applied optics

Ultrafast switching with nonlinear optics in thin films

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Received 12 October 2022; revised 9 December 2022; accepted 15 December 2022; posted 19 December 2022; published 8 February 2023

We demonstrate a novel, to the best of our knowledge, concept for an all-optical switch based on the optical Kerr effect in optical interference coatings. The utilization of the internal intensity enhancement in thin film coatings as well as the integration of highly nonlinear materials enable a novel approach for self-induced optical switching. The paper gives insight into the design of the layer stack, suitable materials, and the characterization of the switching behavior of the manufactured components. A modulation depth of 30% could be achieved, which prepares the way for later applications in mode locking.

https://doi.org/10.1364/AO.478075

1. INTRODUCTION

Thin film coatings play an essential role in modern optical systems and are crucial for further development in this field—for instance, in ultrashort pulsed, high-intensity lasers, where optical coatings are used for management of group delay dispersion and pulse shaping [1], as well as in precision systems such as gravitational wave detectors, where thin film mirrors provide the lowest losses currently achievable [2]. Nonlinear effects in thin film coatings are usually regarded as loss mechanisms or even responsible for the destruction of the optical components, and therefore undesirable [3,4]. However, the requirements for optical systems, especially when considering current trends towards integration and miniaturization, require new approaches for the conception of optical components. By combining the existing, highly developed coating technologies with fundamental nonlinear effects, novel concepts for integrated optical components are possible. Since the materials in dielectric coatings are typically amorphous in structure, only odd-ordered nonlinear effects occur, and because of the decreasing strength of interaction, only third order effects achieve meaningful efficiency [5]. One application of third order nonlinear effects that has been researched in the past years investigates the possibility of frequency tripling in thin film stacks. The efficiency of nonlinear conversion processes in thin films was greatly increased [6]. Dealing with another process resulting from the inherent nonlinearity of coating materials, the impact of the optical Kerr effect on the spectral properties of thin film coatings has been demonstrated. Significant changes of reflectance in the order of several percent could be observed [7,8].

In this work, this seemingly undesirable effect of the optical Kerr effect in film coatings will be applied in an all-optical switch, called the Kerr-band switch (KBS). The switching function is based on a change of refractive index in a thin film interference coating, caused by the optical Kerr effect and high-intensity laser radiation, which in turn changes the transmittance and reflectance of the interference coating. The index change caused by the optical Kerr effect is proportional to the incident intensity and the nonlinear refractive index n_2 of each considered material [5]

$$n = n_0 + n_2 \cdot I. \tag{1}$$

To amplify the possible change in refractive index as much as possible, special coating designs as well as highly nonlinear materials can be applied, and switching amplitudes several tens of percent are possible this way.

A switch working this way would have many advantages over currently available technology. Since materials utilized in optical coatings have a wide spectral range of applications, the KBS concept would be flexible in its switching wavelengths, which would be mainly determined by the design of the thin film stack, which can be chosen for each application. Additionally, because the switching is based on the optical Kerr effect changing the properties of an interference coating, the switching itself is considered as a lossless mechanism, and only losses from the applied materials are taken into account. Lastly, the proposed component would consist of a monolithic coating stack, which makes it easy to integrate into typical optical setups. Possible applications for the proposed KBS are in optical resonators of ultrashort pulsed lasers as a mode-locking device or as an ultrafast safety switch in high-intensity beam lines. The content of this work is divided into three sections. First, the theoretical foundation and design considerations are discussed. A suitable design of the thin film stack is essential for the proper function of the resulting optical switch. Second, suitable materials are discussed and the manufacturing of the coatings is presented. Finally, the manufactured component is tested, and its performance as an optical switch is validated.

2. KERR-BAND SWITCHES

The change of the refractive index of optical materials introduced by the Kerr effect can be enhanced by selected interference coatings. This is achieved by making the coatings especially sensitive to index changes as well as amplifying the external intensity of the laser irradiation causing the shift. This section will investigate thin film designs that combine these properties.

