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Anomalous magnetoresistance in NiMnGa thin films

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The origin of anomalous negative magnetoresistance and its temperature dependence in polycrystalline Ni–Mn–Ga films prepared by pulse laser deposition was studied. The investigation of structural, transport, magnetic, and ferromagnetic resonance properties of the films suggests contributions of different mechanisms in magnetotransport. At low magnetic fields the main contribution to magnetoresistance is due to the transport between the areas with different orientation of magnetic moments, while at high fields it is an electron scattering of in spin-disordered areas.
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I. INTRODUCTION

A considerable attention is attracted to Ni–Mn–Ga alloy system as a magnetic actuator device material with magnetic field induced strain up to 10%.¹ Recently^{2–5} it has been shown that continuous films of NiMnGa and some other Heusler alloys demonstrate negative magnetoresistance effect. Different values of magnetoresistance and different temperature dependences were observed. The values of about -1.2% in magnetic field 10 kOe at $T=23$ K were reported² for Ni–Mn–Ga films deposited by pulse laser deposition on Si wafers. However, the temperature dependence of magnetoresistance was not properly described there. In³ a series of epitaxial films of the full Heusler alloys Ni₂MnGa, Ni₂MnGe and, Ni₂MnAl grown by molecular beam epitaxy on GaAs substrates, were studied for their magnetic and transport properties. It was shown that Ni₂MnGe films demonstrate magnetoresistance of about -1% in a magnetic field of 90 kOe at $T=300$ K, while for other materials the magnetoresistance was very small. It was mentioned that the maximal values of magnetoresistance are observed in the vicinity of the Curie temperature. For other temperature ranges the magnetoresistance decreases to negligible values. Somewhat higher values (-7% at 10 K and -1% at 300 K in a magnetic field of 50 kOe) were found for monocrystalline thin foils of Cu–Al–Mn alloys.⁵ In this case a monotonic decreasing of magnetoresistance with temperature increasing was observed. The origin of the negative magnetoresistance in such systems, as well as the unusual character of temperature dependence of magnetoresistance, is not quite clear so far. Several different mechanisms for different temperature ranges were proposed. The authors of Ref. 2 attributed the appearance of magnetoresistance at the low temperatures to spin-transport effect in inhomogeneous or granular structures. Around Curie temperature the magnetoresistance was found to be very similar to that of mixed valence manganites.³ Also-spin transport mechanism that considered different

magnetization processes in large ferromagnetic clusters and spin reorientation of small clusters and boundary spins was analyzed in Ref. 5.

This work is devoted to the investigation of Ni–Mn–Ga films and analysis of the mechanisms that are responsible for their magnetoresistance properties.

II. EXPERIMENTAL DETAILS

The films were prepared using pulse laser deposition onto Al₂O₃ and Si substrates held at 773 K in vacuum of 10^{-6} Torr using a Custom build system with KrF laser operating at 248 nm. The compositions of targets and obtained films were determined by EDAX and are presented in Table I. Scanning electron microscopy showed the formation of 200 nm thick polycrystalline films with average grain size of ~ 70 nm (see Fig. 1). The structure of the films was investigated by x-ray diffractometry using Cu $K\alpha$ radiation. Magnetic properties were investigated using Quantum Design MPMS 5S superconducting quantum interference device magnetometer in the 5–350 K temperature range. Transport measurements were carried out in Quantum Design PPMS Model 6000 in the 5–350 K temperature range and at the fields (H) up to 90 kOe. Magnetoresistance was measured using standard four points technique in current in the film plane configuration. The magnetic field was applied in the film plane parallel (L geometry) and perpendicular (T geometry) to the current. Ferromagnetic resonance was studied in 10–293 K temperature range using x -band Bruker EMX300 EPR spectrometer.

TABLE I. Composition of the films.

