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Experimental determination of the uncertainty of the absorption coefficient of crystalline silicon

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Abstract

Based on a combined analysis of spectroscopic ellipsometry, reflectance and transmittance measurements as well as spectrally resolved luminescence measurements and spectral responsivity measurements, we present data of the coefficient of band-to-band absorption of crystalline silicon at 295 K in the wavelength range 250 – 1450 nm. A systematic measurement uncertainty analysis according to the “Guide to the Expression of Uncertainty in Measurements” (GUM) is carried out for each method, showing that the relative uncertainty of the absorption coefficient data so determined is of the order of 0.3% at 300 nm, 1% at 900 nm, 10% at 1200 nm and 180% at 1450 nm. The data are consolidated by comparison of measurements carried out independently at different institutions. The uncertainty of solar cell energy conversion predictions by means of simulations due to the uncertainty of the absorption coefficient data is shown to be of the order of 0.1% relative.

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1. Introduction

The absorption coefficient of crystalline silicon is an important material parameter for a variety of applications in the field of photovoltaics, e.g., device simulations aiming at the prediction of energy conversion efficiencies or the analysis of luminescence measurements. It is thus no surprise that the determination of the absorption coefficient has been an ongoing subject of research since 1955 [1,2,3,4,5,6,7]. However, as shown in Fig. 1, significant deviations between the data sets from literature are found, which are of the order of up to 30%. It is unclear whether these deviations are only due to specific properties of the investigated samples such as doping concentration, sample temperature or the surface geometry, and to which extent they can be explained by measurement uncertainty. The accuracy of the literature data is not assessable since measurement uncertainties have not been determined systematically or, as for most of the studies, have not been specified at all. This lack of information casts doubt on the accuracy of combined data sets which have been calculated from other sources. Moreover, deviations between measured and modeled luminescence spectra of crystalline silicon wafers and solar cells have been reported [8,9] when using the absorption coefficient data of Green [7], which is probably the most widely used data set in the photovoltaic community, as input for the model.

We aim at resolving the mentioned discrepancies by presenting new data of the coefficient of band-to-band absorption of crystalline silicon at 295 K using spectroscopic ellipsometry, measurements of reflectance and transmittance, spectrally resolved luminescence measurements and measurements of the spectral responsivity of silicon solar cells. Together, these measurements cover the wavelength range 250 – 1450 nm, as shown in Fig. 2. For each method, a systematic measurement uncertainty analysis is carried out. The analysis is based on an extensive characterization of the measurement setups and follows the *Guide to the expression of uncertainty in measurement* (GUM) [10]. The resulting absorption coefficient data is consolidated by intercomparison of measurement results obtained at different institutions and using different measurement methods. Temperature coefficients are provided, allowing the data to be transformed to other temperatures. Moreover, we analyze the impact of the uncertainty of the absorption coefficient on the accuracy of solar cell energy conversion efficiency predictions by means of device simulations and show that the presented data resolve the aforementioned discrepancies when modeling luminescence spectra.

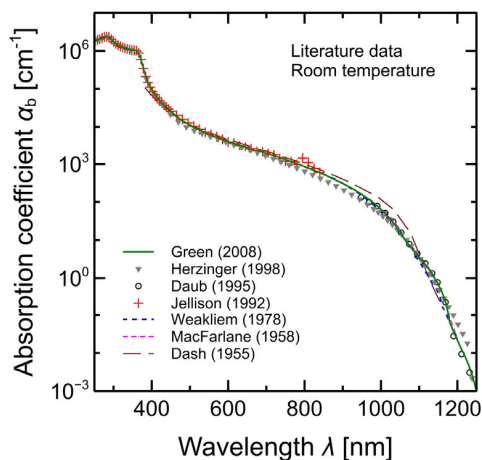


Fig. 1: Literature data of the absorption coefficient of crystalline silicon at room temperature.

