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Anomalous Hall effect in $\text{Ni}_{47.3}\text{Mn}_{30.6}\text{Ga}_{22.1}/\text{MgO}$ (001) thin films

M.I. Blinov¹, V. Chernenko^{2-4,*}, V.N. Prudnikov¹, I. R. Aseguinolaza³, J.M. Barandiaran³,

E. Lahderanta⁵, and A.B. Granovsky¹

¹ Faculty of Physics, Lomonosov Moscow State University, Moscow, 119991 Russia

² Basque Center for Materials, Applications and Nanostructures, UPV/EHU Science Park, 48940 Leioa, Spain

³ University of Basque Country (UPV/EHU), Bilbao 48080, Spain

⁴ Ikerbasque, Basque Foundation for Science, Bilbao 48013, Spain

⁵ Lappeenranta University of Technology, Lappeenranta 53851, Finland

⁶ Institute for Theoretical and Applied Electromagnetics of Russian Academy of Science, Moscow 125412, Russia

⁷ National University of Science and Technology "MISiS," Moscow 119049, Russia

[*volodymyr.chernenko@ehu.eus](mailto:volodymyr.chernenko@ehu.eus)

We investigate the temperature dependences of the magnetic properties, electrical resistivity, magnetoresistance, and Hall effect resistivity, $\rho_H(H)$, in thin film of the Heusler-type $\text{Ni}_{47.3}\text{Mn}_{30.6}\text{Ga}_{22.1}$ (at.%) magnetic shape memory alloys (MSMA) epitaxially grown onto a $\text{MgO}(001)$ substrate. The results reveal martensitic transformation (MT) at about 230 K, premartensitic transition around 285 K and the Curie temperature of austenite around 380 K. We obtained the coefficients of normal Hall effect (NHE), R_0 , and anomalous Hall effect (AHE), R_s , by fitting the total Hall resistivity curves $\rho_H = R_0 B_z + 4\pi R_s M_z$ in several magnetic field ranges (0.1 - 1 kOe, 0 - 5 kOe, 8 - 16 kOe, 0 - 16 kOe), using the experimental magnetization data. Both coefficients R_0 and R_s strongly depend on the magnetic field. We also fit the Hall effect resistivity with the expression $\rho_H = R_0 B_z + 4\pi R_s M_z + \Delta\rho_H$ using the coefficients R_0 and R_s obtained from high field interval (8 - 16 kOe), where the last term $\Delta\rho_H$ was considered to correspond either to the topological Hall effect or to the antiferromagnetic Hall effect. The obtained temperature dependence and magnitude of $\Delta\rho_H$ discard the presence of the skyrmions or antiskyrmions. We conclude that unconventional field dependences of the NHE and AHE coefficients are produced by the antiferromagnetic correlations and the influence of the magnetic field on the electronic structure.

Keywords: Ni-Mn-Ga Heusler alloy, thin film, martensitic and premartensitic transitions, anomalous Hall effect, topological Hall effect.

I. INTRODUCTION

The anomalous Hall effect (AHE) was discovered 140 years ago and since then has attracted increasing attention (see, e.g, Refs. [1,2]). This effect is central in the group of galvanomagnetic and thermomagnetic phenomena, the progenitor of a whole class of Hall effects (quantum, optical, spin Hall effects), the forerunner of spin-dependent transport phenomena. The possibility of AHE applications in spintronics was considered in Ref. [3]. The spin Hall effect, which is of the same origin as AHE, is considered as the most promising phenomenon for creating and detecting spin currents [4]. It should also be emphasized that magneto-optical effects in the infrared region are nothing but frequency-dependent AHE.

The AHE cannot be explained in the framework of the Boltzmann transport equation and non-relativistic quantum theory. Therefore new methods in solid state physics, such the density matrix method and quantum transport equation [5], the Berry phase concept in transport phenomena [1], were first developed to explain AHE.

