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Determination of the Local Thermal Diffusivity of Inhomogeneous Samples by a Modified Laser-Flash Method[1]

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The local thermal diffusivity is of special interest for quality control of materials grown by physical vapor transport. A typical specimen of these materials consists of single crystals with sizes up to 1 mm. The conventional laser-flash method delivers only an average value of the thermal diffusivity of these polycrystalline materials. A local sensitive measurement system is desirable to determine the thermal diffusivity of single grains with diameters of 100 μm and above. In this work a modification of a standard laser-flash apparatus is presented. The key feature is the position control of the specimen in the plane perpendicular to the laser beam and the IR-detection unit. The mechanical precision of the position control is better than 100 μm. The IR-detection unit consists of a MCT-detector, a polycrystalline IR-fiber, and a system to focus on the sample surface. To study the experimental potential of the modified laser-flash method, measurements of the local thermal diffusivity of a multiphase specimen with known microscopic thermal properties are presented. The obtained results are discussed with respect to the energy profile of the laser beam and the alignment of the IR-detection unit. It is shown that the thermal diffusivity of a small specimen area with a diameter of 2 mm can be determined with an uncertainty of ±5%. For a polycrystalline aluminum nitride (AlN) specimen with grain sizes of the order of 1 mm,

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a mean value for the thermal diffusivity of $(72.1 \pm 3.6)\,\mathrm{m^2 \cdot s^{-1}}$ at room temperature is determined. A possible local variation of the thermal diffusivity cannot yet be observed. An improvement of the resolution is in progress.

KEY WORDS: aluminum nitride; laser-flash method; local thermal diffusivity; microscopic specimen.

1. INTRODUCTION

Aluminum nitride (AlN) is a ceramic compound with interesting thermal and electrical properties for technical applications. Although it has a high thermal conductivity λ at room temperature of up to $319\,\mathrm{W \cdot m^{-1} \cdot K^{-1}}$ for single crystals, it has a high specific electrical resistance greater than $10^{11}\,\Omega \cdot \mathrm{m}$ for specimens with only a few impurities. Typical for technical applications are polycrystalline materials with a lower thermal conductivity due to the influence of the arbitrary grain orientation and size on the thermal transport properties [1]. One aim of actual investigations in material research is the preparation of polycrystalline AlN with large grain sizes and low oxygen content by physical vapor transport (PVT) [2]. The knowledge of the thermal conductivity and its possible variation within the resulting polycrystalline material is important for the quality control of the preparation process.

Based on thermal-conductivity measurements of specimens from different positions within a polycrystalline AlN boule performed in Ref. 2, in this work an extension of the well-known laser-flash experiment is presented to determine local values of the thermal diffusivity and to study effects of possible thermal inhomogeneities in the composition of specimens. The aim of this work was to modify an existing laser-flash apparatus [3] and to measure the local thermal diffusivity $a(x, y) = \lambda(x, y)\rho^{-1}(x, y)c_p^{-1}(x, y)$ by moving the specimen perpendicular to the optical axis defined by the laser beam and the detection unit. In this work the experimental setup is described and measurements on a demonstrator specimen made of stainless steel and silver as well as a polycrystalline AlN disk will be discussed.

2. EXPERIMENTAL SETUP

During a standard laser-flash experiment, the specimen is heated at the front side by a short laser pulse. The temperature response at the specimen rear side is detected by an infrared sensor. From the time-dependent temperature increase, the thermal diffusivity is derived by fitting the solution of the one-dimensional equation of heat transfer to the experimental

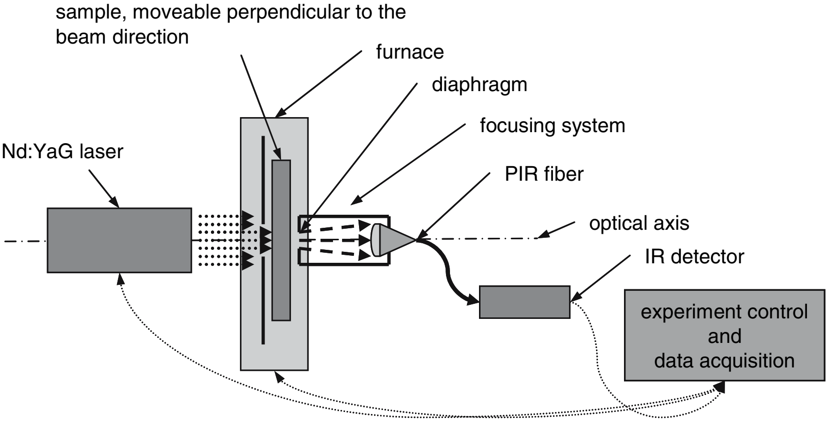

Fig. 1. Sketch of the modified laser-flash apparatus. Specimen is moveable perpendicular to the laser beam.

data. The influence of lateral heat losses is minimized by irradiating the total specimen surface and detecting only the center position of the specimen rear side.

