Interface-induced enhancement of the anomalous Nernst effect in ferromagnetic Mn$_5$Ge$_3$C$_{0.8}$ films

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The anomalous Nernst effect of thin bilayers comprising a thin normal metal film and a ferromagnetic Mn$_5$Ge$_3$C$_{0.8}$ film has been investigated. Epitaxial c-axis grown Mn$_5$Ge$_3$C$_{0.8}$ films have been obtained by e-beam evaporation as well as by magnetron sputtering on Ge(111) substrates. The size of the anomalous Nernst coefficient is independent of the growth method. Pt, Ta, and Pt/Cu bilayers have been used as voltage leads on top of the ferromagnetic film. We show that the interface between the normal metal electrode and the ferromagnetic film contributes significantly to the anomalous Nernst voltage which is suprisingly independent of the choice of the metal. The interface-induced enhancement of the anomalous Nernst effect is verified for samples with varying Mn content or undergoing an additional annealing step. The results suggest that the interface quality has a strong impact on the size of the anomalous Nernst effect.

I. INTRODUCTION

The interactions between charge, spin, and heat currents give rise to several thermoelectric effects that are of recent interest for future caloritronic applications. In conducting magnetic materials, the spin orbit interaction in the presence of heat current and spontaneous magnetization induces the anomalous Nernst effect (ANE). The ANE has obtained great importance in recent times due to its observation in new materials such as chiral antiferromagnets [1, 2], Weyl semimetals [3, 4], group-IV dichalcogenides [5], and many more. In the presence of an in-plane external magnetic field and an out-of-plane temperature gradient, the electric field due to the ANE is given by

\[ \vec{E}_{\text{ANE}} = -S_{\text{ANE}} \nabla T \times \frac{\vec{M}}{|M_s|} \]  

(1)

where $S_{\text{ANE}}$, $\vec{M}$, and $M_s$ are the ANE coefficient at saturation, the magnetization, and the saturation magnetization of the magnetic material, respectively. In spincaloritronics [6, 7] the same geometry with an additional spin detector strip made of a normal metal (NM) with a large spin Hall angle is used to detect the spin Seebeck effect (SSE) via the inverse spin Hall effect. On the one hand, studies show the ANE contribution to be dominant over SSE in metals [8-10]. On the other hand, a significant influence of the interface between the ferromagnetic film and the NM electrode on the ANE has been observed [11]. In alternately stacked multilayers, the ANE voltage is enhanced compared to single layers irrespective of the presence or absence of proximity induced ferromagnetism in the heavy NM [12]. Furthermore, an increase of the ANE coefficient with decreasing thickness of the ferromagnetic film or multilayer has been observed [13]. These results show a pathway towards an enhancement of the thermoelectric performance of devices and motivate our present work on ferromagnetic Mn$_5$Ge$_3$C$_{x}$ films.

Mn$_5$Ge$_3$C$_x$ is of particular interest for future spintronic devices due to its compatibility with complementary metal-oxide-semiconductor (CMOS) device technology [14, 15]. It has an uniaxial magnetic anisotropy along the c axis of the hexagonal D$_{3h}$ structure belonging to space group P6/mcm. Addition of carbon to the parent Mn$_5$Ge$_3$ compound strongly enhances the Curie temperature $T_C$. In particular the composition Mn$_5$Ge$_3$C$_{0.8}$ has the highest saturation magnetization $M_s$ with a $T_C \approx 450$ K [16, 17]. The structural, magnetic, and transport properties of this particular material have been previously investigated in detail [15, 16, 17].

In this work, we investigate the ANE of NM/Mn$_5$Ge$_3$C$_{0.8}$ bilayers on Ge(111) and explore ways to vary and increase the ANE. The Mn$_5$Ge$_3$C$_{0.8}$ films have been prepared by two different growth methods - sputtering and e-beam evaporation - to study the effect of film quality and electrical resistivity on the ANE.