Sample	Bulk target	Film
1	Ni _{53.7} Mn _{26.4} Ga _{18.9}	Ni ₅₅ Mn ₂₃ Ga ₂₂
2	Ni _{52.3} Mn _{27.4} Ga _{20.3}	Ni ₅₃ Mn ₂₄ Ga ₂₃

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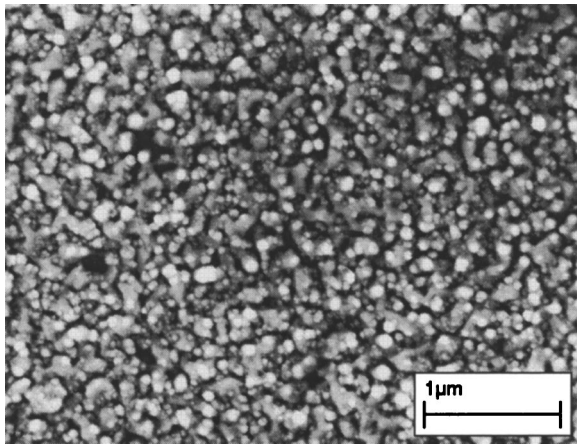


FIG. 1. Scanning electron microscopy photograph of the film 2.

III. RESULTS AND DISCUSSION

The magnetic susceptibility χ data for both samples are presented in Fig. 2(a). The more rapid increased of χ with the temperature T in the vicinity of room temperature is associated with the martensite-austenite transformation and an appearance of a soft magnetic austenite phase. For the films there is no abrupt χ change as it was observed in homogeneous bulk Ni-Mn-Ga alloys.⁶ The first reason is that the films are slightly inhomogeneous in composition and different areas have different transformation points. The second one is that the martensite-austenite transformation is hindered by the interaction between the film and the substrate.

The diffraction patterns for both of the films look similar (Fig. 3). In both cases one can find that at room temperature cubic austenite phase ($a \approx 0.581$ nm, which is very close to $a = 0.582$ nm observed in bulk materials), some tetragonal one, and also residual Mn(II) oxide coexist in the film. Though composition of the samples is very close, the amount of tetragonal phase is different for the samples 1 and 2. It is

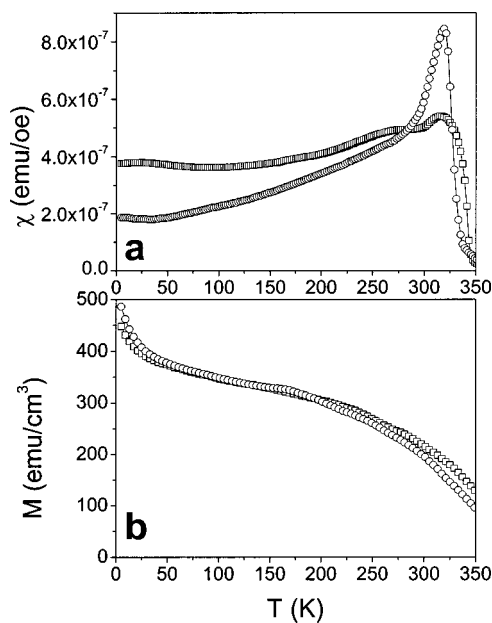
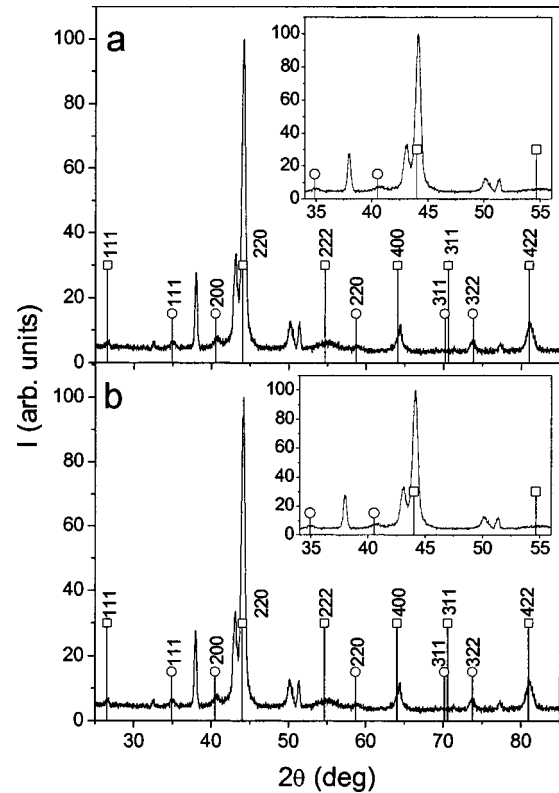


