Research Article

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Light scattering of interference coatings from the IR to the EUV spectral regions

Abstract: Light scattering of optical components caused by residual imperfections can be a critical factor for their practical application. In particular, the scattering properties of optical interference coatings are rather complex. Yet, simple theoretical models and comparisons with experimental results provide valuable insight into the main impact factors and mechanisms. The magnitude of scattering and the dominating factors strongly depend on the wavelength of application in connection with the types of coatings used in the corresponding ranges. The paper, therefore, gives an overview of the scattering properties of coatings in different spectral regions including the visible, deep ultraviolet, and extreme ultraviolet and discusses strong in-band variations of the scattering characteristics that have been neglected so far.

Keywords: light scattering; roughness; thin film coatings.

OCIS Codes: (310.0310) thin films; (290.0290) scattering; (240.5770) optics at surfaces; roughness.

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1 Introduction

Light scattering from thin film coatings can be a critical issue depending on their application. This is, in particular, true for applications at short wavelengths in the deep and extreme ultraviolet (EUV) spectral ranges. However, for certain applications in the visible and infrared regions, light scattering can be problematic, too, if lowest optical losses are required. Measuring the light scattering properties of thin film coatings is, hence, essential in order to thoroughly assess the performance of optical components. Scatter modeling is the key to identify and understand the critical impact factors.

The scattering properties of thin film coatings are substantially more complex than those of single rough surfaces. In contrast to single surface scattering, scattering from multilayers is influenced by the nanostructural properties of all interfaces, their cross-correlation properties, as well as the interference properties of the coating. Modeling multilayer scattering, therefore, requires using reasonable simplifications and approximations.

In this paper, two aspects of thin film light scattering with respect to wavelength will be discussed. First of all, there is the widely recognized general trend that coatings exhibit drastically increased scattering as the wavelengths of application become shorter. This issue is sometimes even more amplified by the fact that coatings for short wavelengths are usually more difficult to fabricate and tend to have more imperfections. The second aspect is usually more or less neglected, although we believe it to have highest practical relevance for many applications: it will be shown that simple estimates of the scatter loss of a coating using a single-surface approximation do not sufficiently describe the scattering properties within the entire application range but can lead to a critical underestimation of the in-band scattering properties.

2 Theoretical models

For most interference coatings, the residual roughness of the interfaces within the multilayer is the dominating source of light scattering. The angle-resolved scattering (ARS) can be calculated using multilayer vector scattering theories [1–3]:

$$\mathrm{ARS}(\theta_s)=\frac{\Delta P_s(\theta_s)}{\Delta\Omega_s P_i}\propto\frac{16\pi^2}{\lambda^4}\sum_{i=0}^{N}\sum_{j=0}^{N}F_iF_j^*\mathrm{PSD}_{ij}(f). \quad (1)$$

ΔP_s is the power scattered into a certain direction, $\Delta\Omega_s$ is the detector solid angle, P_i is the incident power, and

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θ_s denotes the polar angle of scattering measured with respect to the sample normal. N is the number of layers, the F_i are optical factors containing information about the optical properties of the perfectly smooth multilayer (design, dielectric functions, etc.) and the conditions of illumination and detection (angles, polarization, etc.). The roughness factors PSD_{ij} comprise the power spectral density functions of all interfaces (for i=j) and their cross-correlation properties (for i≠j).

Equation (1) can be integrated to get the total scattering (or scatter loss). For normally incident light and isotropic scattering, the integral is rather simple [4]:

$$TS = \frac{P_s}{P_i} = 2\pi \int_{2°}^{85°} ARS \sin\theta_s d\theta_s. \quad (2)$$

The model explains the main factors influencing the light scattering of interference coatings: (i) the nanostructural properties of all interfaces in the coating starting with the substrate, (ii) the cross-correlation properties of the roughness at different interfaces, and (iii) the optical factors, which are closely connected to the field strengths within the coating. It is crucial to understand that the fields at the interfaces together with the roughness of the individual interfaces produce the light scattering. The design of the coating together with the cross-correlation properties of the interfaces finally determine how much scattering is emitted into a certain direction.

