

Ultrathin Transparent Nickel Electrodes for Thermoelectric Applications

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In this work, ultrathin nickel films are developed for application as transparent electrodes in thermoelectric devices. The quality of the films is determined systematically by electrical, optical, and morphological characterization in a series of samples with different thickness. The thermal properties of the films show a dramatic dependence of the Seebeck coefficient on the film thickness. This dependence, with values ranging from -16 to $+5 \mu\text{V K}^{-1}$ for thicknesses from 10 to 2 nm , includes a change in the behavior of the thermoelectric response from n - to p -type. It has also been demonstrated that the accurate estimation of the thermal conductivity in thin films is challenging due to substrate effects. In this situation, a differential measurement method based on scanning thermal microscopy is proposed, as in these conditions the measurements are less sensitive to the substrate effects. In further works, the dependence of the thermal properties of ultrathin nickel films can be exploited as a tuning parameter for the design of thermoelectric devices.

1. Introduction

Thermoelectric (TE) materials are extensively applied for heat-to-energy conversion given their capabilities to generate an electric potential (thermoelectric voltage) in the presence of a temperature gradient, a phenomenon called Seebeck effect.^[1] This removable, clean, sustainable, and safe technology will contribute to reduce fossil fuel dependence and CO_2 emission.^[2,3] Indeed,

thermoelectric energy conversion is considered a key subject for achieving net zero emissions in 2050.^[4] Thermoelectric devices can operate individually (e.g., exposed to solar light) or embedded into a more complex device where they can harvest thermal loss into energy, thus improving the resulting efficiency.^[5] The origin of the thermoelectric voltage is the migration of charges from the hot to the cold regions of the material. The electrical conductivity (σ), the Seebeck coefficient (S), and thermal conductivity (κ_T) are the most important parameters of a TE device, as stated by the dimensionless figure of merit $ZT = \sigma S^2 T / \kappa_T$, being T the absolute temperature.^[6]

Most efficient thermoelectric materials are heavily doped semiconductors with an energy bandgap (E_g) near $10 k_B T$ (in

the other of 250 meV at room temperature), where k_B is Boltzmann's constant, and T is the operating temperature.^[7] Furthermore, due to the small bandgap ($E_g \ll 2 \text{ eV}$), many of these materials tend to be opaque, like the $(\text{Bi}, \text{Sb})_2\text{Te}_3$ family,^[8,9] tellurides,^[10] half-Heuslers^[11] and silicides.^[12] The development of optically transparent thermoelectric materials could open new fields in a range of novel applications, such as smart windows (or screens) with energy harvesting, cooling, and thermal sensing functionalities. Another potential application of transparent thermoelectric elements is the fast on-chip cooling and power recovery for optoelectronic devices, including solar cells, infrared photodetectors, and fully transparent electronic devices.^[5,13,14] In order to develop completely transparent thermoelectric generators, it is necessary to electrically connect in series n - and p -type transparent thermoelectric materials. On the one hand, there is a great diversity of n -type transparent conductors such as heavily doped ZnO ,^[15] In_2O_3 ,^[16] and SrTiO_3 ,^[17] being ITO (Sn -doped In_2O_3) the one with highest thermoelectric efficiency at room temperature ($ZT \approx 0.14$). However, in general, transparent p -type conductors exhibit low electrical conductivities – several orders of magnitude lower than transparent n -type conductors – leading to poor thermoelectric performance.^[18,19] Consequently, the lack of transparent p -type conductors with good thermoelectric properties is the main obstacle to developing completely transparent thermoelectric generators.

Ultrathin metal films (UTMFs), metal films with thickness lower than ten nanometers, are a promising alternative to conducting oxides for the fabrication of transparent

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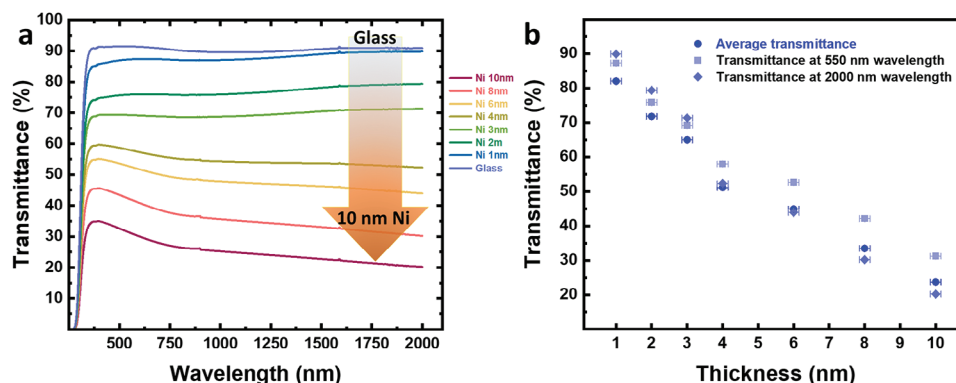


