Optical modeling of conductive layers

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Abstract
Many coatings for technical applications contain one or more conductive layers. These are included in the stack in order to provide either electrical conductivity or high reflectivity which is typical for conductive layers in some spectral ranges. Although modeling the optical response of freely moving charge carriers is quite easy and straightforward applying the classical Drude model, comparing measured and simulated optical and electrical properties often ends up with disagreement. This is irritating and unsatisfying, especially since optical models are likely to be used in the design of new coating products. For technically relevant silver, NiCr and TCO layers typical modeling problems are discussed. We present solutions which are capable of reproducing both optical and electrical measurements.

Introduction
Electrically conducting thin films are applied in many coating products. The most obvious reason to use a metallic or semiconducting film is the requirement of an electrical contact layer. If visible light is going to pass through the layer a transparent conductive oxide (TCO) may be applied – otherwise a metallic layer (which is usually not transparent for light in the visible) can do the job as well.
For some types of coating products metallic layers are used because of their high reflectivity. Depending on details of the final application, good candidates for this purpose are silver, gold and aluminum. In order to suppress the emission of infrared radiation by window glass so-called low-e coatings are used with silver providing the required high reflectance of the coating in the mid infrared. If a thin film's task is to absorb a large fraction of incident light then less noble metals are used because of their low reflectivity combined with high absorption. NiCr is a typical material of this kind.

Optical modeling is a well-established method in the design phase of thin film coatings products. Good models reproduce measured spectra of thin film systems and predict the optical properties of new layer stacks with high precision, reducing the experimental effort to just verifying the predicted properties. Satisfying models for TCO and absorber layers are discussed below.

There are cases, however, for which the agreement of simulated and measured optical and electrical properties is poor. Obviously the model misses a feature of the real coating which leads to the observed mismatch. It is shown in the following that a depth gradient of the electron mobility can remove a typical discrepancy between optical and electrical measurements of silver layers. The extended layer model successfully reproduces all measured optical and electrical quantities for different thicknesses.

Developing an optical model for silver layers
The development of optical models for conductive thin films is demonstrated for sputtered silver layers on glass. Optical spectra of the glass have been measured before the deposition, and a suitable model for the optical constants of float glass has been adjusted to nicely match the measured data. Silver films with nominal thicknesses of 10, 20 and 120 nm have been deposited. Their optical spectra and the sheet resistance were recorded, as well as the normal emissivity in the mid infrared.
In a first approach literature data for the optical constants of silver [1] were tried to generate reflectance (film side and glass side) and transmittance spectra. The only unknown in the model is the Ag thickness which can easily be automatically fitted. However, as shown in fig. 1, the agreement of model and

measured data is poor. A more flexible optical model is needed to account for the properties of this sample.