The number of parameters required to model multilayer scattering is proportional to N^2. Therefore, simplified models have been proposed. The approach described in [5] approximates the coating structure by introducing two parameters describing the average thickness deviation from the theoretical design and an exponent describing the roughness evolution inside the coating. An even simpler approximation is the following: if all interfaces of a highly reflective multilayer can be assumed to have identical roughness properties and are fully correlated, then the multilayer scattering can be approximated by the scattering of a single surface. For a multilayer mirror with effective reflectance R, combining Eqs. (1) and (2) with this assumption leads to [6, 7]:

$$TS_b = R\left(\frac{4\pi\sigma_{rel}}{\lambda}\right)^2. \quad (3)$$

σ_{rel} is the relevant rms roughness. The $1/\lambda^2$ scaling of Eq. (3) is often used to explain the dramatic increase in scattering at short wavelengths and turns out to be a good rough estimate in many cases. One must, however, be very careful when generally applying this simple model to real coatings. Tremendous deviations will occur in particular if (i) there is a real roughness evolution from the substrate through the coating, (ii) the application extends over a certain spectral bandwidth, or (iii) the coatings exhibit spectral shifts over time or during exposure, as will be discussed below. Nevertheless, Eq. (3) can be used to roughly estimate the scatter levels achievable with a certain roughness in many cases. Because of the complexity of scattering from thin film coatings and possible additional sources of scattering such as defects and contaminations, reliable information about the scattering properties can only be obtained through direct measurements at all wavelengths relevant for the application.

3 Instruments for light scattering measurements

The measurement of light scattering from high-quality optical coatings requires special instruments with challenging demands regarding noise levels, dynamic ranges, and linearity. There are only a small number of instruments that meet these requirements [3, 5, 8–11]. At Fraunhofer IOF, instruments have been developed for angle resolved and total scatter measurements at wavelengths ranging from the visible spectral range up to 10.6 µm in the infrared and down to 193 nm [12] and even 13.5 nm [13] in the deep and EUV, respectively.

A schematic and a picture of the table-top scatterometer ALBATROSS TT, developed at IOF, are shown in Figure 1 [9]. The instrument allows highly sensitive ARS measurements to be performed in the visible spectral range at arbitrary incident angles and within the entire scattering sphere. Three lasers (1), operating at 405 nm, 532 nm, and 640 nm are guided into the beam preparation system (2–4). A variable attenuator (5) is used to adjust the power of the incident beam. The beam is expanded using telescope mirrors (6, 7), and an aperture (8) is used to adjust the beam diameter. The spatial filter system (9–14) is crucial to achieve a clean core beam illuminating the sample (15). The polarizer (11) is placed within the spatial filter to reduce stray light. The spatial filter unit (13) can be translated to adjust the focal length of the illumination system. This is essential if measurements of curved samples are performed. The detection system, which is based on a photomultiplier and lock-in signal processing, can be moved within the entire scattering sphere around the sample. A dynamic range exceeding 14 orders of magnitude and a sensitivity that corresponds to a total scatter level below 1 ppm have been achieved.

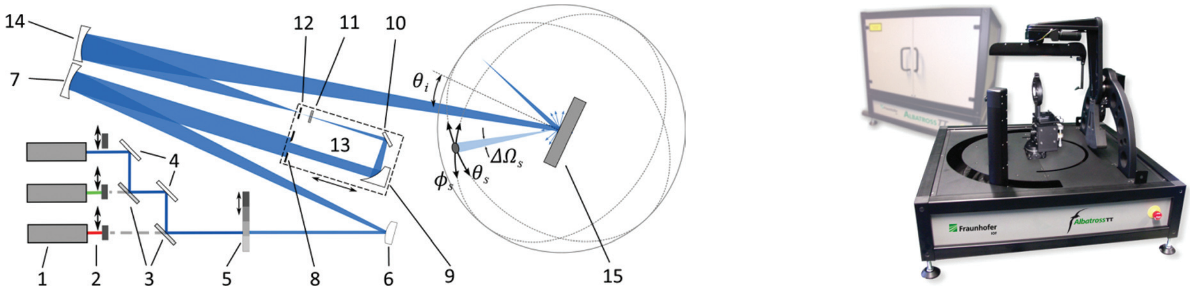

Figure 1 Instrument ALBATROSS TT for angle-resolved light scattering measurements.