Figure 1. a) Transmittance spectra of nickel films of different thicknesses. b) Average transmittance between 300 and 2000 nm (circle scatters, at 550 nm wavelength (squares), and 2000 nm (diamond scatter)).

and flexible electronics.^[20] The resulting electrodes present a good balance between optical transparency and electrical conductivity with a range of applications for energy harvesting from smart windows,^[21] to nanosatellites.^[22] Among the different metals used as transparent electrodes, Ultrathin Nickel Films (UTNFs) present important advantages for thermoelectric applications.^[23,24] As a difference to other metals, UTNFs are stable in air, they can be exposed to nanolithography methods and solvents, and present compatibility with most semiconductors and organic molecules.^[25] As an advantage with respect to gold and silver, the lower percolation point in Ni films allows the growth of very thin films with continuity along the plane.^[26] On top of that, among the metals, Ni exhibits the higher thermoelectric.^[27]

However, current limitations in the characterization of thermal properties prevent an accurate determination of the thermoelectric voltage in UTMFs. This issue is also found in the growing field of 2D thermoelectric materials,^[32] where theoretical models foresee an important deviation of the Seebeck and thermal conductivity of few-layer crystals with respect to their bulk counterparts.^[33] In this line, SThM (Scanning Thermal Microscopy) is an emerging method for obtaining local thermal information, which has been used for measuring the thermal conductivity of 2D materials such as graphene and transition metal dichalcogenides. This technique provides a powerful tool to correlate local variations on the thermal conductivity and surface motifs, which is crucial for understanding heat transfer phenomena at the nanoscale.

In this work, the potential of UTNFs for the application as transparent electrodes in thermoelectric devices is demonstrated. We have experimentally measured the dependence of the electrical resistivity, the Seebeck coefficient, and the thermal conductivity on the film thickness. The transport measurements have been compared with the model proposed by Fuchs and Sondheimer (FS model) to illustrate the impact of the electron scattering at the interfaces. In the case of the Seebeck coefficient we observe an unexpected linear thickness dependence including a transition from n- to p-type. We attribute this behavior to the presence of native nickel oxide whose influence cannot be neglected in UTNFs. Finally, we point out the difficulty of determining the thermal conductivity in UTMFs by means of regular methods, and we propose differential measurements by means of scanning ther-

mal microscopy as a promising solution to characterize UTNF and 2D materials. Our results confirm that the differential measurements combined with planar geometry of our samples favors the quantification of the SThM measurements.

2. Results and Discussion

Solid source electron-beam thermal evaporation of nickel was optimized to obtain ultrathin films with thicknesses ranging from 1 to 10 nm on BOROFLOAT substrates. The growth rate has been calibrated by means of ellipsometry, profilometry, and AFM. The experimental conditions allow growing films of 1 nm with an accuracy of 0.3 nm. **Figure 1a** shows the dependence of the optical transmittance on the film thickness measured in a series of samples. The results are compared to the substrate transmittance (cyan curve). All the samples exhibit good transparency in the visible and two different behaviors can be distinguished depending on the thickness. For films thicker than 3 nm, the maximum transmittance appears close to the UV while for thinner films the transmissivity is almost flat, increasing slowly but surely as approaching mid-IR.

For a better comparison, in **Figure 1b** we plot the average transmissivity (from 300 to 2000 nm) as a function of the nickel thickness with circle scatters. Identical plots are obtained by considering the transmittance for specific wavelengths. For example, the transmittance at 550 and 2000 nm for samples of different thicknesses is represented with rectangular and diamond scatters, respectively. The linear behavior at different wavelengths coincides with the previous results in the literature.^[34] Only in the case of the four-nanometer thick film, the transmissivity deviates from the linear trend. Quantitatively, the maximum transmittance is in the range of 80–70% (including the substrate contribution) for film thicknesses of 2 and 3 nm, in agreement with other published results.^[35] The transparency of the films is reduced for samples in the range from 5 to 8 nm thick, decreasing the transmissivity below 50% as previously published.^[36] Indeed, transmittance measurements are used to double-check the reproducibility of the growth process as the transmissivity of different batches can always be fitted to a straight line. In all the samples prepared, the four-nanometer thick samples deviate from the linear fitting.