II. EXPERIMENTAL DETAILS

The first step of the device fabrication involved dipping the clean Ge(111) substrate in 30% H$_2$O$_2$ for 25 s before mounting in the deposition chamber. Films were obtained by co-deposition from independent Mn, Ge, and C sources using e-beam evaporation in an ultra-high vacuum chamber with a base pressure of $10^{-10}$ mbar or magnetron sputtering in a high vacuum system with a base pressure of $10^{-7}$ mbar. The substrate was maintained at a temperature of 420°C during deposition. After deposition, the sample was allowed to cool down to room tem-
temperature. In the following step, a NM layer strip was deposited in-situ through a mechanical shadow mask with a width of 200 μm and a length l_y = 6 mm. We have used various metals such as Pt, Ta, and Pt/Cu bilayers as NM electrodes. The crystal structure was studied in a Bruker D8 Discover X-ray diffractometer with Cu Kα radiation (λ=1.542 Å). Magnetic hysteresis curves were acquired by means of the transverse magneto-optical Kerr effect (t-MOKE) employing a linearly polarized laser beam of wavelength λ = 633 nm and a Helmholtz coil providing a maximum magnetic field of B = 0.3 T.

Figure 1 shows a schematic of the device geometry. The thermovoltage was measured by Pt wires of 50 μm diameter which were glued to the ends of the NM strips with conducting silver epoxy. A 2-kΩ resistor separated by a thin Kapton foil was glued on top of the device for generation of a temperature gradient $\nabla T$. The device was inserted into a physical property measurement system (PPMS, Quantum Design) with a Keithley nanovoltmeter for voltage measurement. Resistive thermometers were connected in a separate run to determine the temperature of the top and the bottom surfaces of the chip with the same heater currents used during the thermovoltage measurement. The transverse thermovoltage $V_y$ was measured for an applied temperature gradient $\nabla T_z = \Delta T_z/l_z$ along the z-axis, where $l_z$ is the depth across which $\Delta T_z$ is measured. All measurements were performed at 300 K.

Figure 2 shows the thermovoltage vs. magnetic field of a Pt/Mn$_2$Ge$_3$Co$_{0.8}$ bilayer for five different applied heater powers. In all our experiments, $\Delta T_z$ was kept within 1.0 to 6.0 K and below 5% of the bath temperature to avoid overheating effects. Magnetic field independent offsets have been subtracted to center the curves around zero voltage. Magnetic fields up to 0.7 T were applied during the measurement in order to capture the anomalous part of the Nernst effect. At higher fields the ordinary Nernst effect dominates over the anomalous part, the result of which is not shown here. The difference between the voltages at the right and left maximum-field values $2\Delta V_y = V_y(+0.7 \text{ T}) - V_y(-0.7 \text{ T})$ shows a linear dependence on the heater power, see inset Fig. 2. $S_{yz}$ is calculated as

$$S_{yz} = \frac{\Delta V_y}{l_y} \cdot \frac{l_z}{\Delta T_z}$$

(2)

In our experiments, the 0.4 - 0.5 mm Ge (111) substrate is four orders of magnitude thicker than the ferromagnetic film and we can only measure the temperature difference between the top and the bottom surface of the entire sample. In the above expression of $S_{yz}$, we therefore use a constant scaling factor of $l_z/l_y=0.092$ for all the results discussed below.

