

Lasers in Manufacturing Conference 2015

Theoretical and experimental determination of the polarization dependent absorptance of laser radiation in carbon fibers and CFRP

Christian Freitag^{*,a}, Lukas Alter^a, Rudolf Weber^a, Thomas Graf^a

^aInstitut für Strahlwerkzeuge IFSW, Universität Stuttgart, Pfaffenwaldring 43, 70569 Stuttgart, Germany

Abstract

A detailed consideration of the fundamental mechanisms of interaction between laser radiation and carbon fibers is required to explore the potential of the laser as a tool for processing of carbon fiber reinforced plastics (CFRP). A key factor for laser materials processing is the absorptivity of laser radiation in the material at the laser wavelength that determines the fraction of laser energy coupled into the material. In the case of CFRP, the complex composite structure of the material requires sophisticated theoretical and experimental investigation of the absorptance.

The absorptance was calculated modelled for a wide range of wavelengths and for an orientation of the electric field perpendicular and parallel to the symmetry axis of the carbon fibers. The model includes the nearly circular cross-section and the birefringent properties of carbon fibers as well as multiple reflections. For carbon fibers it was found, that the total absorptance is larger than 70% for wavelengths in the UV, VIS and NIR and drops to less than 40% for a wavelength of 10.6 μm (CO₂-Lasers). The absorptance for light polarized perpendicular to the carbon fibers was shown to be larger than for light polarized parallel to the fibers. For CFRP the absorptance is increased to values around 80% for wavelengths in the VIS and NIR.

Experimental measurements of the absorptance of carbon fibers and CFRP for laser radiation with a wavelength of 532 nm and 1047 nm were performed to validate the model predictions. Polarization states of the laser radiation perpendicular and parallel to the carbon fiber symmetry axis were investigated. The absorptance was measured to be about 90% for both wavelengths. The calculated higher absorptance for an orientation of the electric field perpendicular to the carbon fiber axis was verified.

Keywords: Micro Processing; Fundamentals and Process Simulation; CFRP; Carbon fibers; Absorptance

* Corresponding author. Tel.: +49 711 685 69759; fax: +49 711 685 66842.
E-mail address: christian.freitag@ifsw.uni-stuttgart.de.

1. Introduction

The laser as a well automatable, noncontact tool without wear offers great potential for processing of carbon fiber reinforced plastics (CFRP). To utilize this potential a detailed consideration of the fundamental process mechanisms between laser radiation and carbon fibers/CFRP is necessary. One fundamental mechanism is the polarization- and wavelength-dependent absorptance of laser radiation in the material. In the special case of carbon fibers and CFRP this needs to take the unique structure of the material into account. We propose a simplified model for carbon fibers and CFRP and derive the optical properties of the carbon fibers from the well-known optical properties of graphite, see also Freitag et al., 2014. With this model the absorptance can be calculated. The calculations are complemented by experimental investigations of the absorptance of carbon fibers and CFRP for laser radiation with a wavelength of 532 nm and 1047 nm.

2. The material

As carbon fibers we considered High Tensity (HT) carbon fibers based on Polyacrylnitril (PAN) which are composed of turbostratic carbon, see Hoffman et al., 1991. A network of regular hexagons arranged in almost parallel layers is formed by the carbon atoms. This structure is similar to the crystal structure of graphite although there are some differences. The interlayer distance is 0.0196 nm larger for turbostratic carbon than in graphite due to additional types of bonds and the atomic lattice of turbostratic carbon may include some cluster defects, see Hoffman et al., 1991. Due to the close similarity of the crystal structure of turbostratic carbon and graphite, we derived the optical properties of turbostratic carbon by the well-known optical properties of graphite.