Starting from the 30s of the last century, the dependence of the Hall resistivity on the magnitude of the magnetic field has been expressed as the sum:

$$\rho_H = R_0 B_z + 4\pi R_s M_z, \quad (1)$$

where the first term describes the normal Hall effect (NHE) due to the action of the Lorentz force, and the second one characterizes the AHE associated with the influence of spin-orbit interaction (SOI), R_0 and R_s are the coefficients of NHE and AHE, respectively. M_z is the component of the magnetization along the z-axis, B_z is the z-component of the magnetic induction

$$B_z = H_z + 4\pi M_z(1 - N), \quad (2)$$

where $0 \leq N \leq 1$ is the demagnetization factor, R_0 and R_s are the coefficients of NHE and AHE, respectively. By definition

$$R_s = \frac{\sigma_{xy}(M_z)}{(\sigma_{xx}^2(B_z) + \sigma_{xy}^2(M_z)) 4\pi M_z}, \quad (3)$$

where the off-diagonal (Hall) conductivity σ_{xy} , as a rule, is much smaller than the diagonal conductivity, σ_{xx} ($\rho = 1/\sigma_{xx}$ is the electrical resistivity). In most cases, when the magnetoresistance (MR) is small, the off-diagonal conductivity of the AHE is a linear function of the magnetization, and the AHE coefficient does not depend on the external field or on the spontaneous magnetization.

Three basic AHE mechanisms, namely, the intrinsic mechanism, skew scattering and the side-jump mechanism determine the concentration and temperature dependences of the AHE in various ferromagnets [1,2]. The intrinsic mechanism is also known as the Karplus-Luttinger contribution associated with velocity corrections due to SOI and interpreted recently through the Berry phase [1]. The discussions about the relative role of each of these mechanisms do not cease till present. For example, as early as in 1958 [5], it was shown by a simple model with a low concentration of impurities that the contributions of the intrinsic mechanism and the side-jump are of the same magnitude but opposite sign, while many researchers believe that the intrinsic AHE mechanism is dominant [1] (see also the discussion in [6]). Moreover, the AHE behavior in Ni-Mn-In based Heusler alloys [6-8] cannot be explained by any of these mechanisms.

Recently, it was shown that in the topologically nontrivial structures (see, e.g., Refs. [9-12]), as well as in the non-collinear antiferromagnets [13,14], an additional contribution to Eq. (1), $\Delta\rho_H$, is possible

$$\rho_H = R_0 B_z + 4\pi R_s M_z + \Delta\rho_H, \quad (4)$$

which can be produced either by the so-called topological Hall effect (THE) or by the antiferromagnetic Hall effect. The field dependence of this term is not known explicitly. In the case of skyrmions $\Delta\rho_H \propto \overline{M} \left(\frac{\partial \overline{M}}{\partial x} \times \frac{\partial \overline{M}}{\partial y} \right)$, the corresponding topological contribution is of the order of the NHE contribution [15], that, as a rule, is much smaller than the second term in Eq.(4). The topological contribution is absent in the saturation (collinear) state and at zero field, reaching a maximum at some intermediate field. If the chirality of the structure or the noncollinearity of the magnetic moments is related to SOI, then this additional contribution does not obviously exceed the usual collinear AHE. However, in some cases, chirality can be created by mechanisms others than SOI, and then giant effects can be detected. For example, a giant AHE was observed in Co_2MnGa characterized by Weyl fermions [16].

The competition of various types of exchange interaction in the presence of distortions of the crystal structure can lead to non-collinear spin configurations [17] or to the formation of skyrmions in the Heusler-type $\text{Ni}_{50-x}\text{Mn}_{25+x}\text{Z}$ ($\text{Z} = \text{Ga, In, Sn, Sb}$) magnetic shape memory alloys (MSMAs) [17]. Such a competition was confirmed by the direct observations using LTEM [18,19] and MFM [20] as well as by the indirect results of AHE measurements [20]. However, when interpreting the additional field dependence of the Hall resistivity as a consequence of skyrmions, it is assumed that the AHE coefficient is field independent, which is obviously unjustified in the case of Heusler-type MSMAs undergoing martensitic transformation (MT), since the application

of the magnetic field can affect their microstructure and, therefore, can change the off-diagonal and diagonal conductivities present in Eq.(3). In particular, it was shown that the AHE coefficient R_s depends on the magnetic field in Ni-Mn-In-Si [6] and Ni-Mn-In-B alloys [8].