III. RESULTS

A. Effect of deposition method

The dependence of the ANE on the substrate material has been previously investigated for magnetite where strain plays an important role [11]. Moreover, the dependence of the ANE on the crystal orientation of Fe$_3$N antiperovskite films on MgO has been investigated [20]. In this section, we discuss the effect of the deposition method on the ANE in ferromagnetic Mn$_2$Ge$_3$Co$_{0.8}$. Figure 3 shows the X-ray diffraction pattern of films with 56-nm thickness deposited by e-beam evaporation or magnetron sputtering. For both films, the (0002) and (0004) reflections of Mn$_2$Ge$_3$Co$_{0.8}$ and of Ge(111) are observed in the 2θ scans shown in Figs. 4(a) and (b). φ scans around the (1123) reflection [Figs. 4(c) and (d)] exhibit regularly
FIG. 3. X-ray diffractograms (2θ scans) of 56-nm thick (a) magnetron sputtered and (b) e-beam deposited Mn$_5$Ge$_3$C$_{0.8}$ films on Ge(111). Minor peaks originate from multiple reflections of the single-crystalline substrate. (c,d) φ scans around the [1123] direction of the films.

Peaks separated by 60° degree. This confirms the hexagonal crystal structure and the fiber texture along the crystallographic c axis of the films. It is remarkable that the magnetron sputtered Mn$_5$Ge$_3$C$_{0.8}$ film exhibits similar texture and epitaxial quality as the film prepared by e-beam evaporation when deposited on single-crystalline Ge(111).

Figure 4(a) shows $S_{yx}$ measured on a sample with a 10-nm Pt electrode on top of Mn$_5$Ge$_3$C$_{0.8}$ which almost saturates at a field of 0.7 T. For the evaporated and the sputtered films we obtain $S_{yx} = 11$ and 9 nV/K, respectively. Similar values have been reported for conventional ferromagnetic films of comparable thickness such as Co, Ni, Fe, and permalloy (Py) [13].

Fig. 4(b) shows magnetic hysteresis curves of the same films measured with t-MOKE. Apart from the fact that both magnetizations do not saturate at the highest available magnetic field of 0.2 T we do not observe a significant difference in the MOKE signal. Furthermore, the magnetization curves show a behavior very similar to the thermovoltage $S_{yx}$ in Fig. 4(a) as expected from the linear dependence of the electric field or ANE voltage on $\vec{M}$ (Eq. 1) with constant $S_{ANE}$.

We conclude that [0001]-oriented Mn$_5$Ge$_3$C$_{0.8}$ films of similar structural quality can be obtained by magnetron sputtering or e-beam evaporation on Ge(111) substrates, but the deposition method does not play a significant role for the overall thermoelectric conversion efficiency.

FIG. 4. (a) Comparison between $S_{yx}$ at 300 K for e-beam deposited and magnetron sputtered films of thickness 28 nm. (b) Magnetic hysteresis at 300 K measured by transverse-MOKE.

B. Effect of normal-metal electrode

For the investigation of the effect of the NM electrode on the ANE, we prepared three different sets of samples with different NM electrodes made from Pt, Ta, and Cu/Pt on 28-nm thick sputter deposited Mn$_5$Ge$_3$C$_{0.8}$
films. For comparison, on one sample the voltage was detected by two point contacts at the edges of the FM film on Ge(111) without a Pt electrode ($t_{\text{Pt}} = 0$). The investigated samples are listed in Table I.