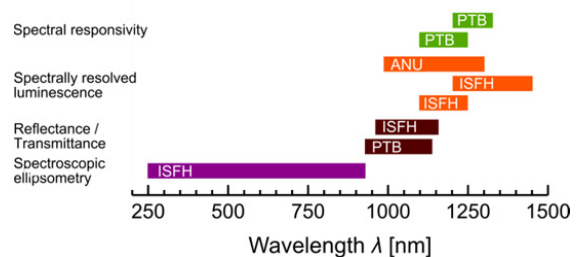


Fig. 2: Overview of the methods applied in this work for the determination of the absorption coefficient.

2. Theory

This paper aims at the determination of the coefficient of band-to-band absorption α_{bb} , which describes the absorption of photons leading to the generation of electrical charge carriers. This is the fundamental absorption process for photovoltaic devices. Beside band-to-band absorption, free carrier absorption (FCA) may be a significant absorption process and must thus be considered. FCA is described by the coefficient of free carrier absorption α_{fc} . A parametrization of α_{fc} is given by Green [11]. In the wavelength range which is of interest for silicon photovoltaics, other absorption processes such as band-impurity absorption can be neglected. The absorption coefficient α is thus the sum of band-to-band and free carrier absorption:

$$\alpha = \alpha_{bb} + \alpha_{fc} . \quad (1)$$

2.1. Spectroscopic ellipsometry

Ellipsometry determines the change of the degree of polarization of a light beam after reflection at a surface. This change is usually described by two parameters, Ψ and Δ , which refer to the change in phase difference between the parallel and perpendicular components and to the ratio of the reflection coefficients of the incoming and outgoing light wave, respectively. From this polarization data, the complex index of refraction is obtained by fitting the data with a suitable optical model for the layer stack. The fit is carried out separately for each wavelength. The absorption coefficient is related to the extinction coefficient κ , which forms the imaginary part of the complex index of refraction, by

$$\alpha(\lambda) = \frac{4\pi\kappa(\lambda)}{\lambda} . \quad (2)$$

In order to obtain Kramers-Kronig consistent data curves, the data so obtained can be fitted with a dispersion relation, which represents the dielectric function of the sample. Further details on ellipsometry can be found in the literature, e.g., [12]. In the wavelength range where ellipsometry is applied in this work (250-930 nm), the coefficient of band-to-band absorption is several orders of magnitude larger than the coefficient of free carrier absorption. The latter can thus be neglected, i.e., these measurements directly determine the coefficient of band-to-band absorption.

2.2. Measurements of reflectance and transmittance

Measurements of reflectance and transmittance are a straight-forward method for the determination of the absorption coefficient. For a planar sample, reflectance R and transmittance T are given by

$$R = R_s \left(1 + \frac{(1 - R_s)^2 \exp(-2\alpha W)}{1 - R_s^2 \exp(-2\alpha W)} \right) \quad (3)$$

and

$$T = \frac{(1 - R_s)^2 \exp(-\alpha W)}{1 - R_s^2 \exp(-2\alpha W)} . \quad (4)$$

In these equations, R_s denotes the surface reflectance and W the thickness of the sample. The absorption coefficient follows from the solution of both equations for α . Note that measurements of reflectance and transmittance are sensitive to both band-to-band and free carrier absorption. In order to obtain the coefficient of band-to-band absorption, a correction may thus be necessary, depending on the wavelength.

2.3. Spectrally resolved luminescence measurements

For typical luminescence measurement conditions, the emitted luminescence photon flux per wavelength interval and surface area $\Phi(\lambda)$ can be approximated by [8,13,14,15]

$$\Phi(\lambda) \approx \alpha_{\text{bb}}(\lambda) \frac{8\pi c n_{\text{Si}}(\lambda)}{\lambda^4} \int_0^W dz f_{\text{esc}}(\lambda, z) \exp\left(\frac{\mu_{\text{ph}}(z) - hc/\lambda}{kT}\right) \quad (5)$$

where c is the speed of light, n_{Si} is the refractive index of silicon, μ_{ph} is the chemical potential of the photons, which is given by the splitting of the quasi-Fermi levels, h is the Planck constant, k is the Boltzmann constant, T is the sample temperature and f_{esc} is the luminescence photon escape probability. In two cases, the relation simplifies [5]:

- If the charge carrier concentration is homogenous, μ_{ph} is independent of z and can be pulled out of the integral. The remaining integral is then proportional to the luminescence spectrum and equals the absorptance of the sample.
- If the absorption coefficient is small, the photon escape probability becomes independent of z . The integral is then a constant with respect to wavelength and the luminescence spectrum becomes proportional to the absorption coefficient.

