

Nernst-Ettingshausen effect in thin Pt and W films at low temperatures

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Abstract

As spin caloritronic measurements become increasingly common techniques for characterizing material properties, it is important to quantify potentially confounding effects. We report measurements of the Nernst-Ettingshausen response from room temperature to 5 K in thin film wires of Pt and W, metals commonly used as inverse spin Hall detectors in spin Seebeck characterization. Johnson-Nyquist noise thermometry is used to assess the temperature change of the metals with heater power at low temperatures, and the thermal path is analyzed via finite-element modeling. The Nernst-Ettingshausen response of W is found to be approximately temperature-independent, while the response of Pt increases at low temperatures. These results

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are discussed in the context of theoretical expectations and the possible role of magnetic impurities in Pt.

While spin caloritronic measurements^{1,2} are increasingly common, distinguishing between different physical mechanisms of generating voltages in structures containing magnetic materials and strong spin-orbit coupling metals can be challenging. For example, in the experimental geometry commonly used for local measurements of the spin Seebeck effect (SSE)³, care must be exercised to distinguish between the SSE and the ordinary Nernst-Ettingshausen response^{4,5} of the strong spin-orbit metal used as an inverse spin Hall detector in the SSE measurement. The Nernst-Ettingshausen effect is the generation of a transverse electric field by a longitudinal thermal gradient in the presence of a mutually perpendicular magnetic field, and it has the same dependence on in-plane magnetic field orientation as the SSE in the local geometry. Particularly as spin caloritronic measurements are extended to lower temperatures and more exotic magnetic systems such as candidate spin liquids⁶, it is important to have a quantitative sense of the relative magnitude of the Nernst-Ettingshausen response and its temperature dependence.

Here we report measurements of the Nernst-Ettingshausen response in the commonly employed strong spin-orbit metal W and Pt, using a device geometry typically used in local SSE measurements, but in this case fabricated on an inert SiO₂/Si substrate, to eliminate any SSE contribution to the measured voltage. We find that the Nernst-Ettingshausen response, linear in magnetic field and heater power, is approximately temperature independent from room temperature to 5 K at fixed heater power for W. In contrast, the response for Pt shows a strong

temperature dependence at low temperatures. We discuss a possible explanation for this in the context of prior measurements performed on coinage metals with dilute magnetic impurities. Johnson-Nyquist noise thermometry is used to assess the temperature rise of the metal layers with the application of heater power at low temperatures, and finite element modeling is used to assess the heat flow in the experimental geometry. These results show the need for care when considering local SSE measurements at low temperatures.

Our devices consist of the multilayer structure as shown in Fig. 1a. Photolithography, magnetron sputtering, and liftoff were used to prepare the Pt (W) wire (800 μm long, 10 μm wide, 10 nm thick) on a silicon wafer capped with 300 nm oxide. A lithographically defined SiO_x layer with a thickness of 100 nm was fabricated on the top of the Pt (W) wire by e-beam deposition and liftoff. Finally, a Au wire (1300 μm long, 10 μm wide, 50 nm thick) was deposited on the SiO_2 layer, just above the Pt (W) wire, to serve as a heater. The SiO_x layer electrically isolates the Au heater and the Pt (W) wire.

For the Nernst-Ettingshausen response measurements, an AC current at angular frequency $\omega = 2\pi \times (7.7 \text{ Hz})$ is driven through the Au wire and generates a vertical temperature gradient across the whole device that oscillates at angular frequency 2ω . The voltage across the Pt (W) wire is measured at 2ω using a lock-in amplifier, to detect the voltage response due to the temperature gradient. The measurements are performed as a function of temperature and field in a Quantum Design Physical Property Measurement System (PPMS) equipped with a rotation stage. Example results are shown in Fig. 1. When the device stays at 5 K and the heater power of 1 mW is

