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Phys. Rev. Lett. **107**, 216604 — Published 17 November 2011

DOI: 10.1103/PhysRevLett.107.216604

Intrinsic spin-dependent thermal transport

S. Y. Huang[1,2†], W. G. Wang[1], S. F. Lee[3], J. Kwo[2,4], and C. L. Chien[1,3,4†]

1. Department of Physics and Astronomy, the Johns Hopkins University, Baltimore, Maryland 21218, USA
2. Department of Physics, National Tsing Hua University, Hsinchu 300, Taiwan
3. Institute of Physics, Academia Sinica, Taipei 115, Taiwan
4. Center for Condensed Matter Sciences, National Taiwan University, Taipei 106, Taiwan

Abstract

Most studies of spin caloritronic effects to date, including spin Seebeck effect, utilize thin films on substrates. We use patterned ferromagnetic thin film to demonstrate the profound effect of a substrate on the spin-dependent thermal transport. With different sample patterns and on varying the direction of temperature gradient, both longitudinal and transverse thermal voltages exhibit asymmetric instead of symmetric spin dependence. This unexpected behavior is due to an out-of-plane temperature gradient imposed by the thermal conduction through the substrate and the mixture of anomalous Nernst effects. Only with substrate-free samples have we determined the intrinsic spin-dependent thermal transport with characteristics and field sensitivity similar to those of anisotropic magnetoresistance (AMR) effect.

PACS numbers: 72.15.Jf , 72.20.Pa, 85.80.-b, 85.75.-d

Thermally induced electron transport in ferromagnetic materials has attracted a great of attention recently [1-12]. The difference in the chemical potentials of the spin-up and the spin-down electrons can cause a pure spin current without the accompaniment of a charge current. On the heel of spintronics, we now have "spin caloritronics", where one exploits the interaction between heat transport and the charge/spin degree of freedom [1, 2]. Devices that manipulate pure spin currents can be highly beneficial to traditional charge-based electronics, which has been plagued by Joule heating as the size of the devices continues to shrink. While experimental [3-9] and theoretical studies [10-12] involving thermoelectricity and magnetoelectronics have been rapidly advancing, some key aspects remain poorly known.

In thermoelectrics, the charge current driven by a temperature gradient ∇T is balanced by a backflow current produced by an electric field E, which is measured as the thermopower, or Seebeck coefficient, $S = E/|\nabla T|$. The conservation of charge current density J_e and the heat current density J_Q in the presence of a temperature gradient in a non-magnetic sample can be expressed as

$$J_e = \sigma \bullet E + \sigma S \bullet (-\nabla T)$$

$$J_Q = \sigma S T \bullet E + \kappa \bullet (-\nabla T) \qquad (1)$$

with σ and κ as the electrical and thermal conductivity respectively [13] and with more complicated expressions for magnetic samples [14, 15]. Along with the Onsager reciprocal relations, the different transport coefficients can be linked through the Mott relation, $S = \frac{\pi^2 k_B^2 T}{3e\sigma}(\frac{\partial \sigma}{\partial E})_{E_F}$, where the Seebeck coefficient S is related to the energy (E) derivative of σ at the Fermi level [16, 17]. Comparing with electrical measurements, it is usually more challenging to measure the weaker heat currents and to establish the temperature gradient.

Recently, spin Seebeck effect has been reported [3, 4], where a thermal gradient in a ferromagnetic metal generates a pure spin current, which is detected by Pt strips via the inverse spin Hall effect. In addition, spin Seebeck effect has also been reported in ferromagnetic semiconductors [5] and insulators [6], although not without complexities and subtleties. For example, the spin voltage in the metal has been detected over macroscopic lengths, many times the spin diffusion length [3-5]. This dilemma might be resolved by the transmission of spin current by magnons [18]. In the spin Seebeck study using GaMnAs/GaAs, when the GaMnAs film was intentionally cut (but leaving the GaAs substrate intact) thus blocking all currents, the spin accumulation persisted [5]. It has been argued that the spin Seebeck effect is greatly enhanced by phonon drag through the substrate [8], thus suggesting the pivotal role of the substrate.