A. Design Considerations

Designs suitable for use as a KBS as defined in this work were targeted to have three fundamental properties: first, they need to have the desired optical characteristics in their "undisturbed" state, e.g., under low-intensity irradiation. Second, this characteristic should be sensitive to a change of the optical thickness in at least one layer of the coating stack. This change of optical thickness then is introduced by the refractive index change caused by the Kerr effect. The third property concerns the "selectivity" of the coating design. When the design interacts with intense laser radiation, only the desired layers should change their optical thickness significantly. This can be achieved in two ways: first, the design also needs to exhibit a large intensity enhancement in said layers. This will ensure that these layers see a much stronger impact of the Kerr effect due to the higher intensity. Second, the materials of the sensitive layers can be chosen to have a much higher nonlinear index of refraction n_2 than the other layers of the design. This makes the layers, where these highly nonlinear materials are applied, "active layers" in the design and ensures, even at similar intensities, a stronger change of the optical thickness in the desired layers. Both approaches should be combined for greater effect.

B. KBS Designs

The requirements defined in the previous section can be fulfilled largely by relatively well-known band pass designs. In their simplest form, these designs consist of two quarter wave optical thickness (QWOT)-stacks with a "spacer layer" of two QWOTs in the center of the form $(HL)^N H^2 (LH)^N$. The number of layer pairs N in the QWOT-stacks can be chosen to define the width and contrast of the pass band. This type of coating stack is very similar to a classical Fabry–Perot cavity. The outer QWOT-stacks form the end-mirrors of the cavity, while the distance is controlled by the spacer layer, which has an optical thickness of integer multiples of half the targeted application wavelength (1030 nm in this case). Therefore this approach features properties very similar to a classical cavity. When the optical distance between the "mirrors" is changed, the resonant wavelength shifts

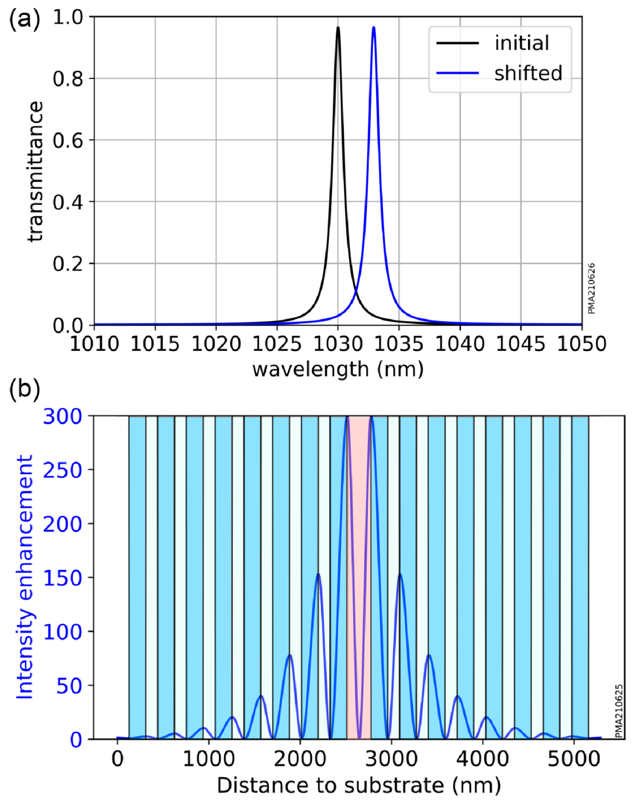

Fig. 1. (a) Original transmittance spectrum (black) of the KBS design compared to the blue spectrum, which results from a 1% change of the refractive index of the central spacer layer. (b) Internal field enhancement of the design (blue). To illustrate the layers, the different materials with alternating refractive indices are highlighted by color. The active material is depicted in pink.

accordingly. Figure 1 illustrates this property for a design of this type with $N = 8$, with the refractive indices $n_H = 2.03$ and $n_L = 1.45$, and an initial central wavelength of 1030 nm. A 1% increase of the optical thickness of the central spacer layer causes a shift of the resonance to higher wavelengths and therefore a reduction of transmittance and increase in reflectance at the application wavelength. Analogous to a classical cavity, the intensity of the incident light is greatly enhanced inside of the spacer layers when in resonance. This intensity enhancement for the investigated example is displayed as a relative factor in Fig. 1(b). As expected, the enhancement is highest in the central spacer layer. The intensity enhancement of the design as depicted in Fig. 1 is at its maximum approximately 300, which, in combination with suitable materials as presented in Section 3 and the intensities employed for the characterization in Section 4, leads to a potential maximum shift of the refractive index of approximately 6.7%.

This design type fulfills all the requirements for a KBS design. This first concept is very simplified: by adding more "spacer layers" or changing the overall layer sequence, the spectral properties as well as the intensity enhancement profile can be specially tailored for different applications.