FIG. 2. The dependence of magnetic susceptibility (a) and magnetization (b) of 1 (squares) and 2 (circles) films on temperature.

FIG. 3. X-ray diffraction spectra of the films 1 (a) and 2 (b). The positions on the lines corresponding the Ni_2MnGa austenitic $L2_1$ phase MnO are denoted by squares and circles, respectively.

known that the structure of Ni-Mn-Ga alloys is very sensitive to composition and preparation technique. In our case the films were prepared in the same technological conditions, thus the difference of the structure is caused by the composition. The structural and composition differences cause differences of magnetotransport properties discussed below. FMR (Fig. 4) at room temperature showed the presence of two magnetic phases with the difference in saturation magnetizations of several percent. Taking into account that the magnetization of the martensitic phase of Ni-Mn-Ga alloys is

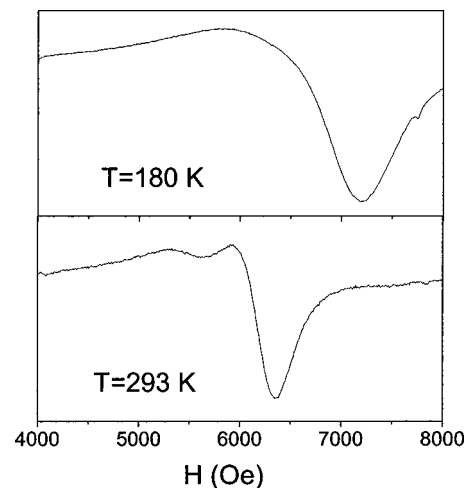


FIG. 4. FMR spectra for the film 2 at 180 and 300 K. The magnetic field is perpendicular to the film plane. The high field peak is corresponded to martensitic phase while the low field one is attributed to austenite.

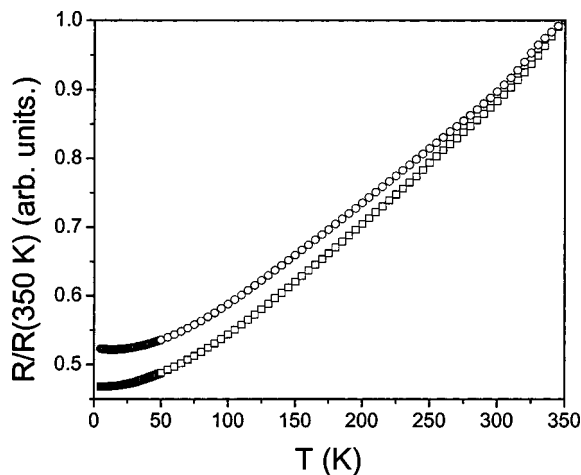


FIG. 5. The dependence of the resistance of 1 (squares) and 2 (circles) films on temperature.

a little bit higher than austenitic one (see, for instance, Ref. 6) the tetragonal phase can be identified as a martensite. The martensite FMR peak grows at the expense of the austenite one with the temperature decrease. But even at 180 K the austenite peak can be observed, which confirms the expanded character of austenite-martensite transformation. The temperature decrease also leads to the broadening of resonance lines, which can be related to the increase of stresses in the film. Contrary to the bulk material⁶ due to the wide temperature range of martensite-austenite transformation in the films, no peculiarities are observed on temperature dependence of magnetization [Fig. 2(b)].