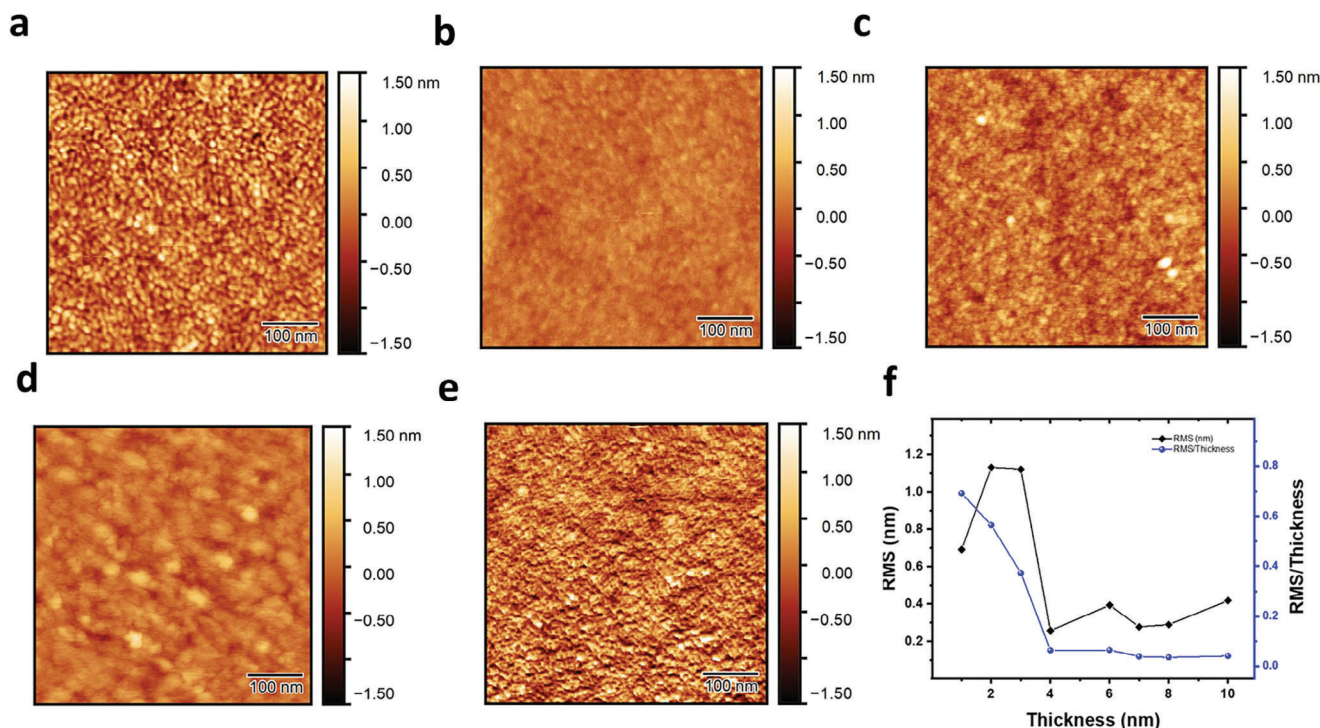


Figure 2. Surface characterization by AFM. a–e) AFM images of the surface of thin films with thicknesses 10, 8, 6, 4, and 2 nm, respectively. f) RMS and RMS versus thickness of the whole set of samples studied.

In connection to the optical properties, our samples also present excellent surface quality with a root mean square (RMS) of the order of 1 nm, as estimated by means of atomic force microscopy (AFM) measurements (see **Figure 2**). For the sake of comparison all the images have been acquired with identical acquisition conditions, under slow scanning rates. As shown in **Figure 2f**, the highest surface roughness is found for the films below 4 nm with a maximum for the case of the 2 nm thick samples, with an RMS ≈ 1.1 nm. Above 4 nm, the RMS is comparable to the substrate roughness (i.e., $\text{RMS}_{\text{substrate}} \approx 0.3$ nm), being the minimum roughness found for the 7 nm thick sample.

In most cases, the growth of metals on transparent substrates like glass or silica needs an adhesion layer.^[37–40] However, nickel can form a continuous film without an adhesion layer. For example, the percolation points on nickel grown by RF sputtering on high pyrolytic fused silica (HPFS) substrates are estimated ≈ 2.5 nm. According to our RMS estimations we can achieve similar qualities on BOROFLOAT. In our case, the coalescence point would be below one nanometer as there are no insights about forming metallic islands (Volmer–Weber growth mode).^[39,40] We can identify some grains mainly in the 2 nm thick sample despite the continuity exhibited by the film. The situation is improved above the 4 nm thick sample with thinner film. Indeed, we can estimate the percolation ≈ 4 nm coinciding with the important drop in the RMS. To double-check this trend, the RMS has been also estimated by means of power spectral density (PSD) analysis on the same AFM images (not shown). By means of this method, we obtained RMS values slightly lower than the ones estimated from the height statistics, but also, the PSD spectra point to the presence of differ-

ent kind of features in the case of two- and ten-nanometer thick samples.