In both cases, the absorption coefficient can be determined from relative luminescence measurements. The proportionality factor can be determined experimentally by scaling the data to previously determined absolute values of the absorptance or absorption coefficient, respectively.

2.4. Spectral responsivity measurements

The spectral responsivity SR of a solar cell, defined as short circuit current per incident power of light, is related to the external quantum efficiency EQE by

$$EQE(\lambda) = SR(\lambda) \frac{hc}{q\lambda}, \quad (6)$$

where q is the elementary charge and the other symbols have the meaning as introduced above. The EQE is connected to the luminescence spectrum by an optical reciprocity theorem [16]. Both quantities can thus be transformed vice versa and the same theory as for the determination of the absorption coefficient from luminescence spectra also applies to measurements of the spectral responsivity.

2.5. Systematic measurement uncertainty analysis

The measurement uncertainty analysis presented in this work is based on an extensive characterization of the measurement setups and follows the procedure outlined in the *Guide to the expression of uncertainty in measurement* (GUM) [10]. The GUM defines the international standard for the systematic evaluation of measurement uncertainties. Basically, it provides a framework for the calculation of the uncertainty of an *output quantity* Y , which is related to *input quantities* X_i by the process equation $Y = f(X_1, \dots, X_N)$, from the uncertainties of the input quantities. The *combined standard uncertainty* u_c of the output quantity usually refers to a *coverage probability* of about 68%. The coverage probability is the probability that a measurement yields a result which deviates from the expectation value of the measurement by not more than the specified uncertainty. Throughout this paper, the *expanded uncertainty* for a *coverage factor* $k = 2$ is indicated, which corresponds to a coverage probability of about 95%.

The conformity of two measurement results x_1 and x_2 with respect to their uncertainty can be assessed quantitatively by calculating the E_n -number [17] which is also used in international key comparisons as a criterion for measurement compatibility. It is defined by

$$E_n = \frac{1}{k} \frac{|x_1 - x_2|}{\sqrt{u_c^2(x_1) + u_c^2(x_2)}}. \quad (7)$$

In this paper, compatibility is accepted if $E_n \leq 1$ holds, i.e., the difference between the two results is smaller than the uncertainty of this difference for $k = 2$.

3. Measurements

The measurements presented in this paper are carried out at ISFH and PTB on various samples. For spectroscopic ellipsometry and reflectance/transmittance (RT) measurements, chemo-mechanically double side polished monocrystalline Czochralski grown *p*-type silicon wafers with a resistivity of 4 to 6 Ωcm are used. The samples are RCA cleaned after polishing. The sample thickness is $(653.0 \pm 1.9) \mu\text{m}$ (sample A) and $(1284.0 \pm 1.9) \mu\text{m}$ (sample B). These values are determined by measuring the thickness at four positions on the sample using a dial gauge. The area of both samples is $3 \times 3 \text{ cm}^2$. During ellipsometry and reflectance/transmittance measurements, the sample temperature is $(295 \pm 1) \text{ K}$. Together, both methods cover the wavelength range 250-1160 nm. Ellipsometry is performed at ISFH, RT measurements are performed both at ISFH and PTB. For the latter, $E_n \leq 1$ is found for all wavelengths, which means that the data are in agreement with respect to their uncertainty.