A rapid decrease in susceptibility and magnetization (Fig. 2) near 350 K corresponds to a ferromagnetic-paramagnetic transition. At low temperatures a rapid increase of magnetization with the decrease in temperature is observed. It can be linked with the presence of paramagnetic phases. The formation of these phases partially can be caused by the oxidation of film surface. X-ray diffraction showed the presence of FCC phase with the lattice parameter $a \approx 0.444$ nm, which corresponds to MnO. Another possibility of paramagnetic phase formation is that in nonstoichiometrical NiMnGa alloys there is a tendency of segregation with the appearance of Ni₂MnGa nanophase.^{6,7} Such segregation can be accompanied by no or small change of lattice parameters.⁷ If there is a lack of Mn, which is responsible for magnetism in NiMnGa compounds,⁸ a part of the material can be in paramagnetic state.

The temperature dependencies of resistance R are presented in Fig. 5. Metallic type conductivity is observed in the whole temperature range. An appearance of a shallow minimum at low temperatures in nonordered metallic alloys as well as the nonlinearity of temperature dependence below 100 K is out of the scope of the present paper and was discussed in many works (see, for instance, Ref. 9 and references therein). The relatively small variation of the resistance with the temperature is also typical for disordered alloys.⁹ The martensite-austenite transition does not lead to pronounced peculiarities of R vs T dependence as in the case

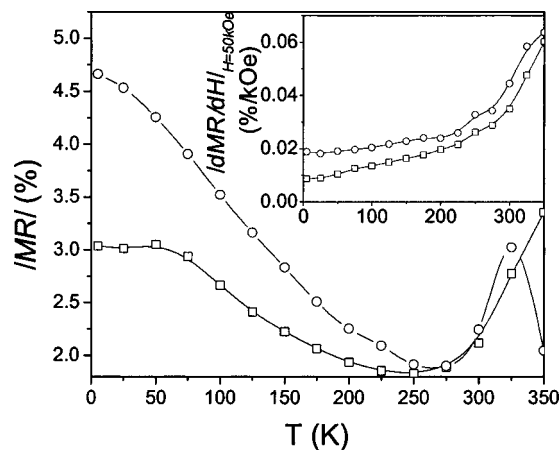


FIG. 6. The dependence of the MR (T geometry) and high magnetic field $\partial MR/\partial H$ of 1 (squares) and 2 (circles) films on temperature.

of NiMnGa bulk alloys⁶ due to the expanded character of the transformation discussed above. Only small disturbance are observed in the transition area.

Both of the films demonstrate negative magnetoresistance (MR) in 5–300 K temperature range (Fig. 6). A maximum appears on the MR vs T dependence at high temperatures. Such a maximum is typical for colossal magnetoresistance (CMR) ceramics in the metal-semiconductor (ferromagnetic-paramagnetic) transformation area,¹⁰ but in the NiMnGa films the character of conductivity does to change and remains metallic in the whole temperature range. The value of MR is higher for the sample 2 and maximum occurs at the lower temperature. These differences are caused by structure and composition differences of the films. The experiments on bulk material with practically the same composition showed that the appearance of the maximum cannot be ascribed to the critical scattering due to the spin fluctuation near the Curie temperature.¹¹ The bulk samples had also negative magnetoresistance effect, but it was pretty low (less than 0.2% at 50 kOe) and no pronounced peak was observed near the Curie temperature. Thus the contribution from this mechanism cannot be very big.

The dependence of MR and M vs H are presented in Fig. 7 (left panel). It is to be noted that the shapes MR vs H curves measured in L and T geometries coincide for the whole temperature range within experimental errors. Moreover, no obvious contribution from anisotropic magnetoresistance was found. It is easy to understand the negligible contribution of the anisotropic magnetoresistance taking into consideration fine grain structure of the films (see Fig. 1). In this case the orientation of magnetization inside a grain is determined not only by the magnetocrystalline anisotropy but the exchange interaction with the nearest grains and, as a result, in most cases the direction of the magnetization does not coincide with the orientation dictated by the structure of the grain.