The complexity of the spin Seebeck effect notwithstanding, even the simpler spin dependent thermal transport in ferromagnetic metals remains poorly known. Another important issue in thermal transport is the temperature gradient, which is often taken to be dictated by the locations of heat source/sink. In this work, we systematically study the interplay between thermoelectricity and spin dependent transport using different sample geometries and temperature gradients. We show that spin dependent thermal transport can be dramatically altered by the presence of the substrate and masked by other spin thermoelectric effects. The temperature gradient can be quite different from that intended. The studies of intrinsic properties require substrate-free samples, with which we have determined the intrinsic spin dependent thermal transport in ferromagnetic metals.

Similar to previous spin transport studies, we have used patterned ferromagnetic thin films on substrates. The pemalloy (Py) films, 20-300 nm in thickness, have been deposited at room temperature by magnetron sputtering on Si substrates 500 μm in thickness with a 1 μm thick surface oxide layer. The wire and Hall bar samples have been patterned by photolithography with a length of about 5 mm and widths of 50 – 100 μm. We have placed a 100-Ω heater (3 mm x 1.5 mm) and a Cu block heat sink 4 mm from the two ends of the wire sample intended to create a uniform temperature gradient ∇T along the wire. We used thermal grease to improve thermal contact. We have used a step-heating method to generate the temperature gradient and measured the thermal voltage at room temperature by a nanovoltmeter after a stabilization time of about 30 minutes. A magnetic field up to 0.3 T aligns the magnetization in the film plane and at an angle θ with respect to the wire direction. The measured voltage across the length of the Py wire is $V_{th} = V_o + \Delta V_{th}(H,\theta)$, where V_o includes the ordinary thermal voltage across the contacts and $\Delta V_{th}(H,\theta)$ is the dependence of the spin-dependent thermal voltage on magnetic field H applied at angle θ.

We denote the sample plane as the *xy*-plane with the wire direction as the *x*-axis. The schematic of a Py wire sample 5 mm in length with two 45° segments is shown in Fig. 1(a). Ordinary thermal voltage causing a constant offset may be eliminated when the two 45° segments are also Py. The open-circuit thermal voltage, corresponding to the longitudinal signal V_x, has been measured across the two ends of the Py wire when a uniform ∇T_x has been set up by a heater power of about 1 W. At different angle θ, V_x varies systematically and asymmetrically with H [Fig. 1(b)] with the saturating V_x exhibiting a sinθ dependence [Fig. 1(c)]. The sinθ behavior has been consistently observed in Py wires of different lengths, in samples with two measuring segments at angles other than 45°, and those with Cu as the measuring segments. The sinθ behavior depends on the existence of a temperature gradient and ferromagnetic thin film.

We have also used a 5-mm long Hall bar sample, shown in Fig. 2(a), to measure the transverse voltage V_y at three locations (hot, middle, and cold) with ∇T_x along the length. At all locations, V_y shows an asymmetrical field dependence. As shown in Fig. 2(b), there is no sign change in V_y at the hot and the cold ends; the magnitude of $\Delta V \approx 0.2$ μV is linearly proportional to the temperature gradient. In the spin Seebeck effect studies of GaAsMn/GaAs, the voltage also shows no sign change at the hot and the cold sides [3]. This has been interpreted as a mixture of spin-Seebeck and planar Nernst effect [3], and the latter provides no sign change in V_y. This possibility is not applicable here. When the heater has been moved closer to the voltage leads to less than 2 mm, a reversal of V_y occurs on the hot side as shown in the Fig. 2(d). As V_x in the wire samples, V_y is also asymmetric in H with an angular dependence of sinθ as shown in Fig 2(c). The 90° phase shift between the two measurements merely reflects the difference in V_x and V_y. Thus both patterned wire and Hall bar consistently exhibit the sinθ dependence, which does not originate from the planar Nernst effect, which has the sin2θ dependence. [19]