In a standard laser-flash experiment the laser, specimen, and detection unit are mounted in fixed positions along the optical axis. In our experimental setup, the position of a tube furnace with the specimen holder is controlled by two stepper motors (Fig. 1). The mechanical resolution of the position control is about 100 μm. The position measurement is performed with a sliding calliper. The maximum specimen size is 25 mm in diameter.

The laser system consists of a Nd:YAG solid-state laser with a pulse energy of up to 20 J and a pulse length between 0.15 and 20 ms. The used laser pulse length is in the range typically between 0.15 and 0.30 ms. The wavelength of the laser beam is 1064 nm.

The detection unit consists of a liquid-nitrogen-cooled mercury–cadmium–telluride (MCT)-detector and a polycrystalline infrared (PIR)-fiber. The core/clad structure of the fiber consists of AgCl and AgBr, respectively. The diameter of the core is 0.9 mm, and the clad has a diameter of 1 mm. The usable wavelength range of the fiber is from 4 to 18 μm. For the locally resolved detection of infrared radiation, a diaphragm with a variable diameter is used.

The experiment is computer controlled, and the temperature increase as a function of time is recorded by the data acquisition system. For evaluation of the thermal diffusivity, several theoretical solutions can be used [4–6].

All specimens investigated in the framework of this study were coated with a thin layer of graphite (approx. 0.01 mm) with a high emissivity to improve absorption of the laser radiation and to enhance the emission of thermal radiation at the specimen rear side. The specimens were measured at room temperature.

3. SPECIMENS

3.1. Stainless Steel

The stainless-steel specimen has a diameter of 12.5 mm and a thickness of (1.017 ± 0.015) mm and is made of stainless steel of Type X 10 NiCrMoTiB 15 15 (Material No. 1.4970). Its thermal diffusivity was measured as a function of the specimen position perpendicular to the laser beam and as a function of the size of the detection area to study the influence of the energy profile of the laser beam and the detector alignment on the measurement results. The specimen was previously measured by the standard laser-flash technique for an intercomparison test between different laboratories [7]. The recommended value for the thermal diffusivity at a temperature of 20°C derived from these results on eight specimens of the same batch measured by eight independent laboratories is (3.50 ± 0.18) $mm^2 \cdot s^{-1}$.

3.2. Silver/Stainless Steel Demonstration Specimen

A demonstration specimen with a well-known inhomogeneity was prepared from a stainless steel disk (Material No. 1.2799) with a diameter of 12.5 mm and a thickness of (1.290 ± 0.017) mm. For this purpose a hole with a diameter of 2 mm was drilled into the center of the stainless steel disk and filled with pure silver. The specimen surface is depicted in Fig. 2.

The thermal diffusivity a of the stainless steel without the insert was measured by a standard laser-flash technique ($a = (11.0 \pm 0.6)\ mm^2 \cdot s^{-1}$). In Ref. 8 the thermal diffusivity of pure silver is given by $a = (174 \pm 5\ mm^2 \cdot s^{-1})$.

3.3. Polycrystalline Aluminum Nitride

To investigate a specimen with unknown local thermal properties, a polycrystalline AlN specimen was chosen. The specimen was made from a PVT process. General physical properties of the specimen were already investigated and described in Ref. 2. Figure 3 shows a picture of the specimen. Clearly visible are the grains of AlN with sizes up

Fig. 2. Demonstrator specimen made of stainless steel with a silver insert of 2 mm diameter. Diameter of the specimen is 12.5 mm.

