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ARTICLE *in* INTERNATIONAL JOURNAL OF THERMOPHYSICS · SEPTEMBER 2007

Impact Factor: 0.96 · DOI: 10.1007/s10765-007-0161-7

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DETERMINATION OF THE LOCAL THERMAL DIFFUSIVITY OF INHOMOGENEOUS SAMPLES BY A MODIFIED LASERFLASH METHOD

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ABSTRACT

The local thermal diffusivity is of special interest for quality control of materials grown by physical vapour transport. A typical sample of theses materials consists of single crystals with sizes up to 1 mm. The conventional laserflash 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 micron and above. In this work a modification of a standard laserflash apparatus is presented. Key feature is the position control of the sample in the plane perpendicular to the laser beam and the IR-detection unit. The mechanical precision of the position control is better than 100 micron. 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 laserflash method, measurements of the local thermal diffusivity of a multiphase sample 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 sample area with a diameter of 2 mm can be determined with an accuracy of ±5%. For a polycrystalline AlN sample with grain sizes of the order of 1 mm a mean value for the thermal diffusivity of $(72.1 \pm 3.6)\ m^2s^{-1}$ at room temperature is determined. A possible local variation of the thermal diffusivity can not yet be observed. An improvement of the resolution is under progress.

KEY WORDS: laserflash; thermal diffusivity; aluminum nitride

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 $W(mK)^{-1}$ for single crystals, it has a high specific electrical resistance greater than $10^{11}\ \Omega m$ for samples with only few impurities. Typical for technical applications are polycrystalline materials with lower thermal conductivity due to the influence of the arbitrary grain orientation and the grain 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 vapour 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 samples from different positions within a polycrystalline AlN boule performed in [2], in this work an extension of the well known laserflash experiment is presented to determine local values of the thermal diffusivity and to study effects of possible thermal inhomogeneities in the composition of samples. The aim of this work was to modify an existing laserflash apparatus [3] and to measure the local thermal diffusivity $a(x,y) = \lambda(x,y) \cdot \left(\rho(x,y) c_p(x,y)\right)^{-1}$ by moving the sample perpendicular to the optical axis defined by the laser beam and the detection unit. In this work the experimental setup is described and first measurements on a demonstrator sample made of stainless steel and silver as well as a polycrystalline AlN disk will be discussed.

2. EXPERIMENTAL SETUP

During a standard laserflash experiment the sample is heated at the front side by a short laser pulse. The temperature response at the sample 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 data. The influence of lateral heat losses is minimized by irradiating the total sample surface and detecting only the centre position of the sample rear side.

In a standard laserflash experiment the laser, sample and detection unit are mounted in fixed positions along the optical axis. In our experimental setup the position of a tube furnace with the sample holder is controlled by two stepper motors (Figure 1). The mechanical resolution of the position control is about 100 micron. The position measurement is performed with a sliding calliper. The maximum sample 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 ms and 20 ms. The used laser pulse length is in the range typically between 0.15 ms 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)-fibre. The core/clad structure of the fibre 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 fibre is from 4 µm to 18 µm. For the locally resolved detection of infrared radiation a diaphragm with variable diameter is used.

The experiment is computer controlled and the temperature increase as function of time is recorded by the data acquisition system. For the evaluation of the thermal diffusivity several theoretical solutions can be used [4] [5] [6].