However, this robust sinθ could not be the intrinsic spin-dependent thermal transport for it violates symmetry; it implies opposite thermal transport for spin orientation in the +y and the -y directions. This casts doubt on the notion of an in-plane temperature gradient. Interchanging the heater and the heat sink causes a sign change of the slope of voltage vs. power as expected as shown in the inset of Fig. 1(c) and Fig 1(d), but the same sinθ dependence remains as shown in Fig. 1(d). Thus the temperature gradient that causes the observed transport could not be in the film plane. To demonstrate perpendicular temperature gradient, we have made measurements on the same sample, one with the heater placed on the top side of the substrate and the other on the bottom side. As shown in Fig. 3, this change of heater position causes a sign change of V_y at the hot sides, where the heater is located, but not the cold side as shown in Fig. 3(a) and 3(b). Thus, the actual temperature gradient ∇T_Z responsible for measured transport is *perpendicular* through the film thickness. The larger signal in the results shown in Fig. 3(a) is simply due to the proximity of the heater.

With the perpendicular temperature gradient ∇T_Z established, the observed results of Py films on substrates can be well accounted for by the anomalous Nernst effect for ferromagnetic materials as described by

$$\vec{\nabla} V_N = -\alpha \hat{m}_1 \times \vec{\nabla} T_Z \quad (2)$$

where α is the anomalous Nernst coefficient, and $\hat{m}_1$ the unit vector of magnetization. Since $\hat{m}_1$ is in the *xy*-plane at different angle θ with the wire direction and ∇T_Z is in the z-direction, ∇V_N is also in the *xy*-plane at an angle of $\pi/2 - \theta$. Consequently, both V_x and V_y are sinusoidal in θ with a 90° phase shift in between. Any other temperature gradient, such as ∇T_x, does not contribute to the voltage measured along the wire direction.

The perpendicular temperature gradient ∇T_Z in the patterned Py films is the consequence of thin films on substrates. One often employs metal thin films on substrates for electrical or thermal conductivity measurements. Since the substrate is typically five orders of magnitude thicker than the thin film, its respective conductivity must be proportionally smaller by an even larger magnitude to avoid appreciable contribution. This condition is readily fulfilled for electrical transport using common substrates, such as Si and GaAs, but *not* for thermal transport. Despite large differences in their electrical conductivities, the values of thermal conductivity of Si, GaAs, Fe and Py of 125, 56, 80, and 30 W/m-K respectively at room temperature are comparable [7, 20]. Therefore, thermal conduction through the much thicker substrate completely overwhelms thermal transport, thus creating a pronounced perpendicular temperature gradient through the thin metal film.

The value of the anomalous Nernst coefficient α, often expressed as $\alpha = \chi S$, is a fraction of the ordinary Seebeck coefficient S, where χ is a parameter. Using the Seebeck coefficient S = -20 μV/K [3] and $\chi \approx 0.13$ for Py [21] and the measured $\Delta V_y \approx 0.2$ μV across a Py wire 100 μm in width, we estimate a temperature difference $\Delta T \approx 0.2$ mK across the thickness of 300 nm of the Py thin film. As recently reported in Seebeck spin tunneling from Py to Si, the tunnel barrier accommodates a larger $\Delta T \approx 0.1$ K [22].

Materials with a strong spin-orbit interaction, such as Pt, play an essential role in the studies of spin Seebeck effect, where the pure spin current that flows into the Pt strip gives rise to an electric voltage [3-6, 8, 9]. We have also measured the Hall bar Py samples covered with a 10 nm Pt layer, slightly thicker than the spin diffusion length of about 5 nm [23]. We anticipate no inverse spin-Hall voltage because of the continuous Pt film. However, a clear but smaller V_y can still be observed as shown in Fig. 3(c) and (d). Therefore, the anomalous Nernst effect cannot be completely suppressed by the thin Pt to reveal only pure spin current effects, such as spin Seebeck effect.