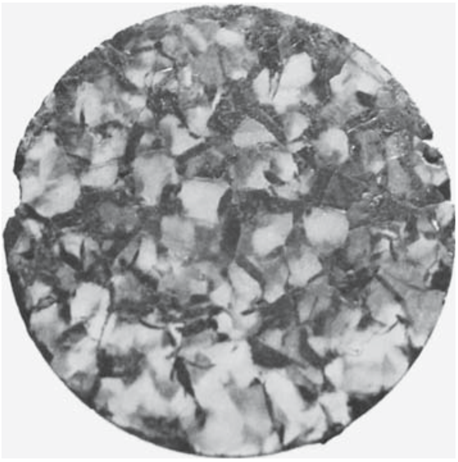

Fig. 3. Polycrystalline aluminum nitride specimen. Diameter of the disk is 12.5 mm.

to 1 mm. The measured density of the specimen is $3220\,\mathrm{kg \cdot m^{-3}}$, which is close to the theoretical bulk density of AlN of $3330\,\mathrm{kg \cdot m^{-3}}$. The derived porosity is about 3–4%. The thickness of the specimen is (0.640 ± 0.007) mm.

4. MEASUREMENT RESULTS AND DISCUSSION

4.1. Influence of Energy Profile and IR-Detection Alignment – Measurements on Homogeneous Stainless-Steel Specimen

The thermal diffusivity of a stainless-steel specimen (Steel No. 1.4970) was measured as a function of the diaphragm diameter (Fig. 1) in a standard laser-flash alignment to study the influence of the size of the detection area and consequently the influence of the signal strength on the measured results. The diaphragm diameter is nearly equal to the detected specimen surface. In this experiment the specimen surface was fully irradiated; the optical axis of the diaphragm was aligned with the center of the specimen surface. In Fig. 4 the values of the derived thermal diffusivity as a function of the diaphragm diameter are shown. It can be seen that, for diaphragm diameters above 2 mm, the derived thermal diffusivity values fit well with the literature data in the given uncertainty interval. For smaller diameters, e.g., 1 mm, the detector signal becomes weaker. Data evaluation then leads to significant lower values of the thermal diffusivity, which are without physical meaning.

Taking into account the aim of the experimental efforts to achieve a high local resolution, it was decided to use the fiber directly without lenses or a diaphragm for detection of the thermal radiation of the specimen

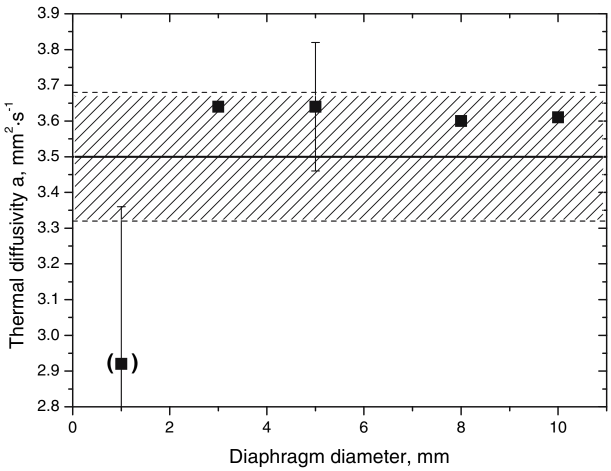

Fig. 4. Measured thermal diffusivity of the stainless-steel specimen 1.4970 as a function of diaphragm diameter. Hatched box indicates the recommended value for the thermal diffusivity of the material with the given uncertainty [7].

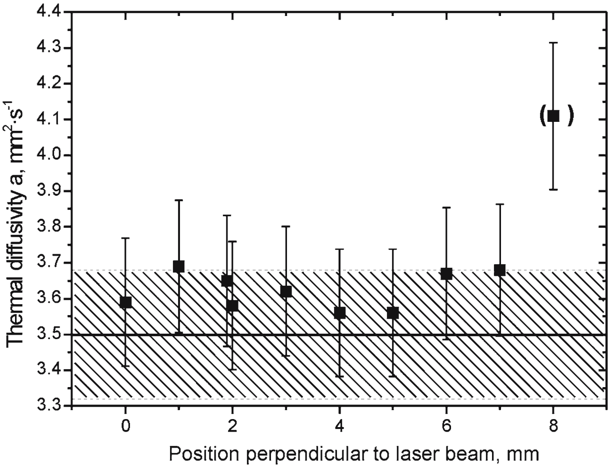

Fig. 5. Measured local thermal diffusivity of the stainless-steel specimen 1.4970 as a function of the specimen position perpendicular to the specimen axis. In the position "0 mm" the specimen is centered with respect to the optical axis given by the detection direction. The hatched box indicates the recommended value [7] for the thermal diffusivity of the material with its uncertainty.

surface. The effective numerical aperture of 0.25 for the PIR fiber and a distance of 1 mm between the fiber and specimen lead to a detection area with a diameter of about 1.5 mm.