The scatterometer thus allows investigations of a large variety of samples ranging from rough technical surfaces to superpolished transparent substrates with rms roughnesses of lower than 0.1 nm as well as high-quality interference coatings.

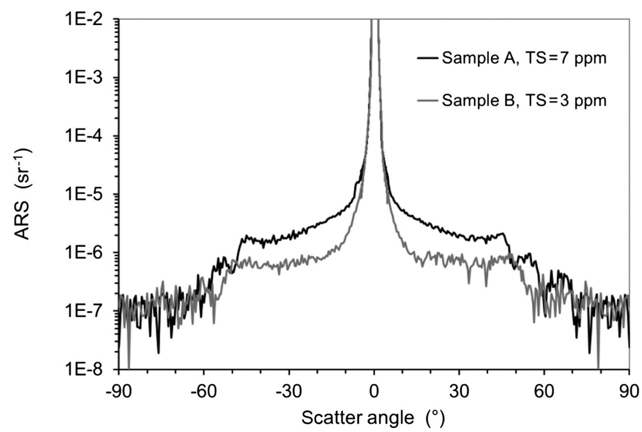

Figure 2 ARS at 1064 nm of HR coatings on fused silica.

4 Results and discussions

4.1 Coatings for the near-infrared and visible spectral ranges

Light scattering is usually considered to be a minor issue for most coatings in the near-infrared and visible spectral ranges. This is mainly because even moderate roughness levels lead to rather low losses according to Eq. (3). Moreover, metal oxides deposited by highly energetic deposition processes that lead to low intrinsic roughness are widely used as coating materials. Yet, for certain applications even in the visible range, light scattering is a critical issue. Prominent examples are low-loss laser mirrors or special filters requiring thick coatings with many interfaces.

Figure 2 shows the ARS of two multilayer laser mirrors for 1064 nm, measured using the instrument described in [5]. The coatings are quarter-wave Ta_2O_5/SiO_2 systems deposited by magnetron sputtering onto superpolished fused silica substrates with slightly different designs [14]. The measured curves exhibit peaks at 0° corresponding to the direction of specular reflection as well as ripple structures at larger angles that are caused by interference effects of waves scattered at different interfaces within the multilayer. The total scatter losses calculated from ARS were as low as 7 ppm and 3 ppm depending on the design.

In addition to losses, the laser stability of optical coatings is a critical issue for high-power applications. Rugate films have been shown to be a promising approach to enhance the laser-induced damage threshold compared to standard stacks [15]. In an attempt to produce dielectric mirrors with enhanced laser stability and low losses, Rugate films composed of mixtures of Ta or Hf and Si oxide were deposited onto superpolished fused silica substrates by pulsed magnetron co-sputtering at Fraunhofer FEP [16, 17]. Angle-resolved scatter measurements were performed at 532 nm using the instrument described in [9]. The results are shown in Figure 3.

The initial $Ta_xSi_yO_z$ coating exhibits a scatter loss of 110 ppm. In a next step, the process parameters were optimized, and Ta was replaced by Hf, which offers better performance in the UV range. The scatter losses of the new coatings were found to be only 7 ppm. Moreover, analysis of the ARS data indicated that replicated substrate roughness has a substantial effect on the observed scattering properties. Depositing the same film on a superpolished Si substrate finally resulted in scatter losses of as low as 3.5 ppm. Modeling the scattering properties using Eq. (1) is actually limited to conventional multilayers with real interfaces. Nevertheless, we believe that the scattering of Rugate films can still be modeled by discretizing the film into a large number of thin layers. In addition, scattering from bulk inhomogeneities is believed to play a significant role. For a more detailed discussion, please refer to Ref. [17].

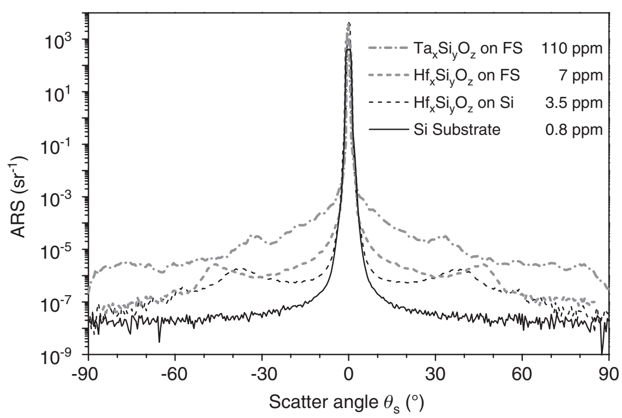

Figure 3 ARS at 532 nm of co-sputtered HR Rugate coatings on different substrates and of an uncoated Si substrate.