In the wavelength range from 1100 to 1250 nm, spectrally resolved electroluminescence (EL) measurements and measurements of spectral responsivity (SR) are carried out at ISFH and PTB, respectively. For both EL and SR measurements, specially designed lab-type solar cells which are made of *p*-type Czochralski grown silicon with a resistivity of 2.5 Ωcm and a thickness of $(711.0 \pm 2) \mu\text{m}$ are used. These solar cells feature chemo-mechanically polished surfaces. The surface metallization is achieved by evaporating aluminium on the rear surface (10 μm) and a grid structure on the front surface (also aluminium, 10 μm). In order to preserve the polished surfaces, neither an anti-reflection coating nor a back surface field is applied. The inhomogeneous charge carrier concentration within the solar cell can be shown not to affect the determination of the absorption coefficient by more than 1% rel. above 1100 nm [18], where the EL data is evaluated.

In the wavelength range from 1200 to 1450 nm, spectrally resolved photoluminescence (PL) measurements are carried out on samples from the same wafers as for the RT measurements. However, these samples are electrically passivated on both sides by 15 nm thick layers of Al_2O_3 (atomic-layer deposited). Moreover, SR measurements are carried out on industrial c-Si solar cells in the wavelength range 1200-1320 nm. The sample temperature for all EL, PL and SR measurements is $(295 \pm 0.5) \text{ K}$. As for the RT measurements, $E_n \leq 1$ is found for the EL/PL/SR data for all wavelengths. From the results of the different measurements, a best estimate for the coefficient of band-to-band absorption is obtained by calculating a weighted average of the data, the weights being given by the inverse squared uncertainty of the data. Moreover, corrections for free carrier absorption are applied as outlined in [19].

The uncertainty analysis includes measurement uncertainty contributions due to measurement noise, spectral bandwidth, wavelength accuracy, nonlinearity of the detectors, effects of the measurement amplifiers, impact of oxide layers on the samples' surfaces, spectral stray light, angle of incidence of light, depolarization of the light beam by the sample and reproducibility of the measurements. The uncertainty of the resulting absorption coefficient

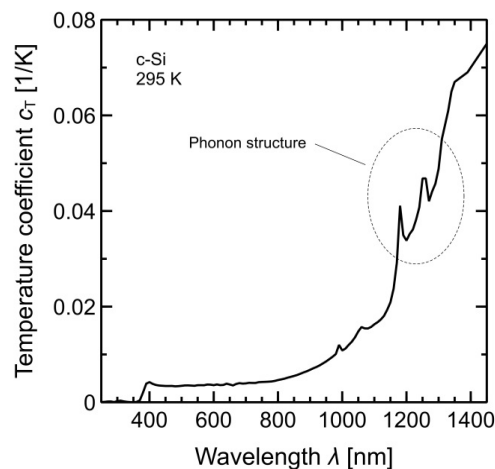


Fig. 3: Temperature coefficient of the absorption coefficient at 295 K. Around 1200 nm, the phonon structure is visible in the data.

data is then evaluated by analytical calculations as specified in the GUM. For the evaluation of ellipsometric measurements, however, this is not possible since the determination of the absorption coefficient involves a fitting procedure and an analytical process equation cannot be formulated. The uncertainty of the absorption coefficient resulting from the ellipsometry data is thus evaluated by performing a Monte-Carlo simulation, which is a numerical approach for the evaluation of measurement uncertainty. This approach also allows to take correlations between the different angles of incidence during the measurements into account. In order to evaluate the uncertainty contribution due to deviations of the sample temperature from the nominal temperature of 295 K during the measurements, the temperature coefficient is determined by carrying out ellipsometry at different sample temperatures around 295 K. Furthermore, PL measurements are carried out at different temperatures at the ANU. From these measurements, the temperature coefficient

$$c_T(\lambda) = \frac{1}{\alpha(\lambda, T_0)} \left. \frac{d\alpha(\lambda, T)}{dT} \right|_{T_0}, T_0 = 295 \text{ K} \tag{8}$$

is determined. Additionally, literature data of the temperature coefficient [7] are taken into account. From these data, the best estimate for the temperature coefficient is obtained by calculating the average of the data, which is shown in Fig. 3.