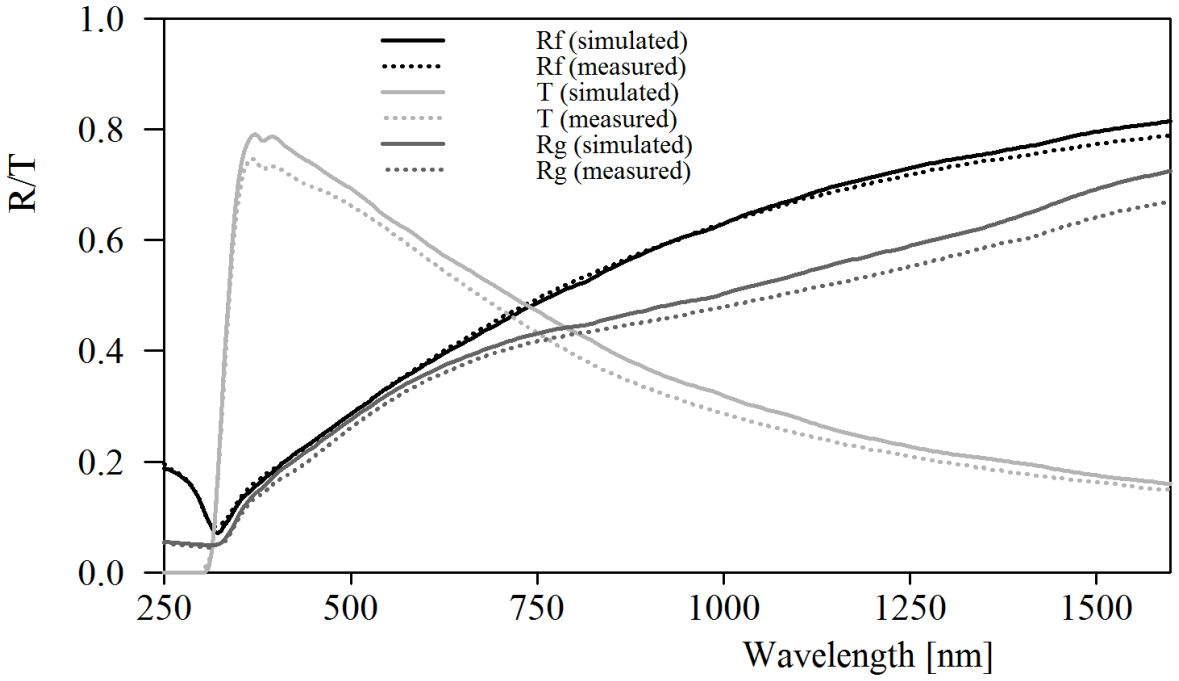

Figure 1. Comparison of measured and simulated spectra, computed using literature data [1] for the optical constants of silver.

The most obvious difference between silver layers is caused by varying mobility of the free electrons in the film. Layers may differ in the density of grain boundaries and surface roughness. Both lead to different scattering rates for the electrons moving in the layer. The electron density as well as interband transitions are not very likely to vary from sample to sample. Hence a mechanism is needed which keeps the electron mobility adjustable while all other optical properties remain constant. In order to achieve this goal it is needed to model interband transitions and free electron properties reproducing literature data in a first step. This was done by adding up a constant susceptibility, two Tauc-Lorentz interband transition models [2] and a Drude model [3] for the free electrons.

The Drude model considers a single representative electron which is accelerated by external electric fields. While it moves through the metal, collisions with defects occur and the electron loses its momentum. The macroscopic response to electric fields is then obtained by scaling up the averaged electron motion. There are two parameters in the Drude theory only, namely the electron density and the collision rate. In the case of semiconductors the electron mass in the Drude expressions is to be replaced by the effective mass. Re-formulations of the Drude terms adapted to optical constant modeling sometimes use the parameters plasma frequency and damping constant. These are directly related to carrier concentration and collision rate.

Applying the model as described above and fitting the electron damping constant in addition to the film thickness, one gets a much better agreement of model and measurements (see fig. 2). The only remaining problem is the transmittance in the range 350 … 450 nm which is too high in the model.