Further proofs about the quality of the film are found in **Figure 3**, where we have represented the dependence of the resistivity on the film thickness. The electrical conductivity measurements are in agreement with the FS model. Only in the case of the one-nanometer thick sample we find a relevant deviation.^[28,29] The FS model was developed to describe the electron transport of ultrathin metals by considering the electron scattering at the interfaces, see Section S11 (Supporting Information). It estimates the impact of the electron scattering at the metal interfaces which would be more relevant when the film thickness is comparable to the electron mean free path (≈ 5.5 nm in the case of nickel). In these conditions, the electrical transport of our films can be estimated according to the next expression:

$$\frac{\rho_f}{\rho_b} = \frac{\lambda_f}{\lambda_b} = \frac{Q_K}{k} \quad (1)$$

where Q_K is the coefficient that establishes the ratio between the electrical resistivity of the thin film (ρ_f) with respect to that of the bulk metals (ρ_b) or between the respective electron mean free paths (λ_f and λ_b , for the film and bulk respectively). The parameter k is the ratio between the film thickness (t) and λ_b .

The numerical solutions of Equation (1) are compared to the experimental measurements in **Figure 3a** consisting of the electrical resistivity for UTNF layers with thicknesses ranging from 10 to 1 nm. Again, two regions with different behavior can be observed, in this case, above and below 5 nm. For thicker films, i.e., 6–10 nm, the electrical resistance is closer to the numer-

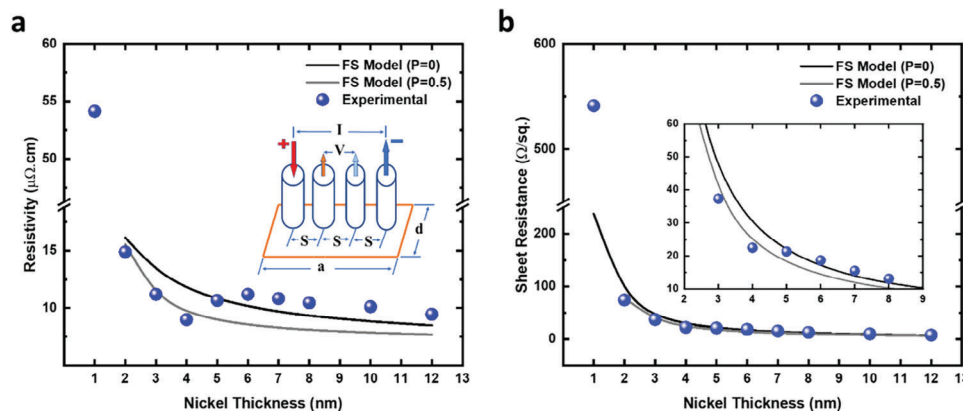


Figure 3. a) Electrical resistivity measured on a batch of samples with different thickness. Inset: scheme of the resistivity measurement for estimation of the sheet resistance. b) Sheet resistance from the same batch of samples.

ical solutions given by Equation (1), considering the specularity parameter $p = 0$ (see Section S11, Supporting Information for more details). Indeed, the experimental measurements are slightly higher than the numerical predictions and both curves are parallel up to 6 nm. Below this thickness the experimental resistivity is slightly reduced approaching the case of $p = 0.5$. Notice that only in the case of one-nanometer thick films the resistivity moves away from the numerical estimation only in the case of one-nanometer thick films. There is a combination of factors to explain this sudden divergence: a reduction of the film continuity in combination with the presence of a native oxide layer, which in the case of the thinnest films will be comparable with the real nickel thickness. The impact of the native oxide on the optical and electrical properties in UTNFs has been previously reported, being the films thinner than 4 nm the more sensitive to the presence of a native oxide.^[35] With the experimental and numerical data of Figure 3a we can calculate the sheet resistance as described in the Methods section. Quantitatively, the sheet resistance of ultrathin nickel films decreases from $74 \Omega \text{sq}^{-1}$ (for 2 nm thick films) to $7.8 \Omega \text{sq}^{-1}$ (for a 12 nm thick sample), see Figure 3b.