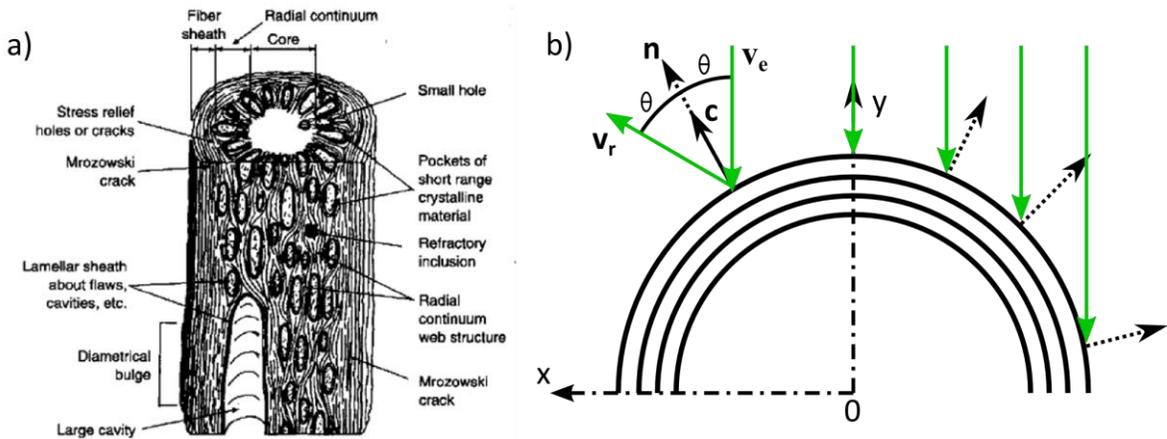


Fig. 1. (a) Structural model of a PAN-based carbon fiber taken from Barnett et al., 1976. (b) Model of a PAN-based carbon fiber. The incident radiation v_e hits the surface of the carbon fiber at different angles of incidence θ depending on the position relative to the fiber axis. The surface normal is n , the optical axis is c and v_r is the vector of the reflected beam.

The inner structure of carbon fibers depends on their type and the fabrication process. The orientation of the carbon layers within the carbon fibers can vary from a radial structure to an onion type structure, see Hamada et al., 1987. In the case of PAN based carbon fibers, the outermost shell can be described as regular concentric cylinders of turbostratic carbon as shown in Fig. 1 (a), see Barnett et al., 1976. We approximated this structure by the model shown in Fig. 1 (b). The outermost shell of the carbon fibers is modeled by regular concentric graphite cylinders. It is sufficient to take only the outer areas of the carbon fiber into

account since the absorption length in graphite in direction of the optical axis c is in the range of 12 nm for wavelengths relevant for laser materials processing.

The uniaxial layered crystalline structure of graphite, where the spacing between two parallel graphite layers is about 2.7 times larger than the distance between neighboring carbon atoms within the layers, leads to a dependence of the optical properties on the orientation of the electric field with respect to the optical axis c . The optical axis is oriented perpendicular to the basal plane, see Borghesi et al., 1991. It describes the only direction in which the optical properties are independent of the orientation of the electric field. For the complex refractive index $N_o = n_o - i \cdot k_o$ for the ordinary beam we used experimental data reported in Borghesi et al., 1991. To calculate the complex refractive index $N_e = n_e - i \cdot k_e$ of the extraordinary beam we used a model described in Djuricic et al., 1999.

The absorptance A of a material can be obtained from $A = 1 - R - T$ where R is the reflected share of the incident laser radiation and T is the transmitted part. Since the absorption length is in the range of 12 nm, the transmitted share of the laser radiation can be neglected and the absorptance is given by $A = 1 - R = 1 - r^2$. The reflection amplitude r can be calculated using equations derived for the reflection at uniaxial birefringent materials by Lekner, 1991. The regarded polarization state was linear polarization oriented parallel and perpendicular to the plane of incidence. It is noted, that a polarization perpendicular to the plane of incidence is parallel to the carbon fiber axis. Accordingly, a polarization parallel to the plane of incidence is perpendicular to the carbon fiber axis.