C. Optimization of Switching Behavior

Although the concept of a KBS illustrated in the previous section is illustrated straightforward, the switching behavior of a design cannot be predicted easily as a consequence of the changing optical properties to the shifting of the design. When the previously established model of a classical cavity is used for explanation, it is intuitively clear that a shift in resonance wavelength caused by the optical Kerr effect comes with a simultaneous reduction of internal intensity, since the resonator and the incident wavelength do no longer match. This in turn limits the optical Kerr effect because it depends on the absolute intensity in the central spacer layer. The behavior of the design for different pulse shapes and intensities therefore becomes nontrivial to predict.

To solve this issue, a simulation approach was developed, which combines the transfer matrix formalism [9], which is an established way of calculating thin film filters, with the nonlinear calculations necessary to include the optical Kerr effect. The main difficulty encountered in this approach is the continuously varying internal intensity in the single layers of a thin film coating, which can also be seen in Fig. 1(b). When the optical Kerr effect is considered, this inhomogeneity of the intensity causes continuously varying refractive indices in the single layers that comprise the layer stack. In the transfer matrix formalism, the layers in a coating stack are assumed to have constant optical properties, so the calculation has to be adapted to accommodate the changes caused by the optical Kerr effect.

The solution chosen in this paper is to split the layers of the original design in so-called sublayers with thicknesses of a few nanometers. This allows the assumption of nearly constant optical properties for each sublayer and therefore a convenient calculation of the optical properties of the stack of all sublayers. In addition to the spatially varying internal parameters in the component, the external intensity of the incident laser is also varying temporally in typical pulsed applications. Therefore, to realistically simulate the behavior of the component, the laser pulse of the intended application has to be modeled and the complete passage of the pulse through the KBS coating has to be simulated.

These requirements are met with the combination of the two different software solutions that execute different parts of the necessary calculations. The overall framework is implemented in a Python software [10] developed in the frame of this work, which loads a design file, splits the original layers into sublayers of appropriate thickness, and saves a design file compatible with the thin film software. The thin film calculations in turn are done by *SPEKTRUM 32* [11], an established, commercial thin film software solution. *SPEKTRUM 32* is called by the Python program and calculates the internal intensity enhancement, as well as transmittance and reflectance of the current design iteration. With the external intensity at the current timestep known from the chosen laser parameters, the absolute, internal intensity is determined and used, in combination with the material parameters, to calculate the refractive index shift caused by the optical Kerr effect for each sublayer. A new "shifted" design is saved and fed back to *SPEKTRUM 32* for the thin film calculations, resulting in a slightly changed internal intensity enhancement as well as changed optical properties. With these new parameters from the design, as well as a slightly changed external intensity corresponding to the temporal pulse shape of the incident laser pulse, new refractive indices for each sublayer are calculated and a new design is handed over to *SPEKTRUM 32*.

This loop continues from the start until the end of the incident laser pulse profile and allows a complete characterization of each KBS's behavior for single pulse interactions. In addition to observing the changes caused by the optical Kerr effect, the simulation also allows the optimization of the switching behavior to increase the switching amplitude of a given design. During optimization, the layer thicknesses are varied randomly within certain limits, and the switching amplitude at a specified pulse energy is evaluated. The bandwidth of the specified laser source is taken into account to avoid increases in the internal intensity enhancement that purely result from a reduction of the filter's bandwidth. It has to be noted, however, that the thin film calculations performed for each intensity level during the simulation are based on the interaction of continous-wave radiation with the layer stack. For extremely short pulses, this approach might lead to errors since the localization of the pulse in the layer stack is not taken into account in the current model. The results of an exemplary design optimization are shown in Fig. 2. Figure 2(a) shows the amplitude of the transmittance-switching of a KBS design before and after optimization of the layer thicknesses for various increasing pulse energies. Both the initial design (black)

