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Determination of the thermopower of microscale samples with an AC method

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ABSTRACT

A modified AC method based on the micro-fabricated heater and resistance thermometers has been developed to measure the thermopower of microscale samples. A sinusoidal current with angular frequency ω is passed to the heater to generate an oscillatory temperature difference across the sample at a frequency 2ω, which simultaneously induces an AC thermoelectric voltage, also at the frequency 2ω. The key step of the method is extraction of the amplitude and phase of the oscillatory temperature difference by probing AC temperature variations at each individual thermometer. The sign of the thermopower is determined by examining the phase difference between the oscillatory temperature difference and the AC thermoelectric voltage. The technique has been compared with the popular DC method by testing both n-type and p-type thin film samples. Both methods yielded consistent results, which verified the reliability of the newly proposed AC method.

1. Introduction

The thermopower, also known as Seebeck coefficient, serves not only as a performance indicator of thermoelectric materials, but also as a sensitive probe to reveal transport properties and electronic structures of semiconductors and metals around the Fermi levels. For bulk samples, the thermopower measurements can be carried out under steady-state (DC) [1–2], quasi-steady-state [3–7] or AC configurations [8–14]. The DC method of measuring the thermopower typically involves generating a small temperature gradient across the sample, and recording the static temperature difference ΔT_{DC} as well as thermally induced thermoelectric voltage ΔU_{DC}. A crucial problem associated with this method is that the spurious DC offset voltage arising from the measurement circuit has to be eliminated. To achieve this, a few pairs of ΔU_{DC}~ΔT_{DC} data should be acquired and the thermopower is actually extracted from the slope of a best-fit line of ΔU_{DC} as a function of ΔT_{DC} [14]. Obviously, it is quite time-consuming to stabilize several increments of ΔT_{DC} during the data acquisition process. To expedite the measurement, Testardi and McConnell [3] and Ivory [4] proposed the quasi-steady-state method in the early 1960s which imposes a continuously increasing ΔT_{DC} while dynamically recording multiple data points of the thermoelectric voltage and the temperature difference. Later, the AC method was introduced by Freeman and Bass [8] and Hellenthal and Ostholt [9] in 1970. It naturally eliminates the effects of the DC offset voltage and makes quick and yet more delicate measurements possible. For example, it allows one to continuously acquire the thermoelectric voltage and the temperature difference by lock-in amplifier to obtain the thermopower [14]. It can also provide sufficient sensitivity at smaller temperature difference (e.g., from 10 to 500 mK) [8,11–12,14] which is an advantage over DC method or quasi-steady-state method. These features are especially useful when one attempts to track variations of the thermopower near critical points (e.g., critical temperature T_c of superconductors) where rapid measurements at small temperature differences are necessary [8,11–12].

It should be noted that the temperature measurement for the bulk samples are routinely accomplished by making thermocouples in close contact with the sample [14]. As the size of the sample shrinks to microscale or nanoscale, it is necessary to load or deposit the samples onto test chips on which micro-sized heating elements and temperature sensing elements can be pre-fabricated or post-fabricated [15–34]. Numerous chip-based measurements have been carried out by applying the DC method [15–27] and the AC method [28–34]. For the latter case, an AC current of angular

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frequency ω is supplied into the heater located in proximity to one end of the sample to create an oscillatory temperature difference along the sample at a frequency 2ω. The amplitude of resulted 2ω thermoelectric voltage $\Delta U(2\omega)$ is detected by the lock-in amplifier. The temperature difference $\Delta T(2\omega)$ across the sample can be sensed by resistance thermometers [29,30] or by scanning thermal microscopy (SThM) [32].

With the AC method, Duarte and co-workers have observed a significant thermopower enhancement for "junctioned" gold nanowires around room temperature [28]. Kirihara et al. have measured the thermopower of single-crystalline boron nanobelts [29]. Kim and co-workers have observed that the thermopower and electrical conductance of two molecular junctions can be simultaneously increased by electrostatic control [32]. As suggested by the authors in supplementary information of Ref. [32], during the measurement a small temperature difference had to be applied to prevent temperature related instabilities in the molecular junctions, which precluded the use of the DC method that has been plagued by a small signal-to-noise ratio. Wu et al. have measured the thermopower of single layer epitaxial graphene by scanning magnetic field which exhibited strong periodic quantum oscillations [30]. In addition, the thermopower of quantum dots embedded in heterostructure nanowires have been investigated by Svensson *et al.* as a function of electrochemical potential [31].