The profound influence of the substrate, from temperature gradient to admixture of other spin dependent thermal effects, amply demonstrates the need of free-standing samples to measure intrinsic spin-dependent thermal properties. We have used a narrow strip of a thin Fe foil 20 μm in thickness suspended at two ends by the Cu blocks, which provide an unequivocal temperature gradient ∇T_x laterally as shown in Fig. 4(a). We have measured the transverse voltage V_x as a function of in-plane magnetic field at different angle θ. At each angle θ, instead of an asymmetric V_x and V_y as previously encountered in thin films on substrates, the behavior of thermal transport is now completely *symmetric*. For increasing H, V_x either increases or decreases depending on the angle between ∇T_x and H as shown in Fig. 4(b). The hysteretic behavior with the voltage peaks occurred at 15 Oe is associated with the switching field. The thermal voltage now exhibits a different angular dependence of $\cos^2\theta$ as shown by the solid curve in Fig. 4(c). This angular dependence, completely different from the $\sin\theta$ dependence observed in thin films on substrate, is the same as that of conventional anisotropic magnetoresistance (AMR) effect. In AMR, the resistivity varies as $\cos^2\theta$ where θ is the angle between current and $\boldsymbol{M}$. The spin-dependent anisotropic thermal voltage is similarly given by $V_{th} = V_{th\perp} + (V_{th\perp} - V_{th\|})\cos^2\theta_M$, where θ_M denotes the angle between the direction of ∇T and magnetization $\boldsymbol{M}$, $V_{th\perp}$ and $V_{th\|}$ for $\boldsymbol{M}$ perpendicular and parallel to ∇T respectively. For Fe at room temperature, the magnitude of anisotropic thermal transport is 0.8 %, similar to that of AMR. Our results also demonstrate that the thermal transport in a ferromagnetic metal can also sense the direction of $\boldsymbol{M}$ with the same sensitivity as AMR. Recently, asymmetric behavior has also reported and speculated the possibility of the presence of a perpendicular temperature gradient in GaMnAs film on a semi-insulating GaAs substrate [5, 24] and in Co_2MnSi and Py system on a MgO substrate [9].

We demonstrated experimentally that the dominant temperature gradient in thin film on substrate is perpendicular to the thin film despite the intended lateral temperature gradient. As a result, the spin-dependent thermal transport properties are obscured by the anomalous Nernst effect. Since most spin caloritronics studies to date [3-9] utilize thin films on substrates, the issue of perpendicular temperature gradient must be addressed. Measurements of intrinsic thermal properties require substrate-free samples, with which we have determined the intrinsic spin-dependent thermal transport, exhibiting a $\cos^2\theta$ angular dependence with a similar magnitude and field sensitivity as those of conventional anisotropic magnetoresistance (AMR).

Acknowledgement

We would like to acknowledge M. N. Ou for helpful discussions. This work is supported by the NSF under Grant No. DMR 05-20491 and the NSC 99-2911-I-007-510 of Taiwan.

† To whom correspondence should be addressed.
ssuyenhuang@gmail.com, clc@pha.jhu.edu.