applied, the second harmonic signal across the W wire has a dominant linear-in-field component that is antisymmetric with the field (Fig. 1b), and this response has the angle dependence expected for the electric field generated by Nernst-Ettingshausen effect, $E_x = \nu B_y \nabla_z T \propto B \cos \alpha$, where ν is Nernst-Ettingshausen coefficient, B_y is y-component of the field, and $\nabla_z T$ is temperature gradient along z-axis. The second harmonic voltage is linear in the heater power up to 2 mW (in Fig. 1d), consistent with a thermal origin. Given the lack of any magnetic response of the SiO₂ substrate layer, we identify the observed voltage as originating from the Nernst-Ettingshausen effect in the thin W wire. The similar results can be found in Pt wire, as shown in SM.

The detailed temperature dependence of the magnitude of the Nernst-Ettingshausen response is shown in Fig. 1c. For a temperature sweep at constant heater power, we find that the response of the W wire is almost temperature independent. In contrast, in the Pt wire the response magnitude at constant heater power decreases as temperature is lowered, falling below detectability below 150 K, and becoming considerably larger as temperature is further decreased below 20 K.

In this experimental geometry, the Nernst-Ettingshausen signal has previously been considered as approximately temperature independent⁷. The Nernst-Ettingshausen coefficient $\nu =$

$$-\frac{\pi^2}{3} \frac{k_B^2 T}{m^*} \frac{\partial \tau}{\partial \varepsilon} \Big|_{E_F} = \frac{\pi^2}{3} \frac{k_B^2 T}{e} \frac{\mu}{E_F} \propto T \text{ for comparatively temperature-independent mobility } \mu \text{ at low}$$

temperatures in thin metal films⁴. At low temperatures, the magnitude of the temperature gradient across the metal layer at a given heater power is inversely proportional to the electronic

thermal conductivity $\kappa_{el} \sim T$. Thus, at a fixed heater power, the expected temperature dependence of $V_N \propto \frac{v}{\kappa_{el}} \propto \frac{T}{T} \propto 1$, consistent with the data in the W wire.

The observed nontrivial temperature dependence of the Nernst-Ettingshausen response in the Pt wire at fixed heater power is therefore surprising. We consider two candidate mechanisms. One possibility is that the Kapitza thermal resistance⁸ at the various metal/dielectric interfaces in the stack, such as between the Pt wire and oxidized silicon substrate, may behave differently with temperature than in the W case, such that the temperature profile at fixed heater power is somehow significantly different between the Pt and W devices. We perform Johnson-Nyquist noise thermometry measurements to test this concern, as described below. Another possibility is that the enhanced Nernst-Ettingshausen response at low temperatures may be due to Kondo scattering by dilute ferromagnetic impurities in the Pt. Prior studies in copper and gold wires containing magnetic impurities have shown a qualitatively similar upturn in Nernst-Ettingshausen response at low temperatures⁹. A study of Nernst response in heavy fermion Kondo materials¹⁰ shows a similar upturn as temperature is decreased toward the heavy fermion coherence temperature scale, which is ascribed to the effect of single-ion Kondo impurity scattering on Nernst response. No obvious Kondo-like response in $R(T)$ of the Pt wire is observed, however (Fig. S2), implying that any magnetic impurity scattering is not contributing noticeably to the longitudinal resistive response.

