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Study of the Nernst-Ettingshausen effect in thin Co/Pt layers at room temperatures

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The results of measurements of the anomalous Nernst-Ettingshausen effect in thin metallic CoPt films formed on various substrates are presented. The similarity of the magnetic field dependences of the Hall and Nernst-Ettingshausen effects is demonstrated. It is assumed that such metallic structures are promising for practical application as thermomagnetic energy converters, and can also be useful for research in the field of thermal spintronics.

Keywords: Co/Pt, thin metal film, Nernst-Ettingshausen effect, Hall effect, Seebeck effect.

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1. Introduction

Thermoelectric Seebeck effect is a phenomenon where a votage is generated between the material faces under the action of temperature gradient. The Seebeck effect is the basis of the operating principle of thermoelectric generators that convert thermal energy into electrical energy. These materials are required for aggressive operating conditions where electric power generation using „classical“ methods is difficult or fundamentally impossible [1]. The coefficient of proportionality between the generated voltage and created temperature gradient is call the Seebeck coefficient. Thermoelectric conversion efficiency is described by the coefficient $ZT = \alpha^2 T/\rho\lambda$ (thermoelectric figure of merit, where α is the Seebeck coefficient (voltage emerging at a temperature difference of 1 K), ρ is the resistivity, λ is the thermal conductivity coefficient). A material is considered to be promising for thermoelectric applications provided that: $ZT \geq 1$. Basic materials for thermoelectric generators are heavily-doped semiconductors with the figure of merit at the level of 1 in various temperature ranges: in a low-temperature region 300−500 K — Bi_2Te_3 [2], in a medium-temperature region 500−700 K — GeTe [3] and in a high-temperature region 600−900 K — Ge_xSi_{1-x} solid solutions [4].

Metals have a low α, therefore, despite a number of obvious advantages of metals as compared to semiconductors with high electric conductivity and radiation resistance being the main of which, these materials are not used in thermoelectric generators, with the only exception for temperature sensors — thermocouples. One of the methods to increase the thermoelectric figure of merit of metals is using thermomagnetic effects that occur in ferromagnetic materials [5]. Of particular interest is the anomalous Nernst-Ettingshausen effect exhibited in thin-film systems with high magnetic anisotropy where transverse or longitudinal difference of potentials occurs with respect to the temperature gradient direction when a sample is exposed to an external magnetic field (transverse and longitudinal effects, respectively).

The objectives of the work were to investigate the Nernst-Ettingshausen effect in thin ferromagnetic Co/Pt films in a practically interesting temperature range near room temperature and to examine the correlation between this phenomenon and the structure magnetism. To achieve the above-mentioned objective, the following problems were solved: developing a measurement configuration to provide simultaneous recording of the Hall and Nernst-Ettingshausen effects; measurement of thermomagnetic and galvanomagnetic effects in similar conditions, comparison of findings concerning the magnetic field dependences of the Hall and Nernst-Ettingshausen voltages.

The paper provides the results of the study of the transverse thermomagnetic Nernst-Ettingshausen effect in thin multilayer Co/Pt films. Earlier in [5], the authors investigated the thermomagnetic Nernst-Ettingshausen effect in a film consisting of ten $[\mathrm{Co}(3\,\text{Å})/\mathrm{Pt}(5\,\text{Å})]_{10}$ layers. It was shown that the anomalous effect was present in a wide temperature range of 10−300 K. The magnetic field dependence of the Nernst-Ettingshausen voltage was in the form of a hysteresis loop. As the temperature increased, the loop width decreased, however, the Nernst-Ettingshausen voltage amplitude increased, which arouses interest in the study of the thermomagnetic effect in this film at temperatures higher than room temperature. Note that the study of the effect particularly above room temperature is of the greatest practical interest because most instrument applications of thermoelectric materials are implemented for this temperature range.

