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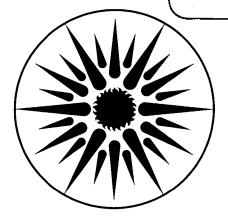
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An Optical Approach to Thermopower and Conductivity Measurements in Thin Film Semiconductors

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Abstract

An optical beam deflection technique is applied to measure the Joule and Peltier heat generated by electric currents through thin film semiconductors. The method yields a spatially resolved conductivity profile and allows the determination of Peltier coefficients. Results obtained on doped a-Si:H films are presented.

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Conductivity and thermopower measurements are widely used methods to study transport properties of semiconductors. The thermopower S is related to the Peltier coefficient Π , which for conduction above a mobility edge E_c may be written as:

$$ST = \Pi = \frac{kT}{e} \left(A + \ln \frac{N_o}{n} \right) \tag{1}$$

Since the quantities A and N $_{\mathrm{O}}$ do not contain the mobility μ , the thermopower provides a measure of the carrier density n alone and is often applied to separate the product $n\mu$ in the conductivity $\sigma = en\mu$. Moreover, the sign of S directly gives the charge of the majority carriers. A common problem with transport experiments is that properties of the metal contacts may influence the results. A current-voltage measurement reflects the conductivity only if the contact resistance is much lower than the sample resistance. In the thermopower experiment, the metal contacts serve as reference for the thermovoltage and contact properties are generally not considered to be a problem in the thermal equilibrium case, i.e., dark transport. In studying photoconductivity, thermopower measurements are much more difficult to perform due to the photovoltage, which may exceed the small thermovoltage. To understand the steady state photoconductivity it is essential to be able to distinguish between the effects due to changes in carrier density and those due to changes in mobility. Photothermopower measurements are of particular importance in the case of a-Si:H, since Hall effect data fail to give even the correct sign of the charge carriers. We present a technique to study transport properties which is based on the photothermal detection of Joule and Peltier heat generated by electrical currents. The method gives a spatially resolved profile of the conductivity along the film surface thus allowing the detection of σ away from any metal contacts. The measurement of the Peltier heat rather than the thermovoltage avoids the influence of photovoltages on the signal. Preliminary results for the photo-Peltier coefficient of doped a-Si:H are presented.

All measurements were done on 1 μ thick a-Si:H films deposited onto quartz substrates. ² An oscillating voltage $U=U_{o}\cos\omega t$ with frequency f=5 to 10 Hz is applied between two evaporated Al-contacts in gap-configuration with a spacing of 1 to 1.5 mm (Figure 1). The heat generated by the current creates a temperature gradient perpendicular to the sample surface, which changes the index of refraction in the surrounding medium (CCl_4) . This change is probed by a laser beam skimming the surface of the sample perpendicular to the current direction. To prevent changes in conductivity caused by the probe laser, an IR-laser $(\lambda = 3.39 \,\mu)$ is used and the deflection is measured by a position sensitive PbSedetector. It was verified that the resistance of the samples is not influenced by immersing the a-Si:H films in CCl_4 , and that CCl_4 itself produces no deflection due to the applied electric field. Both signals appearing at the reference frequency (1f signal) as well as at twice the reference frequency (2f signal) are monitored as the probe beam is scanned along the direction of the electric current (x - direction). It has been demonstrated that the deflection is directly proportional to the heat generated within the interaction region between the probe laser and the sample. The Joule heat U2/R (R-sample resistance) has a component oscillating at twice the reference frequency with amplitude $U_0^2/2R$. The differential Joule heat per unit length in x-direction is given by $\frac{1}{\sigma}(\frac{U_0^2}{2R^2\Omega})$ (Q - cross-section of the sample). The 2f signal therefore, provides a profile of the resistivity $1/\sigma$ along the x-direction with a resolution determined by the beam diameter and the thermal diffusion length. Both quantities are of the order of 0.2 mm. The Peltier heat Π oscillates at the reference frequency with amplitude $\Pi \frac{U_o}{R}$ and appears centered and with opposite sign at each of the contacts.

In Figure 2a the 2f signal is shown for a heavily phosphorous doped sample plotted as a function of x. Both amplitude and phase are nearly constant indicating that the conductivity is uniform throughout the sample. The strong decrease of the signal at both contacts shows that the contact resistance is

negligible compared to the sample resistance. Furthermore, the time delay of the signal at the contacts exhibited as a decreasing phase angle indicates that the tail of the signal is caused by diffusing heat from the gap region rather than by contact properties. It was verified that the 2f signal is linear in the Jouleheat over more than 3 orders of magnitude both by varying the voltage at fixed resistance as well as by varying the resistance in photoconductive samples illuminated with different light intensities at fixed voltages. The minimum detectable signal corresponds to a total Joule heat of 10^{-5} W, which for the given geometric and an electric field of 10^{3} V/cm corresponds to a lower limit of 10^{-6} Ω^{-1} cm⁻¹ for the conductivity.