In the present work, a comprehensive study of the magnetic properties, resistivity, magnetoresistance, and Hall resistivity of epitaxial $\text{Ni}_{47.3}\text{Mn}_{30.6}\text{Ga}_{22.1}$ MSMA films has been carried out in a wide range of temperatures and magnetic fields, encompassing martensitic, premartensitic and austenitic regions. An analysis of the obtained data shows that the additional field dependence of the Hall resistivity is not associated with the formation of skyrmions or antiskyrmions, but with both the antiferromagnetic correlations leading to non-collinear spin structures, and the influence of the magnetic field on the electronic structure.

II. EXPERIMENTAL

A 1 μm -thick $\text{Ni}_{47.3}\text{Mn}_{30.6}\text{Ga}_{22.1}$ (at.%) film was epitaxially grown onto a MgO(001) substrate, heated at 723 K, by magnetron sputtering. Composition of the film was determined with an uncertainty of 0.5 at.% by energy-dispersive X-ray spectroscopy. X-ray diffraction (XRD) at room temperature revealed the out-of-plane 001 crystallographic orientation of the film.

Magnetization was measured in magnetic fields up to 16 kOe and temperature range of 100 - 400 K using a Lake Shore VSM. The magnetic field was applied perpendicularly to the film plane, in the same geometry as in Hall effect measurements. Magnetotransport properties were measured by a four-probe method in magnetic fields up to 20.5 kOe and temperature range of 100-400 K using a custom-made set-up. The temperature dependences of resistance were measured continuously with a cooling-heating rate of about 3 K/min with a resolution of 1 nOhm. Hall effect and magnetoresistance were determined based on a series of measurements for each specific preliminary stabilized temperature. To eliminate the parasitic contributions of MR and other effects in the Hall electromotive force, the measurements of Hall resistivity for each stabilized temperature and magnetic field value were carried out in two different directions of the magnetic field and two directions of the electric current, followed by data processing.

III. RESULTS AND DISCUSSION

Magnetization

The zero-field-cooling (ZFC), field-cooling (FC), and field-warming (FW) thermomagnetization curves, $M(T)$, measured in a magnetic field of 50 Oe are shown in Fig. 1. In the ZFC measurement, the sample was first cooled from 400 K to 70 K in a zero magnetic field and then measured on heating under 50 Oe. In the cases of FC and FW protocols, $M(T)$ was recorded during cooling and

warming the sample in the presence of a 50 Oe magnetic field. A difference between ZFC and FC curves, depicted in Fig. 1 and typically observed for MSMA, stems from the different magnetic domain structure of the film resulting from ZFC and FC protocols: the cooling without magnetic field leads to a disordered domain structure and random orientation of the martensitic variants causing an almost demagnetized state of the sample with a small magnetization, whereas the preferential orientation of domains, giving rise to elevated magnetization, takes place during cooling in the presence of magnetic field. The temperature dependence of magnetization at 50 Oe in Fig. 1 shows that the forward MT occurs at $T_M \approx 230$ K (between 250 and 210 K) and the thermal hysteresis of MT is about 13 K. The Curie temperature (T_C) in austenite, as determined from the inflection point of the $M(T)$ curve (which is a commonly used procedure) is equal to $T_C \approx 380$ K, that is expected for the alloy content [21]. Two irregularities can be found in the thermomagnetization curve under high magnetic field shown in the Inset to Fig. 1. The first one is apparent in the region of MT. The second one, at $T \approx 285$ K, can be tentatively attributed to a pre-martensitic transition (see [22] and references therein), and is further confirmed by the resistivity and magnetoresistance results below.