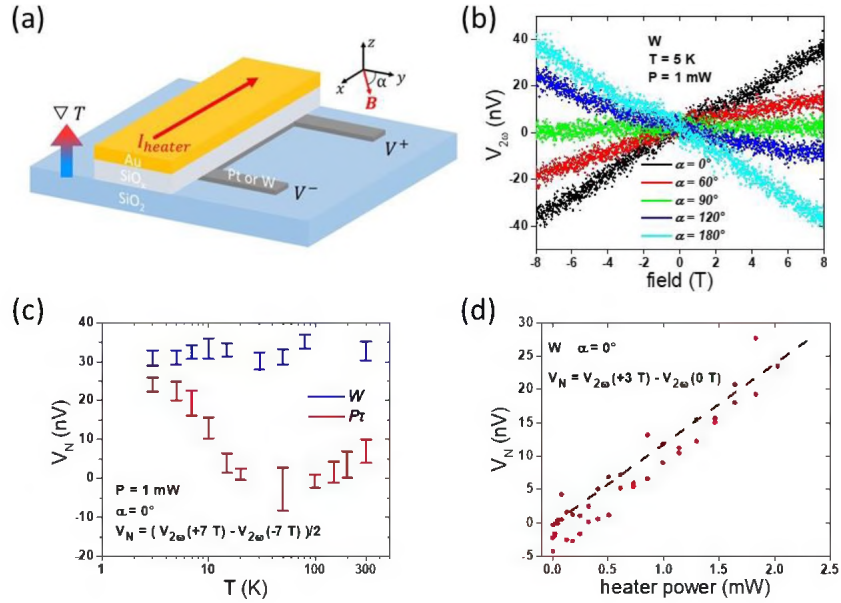


FIG. 1. (a) The device geometry in our measurements, showing the definition of field orientation angle α in the x - y plane. The heating current runs through the Au wire and generates a temperature gradient perpendicular to the silicon wafer. (b) Field dependence of the second harmonic signal at 5 K in W wire for different field orientation angles. The 2ω signal at $0^\circ/180^\circ$ gives the largest responses, and the 2ω signal at 90° gives approximately no response, as expected for the Nernst-Ettingshausen effect. (c) Temperature dependence of the Nernst-Ettingshausen voltage in W and Pt at fixed heater power. The Nernst-Ettingshausen voltage is quantified at $\alpha = 0^\circ$ through the difference of the second harmonic signal between 7 T and -7 T, divided by two. The heater power in (b) and (c) is 1 mW. (d) The Nernst-Ettingshausen voltage (quantified as the difference of the second harmonic signal between 3 T and 0 T at $\alpha = 0^\circ$) is linear in heater power, as expected.

To better understand the temperature profile of the device when applying a steady state temperature gradient with the heater, we measure the thermal noise in the Pt (W) wire to obtain the actual temperature change of the detector wire under different heater powers. Johnson-Nyquist thermal noise^{11,12}, originates from the random motion of charge carriers due to thermal excitation. As a consequence of the fluctuation-dissipation relation, the equilibrium thermal motion of carriers creates a fluctuating voltage on the terminals of an open-circuit resistive element, with a power spectrum density (PSD) given by $S_V = 4k_B T R$, where T and R is the temperature and resistance of the resistive element. As a result, measuring the noise power S_V and the resistance R , we can immediately obtain the temperature $T = S_V / 4k_B R$. When applying heater power, the temperature of the detector wire will unsurprisingly rise, leading to an increase in S_V . Considering that the resistance of the detector wire is approximately temperature-independent below ~ 20 K (Fig. S2), the average temperature increase is $\Delta T = \Delta S_V / 4k_B R$. Fig. 2a shows histogram of inferred ΔT of the Pt wire when the cryostat temperature is fixed at 5 K and a heating power 1 mW is repeatedly applied. Fig. 2b shows that the temperature change ΔT is slightly sublinear in the heater power, suggesting that at large heater powers some part of the thermal path is driven out of the linear regime of heat transport across the various interfaces. The linearity of the Nernst-Ettingshausen response with heater power (Fig. 1d) implies that the thermal conduction across the metal layer itself remains in the linear regime. Fig. 2c shows a comparison of ΔT at different cryostat temperatures between the Pt and W wires when applying 1 mW power. The magnitude of ΔT decreases with the increasing temperature, indicating that thermal conduction of the whole device structure is temperature dependent, as expected when Kapitza-like thermal boundary resistances contribute to the thermal path. The key take-away is

that the values of ΔT in Pt and W under the same heater conditions are quite similar, implying that the difference in temperature dependence of Nernst-Ettingshausen response (Fig. 1c) is not due to drastically different thermal transport profiles.