The flat conductivity profile as shown in Figure 2a is not obtained in all samples. In Figure 3 results of a sample with lower doping concentration are shown. It can be clearly seen that the major part of the Joule heat is generated at the contacts. It follows that the resistance of the film is much lower than indicated by a current-voltage measurement. It should be noted that this influence of the contacts does not show up in the current-voltage characteristics which indicate ohmic behavior and are symmetric in both directions (see insert). We cannot give a conclusive explanation for the origin of this behavior which was found in 2 out of 5 samples; however, it seems at least possible that some published conductivity results may have been influenced by contact effects.

The magnitude and phase of the 1f-signal of the heavily phosphorus doped sample are plotted in Figure 2b. The signal has the predicted shape for the Peltier heat with approximately symmetric peaks appearing at the contacts. The phase of the signal at the peak-positions differs by 180° indicating that the deposited heat has opposite signs at the contacts. Also shown is the phase of the 1f-signal measured in a boron doped sample, which at each contact is shifted by 180° with respect to the phosphorus doped sample. This behavior reflects the change in sign of the Peltier coefficient as the conductivity is changed from n-

to p-type. As in the case of the 2f signal the phase decreases in the tails which is attributed to heat diffusing from the peak position, leading to a time delay of the signal. The Peltier heat was found to be proportional to the current through the sample over more than two orders of magnitude. The Peltier-coefficient Π can be calculated directly from the integrated 2f and 1f signals (S_{2f} and S_{1f}). These quantities are proportional to the total Joule heat ($\frac{U_0^2}{2R}$) and Peltier heat ($\Pi \frac{U_0}{R}$), respectively. To compare the signals quantitatively, we have chosen twice the frequency for S_{1f} than in the case of S_{2f} making sure that both quantities experience the same thermal relaxation. Dividing S_{1f} through S_{2f} and solving the equation for Π we are left with:

$$\Pi = \frac{U_o}{2} \times \frac{S_{11}}{S_{21}} \tag{2}$$

Determining the sign of Π is equivalent to determining the phase angle, which corresponds to heating in phase with the applied voltage. This can be easily done by adding a dc-offset U_1 to the oscillating voltage $U_0\cos\omega t$. Then, besides the signal from the Peltier heat a second 1f signal due to the Joule heat appears with amplitude proportional to $\frac{2U_1U_0}{R}$. Comparing the phases of these two signals allows the determination of the sign of Π .

Table 1 summarizes the results and compares them with literature data from samples with similar doping level. In this case, the Peltier coefficient has been calculated from thermopower data using Onsagers' relation $\Pi=ST$. We note that both the sign and the absolute values of Π are reproduced satisfactorily by the present results within the uncertainty of comparing non-identical samples. The data may also serve as a proof for Onsagers' relation in amorphous materials. Although this is generally accepted, it has been recently pointed out that its validity is by no means obvious for inhomogeneous materials. In case of failure of the Onsager relation the Peltier coefficient would still keep its usefulness as a measure of the mean position of the fermi level whereas

this would not be true for the thermopower.

In Figure 4 results from a photoconductive sample illuminated with various light intensities are shown. Illumination was carried out with a white light source and a 660 nm edge filter. The Peltier coefficient is plotted as a function of the conductivity starting with the dark value. The sign of Π indicates that both dark and photoconductivity are carried by electrons. A clear tendency to lower Π values at increasing conductivity can be observed. A least square fit of the data leads to a slope of 27 mV, which is close to $\frac{kT}{e}$. A slope of $\frac{kT}{e}$ is equivalent to a conductivity, which is only changed by the carrier density whereas the mobility remains constant.

So far most attempts to separate effects due to changes in carrier density from changes in mobility in a-Si:H are based on time resolved studies. 8,9 Conclusions concerning the steady state photoconductivity $\sigma_{\rm ph}$ require specific assumptions about the recombination and transport properties, and are strongly model dependent. While Street suggests that the carrier density has a weaker dependence on the illumination intensity than $\sigma_{\rm ph}$ itself, others assume that the excitation dependence of $\sigma_{\rm ph}$ reflects the recombination kinetics. It should be emphasized that the interpretation of the Peltier coefficient as indicating the position of the Quasi fermilevel is quite general and requires no knowledge of the transport or recombination mechanism. The only assumption being made is that the states contributing to the conductivity are occupied according to Boltzmann statistics. The present results point to a constant mobility, however clearly more data are needed.

We have shown that the photothermal detection of electronic transport yields information about contact influences, macroscopic sample homogeneity, and peltier coefficients of thin film samples. Possible extensions of the method, which are currently under investigation, include the ability to provide two dimensional conductivity images of device structures and the separation of surface from bulk contributions to the conductivity.