FIG.1. ZFC, FC, and FW temperature-dependent magnetization curves of $\text{Ni}_{47.3}\text{Mn}_{30.6}\text{Ga}_{22.1}$ thin film at $H=50$ Oe. Inset shows the temperature dependence of the magnetic moment at $H=16$ kOe.

Magnetization curves shown in Fig.S1, provided in Ref.[23], demonstrate that the coercive force (H_c) in the martensitic phase is equal to about 180 Oe and almost independent of temperature in the range $T = 110\text{--}200$ K; in the austenitic phase $H_c(300\text{ K}) \approx 25$ Oe.

Electrical resistivity and magnetoresistance

Figure 2 shows the temperature dependence of electrical resistivity of the film during cooling and heating.

FIG.2. Temperature dependence of electrical resistivity for $\text{Ni}_{47.3}\text{Mn}_{30.6}\text{Ga}_{22.1}$ thin film.

The behavior of resistivity, $\rho(T)$, in the vicinity of MT is typical of Ni-Mn-Ga alloys [21, 24, 25]. With increasing temperature, the resistivity increases in the martensitic phase, then decreases in the MT region and monotonically increases in the austenitic phase. A mismatch between cooling-heating curves is caused by not perfect control of the sample temperature at low temperatures. The temperature hysteresis of the resistivity at MT correlates well with the temperature hysteresis of the magnetization (Fig.1). The quasilinear $\rho(T)$ dependence in Fig.2 significantly bends at about 285 K indicating a smeared transition from austenite to the pre-martensitic phase. Such a behavior was previously observed for the bulk Ni-Mn-Ga [25].

Figure 3 shows the temperature dependence of MR between 100 and 400 K.

FIG.3. Temperature dependence of magnetoresistance for the $\text{Ni}_{47.3}\text{Mn}_{30.6}\text{Ga}_{22.1}$ thin film.

Figure 3 shows that MR is negative and small, less than 1% at 20.5 kOe, which is typical for Ni-Mn-Ga alloys [24-26]. MT has a weak influence on MR, since the resistivity itself (Fig. 2) changes only slightly across MT ($\approx 3\%$). Nevertheless, the peculiarity at 235 K clearly correlates with MT obtained from magnetization and resistivity behavior.

In strong fields, a deep MR minimum is observed at 360 K, which is below the Curie temperature determined from $M(T)$, $T_c \approx 380\text{K}$. The temperature of the MR minimum correlates with the start of the rapid change of $M(T)$ in the ferromagnetic phase (Fig.1). It means that the magnetic disorder increases greatly well below the Curie temperature, in the range of 360-380 K (Fig.1), a behavior that cannot be described by a Brillouin function.

The negative peak of MR at 285 K is an interesting finding of the present work, and can be an indication of a premartensitic transition at such a temperature in accordance with the results shown in Figs. 1 and 2. Generally, it is very difficult to prove the existence of this weak first-order transition, produced by a soft-mode condensation in Ni-Mn-Ga alloys, because very few physical properties exhibit clear anomalies at the transition [21, 22, 26]. After the present results, MR could be used as a very convenient method for detecting the premartensitic transition in thin films of Ni-Mn-Ga.

Anomalous Hall effect

Fig. 4 shows the field dependences of the Hall resistivity at different temperatures in the martensitic phase (110 K and 200 K), in the MT region (225 K and 230 K), and in the

austenitic phase (260 K, 280 K, and 330 K). The field dependences of the Hall resistivity are also presented in more detail in Fig.2S [23].

FIG.4. Hall resistivity for $\text{Ni}_{47.3}\text{Mn}_{30.6}\text{Ga}_{22}$ thin film at temperatures: (I) below MT; (II) across MT; and (III) above MT.