4.2 Coatings for the deep ultraviolet range

Coatings for 193 nm in the deep ultraviolet spectral range are mainly needed for optical lithography systems and material processing applications. The short wavelength makes light scattering one of the major issues for several reasons: First, the strong wavelength dependence [Eq. (3)] leads to substantial scattering even if coatings with low roughness could be produced. Second, metal fluorides have to be used as coating materials and are usually deposited using classical unassisted thermal evaporation leading to significant intrinsic thin film roughness and porous structures [18–21]. Questions regarding the influence of substrate or thin film roughness or about optical thickness errors and their influence often arise.

The results of ARS measurements of HR coatings for 193 nm performed using the instrument described in [12] are shown in Figure 4. The coatings are AlF_3/LaF_3 quarter-wave stacks deposited onto superpolished CaF_2 substrates with an rms roughness of 0.27 nm (AFM, 1×1 μm²). After coating, an increase in the top-surface roughness to 5 nm was observed and attributed to columnar growth. The measurement results are shown together with modeling results achieved using the procedure described in detail in Ref. [5].

The scatter loss at 193 nm determined by integrating the measured curve is 2.8% and, thus, constitutes the dominating loss mechanism in this type of coating. The modeling results shown in Figure 4 (left) obtained by varying the roughness evolution parameter β reveal that the coating exhibits a rapid roughening from the substrate through the multilayer. This method thus provides quantitative information about the roughness of the inner interfaces in contrast to the AFM top-surface data. The results shown in Figure 4 (right) obtained by varying the optical thickness parameter δ reveal that the coating exhibits a deviation of 3% of the average optical thickness of each layer from the perfect quarter-wave design. This is most likely caused by adhered water in the porous coating structure, which leads to a spectral shift of the coating and, thus, altered interference properties and enhanced scattering at the original wavelength of 193 nm. Remodeling investigations revealed that the scatter loss could be reduced to 1.4% by precompensating the spectral shift even if the coating had the same roughness [12].

4.3 Coatings for the extreme ultraviolet range

The semiconductor industry has always been striving for a continuous reduction of features printable by optical lithography. EUV lithography first at 13.5 nm and later at even shorter wavelengths is the most promising way to follow this trend also in the future. Because of the

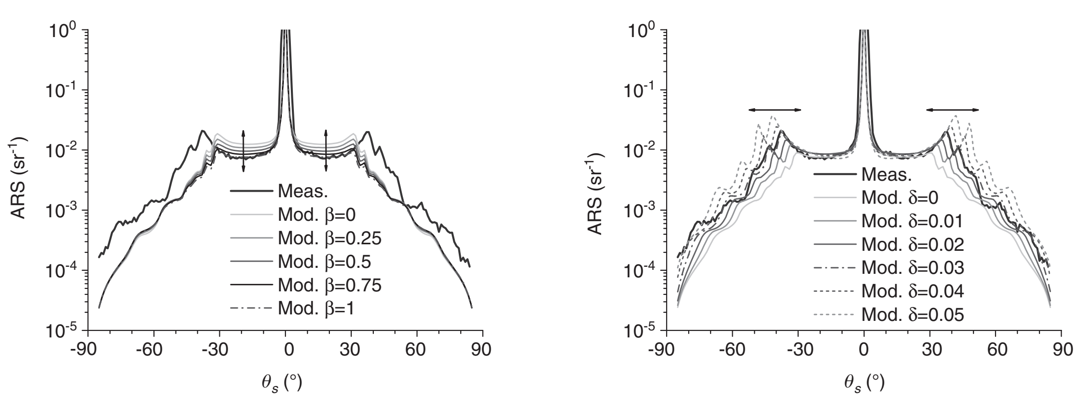

Figure 4 ARS of HR coating for 193 nm. Measurement (meas.) results obtained at 193 nm and modeling (mod.) results by varying the roughness parameter (left) and the optical parameter (right).

strong wavelength dependence in scattering, this leads to extremely challenging demands on optical components. Very often, light scattering issues of coatings already start with the substrate. In Figure 5, results of ARS measurements at 13.5 nm of EUV multilayer mirrors deposited onto different substrates are shown [13]. The coatings consist of 60 periods of Mo/Si deposited using magnetron sputtering [22].