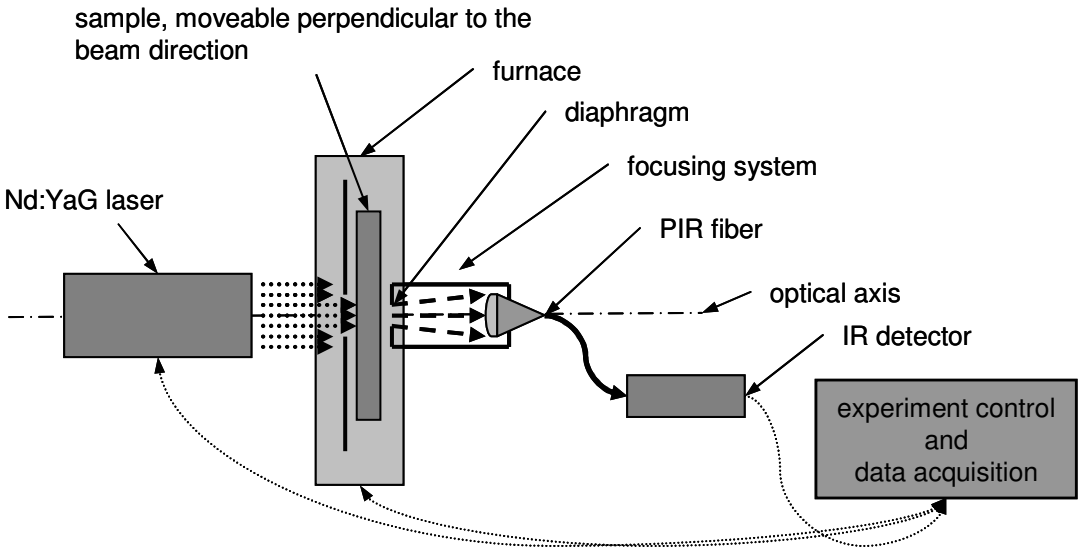

Figure 1: *Sketch of the modified laserflash apparatus. The sample is moveable perpendicular to the laser beam.*

All samples 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 radation and to enhance the emission of thermal radation at the sample rear side. The samples were measured at room temperature.

3. SAMPLES

3.1. Stainless Steel

The stainless steel sample 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 sample 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 sample was previous measured by standard laserflash 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 samples of the same batch measured by eight independent laboratories is (3.50 ± 0.18) mm^2s^{-1} .

3.2. Silver/Stainless Steel Demonstration Sample

A demonstration sample 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. As inhomogeneity served a hole with a diameter of 2 mm near the centre of the stainless steel disk, witch was filled with a wire of pure silver. The sample surface is depicted in Figure 2.

Figure 2: *Demonstrator sample made of stainless steel with a silver insert of 2 mm diameter. The diameter of the sample is 12.5 mm.*

The thermal diffusivity a of the stainless steel without the insert was measured by standard laserflash technique (a = (11.0 ± 0.6) mm^2s^{-1}). In [8] the thermal diffusivity of pure silver is given by a = (174 ± 5) mm^2s^{-1}.

3.3. Polycrystalline Aluminum Nitride

To investigate a sample with unknown local thermal properties, a polycrystalline AlN sample was chosen. The sample was made from a PVT process. General physical properties of the sample were already investigated and described in [2]. Figure 3 shows a picture of the sample. Clearly visible are the grains of AlN with sizes up to 1 mm. The measured density of the sample is 3220 kgm^{-3}, which is close to the theoretical bulk density of AlN of 3330 kgm^{-3}. The derived porosity is about 3 to 4%. The thickness of the sample is (0.640 ± 0.007) mm.

Figure 3: *Polycristalline alluminum nitride sample. The diameter of the disk is 12.5 mm.*

4. MEASUREMENT RESULTS AND DISCUSSION

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

The thermal diffusivity of the stainless steel sample (steel no. 1.4970) was measured as a function of the diaphragm (Figure 1) diameter in a standard laserflash alignment to study the influence of the size of the detection area and consequently the influence of the signal strength on the measuring results. The diaphragm diameter is nearly equal to the detected sample surface. In this experiment the sample surface was fully irradiated; the optical axis of the diaphragm was aligned with the centre of the sample surface. In Figure 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 diameter, 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.

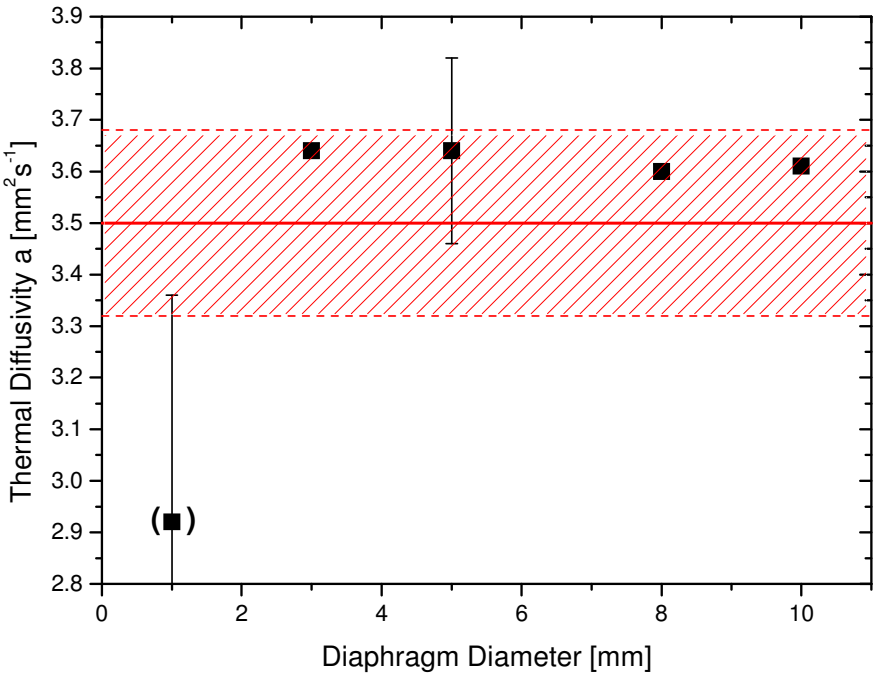

Figure 4: *Measured thermal diffusivity of the stainless steel sample 1.4970 as function of diaphragm diameter. The hatched box indicates the recommended value for the thermal diffusivity of the material with the given uncertainty [7].*