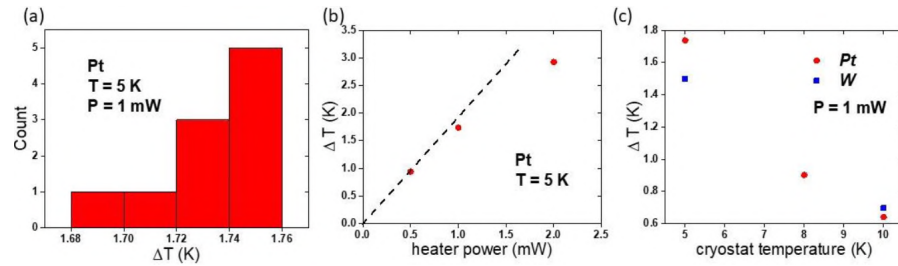


FIG. 2. (a) Histogram of the average temperature increase ΔT for a Pt wire at cryostat temperature 5 K when a heater power of 1 mW is applied. (b) ΔT versus heater power for the Pt device. The black dashed line is a guide to the eye assuming linear response at low heater power. The measured trend is sublinear, suggesting that some component of the complete device thermal path is pushed out of the linear regime of heat transport at high heater powers. (c) ΔT at 1 mW heater power versus cryostat temperature, showing that thermal conduction of the whole device changes with temperature, as expected in the presence of Kapitza-like thermal boundary resistance. The error bar in (b) and (c) are smaller than the data point markers.

Nernst-Ettingshausen coefficients in such thin films have not been reported explicitly; however, some control experiments in LSSE measurements¹³ allow an estimate of the value in Pt to be 1.5×10^{-11} V/(TK) at 10 K (see SM for details). It is worthwhile to do an order-of-magnitude

comparison of the measured response with theoretical expectations for the magnitude of the Nernst-Ettingshausen coefficient. Roughly evaluating the simple Fermi liquid expectation^{4,5} $\nu =$

$$-\frac{\pi^2}{3} \frac{k_B^2 T}{m^*} \frac{\partial \tau}{\partial \epsilon} \bigg|_{E_F} = \frac{\pi^2}{3} \frac{k_B^2 T}{e} \frac{\mu}{E_F}$$

requires values for the Fermi energy E_F and the carrier mobility μ , which may be estimated from the Drude conductivity σ and a free electron density n by $\mu = \sigma/(ne)$. In Pt, from the measured resistance and dimensions we find an electrical conductivity at 5 K of $\sigma = 2.31 \times 10^6$ S/m. A reasonable single-band free-electron model estimate for carrier density for Pt¹⁴ is $n \approx 1.6 \times 10^{28}$ m⁻³. This leads to a mobility estimate of 9.02×10^{-4} m²/(Vs). From relativistic band structure calculations¹⁵, $E_F \approx 10$ eV. The Nernst-Ettingshausen coefficient for Pt at 5 K should then be $\nu \approx 1.1 \times 10^{-11}$ V/(TK). Similarly, for W, a free electron model estimate for carrier density^{16,17} is $n \approx 2.1 \times 10^{28}$ m⁻³, and the Fermi energy¹⁸ is $E_F \approx 9.75$ eV. The measured electrical conductivity of the W film at 5 K is $\sigma = 1.15 \times 10^7$ S/m, giving a mobility estimate of 3.42×10^{-3} m²/(Vs). The Nernst-Ettingshausen coefficient for W at 5 K is then estimated to be $\nu \approx 4.3 \times 10^{-11}$ V/(TK).

Extracting the Nernst-Ettingshausen coefficient from the experimental data requires knowledge of the temperature gradient across the metal layer, which we cannot measure directly. A thermal model with reasonable values for material parameters can act as an order-of-magnitude point of comparison with the theory estimates of the Nernst-Ettingshausen coefficient above. Accurate thermal models require the temperature-dependent thermal conductivity of all the involved materials as well as an understanding of interfacial thermal effects. We can estimate the electronic thermal conductivity from the measured electrical conductivity of the metal films and Wiedemann-Franz law, and we take the phonon contribution to be negligible at low

temperatures. We neglect the Ti adhesion layer between the Au and the SiO_x isolation layer. We discuss these thermodynamic parameters in detail in the SM.