The local thermal diffusivity of the homogeneous stainless-steel specimen as a function of the specimen position perpendicular to the laser beam was measured to study the effect of the detection position on experimental results. The derived values of the thermal diffusivity as a function of the specimen position perpendicular to the laser beam are shown in Fig. 5. At the position "0 mm" the specimen is nearly centered with respect to the optical axis given by the detection direction.

A possible reason for the slight variation of the measured thermal diffusivity for the positions between 0 and 7 mm could be the influence of inhomogenieties of the graphite layer and the small detection area of the IR fiber. Nevertheless, the result is in good agreement with literature values.

The increase of the derived value of the thermal diffusivity at a position of 8 mm is caused by the influence of a small part of the laser beam heating the end of the optical fiber directly, due to insufficent alignment of the diaphragm on the front side of the specimen. This effect can be seen

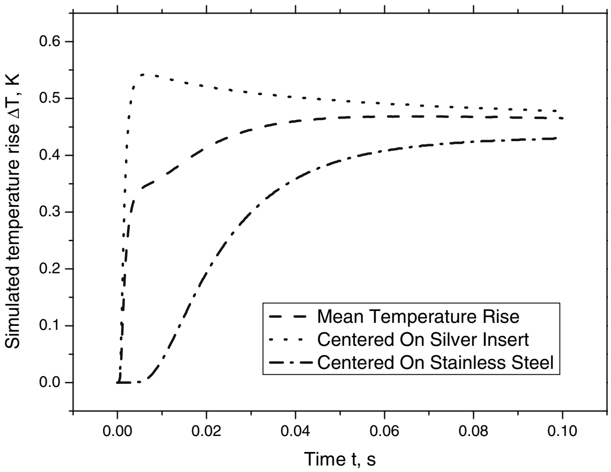

Fig. 6. Simulated temperature rise on the backside of the silver/stainless steel demonstrator specimen for two positions.

in the measurement curve as an instant temperature rise at the time of the laser pulse.

4.2. Measurements on Demonstrator Specimen

A two-dimensional numerical simulation of the laser-flash experiment on the silver/stainless steel demonstrator specimen was performed to study the possibility of the evaluation of local thermal-diffusivity values from the measurements. The simulations were performed with the program HEAT 2 [9]. For the simulation the thermal properties and the physical dimensions of the demonstrator specimen were used. Figure 6 shows the simulation results for the time-dependent temperature rise at two points on the rear specimen surface. The first point is for the center of the silver insert (dotted line). The second point lies 3 mm apart in the region of the stainless steel (dashed/dotted line). A mean time-dependent temperature rise was calculated from four simulated single signals from point sources (dashed line) because in the real experiment the detector signal is correlated with the mean temperature of a finite area.

The evaluation of the mean temperature in the time interval below 0.008 s following the model in Ref. 4 yields a value for the thermal diffusivity of $(168 \pm 17)\,\mathrm{mm^2 \cdot s^{-1}}$. This value is in good agreement with the value of $174\,\mathrm{mm^2 \cdot s^{-1}}$ for the thermal diffusivity of silver used as simulation input. The evaluation of the curve for times above 0.04 s yields a