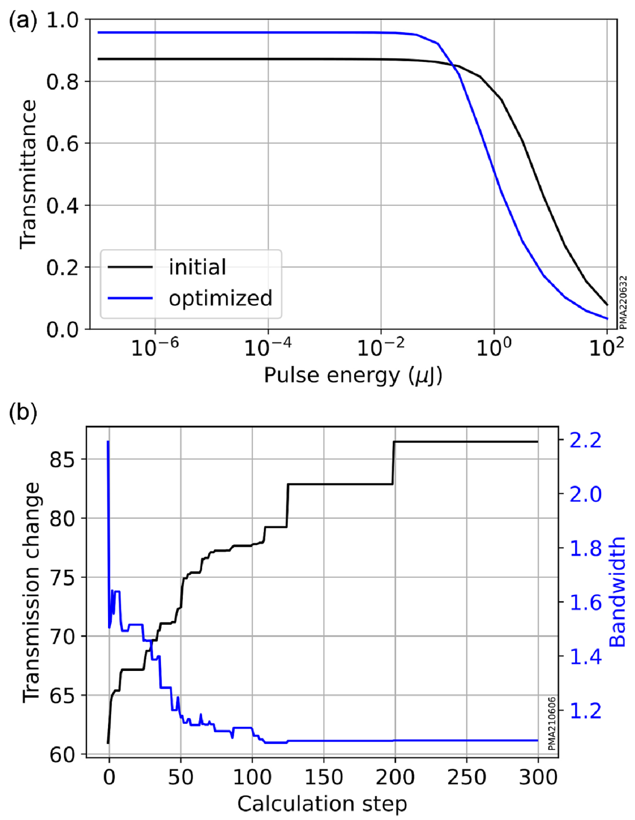

Fig. 2. (a) Change transmittance of the KBS design before and after optimization. Both designs exhibit the desired optical switching of transmittance, with the optimized variant switching at lower pulse energies. (b) Progress of the optimization process over the process steps. An increase in transmission change with a decrease of bandwith can be observed.

and the optimized design (blue) exhibit the desired switching behavior expressed as a reduction in transmittance with the optimized design switching at lower pulse energies. Figure 2(b) shows the development of the switching amplitude and design bandwidth [full width at half maximum (FWHM)] during the optimization process. The switching amplitude is determined by calculating the change in transmission at a fixed pulse energy and clearly improves during the optimization process. The bandwidth of the design initially decreases quickly, but reaches a limit set by the simulation parameters, where it remains for the remaining steps.

Another possibility for improving the switching of KBS is the choice of materials. Variation of the strength of the optical Kerr effect tested with the presented simulation method shows a clear advantage of a high contrast between the central spacer layer and the surrounding mirror coating. Therefore, highly Kerr-active coating materials are desired for these layers.

3. MATERIALS AND MANUFACTURING

As indicated in the previous section, highly Kerr-active materials are advantageous for the switching function of the KBS designs. Typical coating materials show nonlinear refractive indices ranging from $3 \times 10^{-16}\ \mathrm{cm^2 W^{-1}}$ (SiO_2) up to approximately $20 \times 10^{-16}\ \mathrm{cm^2 W^{-1}}$ for (TiO_2) [12,13]. Because this range is already relatively small, and both high- and low-index materials are required for the coating design to function, a large contrast between the spacer layer and the surrounding mirror stacks cannot be achieved with standard materials.

To solve this challenge, indium tin oxide (ITO) was chosen as the active material for the components evaluated in this work. ITO is frequently used in opto-electronic applications such as touchscreens and solar cells, and therefore an established thin film material. ITO belongs to a group of materials commonly known as epsilon-near-zero materials (ENZ materials), which have been of interest for applications in nonlinear optics for a few years due to their strong nonlinear response [14,15]. The mechanisms that the observed large nonlinear effects are based on are not entirely the same as those observed in "classical" optical materials, but rather a combination of free carriers and intrinsic field enhancement [16]. It exhibits a nonlinear refractive index of $2.3 \times 10^{-13}\ \mathrm{cm^2 W^{-1}}$ [17], and thus can deliver a large contrast to typical high- and low-index coating materials. It has to be noted, however, that the nonlinear effects in ITO can be of sufficient strength to exceed the assumption of small perturbations that is typically made in modeling nonlinear optics [18]. In addition to its strong nonlinear interaction with incident radiation, the intrinsic laser-induced damaged threshold (LIDT) of ITO is relatively high [19], which makes it ideal for the integration into KBS.

The generated and optimized designs were manufactured with the well-established ion-beam sputtering (IBS) coating process on fused silica substrates with 1 mm thickness. As materials for the mirror stacks, Ta_2O_5 was chosen as high-index material and SiO_2 as low-index material. All materials were sputtered in a reactive process, with added oxygen to ensure dielectric properties. ITO was sputtered from a metallic target consisting of 90% indium and 10% tin.