4. Results of measurements

The resulting absorption coefficient data is shown in Fig. 4 (left graph). The solid black line visualizes the data as determined in this work, the dashed black line represents the relative uncertainty of these data. For comparison, literature data which are also available for 295 K [5,7,20] are also visualized. For the data of Green, “error limits” are given for three distinct wavelengths (250, 460 and 1200 nm). In order to use this information, it appears reasonable to interpret these data as estimates of the measurement uncertainty for $k=2$. Note that the exact meaning of “error limits” is not specified. Due to the lack of information about its behavior at other wavelengths, the uncertainty is, as a first approximation, linearly interpolated between the given values. The graphs on the right show the ratio of the different data sets and the E_n number. As the uncertainty of the data from [5] and [20] is unknown, the “Pseudo E_n number” is calculated, which is defined analogously but neglects the unknown uncertainty of the literature data. The Pseudo E_n number thus indicates whether the deviations can be explained solely by considering the uncertainty of our absorption coefficient data.

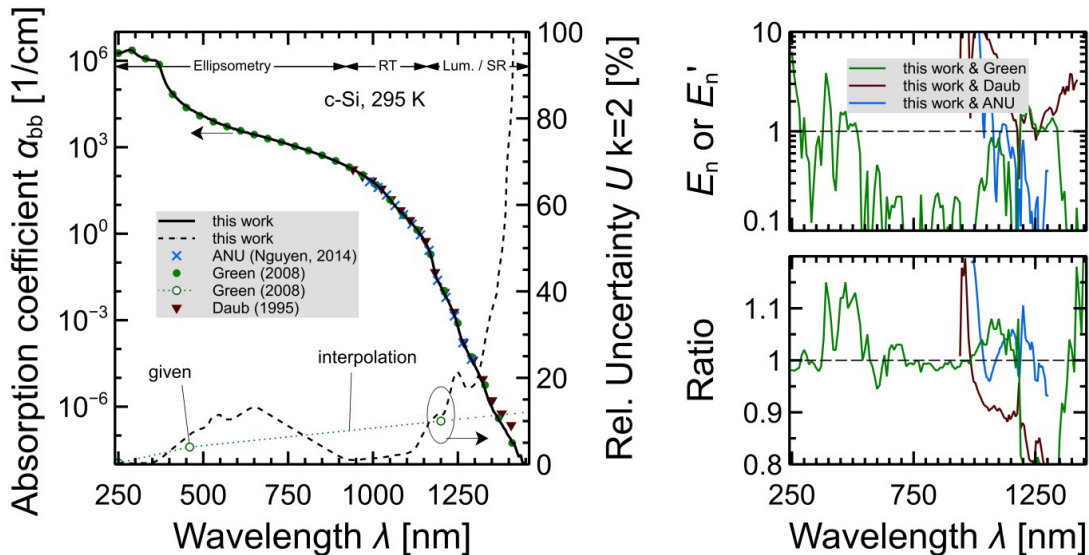


Fig. 4: Resulting data of the coefficient of band-to-band absorption of crystalline silicon at 295 K and its uncertainty.

Good agreement between the data measured at ISFH, PTB and ANU is achieved, as can be seen from the E_n number, which is smaller than unity for most wavelengths. Compared to the data of Green, the absorption coefficient determined in this work is about 20% smaller at wavelengths around 1200 nm. It is exactly this wavelength region where an overestimation of modeled luminescence spectra by about 20% has been reported [8,9] when using the absorption coefficient data of Green. Figure 5 verifies that the absorption coefficient data determined in this work resolve this deviation by comparing a measured PL spectrum of a c-Si wafer to the model curves calculated using both data sets of the absorption coefficient.

5. Impact of absorption coefficient uncertainty on energy conversion efficiency predictions

The optimization of the energy conversion efficiency of solar cells is supported by device simulations. Such simulations require input data such as the absorption coefficient. Uncertainties of these input data consequently lead to uncertainties of the calculated energy conversion efficiency. Therefore we consider the uncertainty of such simulations due to the uncertainty of the absorption coefficient. The following analysis refers to ideal silicon solar cells, which is sufficient for the estimation of the order of magnitude of the uncertainty of the resulting energy conversion efficiency and allows an analytical model to be used.