Much of the total thermal resistance at low temperatures is expected to come from material boundaries⁸. Of particular interest are the metal/dielectric interfaces in the device stack: the boundary between the SiO₂ of the substrate and the evaporated Pt (W), the boundary between the Pt (W) and SiO_x isolation layer, and the boundary between the SiO_x and the Au heater. We include these thermal boundary resistances in the model. Unfortunately, even a crude estimate of such interface effects is difficult: phonon boundary resistances depend strongly on the fabrication method, and good comparisons in the literature are sparse⁸. In the model we treat all metal/dielectric interfaces to have the same boundary resistance (valid to within an order of magnitude), which we vary to duplicate the experimentally measured Pt (W) temperature increases at given total heater powers in Fig. 2c. The thermal boundary resistances found at each substrate temperature (see Table S1) are the same order of magnitude as those previously reported⁸ and grow with decreasing temperature as expected. From the models we calculate the amount of heat that passes through the Pt (W), and, using the Pt (W) thermal conductivity and thickness, we find the temperature gradient across the metal layer. The model geometry and a representative temperature profile are shown in Fig. 3.

The finite-element model also confirms that a uniform gradient approximation perpendicular to the plane of the device is valid, as the total heat current through the sides of the Pt (W) is roughly 1 μ W or less in all cases. Note the Pt (W) wire is only 800 μ m long while the Au heater wire is

1300 μm in length, so at a total heater power of 1 mW, only about 0.615 mW of the heater power is directed toward the metal film.

The data in Table S1 and the device geometry are then used to estimate the Nernst coefficient using Eq. (1)

$$\nu \sim \frac{E_x}{B_y} \left(\frac{dT}{dz} \right)^{-1} = \frac{E_x}{B_y} \left(\frac{\kappa_{Pt(W)} l w}{q_b} \right)^{-1} \quad (1)$$

where q_b is the heat current through the bottom of the metal, $\kappa_{Pt(W)}$ is the Pt (W) thermal conductivity calculated from the measured electrical conductivity, and l and w are the length and width of the Pt (W) wire. For example, the order of magnitude of the Nernst coefficient at substrate temperature 5 K (Pt temperature 6.8 K) is 6.9×10^{-11} V/(TK). Table S1, S2 show the results for the Pt and W devices in this context.

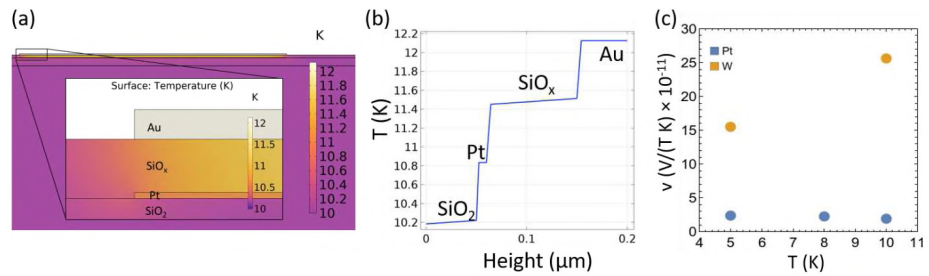


FIG. 3. (a) Map of temperature over the cross section of the Pt device at substrate temperature 10 K. (b) example temperature profile vs z within the device at substrate temperature 10 K; the

steep interfacial temperature changes result from thermal boundary resistances. (c) Estimated Nernst coefficients for Pt and W at select temperatures.