Acknowledgements

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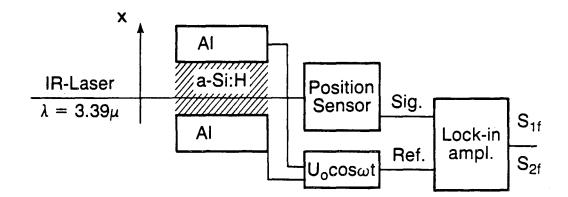
Figure Captions

- Figure 1 The experimental set-up.
- Figure 2 Magnitude and relative phase angle of the 1f and 2f signals for a heavily phosphorus doped sample.
- Figure 3 2f signal for a sample with non-ohmic contacts. The insert shows the current-voltage characteristics.
- Figure 4 Peltier coefficient Π of a photoconductive sample at various light intensities.

Table 1

Peltier Coefficient and Conductivity at Room Temperature

Doping	$\sigma (\Omega^{-1} \text{cm}^{-1})$	П (V)	Doping	$\sigma (\Omega^{-1} \text{cm}^{-1})$	S-T (V)	Ref.
PH ₃ ; 3x10 ⁻²	8x10 ⁻³	-0.21 -	PH ₃ : 3x10 ⁻² PH ₃ : 10 ⁻²	3.2x10 ⁻³ 5x10 ⁻³	-0.28 -0.25	4 5
PH ₃ ; 2x10 ⁻⁴	4.2x10 ⁻⁴	-0.33 -	PH ₃ ; 3x10 ⁻⁴ PH ₃ ; 2.5x10 ⁻⁴	5.2x10 ⁻⁴ 5.1x10 ⁻⁴	-0.34 -0.33	4 5
B ₂ H ₆ : 10 ⁻²	9.5x10 ⁻³	+0.23	B ₂ H ₆ ; 10 ⁻²	2x10 ⁻⁵	+0.30	6



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Figure 1

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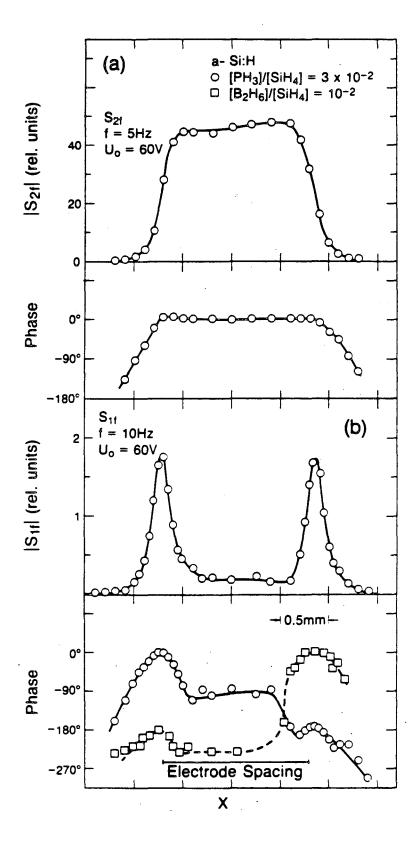
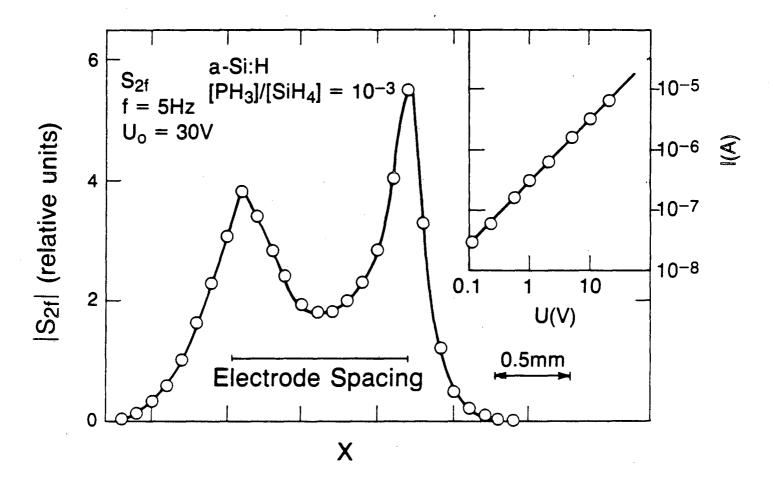


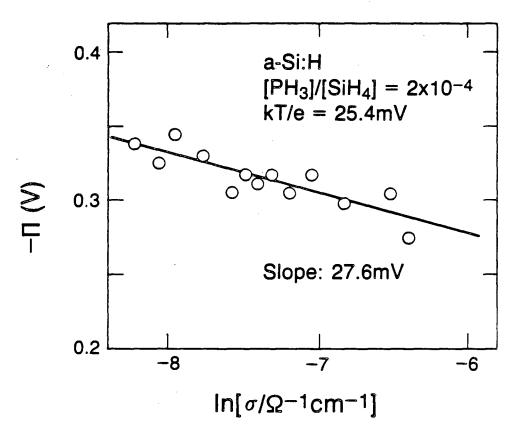
Figure 2



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Figure 3

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Figure 4

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