The sample under consideration in this paper consists of a central ITO layer bordered by QWOT-stacks towards substrate and surface. The design initially had a structure $(HL)^6\mathrm{ITO}(LH)^6L$ with an ITO-layer thickness of 2 QWOT, but was numerically optimized for better switching performance, which leads to slightly different layer thicknesses. The simulation of the nonlinear properties does not at this point include the materials absorption, which may decrease the switching amplitude in real measurements.

The spectral transmittance as well as a spectrophotometric measurement done after the deposition are presented in Fig. 3. The peak transmittance of the design at central wavelength is 36%, and the value measured by the spectrophotometer after deposition of the coating is 27%. The deviation is probably due to the losses in the spacer layer, which can vary slightly from coating to coating and have a large influence on the final transmittance. The FWHM bandwidth of the transmission peak is 17 nm, which is broad enough to not narrow the laser spectrum, which features a bandwidth of approximately 0.5 nm. The wide bandwidth of 17 nm also enables in theory the use of this KBS design with much shorter pulse durations of around to 70 fs.

4. SETUP FOR VALIDATION OF SWITCHING BEHAVIOR

To validate the concept, an optical experiment was performed to measure the transmittance and reflectance of the manufactured coatings at various intensities. The layout is detailed in Fig. 4. A *Trumpf TruLase 5050* laser system is used to irradiate the samples. The system generates output pulse energies of up to 250 μJ at pulse durations of 10 ps, a wavelength of 1030 nm, and repetition rates of up to 200 kHz. The spectrum of the laser was recorded using a *Thorlabs OSA201C* spectrum analyzer. The resulting intensity distribution is presented in Fig. 5. The bandwidth of the system is approximately 0.5 nm. The measurements were performed at 20 kHz to limit the average power and influence of thermal effects on the samples. To exclude the influence of thermal effects, measurements at different repetition frequencies and similar intensities were performed, and no significant change in behavior could be observed. The beam diameter on the sample was 1.5 mm. A polarization-based attenuator regulates the intensity of the laser radiation, and

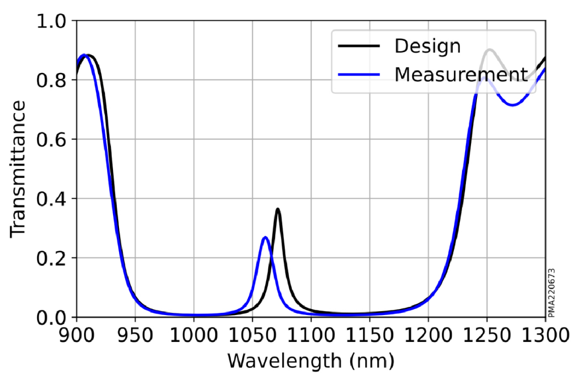

Fig. 3. Designed (black) and measured (blue) transmittance of the deposited KBS. The component was designed for slightly longer wavelengths to allow precise tuning to the laser wavelength by variation of the angle of incidence.

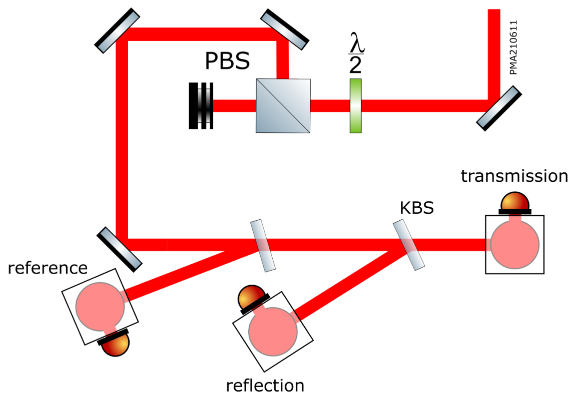

Fig. 4. For validation of the switching behavior of Kerr-band switches, the relative transmittance and reflectance is measured at different laser intensities.