2. Summary of galvanomagnetic and thermomagnetic phenomena

It is known that when electric current I is passed through a sample, a transverse difference of potentials will be induced in the external magnetic field B — the Hall voltage U_H defined as [6]:

$$R = U_H/I = R_H B + R_{AH} M(B), \quad (1)$$

where R is the Hall coefficient, R_H, R_{AH} are the normal and anomalous Hall constants, respectively, $M(B)$ is the field dependence of the sample magnetization. The anomalous component is induced by the presence of spin-dependent scattering of free charge carriers in the sample which leads to distortion of the linear magnetic field dependence $R(B)$. For some materials, particularly for complex ferromagnetic alloys, $R_H \ll R_{AH}$ is satisfied, the form of curve $R(B)$ in this case is defined by the form of $M(B)$ of the film. A similar inequality may be also true for simple ferromagnetic materials, for example, Ni [7].

To observe the Nernst-Ettingshausen effect on the opposite faces of a material, the temperature gradient ΔT shall be provided, which in turn will induce a free charge carrier flow from a „hot“ face to a „cold“ one and vice versa. Similar to the Hall effect, the external magnetic field induces the transverse Nernst-Ettingshausen voltage U_{NE} defined as follows [8]:

$$Q = \frac{U_{NE}}{\Delta T} = Q_{NE} B + Q_{ANE} M(B), \quad (2)$$

where Q is the Nernst-Ettingshausen coefficient, Q_{NE} and Q_{ANE} are the normal and anomalous Nernst-Ettingshausen constants, respectively. From equation (2) it follows that, similar to the anomalous Hall effect (1), the form of the Nernst-Ettingshausen magnetic field dependence is defined by the form of $M(B)$ suggesting that $R(B)$ and $Q(B)$ are similar.

Also similar to the anomalous Hall effect, the anomalous component of the Nernst-Ettingshausen effect in complex ferromagnetic alloys may be much higher than the normal thermomagnetic effect. For example, observations of high anomalous thermomagnetic effect in Fe_3GeTe_2 [9], CoFe [10], MnSb [11], $Li_{0.9}Mo_6O_{17}$ [12], L_{10}–FePt [13], Fe_3O_4 [14] thin films are reported. The anomalous component of the thermomagnetic Nernst-Ettingshausen effect was described by the authors earlier in [5].

3. Formation of thin-film structures for investigation of thermomagnetic effects

An investigation object in the present paper was CoPt film formed on various substrates above the pre-deposited thin (~ 1 nm) Al_2O_3 layer by alternate evaporation of Pt (5 Å) and Co (3 Å) layers using the electron beam deposition in vacuum. The total amount of [Co/Pt] layers was 10, thus, the total CoPt film thickness was ~ 8 nm. Layer thicknesses were controlled during sputtering using the quartz thickness sensor. A pre-formed thin aluminum oxide layer is necessary to prevent the cobalt atom diffusion into the substrate [15]. The film composition corresponded to a sample investigated by the authors in [5] in the temperature range of 10–300 K.

For this study, two samples with the same composition were prepared on semi-insulating gallium arsenide substrates,$\langle 100 \rangle$, and silicon substrates, $\langle 100 \rangle$, whose resistivity was higher than 3000 Ohm $\cdot$ cm. Resistance of substrates is much higher than that of films, therefore, kinetic phenomena induced by external impact may be considered as taking place only in the metal film. The substrates were chosen because of their availability, adequate roughness and resistivity, and the values of the thermal conductivity coefficient λ. Thus, the thermal conductivity of a silicon wafer is three times as high as that of gallium arsenide at room temperature: $\lambda_{GaAs} = 55\,W/m \cdot K$ [16], $\lambda_{Si} = 155\,W/m \cdot K$ [17]. Since the film thickness is much smaller than the substrate thickness, and the thermal conductivity coefficient is relatively high, it is the substrate that governs heat transfer processes.

Previously in [18], the authors performed phase and structural investigations of samples with similar composition by the X-ray diffraction method using the Bruker D8 Discover diffractometer. The investigations found that the individual Co and Pt layers in $[Co(3)/Pt(5)]_{10}$ type films are strongly mixed with one another to form a Co_xPt_{1-x} solid solution with periodically varying composition $x \sim 0.45$.