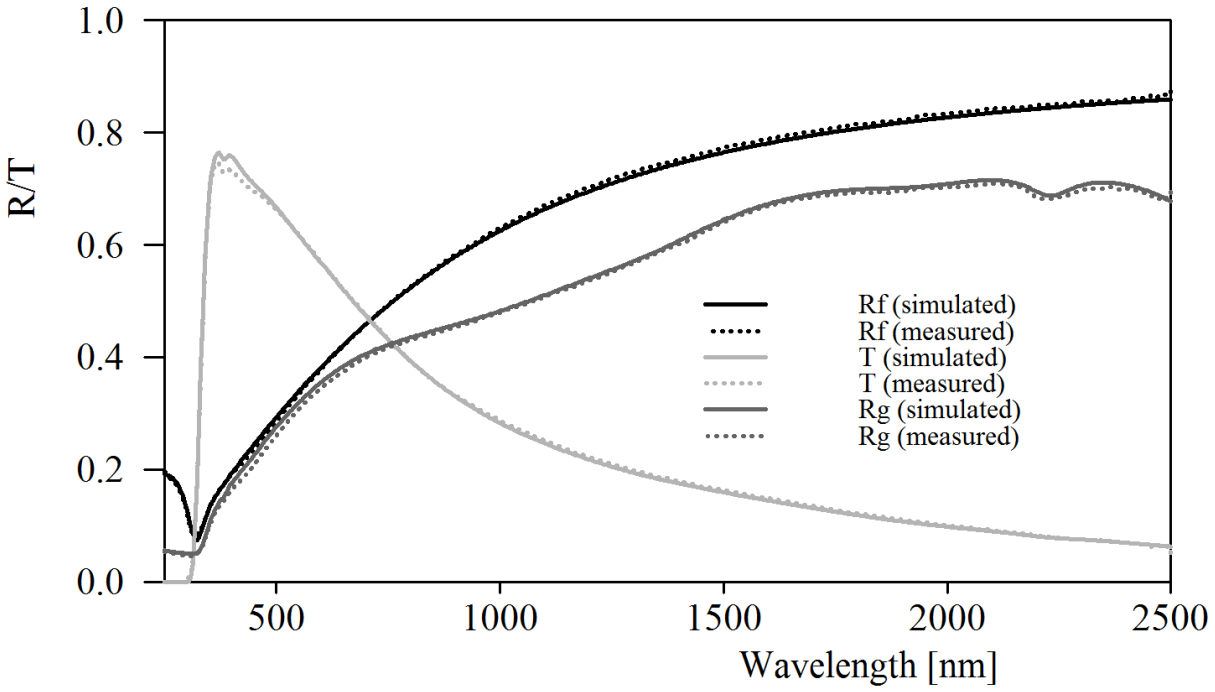

Figure 2. Fit of the optical spectra of a 10 nm silver film on glass, adjusting the electron damping and the film thickness only.

Up to now optical spectra in the UV, visible and near infrared have been taken into account only. However, the Drude model can be used to compute mid infrared as well as electrical properties as well. Unfortunately, the comparison with measured values turns out to be disappointing. The simulated emissivity [4] of 8.1% is much lower than the measured value of 8.6%, and the model sheet resistance of 7.8 Ω differs significantly from the measured value of 6.1 Ω.

In order to further improve the model it is useful to take a look at the deposition method and known properties of the material. Silver layers sputtered on glass exhibit a discontinuous island structure before a continuous silver film develops. This means that the density of grain boundaries is likely to be higher at the bottom of the layer than at the top. Translated into electron properties this suggests to try a depth gradient of the damping constant, with decreasing damping from bottom to top of the film. A 'gradient' of 2 silver sublayers is already sufficient to achieve agreement of the sheet resistance, maintaining the optical fit quality shown in fig.2. The simulated value of the mid infrared emissivity, however, is still too low in this approach.

The disagreement of the transmittance spectra in the range 350 … 450 nm gives a hint in what direction the model needs to be extended. Inhomogeneous silver mixtures with other materials absorb in this spectral range [5]. Assuming that the silver layer is not perfectly smooth at the top, a thin air-silver composite (using the Bruggeman effective medium model [6]) was added to the model. Although this extra layer with a silver volume fraction of 10% leads to a much better agreement of the optical spectra, the mismatch of the emissivity values is not affected, unfortunately. This last problem of the model can be removed by adding a second, very special 'roughness' layer underneath the top roughness: The silver content of this mixture is above 99% but 'air trenches' do not allow the electrons to freely move – there is no percolated silver network in this layer. Silver (and other noble metal) composites of this kind show extreme optical constants and have been studied as candidates for highly sensitive infrared sensors [7][8].

The final model which is capable of reproducing all measured data is shown in fig. 3. The thickness of the two bottom layers with strong and medium damping sums up to 5.6 nm. This is in the thickness range where a continuous silver film is formed [9] – it is very likely that below this 'threshold' the density of grain boundaries is high. Above these two layers the model contains a thin (2.2 nm) defect-free highly conductive film, followed by the previously discussed two discontinuous layers that account for surface roughness.

