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Magnetic and transport properties of NiMnAl thin films

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The magnetic and transport properties of Ni_2MnAl thin films prepared using pulse laser deposition were investigated. It was shown that the films are granular and multiphase. Contrary to the data reported earlier we observe nonmonotonic temperature dependence of resistance with minimum in the vicinity of 100 K for the films deposited on substrates held at 773 K and negative magnetoresistance (MR) values of about 2.5% at 5 K and 1.6% at RT in magnetic field of 50 kOe. These values of MR are the highest reported up to date for Ni_2MnAl films. Large negative MR in our films arises from a nonhomogeneous structure and its origin is the same as for granular films.
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Heusler alloy (HA) thin films attract much attention for their applications in magnetoelectronics. It is due to the fact that some HAs are half metallic ferromagnets with large asymmetry in density of states at Fermi level.[1–6] This allows high spin polarization that leads to high values of tunneling magnetoresistanse in magnetic tunnel junctions (MTJs). For example, tunneling magnetoresistance (TMR) as high as 40% at room temperature was obtained for MTJs using Co_2MnAl alloy.[3] HA films are interesting not only because of their MTJ applications but also their unusual optical,[2,7,8] magnetic, and magnetotransport properties.[6,9–14] However, the origin of MR effect in HA films is not very clear. It is known that the structure and magnetic properties of HA films strongly depend on the composition and preparation procedure.[8,11,12] It was shown that the deposition conditions and postdeposition heat treatment dramatically change the magnetic and transport properties of NiMnGa films.[12] Optimization of these properties is mandatory to achieve high values of TMR in MTJs. Previous study of full epitaxial HA thin-film Ni_2MnAl prepared by molecular-beam epitaxy showed that the films are not ferromagnetically ordered and have very small temperature-dependent negative MR.[9] The origin of the effect was not comprehensible. It is worth to note that up to date there is no clear picture on magnetism in this alloy. It was shown that ferromagnetic and antiferromagnetic states can coexist in stoichiometric Ni_2MnAl alloy and ferromagnetic ordering is typical for off-stoichiometric alloys.[15] The combination of ferromagnetic and antiferromagnetic (or nonmagnetic) phases in the film can produce sufficient negative magnetoresistance effect. In this paper we prepare Ni_2MnAl HA films and study their structure and magnetotransport properties.

Films of ~300 nm in thickness were deposited in vacuum of 10^{-6} Torr on Al_2O_3 substrates held at 773 K using a custom-built pulse laser deposition system. Compositions of the target and the film were determined using energy dispersive analysis of x ray to be $Ni_{50}Mn_{27}Al_{23}$ and $Ni_{54}Mn_{25}Al_{21}$, respectively. The structure of the film was studied by a field-emission scanning electron microscope (FESEM) and x-ray-diffraction technique. Magnetic properties were investigated using a Quantum Design MPMS 5S superconducting quantum interference device (SQUID) magnetometer in the 5–300 K temperature range. Transport measurements were carried out in Quantum Design PPMS Model 6000 in the 5–300 K temperature range and at the fields (H) up to 50 kOe. Magnetoresistance was measured using standard four-point technique in current in the film plane configuration. The magnetic field was applied in the film plane and perpendicular (T geometry) to the current. Ferromagnetic resonance was studied at room temperature using an x-band Bruker EMX300 electron-paramagnetic-resonance (EPR) spectrometer.

FESEM investigations (Fig. 1) show that the film is polycrystalline with a particle size in the range of 20–40 nm. X-ray-diffraction pattern (data not shown) reveals that the

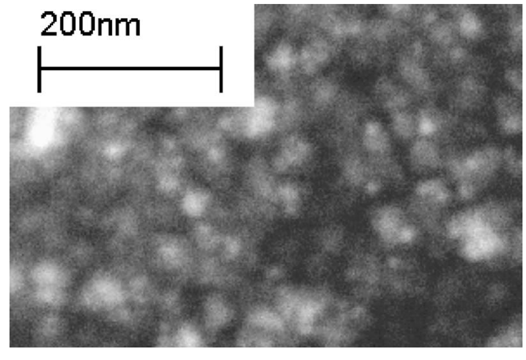

FIG. 1. Field-emission scanning electron microscope image of the film.