As can be seen from these figures, the field dependence of the Hall resistivity, at all temperatures except far away from MT in austenite (see, curves at 280 and 330 K), has an unusual form for ferromagnets: first, it is nonmonotonic, and second, it does not saturate. Even at 330 K, it cannot be saturated in 20.5 kOe, although the curve resembles the behavior of a typical collinear ferromagnet. Obviously, in this case, the standard method for determining the coefficients of NHE and AHE from asymptotes in weak and strong fields does not work. In our case, two possibilities arise: either one considers the NHE and AHE coefficients as field-dependent and use Eq.(1), or

assumes that the NHE and AHE coefficients are field-independent and all the features are associated with the additional term $\Delta\rho_H$ in Eq. (4).

Let us consider, first, the option of field dependent NHE and AHE coefficients. Iterative data treatment of the magnetization and Hall resistivity yields values of the NHE and AHE coefficients for Eq.(1) in different field intervals. The choice of the field intervals is explained in the Supplement Material and shown in Fig. 3S [23]. The results of this approach are depicted in Fig. 5.

Fig. 5. Temperature dependences of the NHE (a) and AHE (b) coefficients for $\text{Ni}_{47.3}\text{Mn}_{30.6}\text{Ga}_{22.1}$ thin film obtained in different magnetic field intervals. The choice of field intervals is explained in Supplement [23]. Bold lines denote curves obtained for high field interval, $8 \text{ kOe} < H < 16 \text{ kOe}$. The temperature ranges I, II and III correspond to the martensitic, premartensitic and austenitic phases, respectively.

One can see from this figure that the NHE and AHE coefficients strongly depend on the range of fields used. This effect is more pronounced between the field ranges: 0 - 5 kOe and 0.1 - 1 kOe, where they have opposite signs. The coefficients determined in the range 0 - 5 kOe, where the magnetic field-induced twin boundaries rearrangements can result in a highly non-monotonic behavior, should be excluded from consideration. In the remaining field ranges the NHE coefficient changes from negative in martensite to positive in austenite, as observed for other alloys

of this family [7, 8]. The AHE coefficient decreases with temperature in the martensitic phase and increases in austenite. Obviously, there is no correlation between the temperature behavior of the AHE coefficient and the resistivity. The relationship between the AHE coefficient, R_s , and resistivity, ρ , is usually presented in the power law form $R_s \sim \rho^\alpha$, where the power-law index $\alpha=1$ in the case of the skew scattering, whereas $\alpha=2$ in the case of the intrinsic or side-jump mechanism [1,2]. For high-resistivity alloys $\alpha=0.4$ [1]. One can easily see from Fig. 5 that none of these relations takes place. For example, the AHE coefficient in martensite decreases with increase of temperature in spite of a resistivity increasing. Therefore, to determine THE, it is not correct to assume that $R_s \sim \rho^2$, as it was done in the most papers (see, e.g., Ref.[20]).

Consider now the second option for determining the coefficients. To do this, we assume that the NHE and AHE coefficients are field independent and equal to the values determined for high fields in the range of 8-16 kOe. Then, an additional contribution to the Hall resistivity $\Delta\rho_H$ will have the form presented in Fig. 6. As seen, the extra term $\Delta\rho_H$ has a consistent behavior as a function of field and temperature, so indicating this fitting procedure as quite a realistic one.

FIG. 6. Magnetic field dependence of the term $\Delta\rho_H$ in Eq. (4) at different temperatures.

In the framework of the well-established theories [7,8,13,18] the sign of topological Hall effect (THE) is opposite to that one of NHE for skyrmions with positive vorticity, while for antiskyrmions the signs are the same, and the field dependence of THE presents a hump (or a