4. Experiment procedure

To study galvanomagnetic and thermomagnetic effects, a special contact layout was created in the formed samples by the photolithography method as shown in Figure 1, *a*.

The lithographic picture was formed by reactive ion etching (Ar^+), as a result four structures of various functional purpose were formed on a single substrate on the basis of a single film: structures *1–2, 7–8* — served as thermoresistors (length — 1 mm, width — 250 μm), system of contacts *3–6* — test sample (distance *3–6* — 7 mm, distance *4–5* — 4.5 mm, strip width — 1 mm), structure *A–B* — meander-shaped heating resistor. Photographic mask is shown in Figure 1, *b*, and the film formed on the *i*-GaAs substrate is shown in Figure 1, *c*. To evaluate the influence of etching on the electrophysical properties

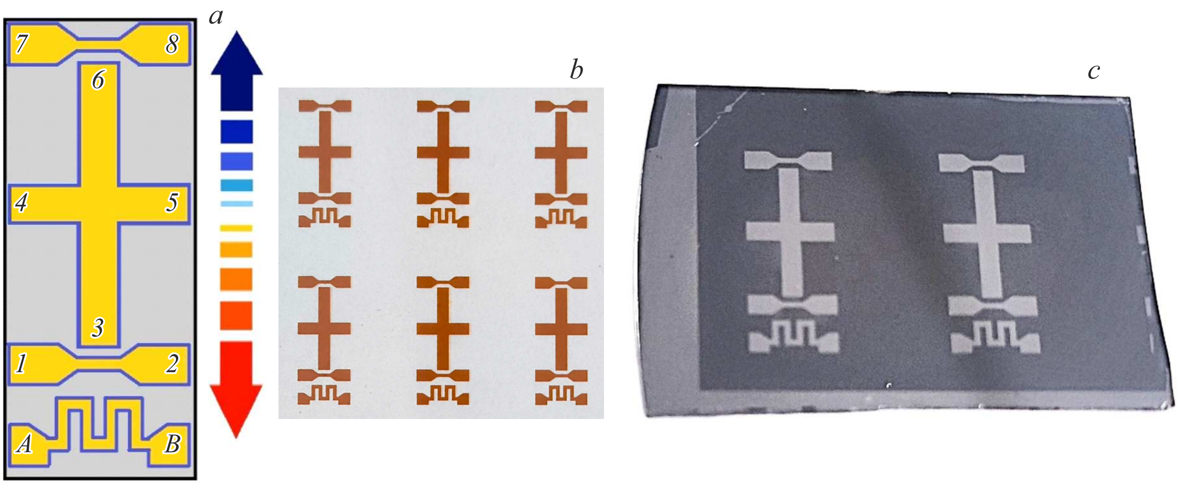

Figure 1. *a* — film topology for measuring galvanomagnetic, thermoelectric and thermomagnetic effects: *1–2*, *7–8* — thermoresistors, *3–4–5–6* — Hall and Nernst-Ettingshausen effect measurement contacts, *A–B* — heating resistor; *b* — photographic mask; *c* — photo of a lithographic picture sample.

of the film, film resistivity was measured before and after etching on a separate wafer using a standard Van der Pauw four-contact configuration. Resistivity before etching was equal to $(5.650 \pm 0.003) \cdot 10^{-5}$ Ohm $\cdot$ cm, resistivity after etching was equal to $(5.652 \pm 0.003) \cdot 10^{-5}$ Ohm $\cdot$ cm, which coincide within the limits of error. Integral resistance of thermoresistors was ~ 350 Ohm, resistance of heating resistor was ~ 1.5 kOhm.

Note that the above-mentioned form of sample *3–6* includes symmetrically placed contacts *4* and *5* which are aligned strictly perpendicular to the created temperature gradient, thus, providing zero temperature difference between these contacts. In this case, the recorded Nernst-Ettingshausen voltage is not affected by „parasitic“ components such as contact thermal voltage between the conductors and sample, and bulk thermal voltage of the sample.