The high-spatial frequency rms roughness of the uncoated substrates were 0.3 nm, 0.2 nm, and 0.1 nm, for samples A, B, and C, respectively. The total scatter losses determined from ARS were as high as 3.9% and 2.3% for samples A and B and still 0.8% for sample C, which can be considered to have the best surface quality possible, in general. Further analysis of the results revealed a dominating impact of substrate roughness replicated through the multilayer structures, in particular, at small scatter angles and especially for samples A and B. The scattering of sample C is dominated by intrinsic roughness of the coating that adds to the replicated substrate roughness. All the observed roughness evolution and replication processes can be modeled very accurately, which allows the roughness, and thus the scatter properties, of EUV coatings to be predicted based on knowledge of the substrate only or to optimize the design with respect to minimum scattering [23].

4.4 Spectral scattering characteristics of thin film coatings

The spectral scattering properties of coatings are not sufficiently described by the simple general rule of thumb that scattering increases proportionally to $1/\lambda^2$. Aside from the fact that real coatings usually exhibit a considerable roughness evolution and partially correlated interfaces, the simple single surface approximation conceals the fact that for a given coating, the scattering can vary dramatically around the resonance wavelength. In Figure 6, modeling results of the total scattering and the specular reflectance of a HR coating for 193 nm are shown as a function of wavelength and compared to the results of a single surface with the same roughness. For the sake of simplicity, we once more assumed perfectly correlated interfaces with identical roughness although the results discussed in Section 4.3 demonstrate that this is a rather unrealistic approximation.

The TS seems to vary proportionally to the reflectance of the coating. In the resonance region, the backscattered radiation constructively interferes just like the specularly reflected partial waves. However, the position of the maximum TS is slightly shifted away from the central wavelength of 193 nm. In Figure 7, we have normalized the TS results to the R results to illustrate this fact.

The single surface results follow the simple $1/\lambda^2$ relationship. The normalized TS of the multilayer coincides with that of the single surface at the central wavelength of 193 nm. This means that the scattering of the HR coating can be approximated if the roughness is known, and the assumption of perfectly correlated interfaces is justified. This single surface approximation is, however, only valid at the central wavelength and if the scattering is concentrated around the specular direction. In particular, near the edges of the central resonance region, substantial scatter enhancement occurs. This can be explained by strong enhancement of the field intensities inside the coating.

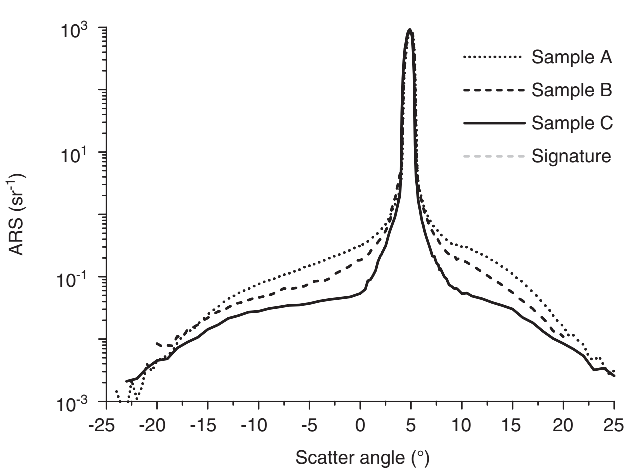

Figure 5 ARS of HR coatings for 13.5 nm deposited onto different substrates.