Figure 5 (a) shows $S_{yz}(B)$ for Pt electrodes. $S_{yz}$ is strongly enhanced by one order of magnitude after deposition of only 2 nm Pt on Mn$_5$Ge$_3$C$_{0.8}$ and further increases with increasing Pt thickness to a maximum $S_{yz}$ = 19.02 nV/K for $t = 12$ nm and drops again to $S_{yz}$ = 5.93 nV/K for $t = 20$ nm. The dependence of $S_{yz}$ at ±0.7 T on the Pt thickness is shown in the inset of Fig. 5. The enhancement of $S_{yz}$ caused by the deposition of 2 nm Pt is not due to the SSE effect, which is negligibly small, as discussed below for Pt/Cu electrodes. Considering the sheet resistances $R = \rho/t$ (\rho: resistivity) of the Pt strips at 300 K (Table II), one would expect the highest thermovoltage from the film with 2 nm Pt layer since this particular strip exhibits the highest sheet resistance. However, among the samples with Pt electrodes, a nonlinear dependence of $S_{yz}$ on the Pt thickness is observed, see inset Fig. 5(a), similar to previously reported behavior observed for bilayers of Pt as well as Cu on Py [11]. In particular, $S_{yz}$ drops at $t_{\text{Pt}} = 10$ nm where the resistivity $\rho$ of Pt drops as well. Furthermore, in multilayers of Pt/CoFeB, a maximum of the transverse thermovoltage was observed for a Pt thickness of 2 nm [21]. In contrast, we observe a maximum of $S_{yz}$ at a Pt thickness of 12 nm. Similar nonlinear dependencies on the NM layer thickness have been also observed for Ta electrodes or Cu(t)/Pt(10 nm) bilayers, see Table I. The sample with a Cu(6 nm)/Pt(10 nm) bilayer strip exhibits the highest thermoelectric voltage $\propto S_{yz}$ but again drops to a smaller value for a total thickness of 20 nm similar to the behavior of Pt electrodes. In contrast, for Ta the thermovoltage drops at $t_{\text{Ta}} = 10$ nm but rises again for $t_{\text{Ta}} = 20$ nm.

Our devices are NM/FM bilayers. Any metal strip used as a voltage lead forms a parallel resistance circuit with the FM film. In order to compare samples with different NM electrodes, the coefficient $S_{yz}$ is usually scaled with the total sheet resistance $R_0$ at the lead $R_0$ is written in terms of the sheet resistances of the FM film $R_0$ and the contact NM layer (or layers) $R_{\text{NM}}$: $R_0^{-1} = R_{\text{FM}}^{-1} + R_{\text{NM}}^{-1}$. The sheet resistance of 28-nm Mn$_5$Ge$_3$C$_{0.8}$ films is $R_{\text{FM}} = 100 \Omega$ and in the following we neglect the contribution from the Ge substrate with a sheet resistance of 910 $\Omega$ at room temperature. In order to eliminate the contribution from the different intrinsic resistivities of the NM, $\rho$ of these NM strips were measured and the ANE was scaled with the total sheet resistance $R_0$ as shown in Figs. 5(b-d) and Table I. Except for the 20-nm Pt electrode we now ob-

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**FIG. 5.** (a) $S_{yz}$ vs. magnetic field for sputtered Pt(t)/Mn$_5$Ge$_3$C$_{0.8}$ bilayers. The data of a single Mn$_5$Ge$_3$C$_{0.8}$ film ($t_{\text{Pt}} = 0$) are scaled by a factor 5. The inset shows the Pt thickness dependence of $S_{yz}$ (left) and $\rho$ (right). (b) $S_{yz}/R_0$ scaled with the effective sheet resistance $R_0$ of the NM layers. The inset shows the monotonic increase of $S_{yz}/R_0$ with total Pt thickness. (c,d) $S_{yz}/R_0$ for Pt/Cu and Ta NM-electrodes, respectively. Insets show the dependence of $S_{yz}/R_0$ on the total NM thickness.
serve a monotonic increase of the ANE with increasing total NM thickness, irrespective of what NM material we use, see insets of Figs. (b-d). This confirms that the main reason for the nonlinear thickness dependence of $S_{yz}/R_0$ is the thickness dependence of the NM sheet resistance or resistivity, as observed, for instance, for Pt in Fig. 4a) (inset). The stronger-than-linear increase of $S_{yz}/R_0$ with the total NM thickness is a consequence of the electronic transport in thin films described by the Fuchs-Sondheimer theory [23,24]. The resistivity of thin films increases with decreasing thickness $t$ which gives rise to an additional increase of $S_{yz}/R_0 \propto t/\rho(t)$.