The energy conversion efficiency is given by

$$\eta = \frac{V_{mpp} I_{mpp}}{P_{in} A_{cell}} \tag{9}$$

where V_{mpp} is the voltage at the maximum power point (MPP), I_{mpp} is the corresponding current, P_{in} is the power of the incident light and A_{cell} is the active area of the solar cell. For an ideal solar cell, current and voltage are related by the well-known Shockley diode equation [21]. This equation provides the relation between the absorption coefficient, which determines the short circuit current, and the energy conversion efficiency. The relative uncertainty of the maximum power point current is given by the relative uncertainty of the short circuit current times the ratio of short circuit current and maximum power point current. For an ideal solar cell, whose performance is only limited by radiative recombination, the saturation current density is 0.27 fA/cm² [22], which leads to a ratio of 1.03. For realistic solar cells, I_{sc} / I_{mpp} is typically of the order of 1.1. In order to obtain conservative estimates of the uncertainty, a ratio of 1.1 is used in the following. The calculation of the uncertainty of the short circuit current requires a model for the absorptance A of the solar cell. In this work, an analytical model based on the works of Basore [23] and Brendel [24] is used. As ideal solar cells are considered, the analysis can be restricted to the base contributions as outlined in [8]. The short circuit current and its uncertainty depend on the spectral distribution of

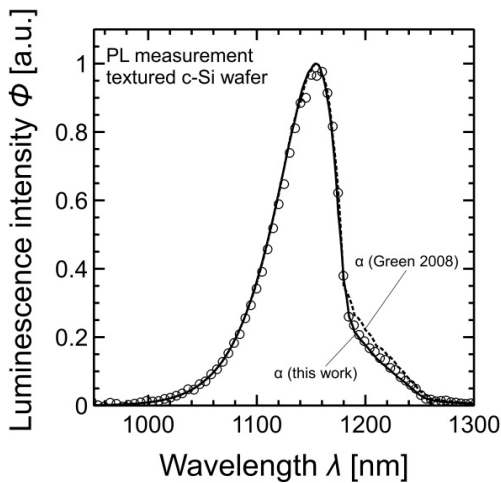


Fig. 5: Comparison of a measured PL spectrum of a c-Si wafer to the model curves calculated using the data set of the absorption coefficient as determined in this work and as published by Green [7].

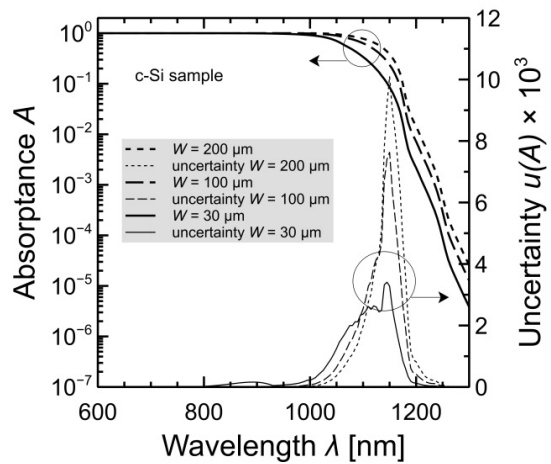


Fig. 6: Absorptance of ideal c-Si samples with different thicknesses. The thin lines show the combined standard uncertainty of the absorptance following from the uncertainty of the absorption coefficient as determined in this work.