dome-shaped curve) with $\Delta\rho_H=0$ at $H=0$ and at $H>H_s$, where H_s is the saturation field [9, 10, 20]. Despite the observed features in the $\Delta\rho_H(H)$ dependence resemble the predicted behavior for THE produced by the skyrmions or antiskyrmions, there are serious doubts that it is the case here for three reasons. First, $\Delta\rho_H$ is negative, while, e.g., for NiMnGa ribbon it is positive [20]. Second, according to Ref.[20], $\Delta\rho_H$ produced by skyrmions is two orders of magnitude larger than in our case. And third, the temperature dependences of $\Delta\rho_H$ recorded in Ref.[20] is different than observed in the present work. One may argue that the strong disagreement between our results and those ones for THE in NiMnGa ribbons [20] might be related to the possible different types of skyrmions and their density in ribbons and films. However, to our opinion, an idea about the presence of conventional skyrmions originated from a Dzyaloshinski-Moriya-type exchange mechanism could be disregarded both in Ni-Mn-Ga films and ribbons, simply because this interaction mechanism has not been confirmed yet in these materials. On the other hand, bubble skyrmions may exist in Ni-Mn-Ga alloys [18-20], but the perpendicular anisotropy, required for a formation of this type of skyrmions, was not found in our thin film samples. Moreover, the observed features of additional contribution to Hall resistivity do not correspond to THE produced by skyrmions.

We think that the disclosed unconventional field dependences of NHE and AHE are due to both the existence of antiferromagnetic correlations and the influence of the magnetic field on the electronic structure rather than due to skyrmions. Indeed, both the sign and the amplitude of observed $\Delta\rho_H$ in our thin film samples disagrees with THE features observed for ribbons [20]. Therefore, based on previous results on the Hall effect in NiMnIn-based alloys [6-8] and on the complicated exchange interactions in Ni-Mn-Ga alloys [27, 28], we propose herewith another explanation. It is well known that an excess of Mn in the atomically ordered MSMAs, as in our case, gives rise to antiferromagnetic (AF) interactions between Mn atoms on their proper sites and those on the Ga sites [27, 28]. Such AF interactions compete with the ferromagnetic interactions

between Mn magnetic moments on their proper sites, and lead to non-collinear magnetic structures [24]. By applying a magnetic field, however, the non-collinear structures disappear. In addition, the non-collinear structures are more pronounced at low temperatures and tend to disappear gradually with the increase of temperature [24]. As can be seen in Fig. 5, the NHE and AHE coefficients determined in the range of 0 - 5 kOe have nothing to do with the same coefficients determined in high fields. They are even opposite in sign at low temperatures. It means that in this range of magnetic field and temperatures there is a quite strong additional contribution of the same order of magnitude as the conventional Hall effect resistivity. Since it is large and gradually disappears with temperature and field, we can attribute it to AF correlations and changes in the electronic structure under an applied magnetic field related to them. The influence of magnetic field on the electronic structure is clearly seen from the difference of the NHE coefficient determined in the ranges of 0 - 5 kOe and 0.1 - 1.0 kOe. It is important to specify that the electronic structure changes under the applied magnetic field not only at MT but in the whole temperature range studied. The behavior of the NHE and AHE coefficients determined in high magnetic fields, 8 - 16 kOe, are typical for MSMA alloys (bold lines in Fig.5), namely, with increasing temperature the NHE coefficient changes its sign from negative to positive value at MT, and the AHE coefficient decreases in martensitic phase and increases in austenitic phase [6-8]. Such behavior was recently discussed in detail for the case of polycrystalline Ni-Mn-In-B alloys [8].

IV. CONCLUSIONS

Magnetic and magnetotransport properties of $\text{Ni}_{47.3}\text{Mn}_{30.6}\text{Ga}_{22}$ thin films grown epitaxially on MgO (100) substrates were studied in a wide temperature range including martensitic and austenitic phases, as well as martensitic transformation. Both the NHE and AHE coefficients were determined by fitting the field dependence of Hall resistivity in different field intervals. The coefficients obtained in weak magnetic fields drastically differ from those in high magnetic fields. They are even different in sign. It means that there is an additional contribution to Hall resistivity, at least, in weak and intermediate magnetic fields. An attempt to describe this contribution as a result of topological Hall effect due to presence of skyrmions or antiskyrmions

was not successful because the obtained temperature dependence of this term and its magnitude were in contradiction with established theories and previous experiment [20]. We argued that unconventional field dependences of NHE and AHE in the considered case of magnetic shape memory alloy are due to both the antiferromagnetic correlations and the influence of the magnetic field on the electronic structure.

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