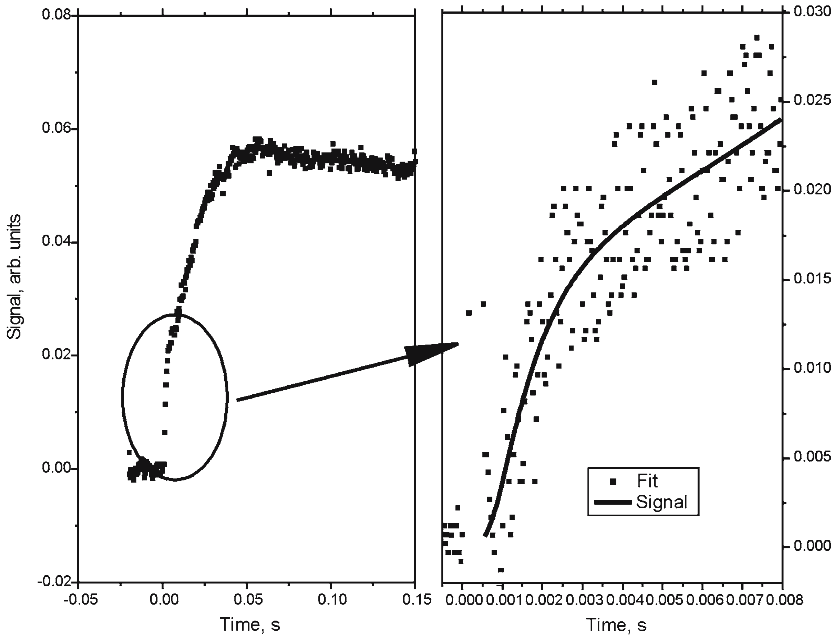

Fig. 7. Resulting detector signal for a laser-flash measurement on the silver/stainless steel demonstrator specimen. Detector was focused onto the silver spot. Two values for the thermal diffusivity can be derived by analyzing different time sections.

significantly lower value of ($17 \pm 3\,mm^2 \cdot s^{-1}$) for the thermal diffusivity. This has to be compared with the value expected for the stainless steel specimen ($11.0 \pm 0.6\,mm^2 \cdot s^{-1}$). It should be mentioned that in this case the uncertainty of the fit is relatively high for this time range: thus, only the order of magnitude can be derived for the diffusivity.

Figure 7 shows a typical laser-flash curve with the detection unit focusing on the region of the silver insert. The measurement curve can be split into two time ranges as expected from the simulation results. These are, first, the fast signal rise caused by the highly conducting silver, and second, the slower, but stronger, signal rise caused from the "slower" stainless steel. The thermal diffusivity in each of the two time ranges, extracted according to the model in Ref. 4, is $(163 \pm 16)\,mm^2 \cdot s^{-1}$ for the fast signal rise and $(16.4 \pm 41.6)\,mm^2 \cdot s^{-1}$ for the slower part. The continuous increase of the signal for the case of the fast rise is treated as heat gain comparable to the heat losses in Ref. 4. By moving the specimen off the centered position, the silver signal vanishes.

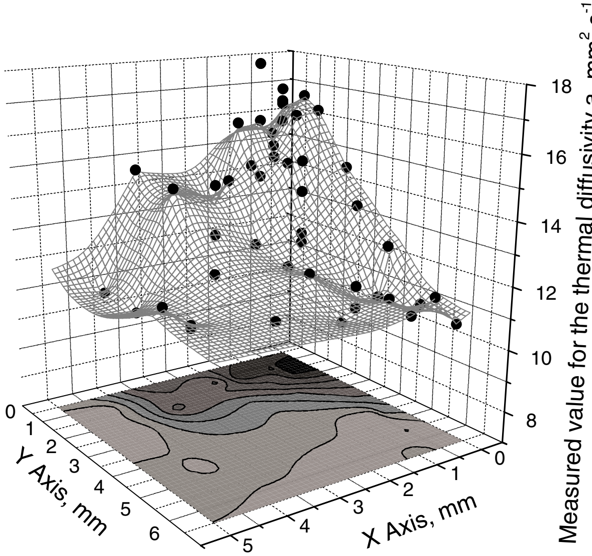

Fig. 8. Measured local thermal diffusivity of the silver/stainless steel demonstration specimen.

In subsequent experiments, only the long-time evaluation of the laser-flash curve was considered. In Fig. 8 the resulting values for the local thermal conductivity of the demonstrator specimen are shown as a function of the specimen position perpendicular to the laser beam. The silver insert is centered at $x, y = (0, 0)$. Clearly visible is the increase of the measured thermal diffusivity by more than 50% close to the silver insert. At 4 mm away from the silver insert, the measured value of the thermal diffusivity is equal to that of the pure stainless steel.

4.3. Measurements on Polycrystalline Aluminum Nitride

In Fig. 9 a typical detector signal as a function of time and the corresponding fit curve is shown for polycrystalline aluminum nitride (AlN). In contrast to the measurements on the demonstrator specimen, the analytical model for homogeneous materials fits well to the complete measurement curve. An influence of the smaller local differences in the thermal diffusivity of the polycrystalline AlN specimen on the measurement curve is not expected. Figure 10 shows the thermal diffusivity along the y-axis from the center of the disk-shaped specimen to the rim.