As matrix material we considered the thermoset material MGS RIM 135 for the calculations. The experimental measurements were performed with CFRP with the thermoset material RTM 6 as matrix material. The volume content of matrix material was about 40 % for the calculations as well as for the experimental measurements. Regarding the thermoset material MGS RIM 135, its absorption coefficient has been measured to be always less than 0.1 mm^{-1} for wavelengths between 400 nm and 1400 nm. This gives an absorbed intensity in the matrix layer on top of the carbon fibers, which has usually a thickness of about $50 \text{ }\mu\text{m}$, of about 0.5% which is therefore neglected. According to the specifications of the manufacturer, the refractive index of the matrix material RIM 135 is $n_M \approx 1.55$. This needs to be considered when calculating the reflection amplitudes of the carbon fibers when surrounded by matrix material as it is the case for CFRP. For non-infiltrated fabrics of carbon fibers, the carbon fibers are surrounded by air with a refractive index of 1.

3. Absorptance of carbon fibers and CFRP derived from the theoretical model

3.1 Absorptance of a single carbon fiber

Carbon fibers have a nearly circular cross section. The diameter of carbon fibers is usually in the range of a few micrometers. We regarded in this study carbon fibers with a diameter of $8 \text{ }\mu\text{m}$. As shown in Fig. 1 (b), light at all angles of incidence ranging from -90° to $+90^\circ$ is absorbed by the cylindrical fibers along their irradiated circumference. As the diameter of the laser beam is typically much larger than $10 \text{ }\mu\text{m}$, the fibers absorb radiation simultaneously along the whole circumference. To calculate the absorptance along the circumference, we approximated the incoming laser beam by a multitude of parallel light rays.

In Fig. 2 (a) the local absorptivity on the surface of a carbon fiber for a wavelength of 515 nm is shown. For a polarization parallel to the carbon fiber axis the highest absorptivity is found in the center of the surface facing the incident beam. The absorptivity in the center of the carbon fiber surface is the same for parallel and perpendicular polarization. However, for a polarization perpendicular to the carbon fiber axis, the absorptivity increases towards the edges of the irradiated surface and has its maximum near the edge of the irradiated carbon fiber. The mean absorptance A_{av} of one single carbon fiber is obtained by averaging the absorptivity over the whole surface of the carbon fiber. The result is shown in Fig. 2 (b) for wavelengths

between 100 nm and 11000 nm. Three wavelengths relevant for materials processing (355 nm, 532 nm and 1064 nm) have been marked in the figure. The absorptance is always higher for a polarization perpendicular to the carbon fiber axis than for a polarization parallel to the carbon fiber axis. For wavelengths in the ultra-violet (UV) and visible wavelengths (VIS), the absorptance is very similar with a slight decrease towards the near infrared (NIR). The absorptance decreases significantly for a wavelength of 10.6 μm which is typical for CO_2 laser radiation.