In Ref. [29] and Ref. [32] (see Eq. S(6) in the supplementary information), the thermopower derived from the AC method S_{AC} is given in the form of

$$S_{AC} = -\Delta U(2\omega)/\Delta T(2\omega) \tag{1}$$

As mentioned later in Section 2, if we assume that the sample is in good thermal contact with the substrate of the chip and the thermal wave propagates mainly through the substrate, the temperature oscillation at hot end and at cold end of the sample should exhibit a phase difference that depends on the frequency ω, distance between the two temperature sensing locations L_{HL}, and thermal diffusivity of the substrate. In many studies [29–32], the substrate of the test chips is made of materials possessing a high thermal diffusivity (e.g., Si or SiC), the frequency of the heating current f_{HC} ($f_{HC} = \omega/2\pi$) is relatively low (often less than 20 Hz) and L_{HC} is small (ranging from several nanometers to several micrometers). Under these circumstances, the phase difference may be ignored. $\Delta T(2\omega)$ can be determined directly from $\Delta T(2\omega) = T_H(2\omega) - T_L(2\omega)$, where $T_H(2\omega)$ ($T_L(2\omega)$) is the amplitude of the temperature oscillation at the hot end (cold end) [32]. However, under certain conditions such as a substrate with low thermal diffusivity being used and L_{HL} being large (>100 μm), the phase difference may become prominent even at f_{HC} <10 Hz, as we will show in this study. In this scenario, in order to properly retrieve $\Delta T(2\omega)$, both the amplitude and the phase of the temperature oscillations at the hot end or cold end of the sample should be taken into account.

In this paper, we present a modified AC method to measure the thermopower of the microscale samples in a general way. The prerequisite that the temperature oscillations on both sides of the sample should have a negligible phase difference is not necessary. Here, the temperature of the samples is monitored by two four-probe resistance thermometers which have been calibrated separately. The amplitude and phase of the AC temperature oscillation are measured for each individual thermometer. The results are used to yield the amplitude and the phase of the 2ω temperature difference through a simple trigonometric function calculation. Also, the amplitude and the phase of the 2ω thermoelectric voltage are directly recorded by the lock-in amplifier. The amplitude and the sign of the resulted thermopower depend on the amplitude ratio and phase relationship between the 2ω thermoelectric voltage and the 2ω temperature difference, respectively. The method, along with the popular DC method, has been applied to the p-type Sb_2Te_3 and n-type Bi_2Te_3 thin film test samples. The results of the two methods agree with each other reasonably well within experimental uncertainty. This method may be useful for carrying out the chip-based thermopower measurements for the samples which only allow a small temperature difference being applied while the phase difference of the temperature oscillations at the two temperature sensing locations can't be ignored.

2. Experimental methodology

As illustrated in Fig. 1, when a sinusoidal current with the angular frequency ω is driven through the heater, a periodic heating wave at the frequency 2ω will be generated via Joule heating and it will propagate towards the microscale sample through the substrate. The sample is assumed to be in good thermal contact with the substrate and with the two thermometers Th1 and Th2, of which Th1 is located closer to the heater. To sense the temperature variation, two outer contact pads of Th1 or Th2 are used for sourcing current, and two inner contact pads for measuring resistive voltage drop.

The instantaneous temperatures at the positions where the two thermometers are located can be expressed as

$$T_i = T_0 + \Delta T_i^{dc} + T_i^{ac} = T_0 + \Delta T_i^{dc} + |T_i^{ac}| \cdot \sin(2\omega t + \varphi_i)\ (i = 1, 2) \tag{2}$$

where subscripts 1 and 2 correspond to Th1 and Th2, respectively. T_0 is the initial temperature of the substrate before turning on the heater. ΔT_i^{dc} represents the DC temperature rise of the thermometer, T_i^{ac} the AC temperature fluctuation with the angular frequency 2ω. $|T_i^{ac}|$ and φ_i are the amplitude and phase of T_i^{ac}, respectively.

If we approximate the heat transfer along x-direction through the substrate as a one-dimensional (1D) process, the AC component of the temperature variation at position x (the heater is assumed to be located at x = 0) can be given as [10,35–36]

$$T_{1D}^{ac} = \frac{j_0}{k}\sqrt{\frac{\alpha}{2\omega}}\exp\left(-\sqrt{\frac{\omega}{\alpha}}x\right)\cos\left(2\omega t - \sqrt{\frac{\omega}{\alpha}}x - \frac{\pi}{4}\right) \tag{3}$$

where j_0 is heat flux amplitude. k and α are the thermal conductivity and the thermal diffusivity of the substrate, respectively. Eq. (3) suggests that the thermal wave propagates away from the heater with a decaying amplitude and a phase shift, which further implies that T_1^{ac} and T_2^{ac} in Eq. (2) should have not only different amplitude ($|T_1^{ac}| > |T_2^{ac}|$), but also different phase ($\varphi_1 \neq \varphi_2$). The

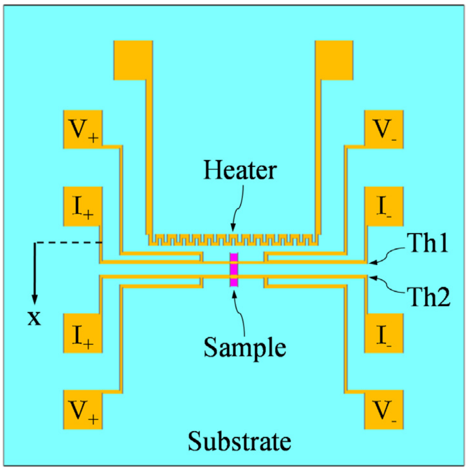

Fig. 1. Layout of the device for measuring the thermopower of a microscale sample.