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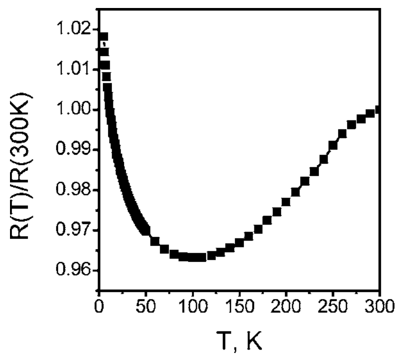

FIG. 2. Temperature dependence of resistance measured without an applied magnetic field.

film is multiphase. The lines of $L2_1$ cubic Ni_2MnAl phase with $a=0.581$ nm (which is very close to the bulk material[15]) with superstructural lines ⟨210⟩ and ⟨332⟩ as well as additional reflexes which can be attributed either to the lines ⟨111⟩ and ⟨200⟩ of Al or to the strongest lines of Al_6Mn compound[16] were found.

Contrary to the data on Ni_2MnAl film prepared by molecular-beam epitaxy[9] that shows metallic temperature dependence of resistance, the temperature dependence of resistance in our case is very weak and has a nonmonotonic characteristic (Fig. 2). A broad minimum appears at temperature ~100 K. Such a behavior is typical for disordered and/or nanocrystalline films (see, for instance, Ref. 17 and references therein) and is caused by inhomogeneous structure of the film. This points that different deposition techniques lead to the formation of the films of different structures and transport properties. Unlike molecular-beam epitaxy that produces monocrystal films[9] the use of laser pulse deposition leads to the formation of granular films with a disordered structure. The latter should provide higher values of magnetoresistance.

The *x*-band FMR spectra (Fig. 3) recorded for the magnetic field in the film plane and perpendicular to the film plane consist of several lines from the substrate (at the fields below 1000 Oe) and broad FMR line which position shifts from 3000 Oe for the field in the field plane to 4000 Oe for perpendicular configuration. The latter can arise either from

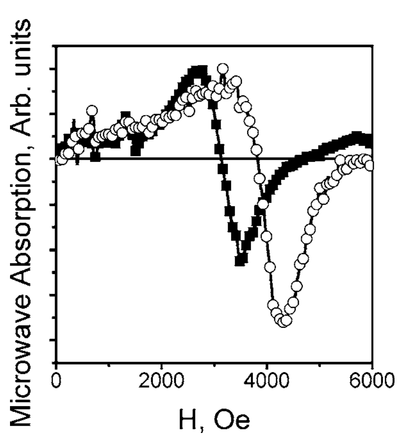

FIG. 3. FMR spectra for the film measured with a magnetic field applied parallel (solid square) and perpendicular (open circle) to the film plane. The sharp peaks at the fields below 1000 Oe correspond to the Al_2O_3 substrate.

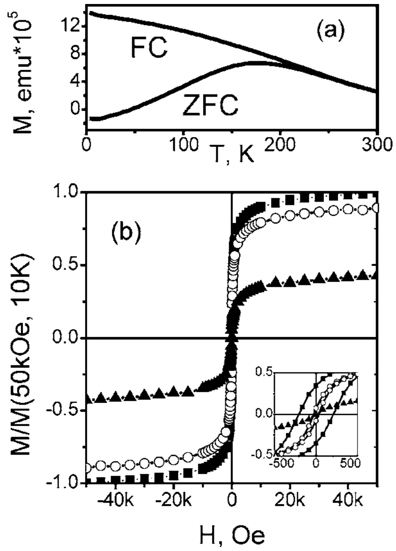

FIG. 4. Field-cooled (FC) and zero-field-cooled (ZFC) magnetic susceptibilities for the film measured at 50 Oe (a) and normalized magnetic hysteresis loops with in-plane applied magnetic field measured at $T=10$ K (square), 100 K (circle), and 300 K (triangle) (b). The inset presents low-field behavior of the hysteresis loops at different temperatures.

a continuous weakly ferromagnetic film (M_s ~40 emu/cm^3) or a granular film with dipole-dipole interaction between granules.[18] However, the temperature dependence of the magnetic susceptibility measured in zero-field-cooled (ZFC) and field-cooled (FC) regimes [Fig. 4(a)] is characteristic for granular films with transformation from ferromagnetic to superparamagnetic state. The size of the magnetic granules was estimated to be about 10 nm. Unfortunately due to the difficulties with the determination of the saturation magnetization, anisotropy, and volumetric filling factor of the granules it is impossible to evaluate the granules size precisely. However, this evaluation is in good agreement with the FESEM data if one suggests that the granules of a ferromagnetic core surrounded with a paramagnetic or nonmagnetic shell.