The resistance of the films is continuously decreasing with the field and even for $H=90$ kOe. No sign of saturation in the whole temperature range was observed. At the higher fields the magnetoresistance becomes proportional to mag-

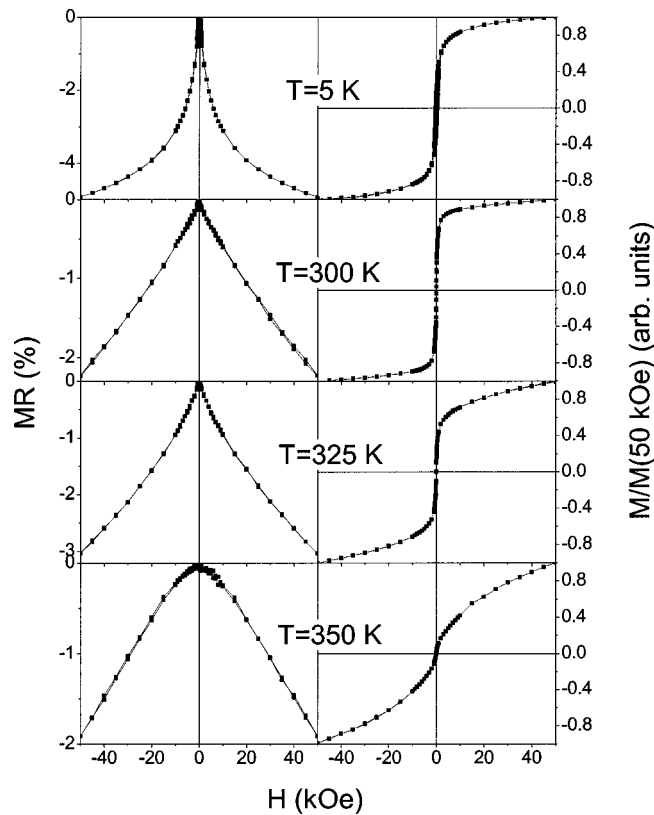


FIG. 7. The dependence of the magnetoresistance in T geometry (left panel) and magnetization on the magnetic field at different temperatures for the film 2.

netic field (Fig. 7). The temperature variation of $\partial MR/\partial H$ at the higher magnetic fields is shown in the inset of Fig. 6. The value of $\partial MR/\partial H$ is monotonously increasing with the temperature.

The dependence of MR vs M^2 are presented in Fig. 8. In our case the dependence shows linear behavior at low fields and a kink for a certain value of the magnetization. Observed features can be explained as follows. It is known that $MR \sim (M/M_s)^2$ behavior is due to the transitions of the electrons between areas with different orientations of magnetization.¹² The contribution of this mechanism is not monotonic. First it gradually decreases with the increase in temperature and becomes practically negligible at room temperature. But near 350 K it becomes substantial again. A kink on MR vs M^2 behavior is usually attributed to the contribution of the very small particles or to the alignment of the disordered surface spin under a very high field. The contribution of this mechanism monotonically increases with temperature.

The appearance of $MR \sim (M/M)^2$ behavior at low temperatures is due to spin transport between grains through the grain boundaries as well as through some nonmagnetic or weak magnetic inclusions. Small value of the effect, comparing with giant magnetoresistance in granular films, is caused by strong dipolar and exchange interaction between grains and correlation of their magnetic moments. Another possible mechanism of magnetoresistance is spin transport between magnetic domains. The decreasing contribution of these mechanisms into the magnetoresistance with the increase in temperature can be explained considering the growth of the