For the film where the voltage was detected by two point contacts at the edges of the FM film on Ge(111) instead of on strips, see Fig. 5(b) for point contacts at the edges of the FM film on Ge(111) sheet resistance of the metallic bilayer.

| NM Material | $t_{NM}$ (nm) | $R_0$ (Ω) | $S_{yz}$ (nV/K) | $S_{yz}/R_0$ (nV/Ω K) |
|-------------|---------------|-----------|----------------|---------------------|
| Pt          | 0             | 123       | 0.7            | 0.0057              |
| Pt          | 2             | 84.5      | 5.07           | 0.06                |
| Pt          | 6             | 60.2      | 14.44          | 0.24                |
| Pt          | 10            | 25.9      | 9.08           | 0.35                |
| Pt          | 12            | 30.2      | 19.02          | 0.63                |
| Pt          | 20            | 12.1      | 5.93           | 0.49                |
| Ta          | 3             | 184       | 33.07          | 0.18                |
| Ta          | 10            | 132.9     | 22.6           | 0.17                |
| Ta          | 20            | 79.1      | 37.07          | 0.48                |
| Cu/Pt(10 nm)| 2             | 31.3      | 28.15          | 0.45                |
| Cu/Pt(10 nm)| 6            | 27.5      | 30.3           | 0.55                |
| Cu/Pt(10 nm)| 10           | 10.5      | 22.55          | 1.07                |

TABLE I. $S_{yz}$ and $S_{yz}/R_0$ values for all the films. $R_0$ is the sheet resistance of the metallic bilayer.

and increases monotonically with the total NM thickness. Experiments on Cu/Pt bilayers have shown that the surface contribution to the ANE is enhanced after deposition of Cu on bare Pt [11]. Hence, in our case, it is likely that the surface conditions are modified after deposition of the NM layer generating an ANE by spin-orbit scattering at the interface. In order to support our assumption, we prepared three distinct devices to study of the effect of composition and sample treatment on the ANE. One device was prepared by using the standard recipe and was annealed in-situ at 400°C for 15 minutes after deposition. For the second and third device, the power of the Mn sputtering target was varied so that the samples contain 10% less or more Mn with respect to the sample discussed above. X-ray diffraction diagrams (not shown) confirm that the crystal structure is retained in all three devices and no additional diffraction peaks were observed. The $S_{yz}$ of these devices is shown in Fig. 6 together with the result of a standard device of Table I. After annealing, the ANE coefficient shows a three-fold enhancement. This indicates an improved atomic order of the Mn$_5$Ge$_3$C$_{0.8}$ alloy and suggests that the structural order of the surface-interface significantly affects the ANE. The alloys with different Mn content exhibit much lower ANE coefficients due to the deviation of the composition from stoichiometry. These results show that with a proper selection of the FM stoichiometry and NM thickness, an interface can be engineered which gives rise to a large ANE voltage. Once the selection of the materials is accomplished, the ANE voltage can be further enhanced by constructing a multilayer instead of a bilayer. Besides, Mn$_5$Ge$_3$C$_{0.8}$, extensive transport studies have been carried out on Mn$_5$Ge$_3$ and Mn$_5$Si$_3$C$_{0.8}$ films [15] which may also serve as candidates for CMOS-compatible thermoelectric devices.
IV. CONCLUSION

The anomalous Nernst effect been studied in thin films of ferromagnetic Mn$_5$Ge$_3$C$_{0.8}$ on Ge (111) covered by different normal metal strips. Measurements have been performed in the transverse configuration with the magnetic field oriented in the film plane and a temperature gradient perpendicular to the plane. We observe a strong enhancement of the ANE by using thin Pt or Ta electrodes on Mn$_5$Ge$_3$C$_{0.8}$ compared to samples without a metallic electrode. The ANE can be further enhanced by insertion of a Cu layer between the normal metal and Mn$_5$Ge$_3$C$_{0.8}$. The enhancement is attributed to the modified surface conditions facilitating the generation of an ANE signal irrespective of the material choice of the NM.

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