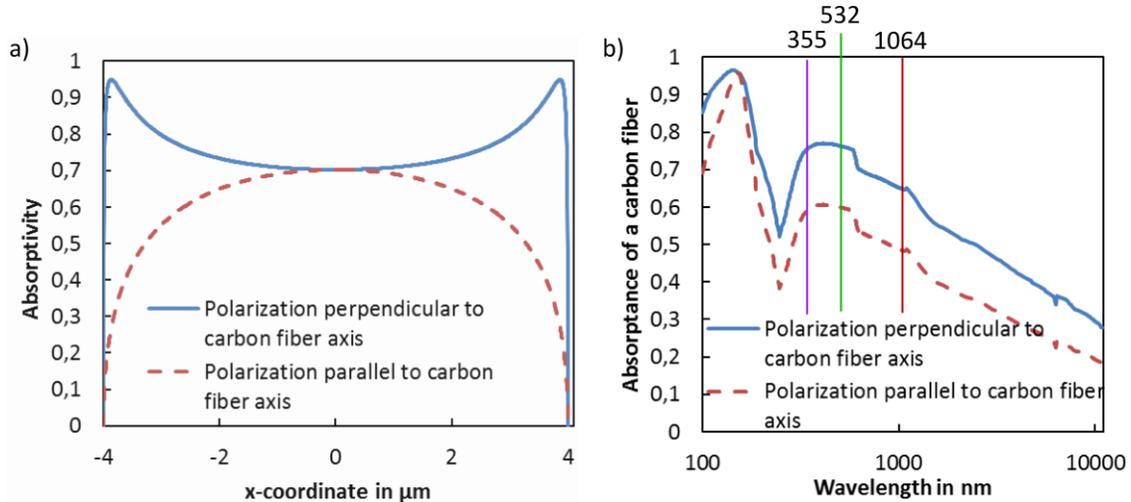


Fig. 2. (a) Absorptivity on the surface of a carbon fiber with a radius of 4 μm for a polarization of the light parallel and perpendicular to the carbon fiber axis, see Freitag et al., 2014. The incident light rays have a wavelength of 515 nm. (b) Average absorptance of a single carbon fiber as a function of the wavelength, see Freitag et al., 2014. The surrounding medium is air.

3.2 Absorptance for multiple carbon fibers

Typically not only one single carbon fiber is being processed but fabrics of several carbon fibers. To calculate the absorptance of laser radiation in multiple carbon fibers, the multiple reflections of the incident radiation between the carbon fibers have to be taken into account. To calculate the absorptance by means of raytracing we consider a model arrangement of carbon fibers which is shown in Fig. 3 (a). The diameter of the carbon fibers is 8 μm . The volume content of carbon fibers in this arrangement is 60% which gives a distance d between the carbon fibers of 1.152 μm . Only light rays between $x = 0$ and $x = r+d/2$ are being considered due to the symmetry of the arrangement. At each reflection of a light ray on a carbon fiber surface, the absorptivity and reflectivity is calculated. The absorptance of one light ray is calculated by summarizing the absorbed shares at each single reflection. Light rays propagating into the material after multiple reflections on the three considered fibers are assumed to be completely absorbed. The other rays are followed until they are reflected back from the assumed sample surface (into the half space $y > 0$). The total absorptance of carbon fibers in air is calculated by averaging the absorptance of multiple equidistant light rays between $x = 0$ and $x = r+d/2$. The result is shown in Fig. 3 (b) for wavelengths between 100 nm and 11000 nm. Compared to the result for a single carbon fiber shown in Fig. 2 (b), the absorptance is increased due to the multiple reflections. The difference between the absorptance for a parallel and perpendicular polarization relative to the carbon fiber axis is less pronounced in the case of multiple carbon fibers (Fig. 3 (b)) than for a single carbon fiber (Fig. 2 (b)).

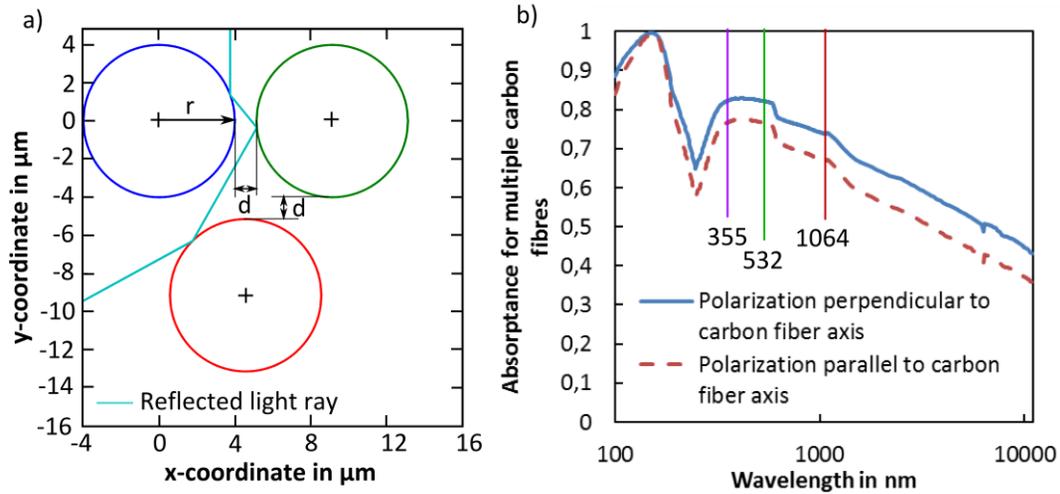


Fig. 3. (a) Exemplary arrangement of carbon fibers used to calculate the absorbance of multiple carbon fibers using ray tracing, see Freitag et al., 2014. The multiple reflections of a light ray incident at $x = 3.764 \mu\text{m}$ is shown. (b) Average absorbance of multiple carbon fibers as a function of the wavelength, see Freitag et al., 2014. The surrounding medium is air.