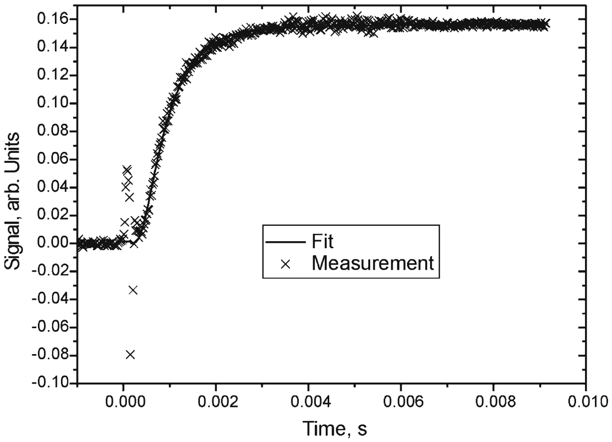

Fig. 9. Typical detector signal for a laser-flash measurement on the polycrystalline AlN specimen. Spikes in the measurement signal at about 0 s are caused by recharging of the capacitor of the laser pump lamp and are omitted in the data evaluation. Also depicted is the fit curve (solid line) to the signal according to the analytical models of Cowan [4] and Azumi and Takahashi [5].

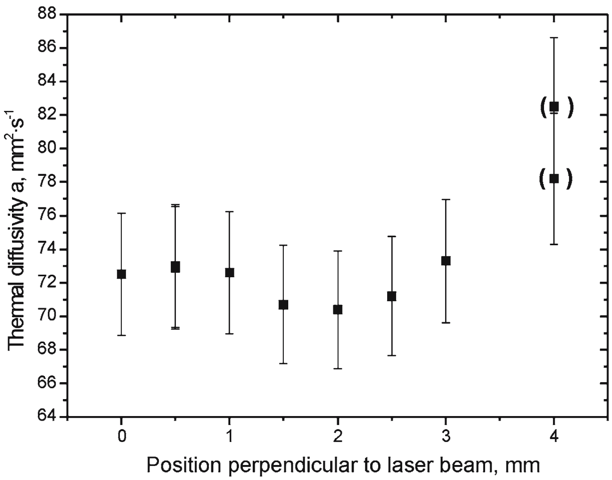

Fig. 10. Measured thermal diffusivity of the polycrystalline AlN specimen (12.5 mm diameter) as a function of the specimen position perpendicular to the laser beam. Increase at 4 mm is caused by direct radiation either transmitted through small holes at the grain boundaries or transmitted between specimen and specimen holder. Note that the zero is highly suppressed.

From thermal-conductivity measurements along the *c*-axis of high-purity single crystals, the upper limit for the thermal conductivity at 300 K of AlN was extrapolated to be $319\,W \cdot m^{-1} \cdot K^{-1}$ [10]. The corresponding maximum value of the thermal diffusivity can be calculated to be $134\,mm^2 \cdot s^{-1}$ at 300 K using a value for the specific heat of $738\,J \cdot kg^{-1} \cdot K^{-1}$ at 300 K [1] and a specimen density of $3220\,kg \cdot m^{-3}$. From Fig. 10 it can be seen that the single crystal value is about 80% higher than the mean value of $(72.1 \pm 3.6)\,mm^2 \cdot s^{-1}$ determined for the AlN specimen investigated in this work. A significant local variation of the thermal diffusivity could not be observed within the present optical resolution.

5. CONCLUSIONS

In this work, measurements of local thermal diffusivities within inhomogeneous specimens were performed. It can be shown that the thermal diffusivity of a small specimen area with a diameter of 2 mm can be deterimend with sufficient accuracy. Measurements on a thermally well-characterized stainless-steel specimen yields a thermal diffusivity value of $(3.6 \pm 0.1)\,mm^2 \cdot s^{-1}$, measured on different positions on the specimen surface, which is in good agreement with the literature value of $(3.50 \pm 0.18)\,mm^2 \cdot s^{-1}$.

Measurements on a stainless-steel specimen with an insert of silver show that for the case of inhomogeneous materials with high differences in the local thermal diffusivity it is possible to derive separate values for the thermal diffusivity by evaluating different time regimes of the measured signal. Also, it is possible to determine the influence of the insert on the measured thermal diffusivity of the adjacent regions.

For the polycrystalline AlN specimen a mean value for the thermal diffusivity of $(72.1 \pm 3.6)\,mm^2 \cdot s^{-1}$ at room temperature was determined. A local variation of the thermal diffusivity could not be observed within the present optical resolution.

Future work has to focus mainly on (i) an improvement of the optical resolution of the apparatus, (ii) three-dimensional numerical simulation of the experiment to study the effects of inhomogenieties in materials and their complex interactions on the temperature response of the specimen, and (iii) development of an automatical data evaluation routine.

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