The sample characterization is completed with thermal measurements at room temperature to assess the potential application of our UTNFs as transparent electrodes in thermoelectric

devices. We focus on the thickness dependence of the Seebeck coefficient and the thermal conductivity. The experimental results of the Seebeck coefficient are shown in Figure 4a, exhibiting an unexpected linear thickness dependence. The thicker sample (10 nm) present a value $\approx -16 \mu\text{V K}^{-1}$ [in the range of the bulk value ($-19.8 \mu\text{V K}^{-1}$)].^[41] The Seebeck coefficient increases as the sample thickness decreases, achieving positive values in the case of the 3 and 2 nm thick samples (≈ 3 and $5 \mu\text{V K}^{-1}$ respectively). This controllable variation of the Seebeck coefficient supposes an important tuning range which enables the use of UTNF to fabricate thermoelectric junctions by combining both p- and n-thermoelectric materials.

In general, this suggests a transition from a regime where electrons dominate the thermoelectric effect to a regime where it is dominated by holes. But as mentioned above, the impact of reducing the thickness will be mainly related to the electron scattering at the interfaces. However, our experimental transport measurements show that the electron scattering at the interface becomes relevant only for thicknesses lower than 6 nm, while the thickness dependence of our experimental Seebeck measurements is clear for all the thicknesses under study. A rough prediction can be obtained from Ref. [42] grey and black lines Figure 4b.

It is also worth noting that only for one-nanometer films the electrical resistivity measurements exhibit a clear deviation from

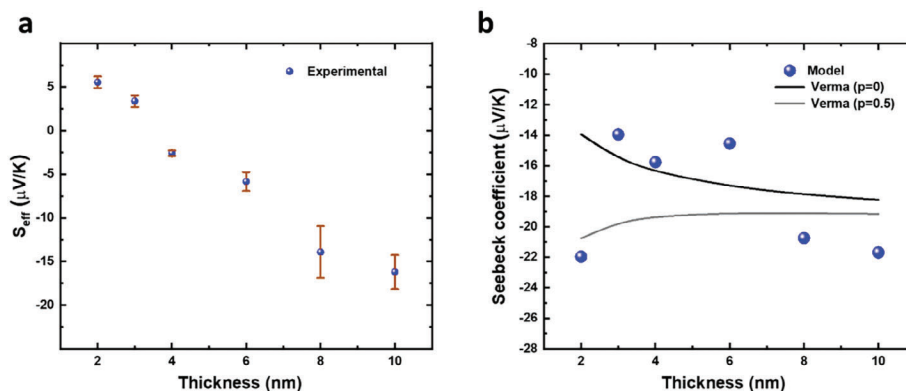


Figure 4. a) Effective Seebeck coefficient of UTNFs. b) The Seebeck coefficient values for UTNFs were measured at two specularity levels ($p = 0$ and $p = 0.5$).

Table 1. Seebeck coefficient predicted with the Model.

Thickness [nm]	S_{eff} [$\mu\text{V K}^{-1}$]	t_{Ni} [nm]	S_{Ni} [$\mu\text{V K}^{-1}$]
2	5.541	1.623	−21.961
3	3.374	2.623	−13.954
4	−2.597	3.623	−15.764
6	−5.840	5.623	−14.541
8	−13.923	7.623	−20.741
10	−16.199	9.623	−21.689

the FS model, which is generally attributed to the morphology of the film (e.g., roughness and loss of continuity). But this could not explain the change in the linear behavior of the thermoelectric response. Thus, the presence of the native oxide layer must be the reason behind the gradual increase of the Seebeck coefficient as the thickness is reduced. Importantly, the Seebeck coefficient of nickel oxide is $\approx 124 \mu\text{V K}^{-1}$.^[43,44] Then, in the case of two- and three-nanometer thick films, the thermoelectric voltage generated by the nickel would be in competition to few-angstrom nickel oxide layer giving an average p-type response.

The thickness dependence is like a previous work Schröder et al. in ultrathin Cr films, Ref. [45] In the case of ultrathin chromium films, the change is from an effective p- to n-type thermoelectric response. This could be understood as the metallic chromium has a positive Seebeck coefficient ($\approx 15 \mu\text{V K}^{-1}$), while the chromium oxide exhibits lower values ($+5 \mu\text{V/K}$ and $-10 \mu\text{V/K}$ in the case of CrO_2 and Cr_2O_3 , respectively).^[42,46,47]

$$S_{\text{eff}} = W_{\text{Ni}} S_{\text{Ni}} + W_{\text{NiO}} S_{\text{NiO}} \quad (2)$$

