after multiple reflection off the parallel faces of the sample. Inverse EMAC was used to record the echo pulses.

The temperature dependence of the low-field susceptibility of Ni_2MnGa was also measured in the present experiment to interpret the temperature and field dependence of the remote acoustic efficiency.

3. MEASUREMENT RESULTS AND DISCUSSION

Both inductive[3] and magnetoelastic[4] interactions can be used to generate sound in magnetically ordered matter.

The EMAC induction mechanism reduces to the interaction of the a.c. current induced by the electromagnetic wave in the skin-layer of the conductor with the external magnetic field H. The efficiency η of this generation mechanism, defined as the ratio of the acoustic and electromagnetic energy fluxes, is proportional to the squared magnetic field strength:[3]

$$\eta = \mu^2 H^2 / \pi \rho S c (1+\beta^2). \qquad (1)$$

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Here $\mu = 1 + 4\pi\chi$ is the magnetic permeability, ρ is the density of the metal, S is the speed of sound, c is the speed of light, $\beta = q^2\delta^2/2$, $\mathbf{q}$ is the acoustic wave vector, and δ is the skin-layer thickness.

The efficiency of the EMAC magnetoelastic mechanism is a nonmonotonic function of the magnetic field; in accordance with Ref. 4 it is given by the equation

$$\eta = S\beta^2\xi(\mu-1)/c\mu^2(1+\beta^2), \qquad (2)$$

where ξ is the magnetoelastic coupling parameter. It is clear that the effectiveness of this electromagnetic and acoustic wave transformation mechanism is determined not only by the electrical and magnetic characteristics but also by the magnetostriction of the metal. It was not difficult to distinguish the acoustic generation mechanisms in experiments on Ni_2MnGa, since the nonmonotonic change in EMAC efficiency attributable to magnetoelastic interaction was observed only in weak magnetic fields when the inductive interaction was not significant.

At low temperatures acoustic generation was observed in the tetragonal phase of Ni_2MnGa only due to inductive interaction. As the martensite transition temperature ($T_m = 270$–280 K) is approached, the amplitude of the recorded signal diminished rapidly, which was due to the rise in acoustic damping at the phase transition.

Acoustic generation due to both inductive interaction in strong magnetic fields ($H \leqslant 8$ T) and magnetoelastic interaction in weak fields ($H \sim 0.1$ T) was observed in the cubic phase up through the ferromagnetic transition temperature. The temperature dependence of the efficiency of the magnetoelastic EMAC mechanism in a field $H + 0.1$ T is shown in Fig. 1. A hysteresis peak η was observed near T_m, while $\eta \propto (T - T_c)^{1/2}$ was found with increasing temperature and as the Curie temperature is approached. It follows from the field relations of EMAC in the ferromagnetic phase of Ni_2MnGa (with the exception of the martensite transformation region) that the acoustic generation efficiency due to the motion of the domain boundaries is small, i.e., acoustic generation is due to spin reorientation processes.

Figure 2 shows the temperature dependence of χ obtained with the oscillatory magnetic field $H \simeq 5 \cdot 10^{-5}$ T)

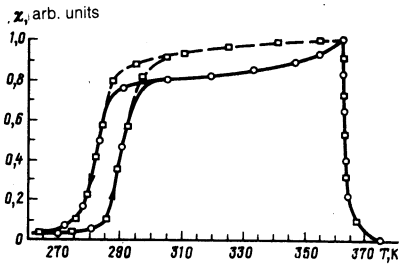

FIG. 2. Temperature dependence of the low-field susceptibility in Ni_2MnGa: ○—$\mathbf{H}\|[100]$, □—$\mathbf{H}\|[110]$.

oriented along the [110] and [100] crystallographic axes. Growth of χ at $T_c = 360$ K accompanies the transition of the compound to the ferromagnetic phase. The observed difference in the height of the Hopkinson maximum on curves *a* and *b* at T_c indicates the magnetocrystalline anisotropy of this compound. In the cubic phase of Ni_2MnGa the susceptibility decays weakly with diminishing temperature and drops off rapidly at the tetragonal phase transition, while hysteresis of χ is observed near the martensite transition. This behavior of χ at T_m is due to the development of martensite crystals and the increasing coercive force of the sample in this case.