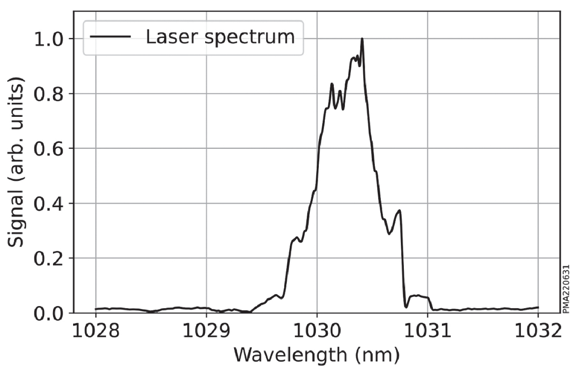

Fig. 5. Spectral intensity distribution of the laser system.

transmitted as well as reflected radiation is monitored by photodiodes with integrating spheres. An additional photodiode monitors the laser power during each step of the irradiation to provide a reference for any changes that occur in transmittance and reflectance. To provide absolute values for reflectance and transmittance, the measurements are absolutely calibrated by referencing to a highly reflective mirror and an "open" measurement without a sample, respectively. During each measurement the waveplate of the attenuator is rotated by 90°, which leads first to an increasing and then a decreasing intensity on the sample. By measuring this way, any hysteresis effects that may occur during the measurement as a result of permanent changes in the sample will be visible immediately.

5. MEASUREMENT RESULTS AND DISCUSSION

With the experimental setup described in the previous section, the manufactured Kerr-band sample was characterized for the behavior of its transmittance and reflectance at various intensities. The measurements were performed for increasing and decreasing incident intensities and were repeated several times to exclude the possibility of permanent changes in the samples being interpreted as switching behavior. Figure 6 shows the reflectance and transmittance of a KBS for rising and falling incident intensities. A clear decrease in transmittance from 26% to 14% with a simultaneous increase in reflectance from 55% to 73% can be observed, and no significant hysteresis is visible for

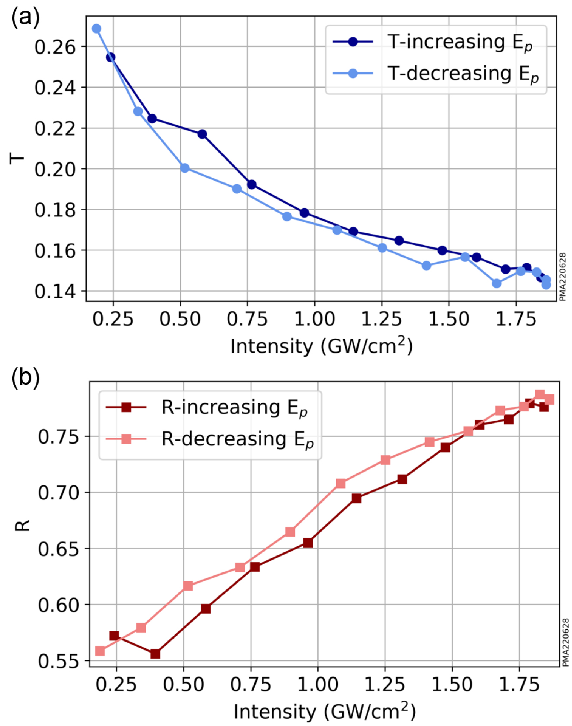

Fig. 6. (a) Transmittance and (b) reflectance of the tested KBS for increasing and decreasing incident intensities. A clear decrease in transmittance and simultaneous increase in reflectance can be observed. No significant hysteresis is visible.

these changes. The transmittance measured for low intensities matches well with the spectrophotometric measurement presented in Section 3. To test the repeatability of the switching of the KBS sample, the measurement was repeated three times on the same spot of the sample. Figure 7 shows the resulting curves for transmittance and reflectance. A small difference between the first and the latter two measurements can be observed, but the overall switching behavior is identical for the three measurements. The small difference that is visible may result from thermal effects, which could cause small changes in the initial transmittance and reflectance of the KBS sample due to heating up.

By adding the transmission and reflection values, it is possible to calculate the losses of the KBS. This reveals that the losses change during switching of the component. This behavior is presented by Fig. 8, which shows the development of the total losses $1-(T+R)$ over the intensity. A clear decrease of the total losses from approximately 20% to below 10% can be observed. The overall high losses and their decrease can be explained by ITO and its role as active material. The material in this component has not been thermally treated after deposition, so it is more absorptive than standard coating materials. Furthermore, the internal intensity in the ITO layer is high due to the nature of the KBS design, as explained in Section 2. These two factors combined create relatively high losses in the ITO layer. When the KBS "switches" the optical thickness of the ITO layer increases due to the optical Kerr effect, which causes the wavelength of resonance of the KBS to shift to longer wavelengths. Analogous to a classical Fabry–Perot resonator, the KBS then behaves more like a mirror, and the intensity in the