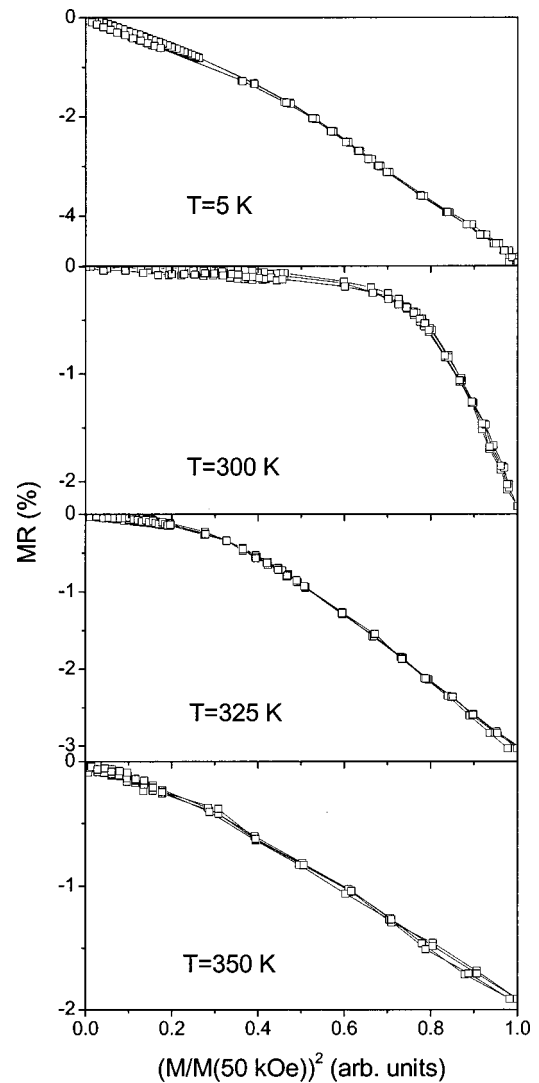


FIG. 8. The dependence of the magnetoresistance on normalized squared magnetization at different temperatures for the film 2.

film resistance (Fig. 5) and decrease of their magnetization [Fig. 2(b)], which results in reduction of spin polarization.

For high temperatures (above 300 K) the increase of $MR \sim (M/M)^2$ contribution can be attributed to slight inhomogeneity of film composition. Different areas of the films have different Curie temperatures and the films become a mixture of magnetic and nonmagnetic material. The second reason is that nonstoichiometrical NiMnGa alloys have a tendency to segregate with an appearance of Ni_2MnGa nanophase. This fact can lead to superparamagnetic behavior of the material near the ferromagnetic-paramagnetic transformation area.⁶

The most interesting is the linear variation of the magnetoresistance with magnetic field. Such a behavior should be observed for the electron scattering off spin-disordered inclusions. The increasing contribution of this mechanism to the magnetoresistance with temperature did not allow ascribing this phenomenon to scattering off paramagnetic or superparamagnetic inclusions. Nonsaturation at high fields excludes the scattering of domain walls like it was observed in CMR ceramics. Another possibility is the formation of spin-

glass-like areas around grain boundaries and near non-magnetic phases (as it was reported in Ref. 13 for granular systems) and/or near antiferromagnetic inclusion, which can appear in Heusler alloys (see, for instance, Ref. 14). It is difficult to saturate these areas even up to 160 kOe.¹⁵ It should be noted that the temperature increase should lead to the growth of these areas in the NiMnGa films due to decrease of magnetization and reduction of anisotropy caused by replacement the magnetically hard martensite phase by the austenite phase with lower anisotropy as well as decrease of strains discussed above. As a result, the contribution of this mechanism into the magnetoresistance should also increase with the temperature and reach the maximum near the Curie temperature. That is precisely what was observed in the experiments.

IV. CONCLUSIONS

In conclusion the NiMnGa films have negative magnetoresistance in wide temperature range and demonstrate an example of spin frustrated magnetic system. The value of magnetoresistance is one of the highest observed in such type of materials. The improvement of the technology can allow to increase this value and to obtain a material with linear dependence of magnetoresistance on magnetic field in a wide temperature range. It opens a possibility for creation of materials with very small temperature variation of resistance and magnetoresistance, which has in combination with linear field dependence of resistance, made them very attractive for applications in magnetic field measurement systems.

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