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Figures with Caption

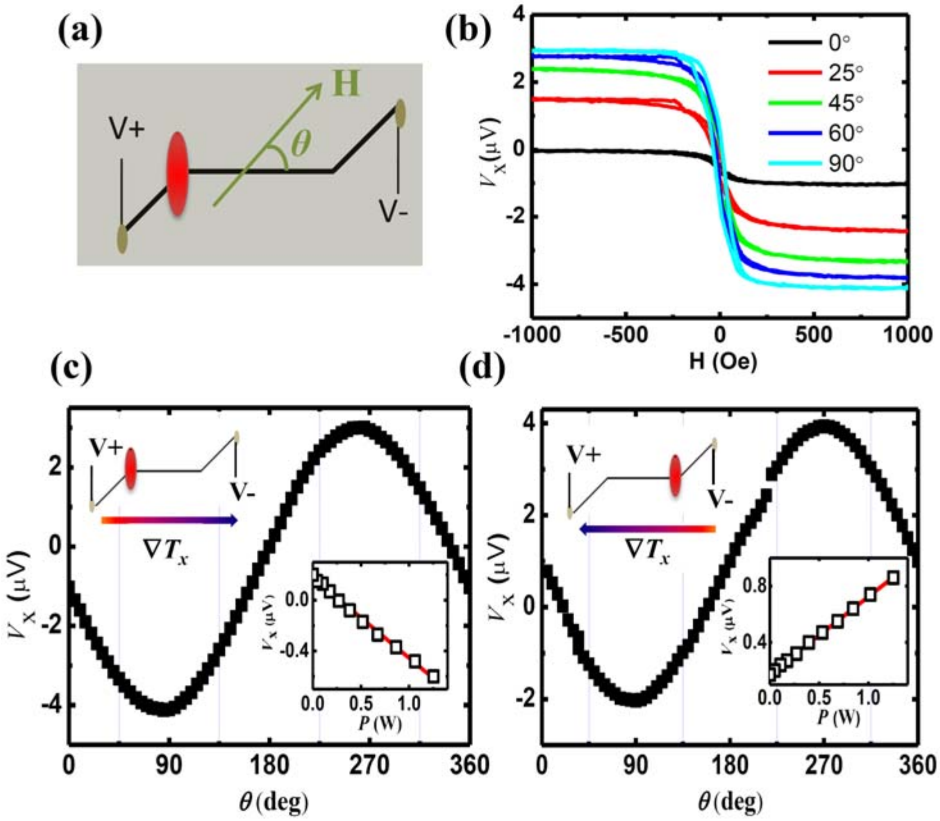

Fig. 1: (color online). (a) Schematic diagram of a Py wire sample for the thermal measurement with voltage leads on two sides, heat source indicated by the large oval, and magnetic field direction at angle θ. (b) field dependence of thermal voltage at different angle θ. Angular dependence of thermal voltage when heater is on the (c) left, and (d) right of the wire. Insets show power dependence of thermal voltage.