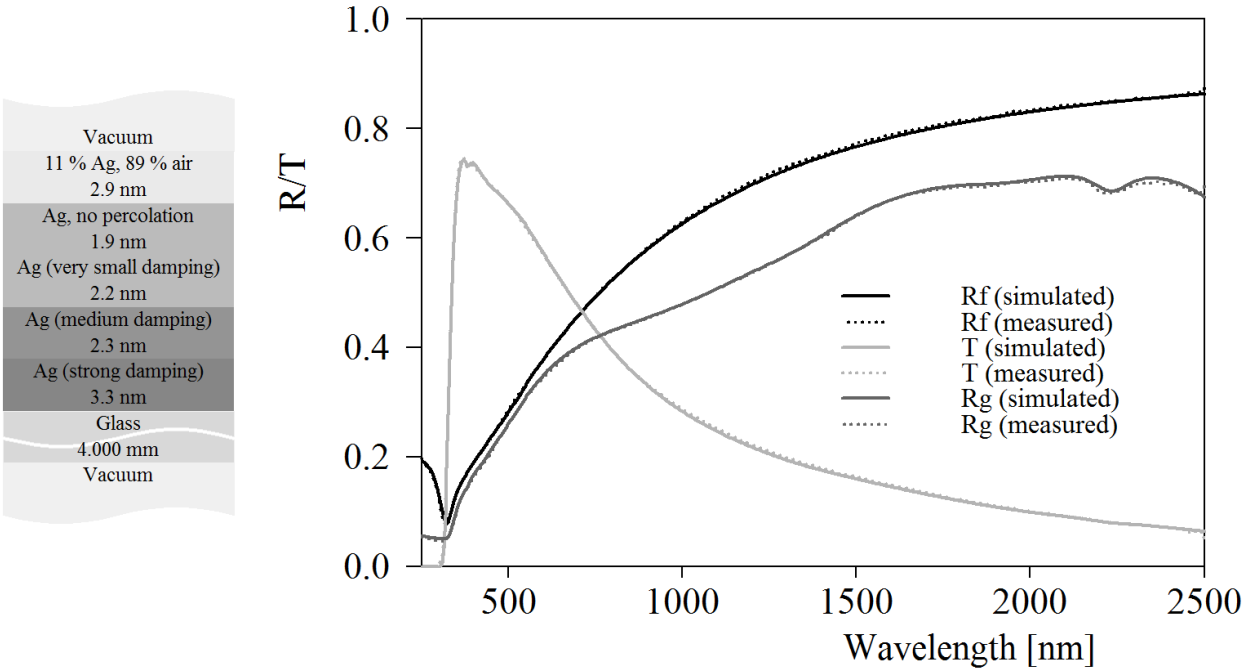

Figure 3. Final optical model which leads to a perfect match of simulated and measured data, including the emissivity of 8.6 % and the sheet resistance of 6.1 Ω. See text for discussion.

The model has been verified with thicker layers (20 nm and 120 nm). Similar optical fit quality has been achieved, and in all cases the emissivity and sheet resistance values of the model matched those that were measured. Figure 4 shows the result for the 20 nm layer.
It should be noted that silver is a metal with extreme optical constants. The high concentration of very mobile electrons cause strong optical effects if the material gets inhomogeneous. Most other metals are less sensitive and easier to model, like the one in the next section.