Measurements were carried out in an electromagnet connected to the computerized voltage source. Magnetic field sweep was performed in the range of ± 150 mT. Magnetic field induction was recorded by a magnetic field sensor based on indium arsenide using the L-CARD E14-140MD computerized data acquisition system.

To measure the thermomagnetic Nernst-Ettingshausen effect in the direction of contact line *3–6*, a temperature gradient was created by passing electric current through the heating resistor. The resistor was heated using the Keithley 6221 current source. The temperature gradient was recorded by measuring thermoresistor resistance variation and comparing this variation with the calibration curve on the basis of the temperature coefficient of resistance. Temperature gradient direction is shown schematically by the colored arrow in Figure 1, *a*. Temperature coefficients of resistance (TCR) of heater *A–B* and thermoresistor *7–8* are the same because both elements are formed from the same CoPt film. TCR was calculated from an additional experiment by analyzing the temperature dependence of the resistance, the estimated value wasequal to $(4.341 \pm 0.013) \cdot 10^{-3}\,K^{-1}$. Thermoresistor magnetoresistance was evaluated experimentally and a value not higher than 0.01% was obtained suggesting that the magnetic properties of CoPt have a minor influence on the resistance variation measurement accuracy when exposed to heating. The temperature measurement accuracy was ~ 0.1 K. To increase the gradient, the opposite face of the substrate with the formed contact system was attached to a solid copper radiator. Typical values of the temperature gradient were $\sim (0.5{-}5.0)\,^{\circ}$C which was defined by the thermal conductivity coefficient of the substrate, on which the film was formed. The voltage generated in the sample was recorded by the Keithley 2401 current/voltage source. Measurements were performed in the room atmosphere at an ambient temperature of 300 K.

When measuring the magnetic field dependence of the Hall effect, the heating resistor stayed on, current was passed between contacts *3* and *6*, and the Hall voltage was recorded between contacts *4–5*. Such scheme allowed the Hall and Nernst-Ettingshausen voltages to be recorded at the same mean temperature of the sample. Thus, the experimental magnetic field dependences of both effects may be compared correctly.

5. Results

Figure 2 shows the experimental magnetic field dependences of the Hall resistance of the test samples at 300 K. The legend indicates the substrates on which the samples were formed: *i*-GaAs is semi-insulating gallium arsenide, Si is silicon.

The measured magnetic field dependences of the Hall resistance are in the form of a hysteresis loop, which is

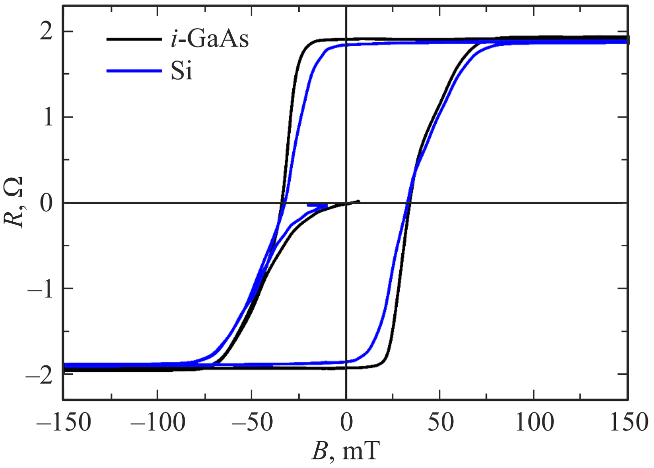

Figure 2. Experimental magnetic field dependences of the Hall resistance at 300 K of the films formed on the semi-insulating gallium arsenide substrates (i-GaAs, black curve) and silicon substrates (Si, blue curve).

indicative of the ferromagnetic properties in the investigated films. The curves adequately correspond to one another Note that the curves are in good agreement with the results obtained earlier [5], which is indicative of high sample-to-sample repeatability of composition and, consequently, of magnetic properties of the films.