3.3 Absorbance of a single carbon fiber surrounded by RIM 135 and CFRP

The matrix material surrounding the carbon fibers has a twofold impact on the absorption behavior of the material. First, the incident laser light is partially reflected at the interface between air and matrix. With the known refractive index of RIM 135 of $n_M \approx 1.55$ the reflectivity of this interface amounts to 4.65%. The rest of the radiation is transmitted through the matrix material. This approximation applies for wavelengths between 400 nm and 1400 nm since the absorbed intensity in the matrix material can be neglected in this spectral range. Second, the higher refractive index as compared to air leads to an increase of the absorptivity on the carbon fibers at every reflection. Accordingly also the averaged total absorbance increases.

In Fig. 4 (a) the local absorptivity on the surface of a carbon fiber for a wavelength of 1064 nm is shown. For a polarization perpendicular to the carbon fiber axis, the birefringent properties of graphite result in a local minimum of the absorptivity near the edge of the carbon fiber. This behavior is only observed with the increased refractive index of the matrix material. This local minimum cannot be observed with air as the surrounding medium with a refractive index of 1. It gets more pronounced for longer wavelengths.

The resulting averaged total absorbance of CFRP with a RIM 135 matrix taking multiple reflections between the carbon fibers into account is shown in Fig. 4 (b) for wavelengths between 400 nm and 1400 nm. In this range of wavelengths the absorbed intensity in the matrix material can be neglected. Compared to the results in Fig. 3 (b) for multiple carbon fibers surrounded by air, the absorbance is increased for CFRP. The difference between the absorbance for polarizations parallel and perpendicular to the carbon fiber axis is reduced by the increased refractive index of the matrix. In general, the absorbance is very high (<70%) and the influence of the wavelength in this spectral range can probably be neglected for materials processing.

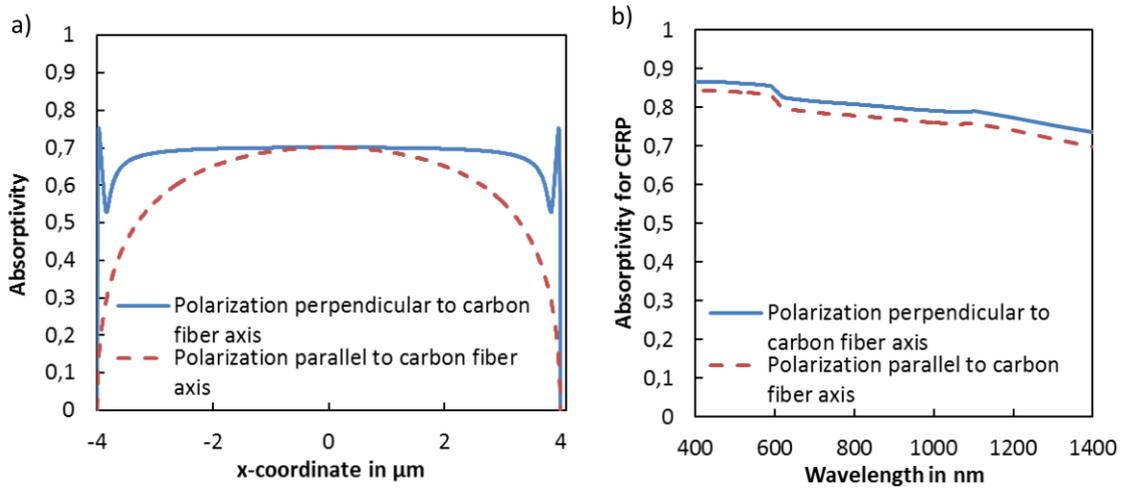


Fig. 4. (a) Absorptivity on the surface of a single carbon fiber with a radius of $4 \mu\text{m}$ surrounded by the matrix material RIM 135 with a refractive index of $n_M = 1.55$ for a polarization of the light parallel and perpendicular to the carbon fiber axis, see Freitag et al., 2014. The incident light rays have a wavelength of 1064 nm . (b) Average absorptance of CFRP as a function of the wavelength calculated with the presented model, see Freitag et al., 2014.