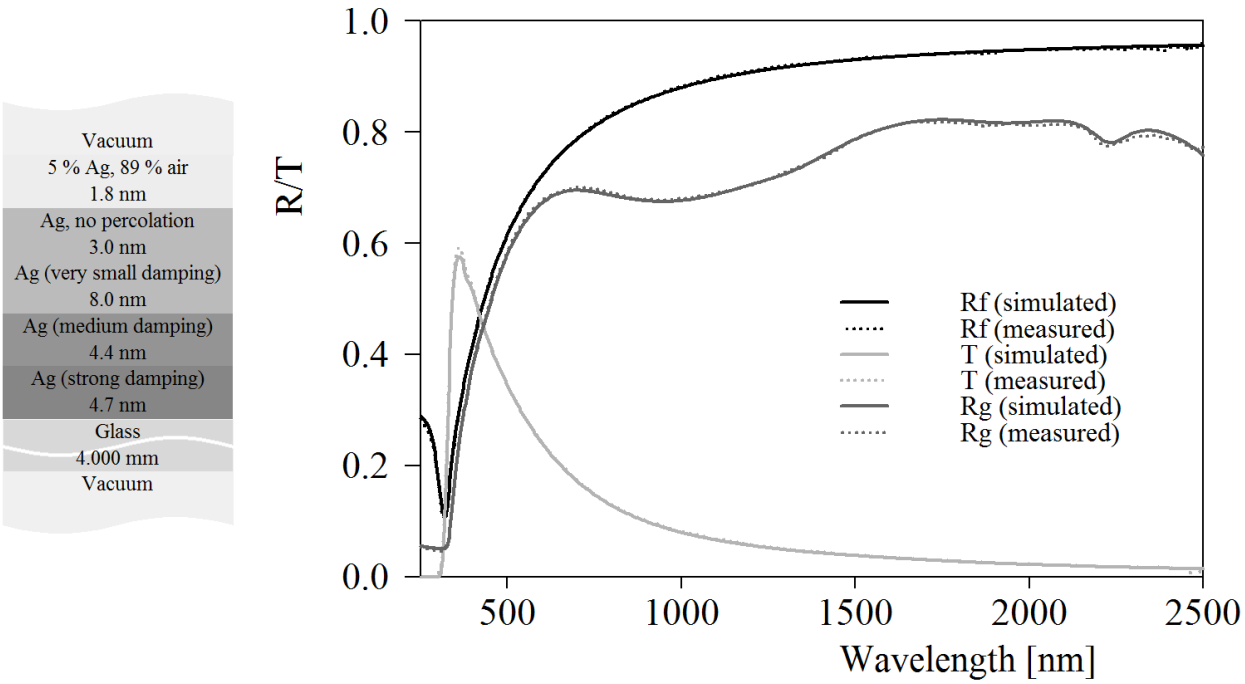

Figure 4. Simulated and measured optical spectra of a 20 nm thick Ag layer on glass. The measured normal emissivity of 3.4 % and the sheet resistance of 2.2 Ω are perfectly reproduced by this model.

Absorber layers

If the task of a thin film in a coating product is to efficiently absorb visible light, a 'non-noble' metal with large electron damping is the material of choice. A good example is NiCr. Like in the case of silver discussed previously, the optical constant model for NiCr consists of interband transitions and a Drude model. However, the damping of the electrons is much stronger and one of the interband transitions is located in the near infrared. The strong damping of both the electrons and the interband transition lead to a very smooth, almost feature-less shape of the refractive index. Its real part n and the imaginary part k have similar size and shape.

The optical constant model for NiCr consists of four susceptibility terms: a constant, an OJL interband transition model in the UV [10], a Kim oscillator [11] in the near infrared representing an interband transition as well, and a highly damped Drude model. In this case it turned out that a simple single layer model is sufficient to reproduce measured optical and electrical values. Figure 5 gives details for a 10 nm layer of NiCr on glass. It was verified that the same model works for other thicknesses as well.

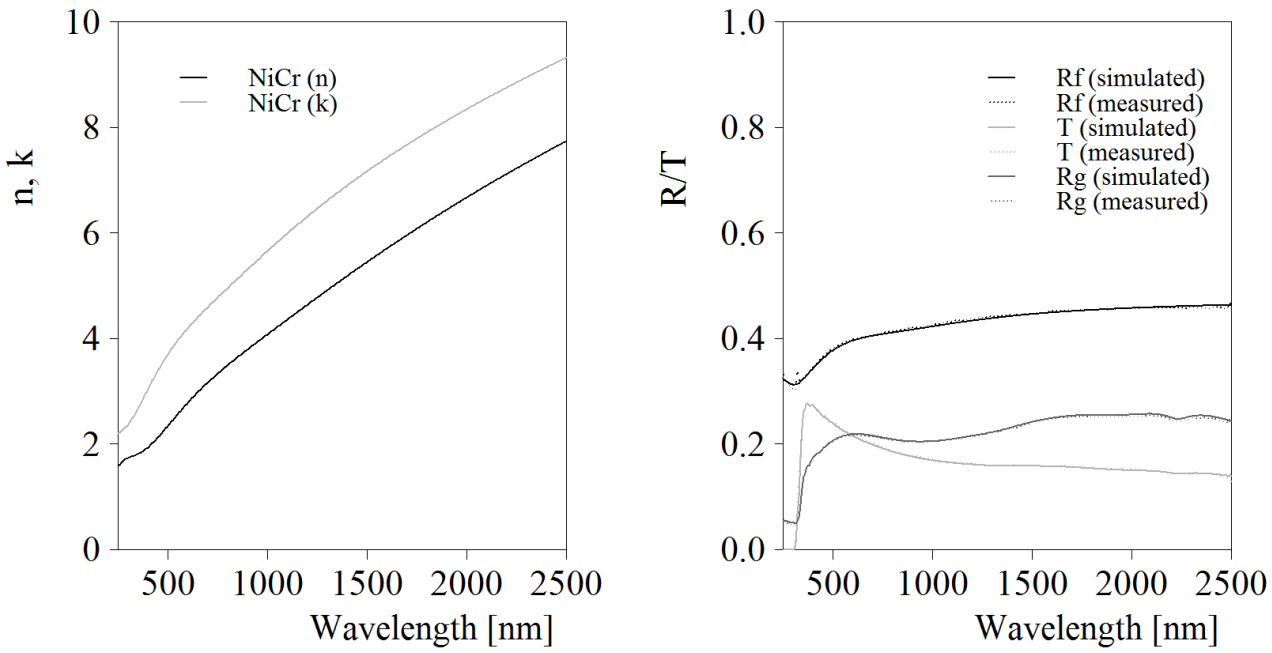

Figure 5. Fit of an optical model for a 10 nm NiCr layer on glass. The complex refractive index obtained by the fit procedure is shown on the left. The sheet resistance of the model perfectly matches the measured value of 96.8 Ω. The difference between measured and simulated emissivity is small (54.8 % and 52.6 %, respectively).