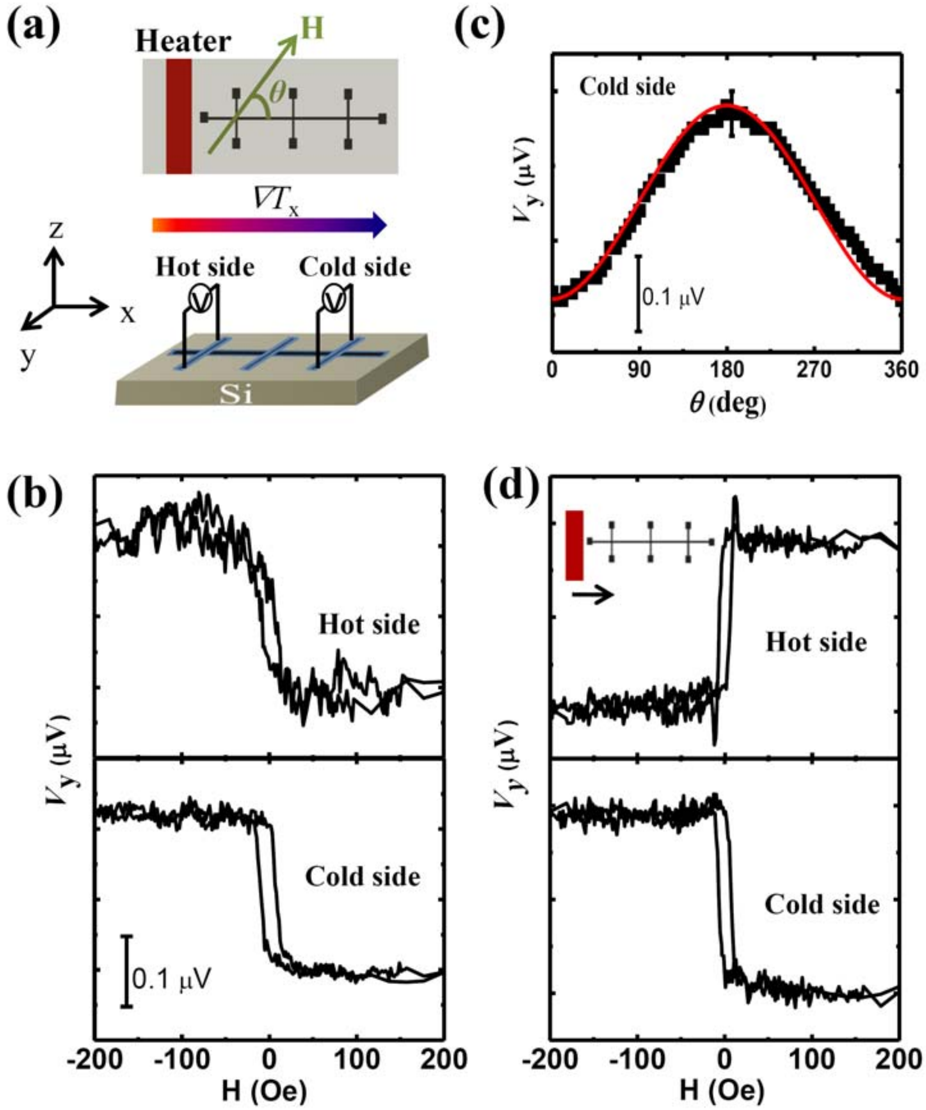

Figure 2

Fig. 2: (color online). (a) Schematic diagram of the Hall bar sample used for the thermal transport measurement with the heater on one side, transverse voltages measured at three locations (hot, cold, middle). (b) Field dependence of thermal voltage at the hot side and the cold side at θ = 0°. (c) Angular dependence of thermal voltage at cold side. (d) Field dependence of thermal voltage at the hot side and the cold side with a closer heater position at θ = 0°.