4. Experimental measurement of the absorptance of carbon fibers and CFRP

The absorptance of carbon fibers and CFRP was measured by means of reflectometry. By measuring the reflected share R of the incident radiation, the absorptance A can again be obtained from $A = 1 - R$ given that the transmitted share can be neglected. The samples used were a unidirectional fabric of carbon fibers and a CFRP sample. The carbon fibers were PAN-based carbon fibers from Tenax "Tenax-E HTS40". The matrix material was the thermoset material RTM 6. Since both matrix materials RTM 6 and RIM 135 are epoxy resins it can be assumed that the refractive index of RTM 6 is similar to the refractive index of RIM 135 for the applied laser wavelengths. The volume content of carbon fibers in the CFRP was about 60%. The thickness of the CFRP sample was about 2 mm, the transmission of laser radiation through the material can therefore be neglected.

Two laser systems with different wavelengths were used for the measurements. The first laser system emits at a wavelength of 532 nm and pulse durations of 8 ps . The second laser system emits radiation of a wavelength of 1047 nm and pulse durations of about 20 ns . The fluence of the incident laser beam on the sample surface was always kept smaller than the ablation threshold of the material. The polarization state of the laser radiation is linear.

A sketch of the experimental setup can be seen in Fig. 5. Due to the circular cross section of the carbon fibers, the material reflects light in every direction within the plane of incidence. This diffuse reflection behavior has been taken into account in the design of the setup. The incoming laser beam is guided towards the sample by two dielectric coated mirrors. A small percentage of the incident laser radiation transmits through each mirror. Behind the first mirror, a photodiode measures the transmitted share and gives an electric signal that correlates linear with the pulse energy of the incoming laser beam. The laser beam is focused by a lens with a focal length of 200 mm . The sample is positioned within an integrating sphere behind the focus to avoid ablation of the material. When the incoming radiation meets the sample, a certain share of the radiation is absorbed and the remaining share is reflected. The diffuse reflected share remains within the integrating sphere. A photodiode beside the integrating sphere measures the radiation within the

sphere and gives an electric signal that correlates linear to the diffuse reflected energy. The directly reflected radiation that leaves the integrating sphere is collimated by the lens in front of the integrating sphere and propagates along the same beam path as the incoming laser beam. The share of the reflected light that transmits through the mirror is being focused into another integrating sphere and is being measured by another photodiode.

With this setup it is possible to measure all directly and diffuse reflected shares of the incoming laser radiation and therefore draw conclusions on the absorbed share.