Transparent conductive layers

As shown before, the simple Drude model is sufficient to describe the properties of metallic materials. It can also be applied to model the properties of charge carriers (electrons or holes) in semiconducting materials. In the case of high doping levels, however, the Drude model fails to deliver high quality optical constants - often fit results are poor. The reason is the large density of ionized doping atoms which cause a frequency-dependence of the electron damping. As suggested in [12] a smooth transition from low damping in the UV to high damping in the infrared nicely matches the electron properties.

This extension of the Drude model is recommended for transparent conductive oxide (TCO) layers. Figure 6 shows a comparison of the performance of the classical and the extended Drude models, together with the shape of the damping constants assumed in these approaches. If the extended Drude model is used to compute electrical values the low frequency (static) value of the damping constant must be taken.

Although not necessary in the example shown in fig.6, optical models for TCO layers frequently require a depth gradient of the optical constants. In contrast to the case of silver layers where only the electron damping was depth-dependent, TCO layers may exhibit a depth gradient of the density of the doping atoms. In such a case both the plasma frequency and the charge carrier damping change from bottom to top of the layer.

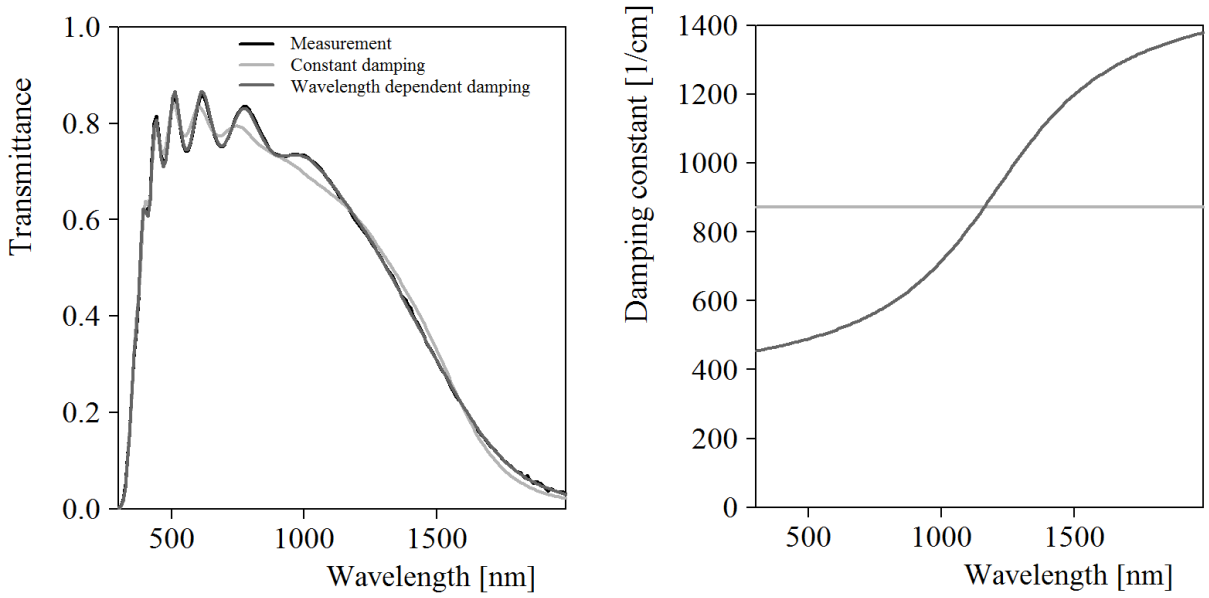

Figure 6. Left: Measured transmittance of a thick (630 nm) ITO layer on glass and two fit results, applying the classical Drude model with constant damping and the extension with wavelength-dependent damping. The values of the damping constants are compared in the right graph.

Summary

The required ingredients for successfully modeling the optical properties of conducting thin films have been discussed. The applied Drude model for free charge carriers allows the computation of both electrical and optical properties. High quality models for NiCr and ITO have been presented. For silver layers a depth gradient of the electron damping had to be introduced to achieve a perfect match of simulated and measured values.

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