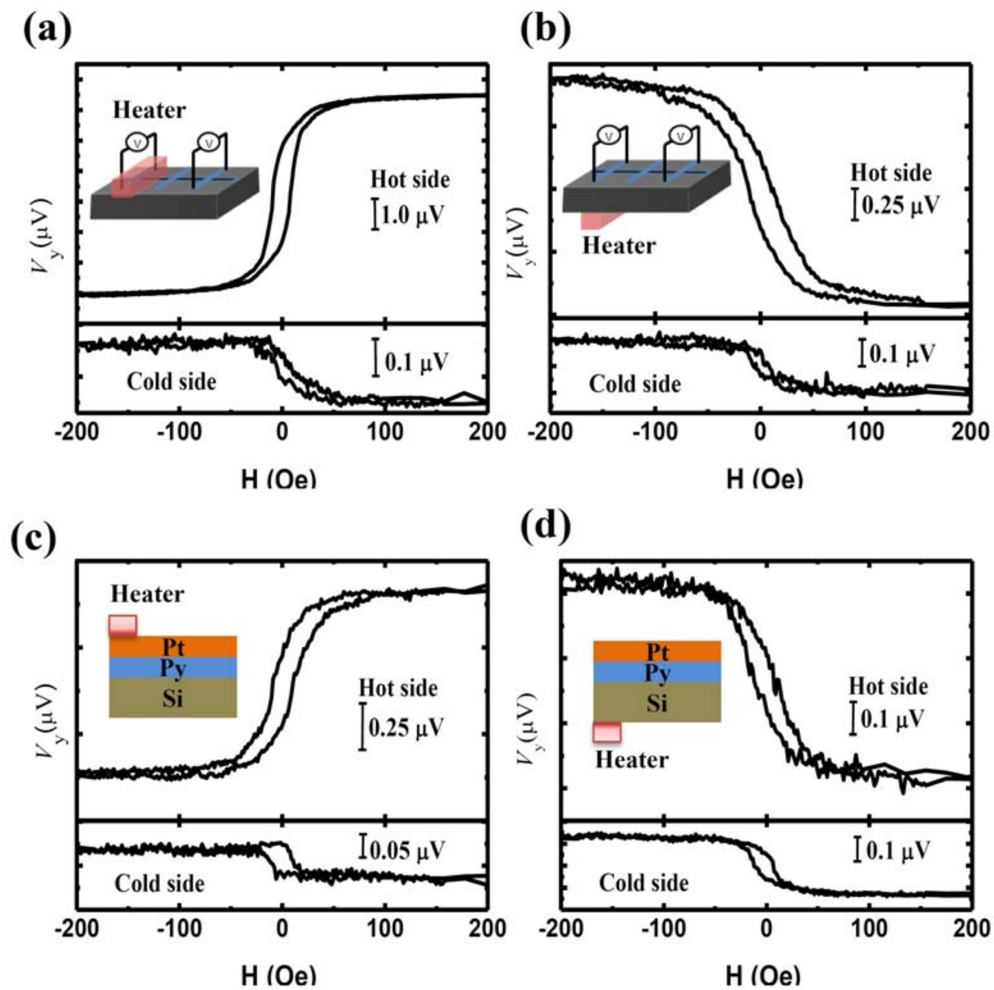

Fig. 3: (color online). Field dependence of thermal voltage on the hot side (top panel) and cold side (bottom panel) of Hall bar Py sample with heater placed on (a) top; and (b) bottom of the sample. Results of same measurements with a 10-nm Pt layer on top of 300-nm Py are shown in (c) and (d).

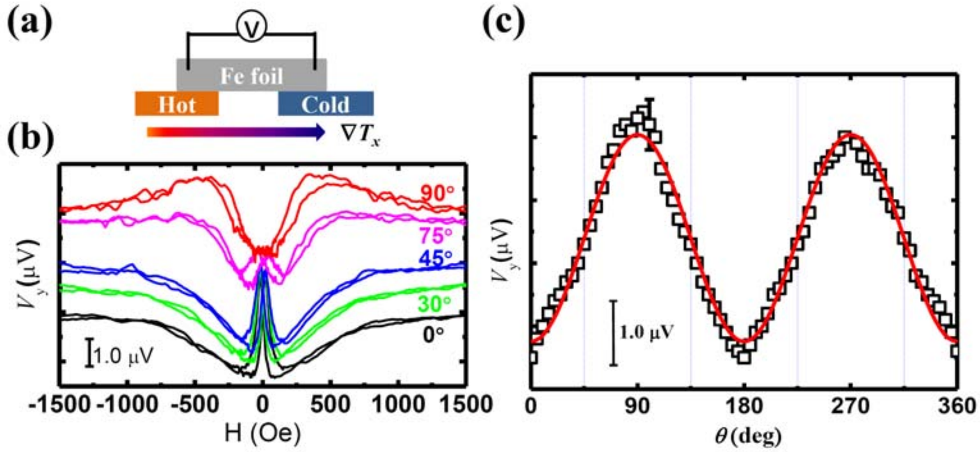

Fig. 4: (color online). (a) Schematic sideview of a suspended Fe foil sample for the intrinsic spin-dependent thermal transport measurement (b) Field dependence of thermal voltage when the field is applied at different angle θ. (c) Angular dependence of saturated thermal voltage with the solid curve of $\cos^2\theta$.