In this way, the resulting response of the film is determined by means of the weighted average of nickel (W_{Ni}) and nickel oxide (W_{NiO}) in the film. The effective Seebeck coefficient (S_{eff}) would correspond to our experimental measurement. As described in the Section S12 (Supporting Information), Equation (2) can be used to estimate the average thickness of the native nickel oxide

$t_{\text{NiO}} = 0.4 \text{ nm}$. Then, the Seebeck coefficient of nickel (S_{Ni}) can be isolated as follows:

$$S_{\text{Ni}} = \left(\frac{-t_{\text{NiO}}}{t} S_{\text{NiO}} + S_{\text{eff}} \right) \frac{t}{t_{\text{Ni}}} \quad (3)$$

This expression can be used to study the dependence of the S_{Ni} with the film thickness. Considering that, $S_{\text{NiO}} = 124 \mu\text{V K}^{-1}$ and $t = t_{\text{Ni}} + t_{\text{NiO}}$, we can observe the dependence of the Seebeck coefficient on the film thickness.

The results are shown in Table 1 and plot as blue scatters in Figure 4b. In general, the experimental values vary around the numerical predictions. However, a better estimation would require a more accurate characterization of the nickel oxide (out of the scope of this work) which here has been considered thickness independent. Also, the continuity and roughness of the sample could drive to certain deviations on S_{Ni} .

About the estimation of the thermal conductivity dependence on the film thickness of the UTNFs a preliminary estimation could be obtained from the electrical conductivity (or resistivity) using the Lorenz Number ($L^{\text{Ni}} = 2.44 \times 10^{-8} \text{ V}^2 \text{ K}^2$),^[48] (see blue scatters in Figure 5a). The results agree with the numerical model proposed by Verma et al.,^[42] (above used for the Seebeck coefficient). Those estimations are double-checked by means of Scanning Thermal Microscopy (SThM) (see scatters in Figure 5b). From the comparison between numerical and experimental values, we can conclude: first, the reduction of the film thickness below the electron mean free path drives to a dramatic reduction of the thermal conductivity of the resulting films; and second, differential measurements with SThM allow the proper characterization of ultrathin metal films.^[33,49,50]

In general, the SThM technique requires an accurate tip calibration. However, because of their planar geometry, our UTNFs can be treated as the case of a 1D multilayer structure. This is because in our experiments, we are not using the SThM probe for obtaining nanometric lateral resolution (in X and Y) but to produce a very little heat flow. Roughly speaking we could compare our measurements with a thermal van der Pauw method with a very tiny probe. Indeed, the samples here studied have not got nanometric lateral size. It is, our films have nanometric thickness (in the Z direction, out-of-plane) but are continuous in the X and Y direction (in-plane), simplifying the geometry of the

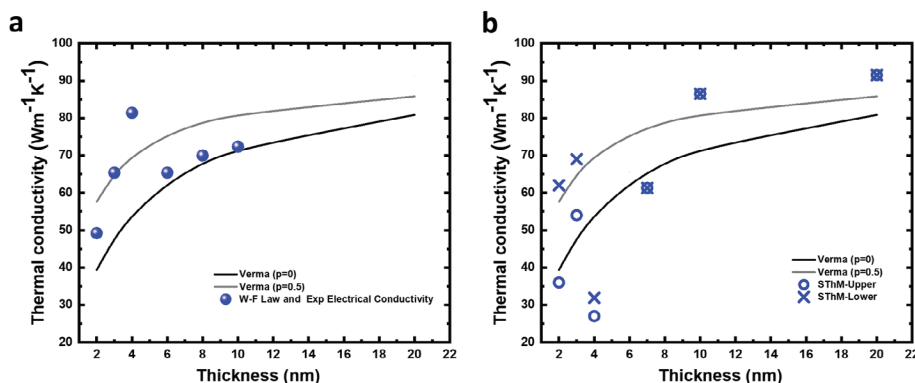


Figure 5. a) Thermal Conductivity coefficients of UTNFs calculated for two different p parameters, as described in ref. [42] (continuous lines), and using the Wiedemann–Franz Law on the experimental transport measurements (spheres). b) Analogous plot compared to values and experimentally determined upper (o) and lower (x) values obtained from the SThM.

problem. In addition, the challenge of quantifying the actual heat flow though the tip is simplified by making differential measurements, see Equations (4) and (5) (more details in SI4, Supporting Information).