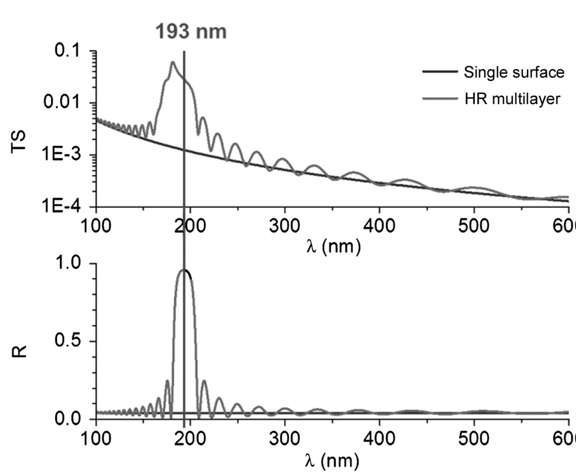

Figure 6 Modeling of total scattering and reflectance of a HR coating for 193 nm as a function of wavelength compared to a single surface with the same roughness.

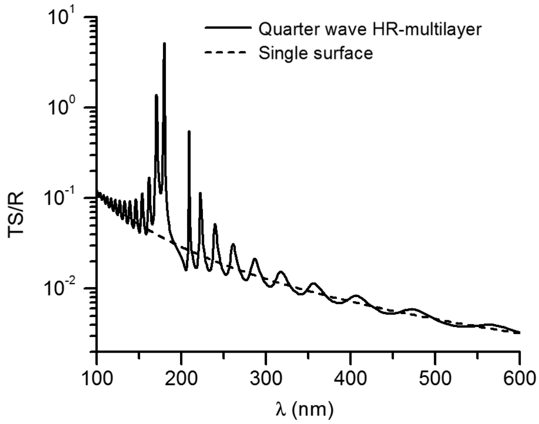

Figure 7 Same data as Figure 5 but TS normalized to R.

The observations provide some valuable insight into the spectral characteristics of scattering from thin film coatings beyond the well-known fact that the scattering goes up as the wavelengths get shorter. Instead, even for a given application in a certain spectral region, dramatic scattering effects can occur, in particular, if the application extends over a certain spectral bandwidth or involves a certain range of incident angles. Particularly interesting effects are expected for narrowband filters as well as broadband and chirped mirrors [24]. These effects can also critically alter the performance of coating during applications, in particular, if spectral shifts caused by environmental or irradiation effects are present. Therefore, light scattering of thin film coatings should not only be assessed at one single wavelength but over the entire range of wavelengths relevant for the application. For this purpose, a new instrument is currently being developed at Fraunhofer IOF that enables sensitive scatter measurements to be performed at arbitrary wavelengths using a continuously tunable narrowband OPO laser light source. A more detailed discussion can be found in Ref. [24].

5 Summary and conclusion

The light scattering properties of thin film coatings are considerably more complex than those of single surfaces. Vector scattering theories allow to accurately predict angle resolved and total light scattering, and simplified models and approximations provide valuable insight into the main factors influencing thin film scattering.

Two general aspects of the spectral properties of light scattering were discussed: (i) the strong increase in scattering when going to shorter wavelengths and (ii) the in-band variations of scattering from coatings. The first aspect is roughly expressed by the simple single-surface approximation and leads to the fact that in the infrared and visible spectral range, scattering is usually only an issue for high-end optical applications like low-loss laser coatings. At shorter wavelengths in the deep and EUV spectral ranges, however, even coatings deposited on superpolished substrates exhibit significant scattering that can be a limiting factor for the application. The second aspect has been more or less neglected so far. It was demonstrated that even for standard multilayer mirrors, the single surface approximation provides a reasonable estimate of the actual scattering properties only at the central wavelength; the scattering in the surrounding region of the reflection band can be substantially higher. Consequently, substantial scattering effects are expected for real applications. Therefore, we concluded that the light scattering of thin film coatings should be assessed over the entire range of wavelengths relevant for the application.

The main mechanism producing scattering that was considered in this paper is interface roughness, which is, in fact, the dominating effect in most cases. Although other imperfections like bulk inhomogeneities or singular surface or bulk defects or contaminations can play critical roles as well. Therefore, a thorough assessment of the light scattering properties of thin film coatings should always be based on measurements. Scatter modeling should then be performed to analyze and interpret the experimental results, gain insight into the relevant scattering mechanisms, and finally improve the performance of the coatings.

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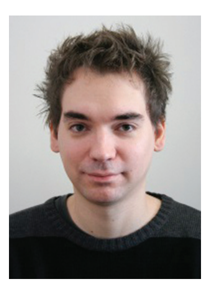

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