Given the temperature behavior of χ the $\eta(T)$ relation can be explained in the following manner. EMAC efficiency in the tetragonal phase of Ni_2MnGa ($T < Tm$) is proportional to H^2 across the entire test magnetic field range, which is due to the small value of χ in this region. The absence of magnetoelastic sound generation for $T < T_m$ indicates that the magnetocrystalline anisotropy of Ni_2MnGa in the tetragonal phase is significantly higher than in the cubic phase. This is consistent with data from direct magnetization measurements.[1] The transformation peak characterized by temperature hysteresis at T_m indicates spontaneous magnetorestrictive strain in the crystal in the vicinity of the structural transition. In the cubic phase the axis of easy magnetization evidently[1] coincides with the [111] crystallographic axis, so that acoustic generation in weak magnetic fields is associated with the magnetic reorientation of the crystal from the [111] direction to the [110] direction.

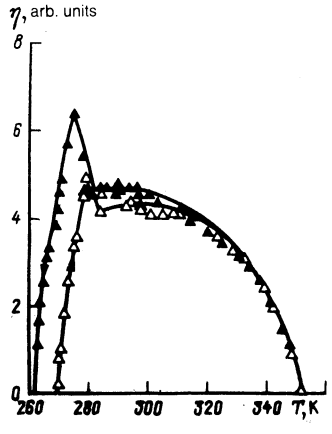

FIG. 1. Temperature dependence of the efficiency of the EMAC magnetoelastic mechanism in Ni_2MnGa in a field $H = 0.1$ T; ▲ and △: Forward and reverse progressions, respectively.

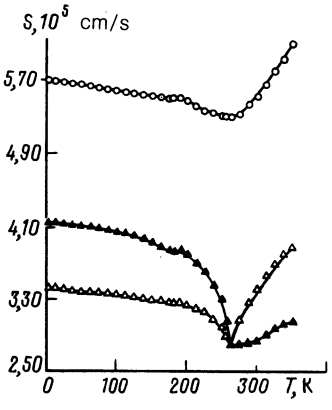

FIG. 3. Temperature dependence of the velocities of the longitudinal S_l (circles ○) ($\mathbf{q}\|[110]$) and transverse S_{t1} (△) ($\mathbf{q}\|[110]$), $\mathbf{p}\|[001]$) and S_{t2} (▲) ($\mathbf{q}\|[110]$, $\mathbf{p}\|[110]$) elastic waves in Ni_2MnGa.

The diminishing EMAC efficiency with rising temperature indicates a reduction in the spontaneous magnetic moment whose oscillations excite elastic waves, while the vanishing of η at T_c indicates the absence of spontaneous magnetorestriction of the ferromagnetic transition in Ni_2MnGa.

The temperature dependences of longitudinal and the two polarizations p of transverse sound are shown in Fig. 3. These measurements made it possible to determine a complete set of elasticity moduli of Ni_2MnGa in the cubic phase. For $T = 300$ K we obtain $C_{11} = 2.13 \cdot 10^{12}$ dyne/cm^2, $C_{12} = 0.87 \cdot 10^{12}$ dyne/cm^2, $C_{44} = 0.92 \cdot 10^{12}$ dyne/cm^2. The Debye temperature corresponding to these values is $T_D = 190$ K. The modulus C_{12} grows rapidly as T_m is approached from the higher temperature range, while C_{11} and C_{44} diminish. This means that the shear modulus $C' = (C_{11} - C_{12})/2$ characterizing the resistance of the lattice to shear strains decays sharply near T_m.

The velocities of the two transverse sound polarizations at $T < T_m$ are significantly different, indicating conservation of the noticeable elastic anisotropy in the martensite state. A weakly-expressed anomaly, whose nature is not yet clear, is quite visible on the temperature dependences of the velocities of all test elastic modes at $T \approx 180$ K.

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