In summary, we measure the Nernst-Ettingshausen voltage in thin film devices based on Pt and W in the geometry commonly used for local spin Seebeck measurements on magnetic insulators. While the W response is approximately temperature-independent, as expected, the Pt response shows a marked increase at low temperatures. Johnson-Nyquist noise thermometry demonstrates that this difference between the responses of the metals is not a result of some difference in thermal path in the devices, but a property of the metals themselves. We therefore suggest magnetic impurity scattering as the likely explanation for the Pt temperature dependence. A finite element thermal model confirms that both metals show Nernst-Ettingshausen coefficients consistent to an order of magnitude with simple Fermi liquid theoretical estimates. These measurements provide useful bounds for the Nernst-Ettingshausen response as a potential confounding effect in SSE measurements, and the Pt results demonstrate that care must be taken in the assumption that Nernst-Ettingshausen response for metals is constant at low temperatures.

Supplemental Material

The supplemental material contains a discussion of the noise thermometry technique, details of the finite element thermal modeling, the transport properties of the Pt and W wires, and additional field-dependence data.

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Author Declarations

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Douglas Natelson: conceptualization (lead), analytical analysis (supporting), funding acquisition (lead), writing (equal); Renjie Luo: investigation (lead), analytical analysis (supporting), writing (equal); Tanner J. Legvold: investigation (supporting), analytical analysis (lead), writing (equal); Liyang Chen: investigation (supporting), writing (supporting).

Data Availability

The data that support these findings are available on Zenodo (get URL).

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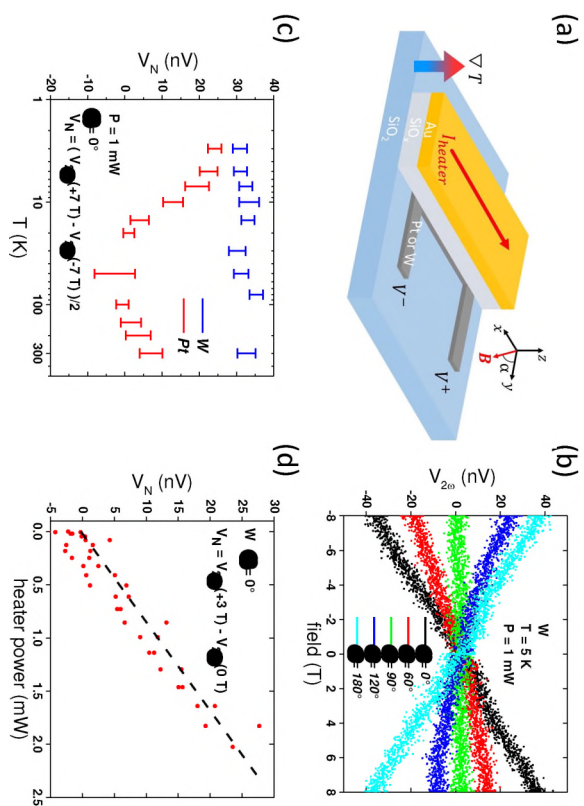
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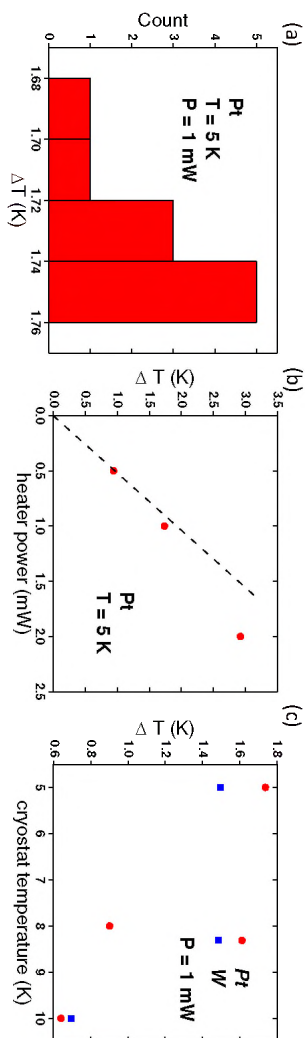
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