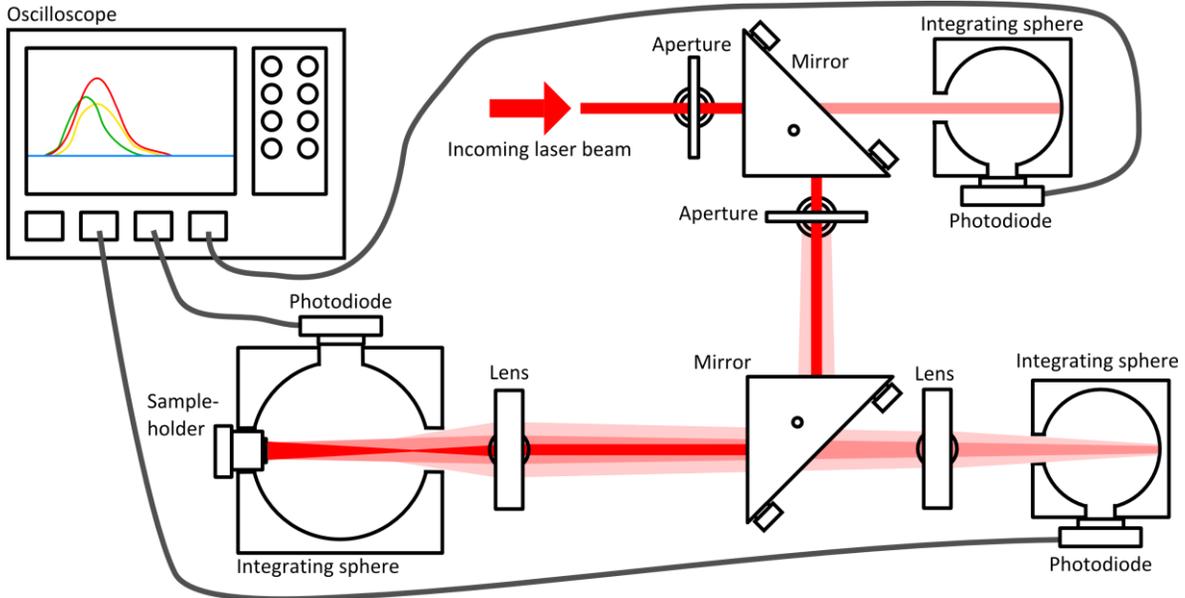


Fig. 5. Sketch of the experimental setup used to measure the reflected share of incident laser radiation from a sample surface.

The measured absorptance of radiation with wavelengths of 532 nm and 1047 nm in carbon fibers and CFRP can be seen in Fig. 6. The angle of incidence of the incoming laser beam on the sample surface was 90° . In Fig. 6 the angle Ω between the carbon fiber axis and orientation of the polarization was varied. An angle of 0° denotes a parallel polarization to the carbon fiber axis, an angle of 90° denotes a perpendicular polarization to the carbon fiber axis. There is a good agreement between the absorptance calculated with the model described in chapter 3 and the measured results. The absorptance is higher in CFRP than in carbon fiber fabrics. For longer wavelengths the absorptance is decreased. For a polarization perpendicular to the carbon fiber axis the absorptance is higher compared to a polarization parallel to the carbon fiber axis.

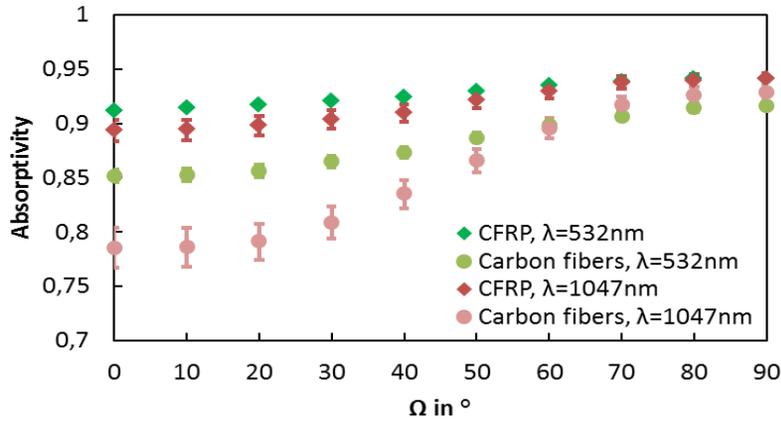


Fig. 6. Measured absorptance of light with a wavelength of 532 nm and 1047 nm in carbon fiber fabrics and CFRP with RTM 6 as matrix material as a function of the angle Ω between the carbon fiber axis and the orientation of the polarization. The polarization is parallel to the carbon fiber axis for $\Omega = 0^\circ$ and perpendicular for $\Omega = 90^\circ$.

A comparison between the calculated values and the experimentally measured values of the absorptance is given in Table 1. The experimentally measured values are up to 20% larger compared to the calculations. Possible reasons for the observed deviation are:

- Approximation of the optical properties of turbostratic carbon with the optical properties of graphite.
- The model assumes an ideal orientation of the carbon layers in the carbon fibers.
- The model assumes a smooth surface of the carbon fibers.
- The model regards only one arrangement of carbon fibers.
- In reality CFRP and carbon fiber fabrics show a lot of irregularities like additional polyester threads, roughness of the CFRP sample surface, irregular arrangement of carbon fibers, varying thickness of the top matrix layer and so on which may increase the absorptance
- Inaccuracy of the experimental measurement by e.g. light that leaves the sample holding integrating sphere and is not detected by the photodiode that measures the directly reflected share, inaccuracy in the calibration of the setup, ...