The thermal conductivity can be determined by comparing the temperature gradients of our films with respect to a silicon sample. The procedure starts applying a moderate voltage to the SThM tip during the measurements (e.g., 100 mV). This voltage fixes a tip temperature slightly higher than room temperature (RT). Then, the tip reaches its maximum temperature in air (T_1), before contacting the sample. After getting into contact, the tip temperature is reduced to T_2 and the heat flow (q) could be defined as:

$$q^i = \frac{T_2^i - T_1^i}{R_c} = \frac{\Delta T_2^i}{R_c} \quad (4)$$

where R_c represents the contact thermal resistance, and the index i ($i = \text{Si}$ or Ni) refers to the material in contact with the tip. It is for nickel in the case of UTNFs and it is for silicon in the case of the reference sample. From Equation (5) we can determine the heat flow rate between a given UTNF and the silicon substrate as follows:

$$\frac{q^{\text{Si}}}{q^{\text{Ni}}} = \frac{T_2^{\text{Si}} - T_1^{\text{Si}}}{T_2^{\text{Ni}} - T_1^{\text{Ni}}} \quad (5)$$

Importantly, the heat flow rate will be the same for all the interfaces on the same sample. As described in the Section SI4 (Supporting Information), this will be exploited for determining the extension of the temperature gradients over the different materials (i.e., the thermal penetration depth). As an advantage, the problem of calculating the thermal conductivity is reduced to the estimation of thermal penetration depths due to flat geometry of our samples. Under the described experimental conditions, the thermal gradient into the nickel films is extended up to 4–7 nm (into the nickel) (see SI4, Supporting Information for a detailed description).

The determination of the thermal gradients allows the estimation of the thickness dependence of the thermal conductivity of nickel as shown in **Table 2**. The experimental SThM measurements are close to our numerical prediction. For example, a thermal conductivity of $\approx 54 \text{ W m}^{-1} \text{ K}^{-1}$ is obtained in the three-nanometre thick film, by considering a thermal penetration depth of 7 nm in nickel (upper limit). This value increases

Table 2. Thermal conductivity coefficient for UTNFs of different thickness estimated from SThM measurements.

Nickel thickness (nm)	T_2^{Ni} [mV]	ΔT_2^{Ni} [mV]	ΔT_3^{Ni} [mV]	K [$\text{W m}^{-1} \text{ K}^{-1}$]	
				Upper	Lower
20	912	−688	−512	91	91
10	922	−678	−522	86*	86
7	1025	−575	−625	61	61
4	1212	−388	−812	27	32
3	822	−778	−422	54	69
2	813	−787	−413	36	62

close to a 10% (up to $69 \text{ W m}^{-1} \text{ K}^{-1}$) if we consider the lower limit of 4 nm for the thermal penetration depth. In the case of thicker films, where the heat is not expected to reach the silicon substrate, we obtain values close to the nickel bulk, slightly higher than the numerical predictions (see Figure 5b). Only in the case of the four-nanometre thick sample we found an important deviation. As this thickness is close to the percolation point, the deviation could be related with a low surface quality. It is also worth noting that we have considered that the thickness of native oxide is constant for all the films, and this could be only true for thick samples. However, in the case of ultrathin films the native oxide is likely dependent on the film thickness and eventually on the surface morphology. Indeed, an increase of the native nickel oxide ratio around percolation would lead to the smoothing of the surface and could explain a low surface quality with a low RMS value.

Out of the thermal conductivity estimated for the four-nanometre thick sample, the results in Table 2 describe a gradual thickness dependence of the thermal conductivity in our UTNFs. This, in combination with thickness dependence of the Seebeck coefficient provides an interesting tuning range of the thermal properties of the UTNFs, as required for the design of thermoelectric nanodevices. The results demonstrate the potential of the SThM for the characterization of the thermal conductivity at the 2D limit. While the estimation of thermal conductivity on nanostructures results quite challenging, the planar geometry of our films allows an accurate quantitative estimation. This could be extrapolable to mechanically exfoliated 2D materials where the thermal conductivity is typically obtained from thermo-optical measurements with the corresponding limitations.^[51,52]

3. Conclusion

Ultrathin nickel films have been developed for the application on thermoelectric devices. The electrical, optical, and morphological characterization of the samples have been studied to determine the quality of the films. The characterization has been completed by measuring thermal properties of the films which presents a dramatic deviation with respect to those values reported for bulk nickel. In the case of the Seebeck coefficient, experimental values vary from -16 to $5 \mu\text{V K}^{-1}$ as the thickness is decreased from 10 to 2 nm. This is attributed to the existence of thin native oxide capping on the films ca. 4 Å. In the case of the thermal conductivity, regular characterization methods drive an important underestimation of the values due to the influence of the substrate. In this situation, differential measurements carried out by means of SThM can improve this estimation and reveal a clear thickness dependence. In further works, this tunability of the Seebeck and thermal conductivity will be exploited for the fabrication of thermoelectric nanojunctions. Importantly, our results will be essential to identify current limitations in the design of nanodevices based on emergent 2D materials whose thermoelectric properties cannot be properly characterized with conventional methods.