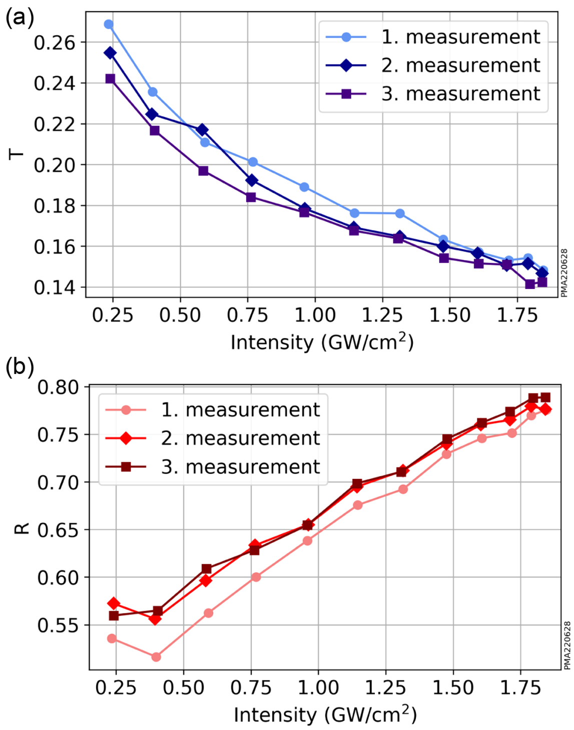

Fig. 7. (a) Transmittance and (b) reflectance of the tested KBS for subsequent measurements of the KBS sample. The switching behavior in reflectance and transmittance is clearly reproduced.

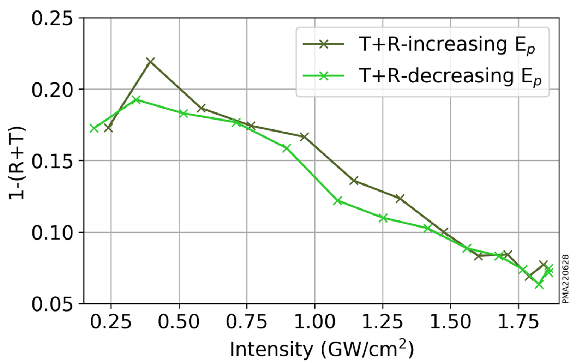

Fig. 8. Total losses calculated from the determined reflectance and transmittance. A clear decrease of losses with increasing intensity can be observed.

ITO layer decreases. Hence, this decreased intensity in the ITO causes smaller optical losses. This leads to the conclusion that the reduced losses at high intensities are a direct indicator of the spectral shifting of the KBS's transmission wavelength.

6. CONCLUSION

In this work, a new approach for ultrafast optical switching, the so-called KBS, is presented. The concept is based on the optical Kerr effect, which causes a change in the refractive index in specially designed, resonant interference coatings, so that the component switches between reflection and transmission are dependent on the incident intensity.

The switching is achieved by using selected active materials with a high Kerr-type nonlinearity in combination with designs, which were optimized for their switching behavior by a self-developed software solution. An IBS process was applied to deposit the developed coating designs. The switching behavior of the produced samples was tested with a combined measurement of transmittance and reflectance at different intensities. A clear decrease in transmittance of 12% with a simultaneous increase in reflectance of more than 20% can be observed. This behavior shows no clear hysteresis and is reproducible, and therefore does not result from damage of the sample under test. The losses in the optical switch were calculated from reflectance and transmittance values, and the switching of these properties also leads to an overall reduction of total losses in the tested sample, which matches the expectations for the optical switching process and indicates the component functions as intended.

For future improvements it is intended to further optimize the utilized active material and its post-deposition treatment to reduce the initial losses and increase the initial transmittance. Further optimizations of the layer design also promise gains in the switching amplitude. Similarly, the characterization of the samples could be improved. In particular, a pump–probe style experiment and *in situ* temperature monitoring would be advantageous to document the behavior of the presented components in more detail. With these improvements to the now proven concept, the KBS is then planned to be tested as an alternative to current mode-locking devices, where its possible advantages can be used to their fullest potential.

Funding. Bundesministerium für Wirtschaft und Technologie (03THW05K12); Niedersächsisches Ministerium für Wissenschaft und Kultur (03THW05K12); Deutsche Forschungsgemeinschaft (390833453, Cluster of Excellence PhoenixD, EXC 2122).

Disclosures. The authors declare no conflicts of interest.

Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

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