Figure 3, *a* and *b* shows comparison of magnetic field dependences of the Hall resistances (HALL) and Nernst-Ettingshausen voltages (NE) of the films on the gallium arsenide and silicon substrates, respectively. The temperature gradient was created by passing the current of 20 mA through the heating resistor. The mean temperature of samples during the measurement of the magnetic field dependence of the Nernst-Ettingshausen voltage was 345 K. Temperature difference between the opposite film faces on gallium arsenide and silicon was equal to (1.35 ± 0.10) K and (0.55 ± 0.10) K, respectively.

The magnetic field dependences of the Hall resistance and Nernst-Ettingshausen voltage are similar for both investigated structures, which correlates with the above-mentioned theory of proportionality of the anomalous component of both effects to the magnetization. $U_{NE}(B)$ of the film formed on Si (Figure 3, *b*, black curve) is more than twice as low as that of the film formed on i-GaAs (Figure 3, *a*, black curve). Since the thermal conductivities of silicon and gallium arsenide are different, within the same heating power of the heating resistor, the temperature gradient on the silicon substrate is lower than that on gallium arsenide substrate, as was expected. To compare the thermomagnetic effect between two films, Figure 4, *a* shows magnetic field dependences of the Nernst-Ettingshausen coefficient recalculated using equation (2), Figure 4, *b* shows magnetic field dependences of the Hall resistance.

Figure 4, *a* shows that the magnetic field dependences of the Nernst-Ettingshausen coefficient for both samples are close to each other which is similar to the magnetic field dependences of the Hall resistances (Figure 4, *b*). Therefore the substrate has a little influence on the thermomagnetic effect. The main substrate's influencing factor that affects the measurements is probably the thermal conductivity that governs the magnitude of the created temperature gradient provided the other conditions of heat flux supply and removal are equal. From the practical point of view, it is reasonable to use thin substrates with low thermal conductivity coefficient. Such approach will make it possible to increase the temperature gradient and, thus, to increase the thermomagnetic potential, and to prevent the substrate conductivity that occurs inevitably during further heating of the entire structure. It is reasonable to use ultrathin substrates also from practical point of view because they provide higher thermomagnetic potentials due to a higher temperature gradient.

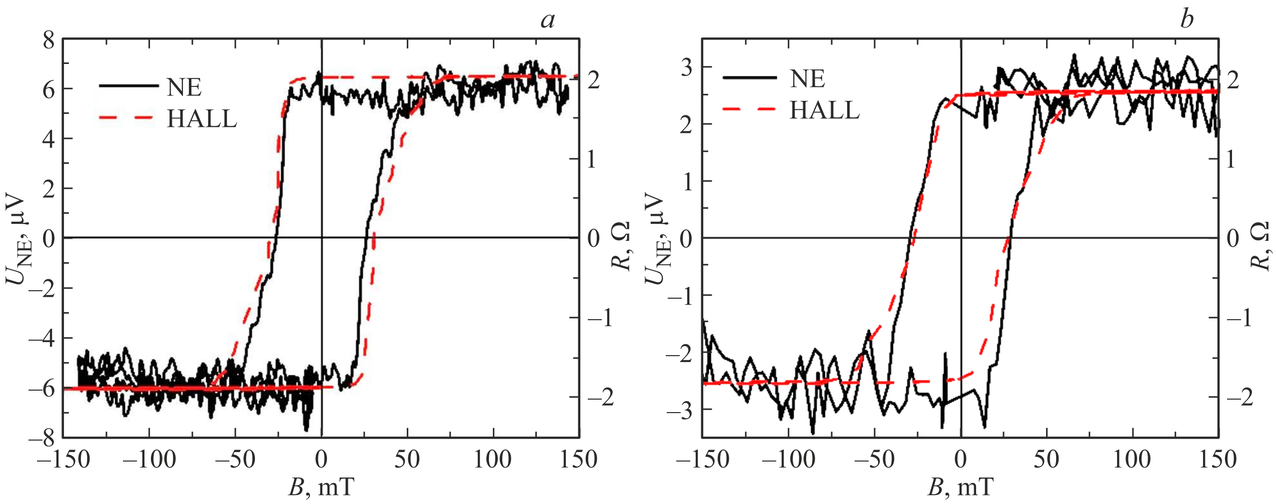

Figure 3. Comparison of the experimental magnetic field dependence of the Hall resistance and Nernst-Ettingshausen voltage of the film formed on the i-GaAs (a) and Si (b) substrates.