4. Experimental Section

Thin Film Growth: Solid source electron-beam thermal evaporation of nickel was optimized to obtain ultrathin films with thicknesses ranging

from 1 to 10 nm on BOROFLOAT substrates. The deposition was carried out into an ultrahigh vacuum chamber with a working pressure of $\approx 10^{-8}$ Torr with a growth rate of $1.5 \text{ \AA s}^{-1}$. The resulting high-quality films presented low roughness [$\approx 2 \text{ nm RMS}$] and non-oxidation insights beyond the native surface oxide. Previously to the growth, the substrates were exposed to a standard cleaning process consisting of three ultrasonic baths (of deionized water, acetone, and ethanol) followed by an overnight annealing at 450 K inside a glove box. The thickness of the films was determined using a quartz microbalance calibrated.^[53] The experimental conditions allow growing films of 1 nm with an accuracy of 0.3 nm.

AFM: Surface characterization have been carried out using the multimode Scanning Probe Microscope Nanoscope IV (from Bruker, former Veeco). The resolution of all the images was 512×512 pixels and they were acquired in Non-Contact Mode over a $500 \times 500 \text{ nm}^2$ area with a scan rate of 0.25 Hz lines per second. Thanks to the low surface roughness very similar feedback parameters during acquisition could be used.

Transport Measurements: Electrical conductivity measurements were carried out using a hand-made four-point probe, controlled by a Keithley 2400 multimeter under LabView interface. Films with well-known lateral sizes have been fabricated to estimate the corresponding sheet resistance using the geometrical approaches described in refs. [34,54,55] For measurement of the Sheet resistance, the expression $R_s = CV/I$ was used, being I the applied current, V the measured voltage, and C a correction factor considering the finite size of the sample. For the samples (of $30 \times 30 \text{ mm}^2$) $C = 3$. Importantly, the sheet resistances from different batches of samples were compared to double-check the reproducibility during the optimization of the growth process.

Transmittance: Optical transmittance measurements were carried out using a JASCO V-650 spectrophotometer in the range from 300–2000 nm. The transitivity of the samples including the substrate contribution was obtained by differential measurement after normalization by the sensitivity of the detectors.^[56]

Seebeck Measurements: A homemade system was assembled to measure the Seebeck coefficient. It was composed of two copper blocks thermally connected by the films under measurement. Two Peltier cells were used to generate, and a Lakeshore 340 temperature controller was used control the temperature gradient between the copper blocks, along the sample. The system was provided of additional electronics to measure the voltage generated by the thermal gradient due to the thermoelectric response of the sample. $S = -\Delta V / \Delta T$.^[57]

Thermal van der Pauw: A homemade system was used to measure the thermal conductivity based on the Van der Pauw method.^[58] In this system, the sample surface temperature was monitored by means of four standard PT-100 resistors in combination with two standard PT-1000 resistors (which were employed as heaters). The temperature of the heat sink was fixed using a Peltier cell controlled with another Lakeshore 340 temperature controller and cooled with a laser heat exchanger. Additionally, a Keithley DAQ6510 switching system was used to measure the voltage for calculating the heating power applied to the sample. All the equipment were controlled using a LabVIEW interface. These systems, widely used in-house for thicker films, illustrate the challenge of carrying out thermal measurements in thin films, and then, justify the use of SThM.

SThM: A Scanning Thermal Microscope VITA module, from Bruker Corp., was employed to estimate the thermal conductivity of ultrathin nickel films. Using a special AFM tip (VITA-HE-GLA-1) with spring constant 0.5 N m^{-1} and resonance frequency 50 kHz that it was same role as a heater, and a thermal probe, the heat diffusion was monitored during a force curve approach–retract curve in nickel films of different thicknesses.^[59] The experiments were carried out on both BOROFLOAT and silicon samples to estimate the dependence of the thermal conductivity on the nickel thickness by differential measurements. This way, the impact of the substrate could be checked in the differential measurements carried out on a low thermal conductivity borosilicate and a high thermal conductivity silicon substrate. Final values were obtained using silicon. The optical, electrical, and morphological characterization of the films was carried out on BOROFLOAT as this is extensively used for the design of optoelectronics and for lab on chip devices. The growth process can be extrapolated to silica and silicon substrates with minor variations of the

resulting properties of the films which in addition present the same morphology.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available in the supplementary material of this article.

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electrical conductivity, scanning thermal microscopy, Seebeck effect, semi-transparent metal films, thermal conductivity, ultrathin metal films

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