the incident light. For the calculations in this work, the tabulated AM1.5G spectral distribution [25] is used. Figure 6 shows the uncertainty of the absorptance caused by the uncertainty of the absorption coefficient for samples with different thicknesses under illumination with the AM1.5G spectrum. Figure 7 shows the resulting relative uncertainty of the energy conversion efficiency as a function of the device thickness. The solid line represents the uncertainty following from the absorption coefficient data presented in this work, the dashed line represents the uncertainty following from Green's absorption coefficient data [7], for which uncertainty estimates are given as described in the last section. For comparison, the theoretical efficiency limits calculated by Richter et al. [26] for intrinsic silicon and p -type silicon ($1 \Omega\text{cm}$) are shown on the right axis. The figure shows that the uncertainty of the energy conversion efficiency due to the uncertainty of the absorption coefficient is generally small. For the thickness of typical c-Si solar cells of about $180 \mu\text{m}$, it is of the order of 0.07% . Note that a relative uncertainty is given in Fig. 7. Hence, for solar cells with an energy conversion efficiency of 20% , the absolute uncertainty of this value is of the order of 0.02% . Other simulation parameters are expected to cause larger uncertainties, so that the uncertainty of the absorption coefficient does not limit the accuracy of such simulations. Compared to Green's absorption coefficient data, the uncertainty of the energy conversion efficiency is reduced by a factor of about 2. This is a consequence of the "dip" of the uncertainty of the absorption coefficient around 1000 nm (see Fig. 4). As can be seen in Fig. 6, contributions to the uncertainty of the absorptance and thus to the uncertainty of the energy conversion efficiency arise mainly from the wavelength region around 1000 nm , where the absorptance varies between its saturation values. The increase of the uncertainty towards thin devices results from uncertainty contributions also at wavelengths below 1000 nm , as shown in Fig. 6.

6. Summary

Based on the combined evaluation of spectroscopic ellipsometry, measurements of reflectance and transmittance, spectrally resolved luminescence measurements and spectral responsivity measurements, we determine the coefficient of band-to-band absorption of crystalline silicon in the wavelength range $250\text{-}1450 \text{ nm}$ and analyze its uncertainty. The uncertainty analysis is carried out in accordance with the *Guide to the expression of uncertainty in measurement* (GUM), which defines the internationally accepted procedure for the systematic evaluation of measurement uncertainties. We determine relative uncertainties of the order of 0.3% at 300 nm , 1% at 900 nm , 10% at 1200 nm and 180% at 1450 nm and show that our data resolve discrepancies which have been reported when using literature data of the absorption coefficient for the modeling of measured luminescence spectra. The data will

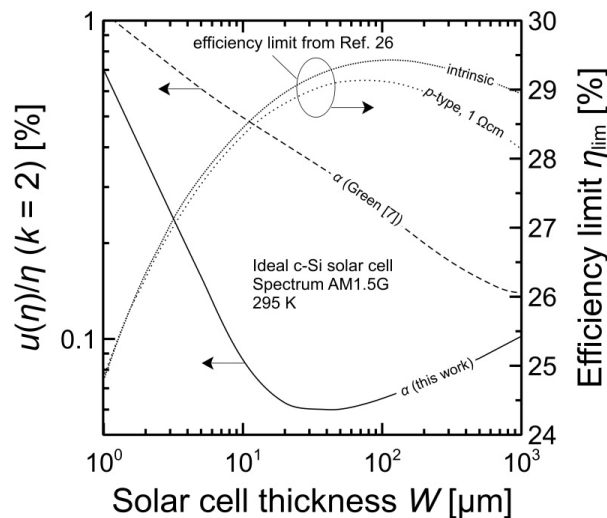


Fig. 7: Relative uncertainty ($k=2$) of the energy conversion efficiency η of an ideal c-Si solar cells as a function of the device thickness W . For comparison, the theoretical efficiency limits for intrinsic silicon and p -type silicon with a resistivity of $1 \Omega\text{cm}$ (according to [26]) are shown on the right axis.

also be published in digital form [19]. Moreover, we analyze the impact of the uncertainty of the absorption coefficient on the accuracy of solar cell energy conversion efficiency predictions by means of device simulations. The contribution of the absorption coefficient to the uncertainty of such simulations is found to be of the order of 0.1% relative (corresponding to 0.02% absolute for solar cells with an efficiency of 20%). The absorption coefficient is thus not expected to limit the accuracy of energy conversion efficiency predictions.

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