The proposed model gives a conservative value for the absorptance of laser radiation in carbon fiber fabrics and CFRP. It predicts very well the influence of the wavelength and the orientation of the polarization relative to the carbon fiber axis on the absorptance.

Table 1. Comparison of the experimental measured and calculated values for the absorptivity of radiation in carbon fiber fabrics and CFRP for wavelengths of 532 nm and 1047 nm.

	532 nm				1047 nm			
	Carbon fibers		CFRP		Carbon fibers		CFRP	
	Perp.	Parallel	Perp.	Parallel	Perp.	Parallel	Perp.	Parallel
Model	0.82	0.77	0.86	0.84	0.74	0.67	0.79	0.76
Experiment	0.92	0.85	0.94	0.91	0.93	0.79	0.94	0.89

5. Conclusion

A model based on raytracing was presented which allows the calculation of the absorptance of carbon fibers and CFRP. This model is based on the assumption, that the optical properties of turbostratic carbon can be approximated by the optical properties of graphite. Since graphite is a uniaxial crystal with birefringent properties, equations derived by Lekner, 1991, were used to calculate the reflection amplitudes. Due to the circular cross section of the carbon fibers, the absorptance was calculated as an average over multiple, equidistant, parallel light rays. In the UV, VIS and NIR the absorptance was found to be larger than 65% for multiple carbon fibers and larger than 75% for CFRP. For a wavelength of 10.6 μm (CO_2 -Lasers), the absorptance drops to less than 40% for multiple carbon fibers. The absorptance for light polarized perpendicular to the carbon fibers was shown to be larger than for light polarized parallel to the fibers. The additional matrix material in the case of CFRP leads to a reduction of this difference.

Additional experimental measurements of the absorptance of laser radiation with wavelengths of 532 nm and 1047 nm in carbon fibers and CFRP confirm the above findings. However, the absolute values of the measured absorptance are up to 20% larger compared to the calculations. The theoretical model gives a conservative value for the absorptance whilst the statements made regarding the influence of different polarization orientations and wavelengths on the absorptance were confirmed.

Acknowledgements

The authors would like to thank the Graduate School of Excellence advanced Manufacturing Engineering GSaME of the University of Stuttgart for the funding of this work and the Institute for Laser Technology in Medicine and Measurement Technique (ILM) of the Ulm University for the measurement of the absorption coefficient of the matrix material RIM135.

References

- Barnett, F. R., Norr, M. K., 1976. A three-dimensional structural model for a high modulus pan-based carbon fiber, *Composites* 7(2), pp. 93-99.
- Borghesi, A., Guizzetti, G., 1991. Graphite (C), in "Handbook of Optical Constants of Solids II" E. D. Palik, Editor, Academic Press, San Diego, p. 449.
- Djurisic, A. B., Li, E. H., 1999. Optical properties of graphite, *Journal of Applied Physics* 85(10), pp. 7404-7410.
- Freitag, C., Weber, R., Graf, T., 2014. Polarization dependence of laser interaction with carbon fibers and CFRP, *Optics Express* 22(2), pp. 1474-1479.
- Hamada, T., Nishida, T., Sajiki, Y., Matsumoto, M., 1987. Structures and physical properties of carbon fibers from coal tar mesophase pitch, *Journal of Materials Research* 2(6), pp. 850-857.
- Hoffman, W. P., Hurley, W. C., Liu, P. M., Owens, T. W., 1991. The surface topography of non-shear treated pitch and PAN carbon fibers as viewed by the STM, *Journal of Materials Research* 6(8), pp. 1685-1694.
- Lekner, J., 1991. Reflection and refraction by uniaxial crystals, *Journal of Physics: Condensed Matter* 3(32), pp. 6121-6133.