The shapes of the magnetic hysteresis loops also confirm the presence of superparamagnetic particles in the film [Fig. 4(b)]. The magnetization saturates relatively slow. The width of the hysteresis loops and the value of saturation magnetization strongly depend on temperature. This is characteristic of an ensemble of superparamagnetic particles. The loops are similar to those observed for magnetic granular films below the percolation threshold of magnetic material.[19]

The dependences of magnetoresistance on temperature and applied field are shown in Figs. 5(a) and 5(b), respectively. The dependence on field is typical for granular structures, i.e., the resistance of the film decreases with a field increase. Also the value of MR decreases with temperature that reflects effect of thermal excitation on small superparamagnetic particles. The value of MR changes from ~2.5% at 10 K to ~1.6% at room temperature in magnetic field of 50 kOe [Fig. 5(a)]. These values are the highest reported for Ni_2MnAl films up to date. It is interesting to point out that the temperature dependence of MR below 175 K is relatively weak. It is in good agreement with the ZFC/FC measurement that show blocking temperature of the magnetic particles in

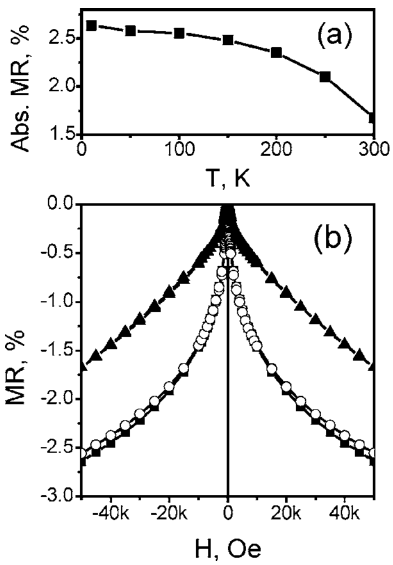

FIG. 5. Temperature dependence of MR (a) and dependence of MR on the applied magnetic field measured at $T=10$ K (square), 100 K (circle), and 300 K (triangle) (b).

the same temperature range [Fig. 4(a)]. Above this temperature MR rapidly decreases as magnetic particles are not blocked and very high magnetic fields are required to line up their magnetic moments.

Comparing the data on the structure and magnetic and transport measurements, one can derive a conclusion that the films are an inhomogeneous mixture of ferromagnetic and nonmagnetic materials. They contain small ferromagnetic particles embedded in nonmagnetic matrix, i.e., they represent a typical magnetic granular structure. The granularity of our films arises from local inhomogeneity. Though the overall composition of the film is close to stoichiometric alloy, there are local areas of different compositions with different magnetizations, which are reflected by x-ray and magnetic data. Spin-dependent transport will occur between those areas through nonmagnetic matrix. The presence of the areas with different magnetizations is also obvious from MR versus $(M/M_s)^2$ curve (Fig. 6). In the case of monodisperse noninteracting ensemble of superparamagnetic particles MR versus $(M/M_s)^2$ should be linear.[11] However, when the system is polydisperse or/and the magnetic particles have different magnetizations this behavior will be nonlinear or a kink should be observed. The deviation from linearity is attributed to the alignment of small particles or disordered surface states under high fields. It is to be pointed out that the value of MR is small compared to the traditional transition-metal–nonmagnetic metal granular structures with composition of magnetic material below the percolation threshold.[19] It seems that in HA films MR is strongly suppressed by dipolar interaction between ferromagnetic particles in the film. As a result the orientation of magnetic moment in individual particle will not only be determined by the magnetocrystalline anisotropy but also by the exchange with the nearest granules.[18] The detailed analysis of the peculiarities of MR involving spin-dependent transport through the areas with different magnetizations in HA thin films and foils was provided earlier in Refs. 11 and 20.

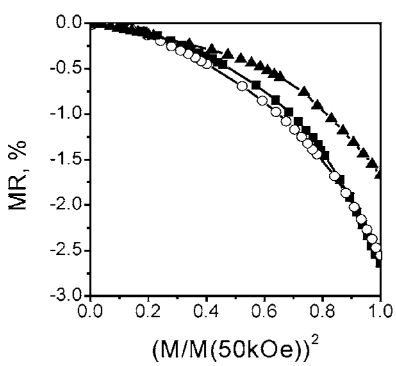

FIG. 6. The dependences of the MR on normalized squared magnetization at $T=10$ K (square), 100 K (circle), and 300 K (triangle).

In conclusion we show that it is possible to achieve large negative magnetoresistance in Ni_2MnAl films. The prepared films were disordered multiphase granular structures. They contain areas with different compositions and magnetizations, which give rise to negative magnetoresistance due to spin-dependent scattering typical for traditional granular films. Additional experiments should be carried out to clarify the magnetic phase diagram of the HA films and improve technology to achieve higher values of magnetoresistance.

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