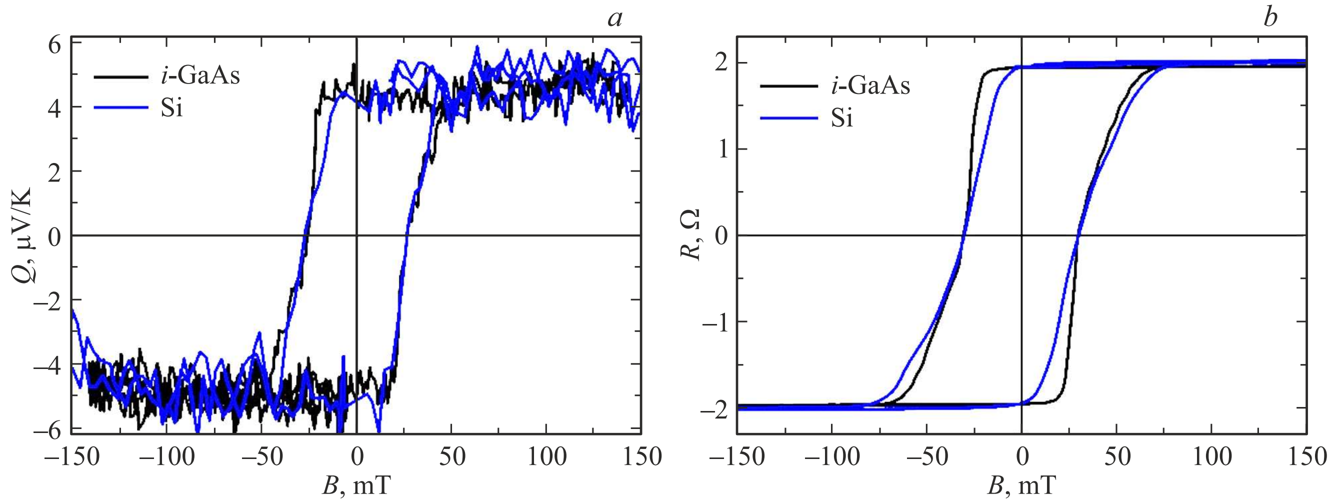

Figure 4. Comparison of the experimental magnetic field dependences of the Nernst- Ettingshausen coefficients (*a*) and Hall resistances (*b*) of the test samples at the mean temperature of 345 K.

From comparison of the magnetic field dependences of the Hall resistance of the samples measured at room temperature (Figure 2) and 345 K (Figure 4, *b*), it can be seen that, as the temperature increases, the coercive field strength decreases, whereas effect amplitude is growing.

It is remarkable that, according to the form of magnetic field dependences of the Nernst-Ettingshausen coefficient, one can draw a conclusion that the film may be considered as a „thermal memory cell“. The structure state can be „recorded“ without an external magnetic field, whereby the temperature gradient, rather than some voltage source, serves as an external power source. This experiment is a remarkable demonstration of thermal spintronics — an area that has been actively discussed in the literature since 2008 [19].

6. Conclusion

The paper describes the results of the investigation of galvanomagnetic and thermomagnetic properties of Co/Pt multilayer thin films formed on the semi-insulating gallium arsenide and silicon substrates. High process repeatability of the film properties is demonstrated regardless of the selected substrate. It was found that, as the temperature increased with respect to room temperature, magnetic field dependences of the considered effects maintain the form of hysteresis loop, but with reduced coercive force. Strong influence of the substrate's thermal conductivity on the temperature gradient formed throughout the structure was detected. Further efforts are suggested to be made for investigating the thermomagnetic effect at a higher temperature in Co/Pt thin films deposited on the substrates that have extremely low thermal conductivity coefficient.

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Conflict of interest

The authors declare that they have no conflict of interest.

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