Having in mind the aim of the experimental efforts to receive a high local resolution it was decided to use the fibre directly without lenses or diaphragm for the detection of the thermal radiation of the sample surface. The effective numerical aperture of 0.25 for the PIR fibre and a distance of 1 mm between fibre and sample lead to a detection area with a diameter of about 1.5 mm.

The local thermal diffusivity of the homogeneous stainless steel sample as a function of the sample 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 sample position perpendicular to the laser beam are shown in Figure 5. In the position "0 mm" the sample is nearly centered with respect to the optical axis given by the detection direction.

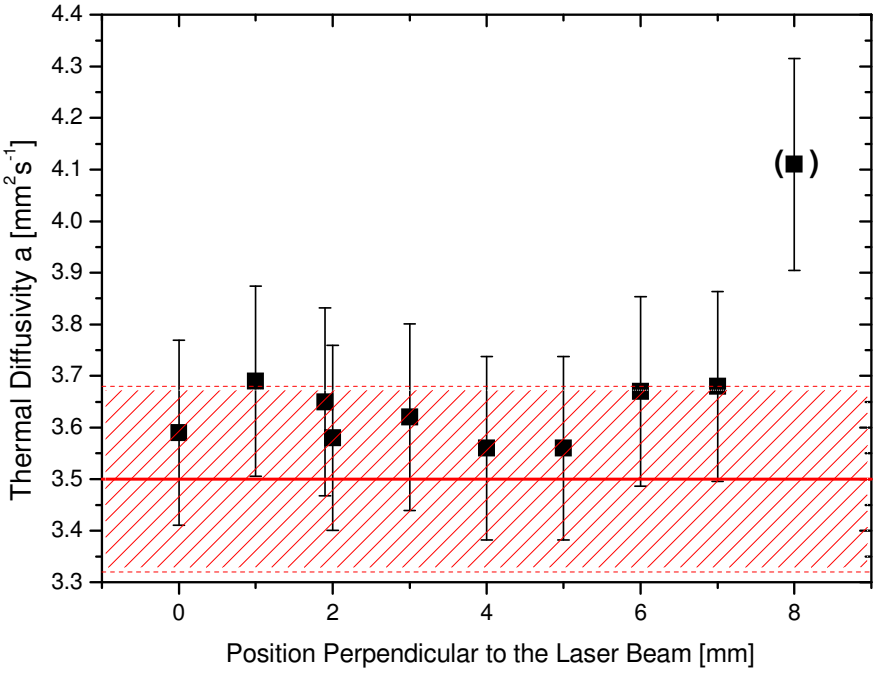

Figure 5**:** *Measured local thermal diffusivity of the stainless steel sample 1.4970 as a function of the sample position perpendicular to the sample axis. In the position "0 mm" the sample is centered with respect to the optical axis given by the detection direction. The hatched box indicates the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

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

The increase of the derived value of the thermal diffusivity at the position of 8 mm is caused by the influence of a small part of the laser beam heating the end of the optical fibre directly, due to insufficent alignment of the diaphragm on the front side of the sample. This effect can be seen in the measurement curve as instant temperature rise at the time of the laser pulse.

4.2. Measurements on Demonstrator Sample

A two dimensional numerical simulation of the laserflash experiment on the silver/stainless steel demonstrator sample 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 sample were used. Figure 6 shows the simulation results for the time dependent temperature rise at two points on the rear sample surface. The first point is for the centre of the silver insert (dotted line). The second point lies 3 mm apart in the region of the stainless steel (dash / dot 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 [4] yields a value for the thermal diffusivity of (168 ± 17) mm^2s^{-1}. This value is in good agreement with the value of 174 mm^2s^{-1} for the thermal diffusivity of silver used as simulation input. The evaluation of the curve for times above 0.04s yields a significantly lower value of (17 ± 3 mm^2s^{-1}) for the thermal diffusivity. This has to be compared with the value expected for the stainless steel sample ((11.0 ± 0.6) mm^2s^{-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.

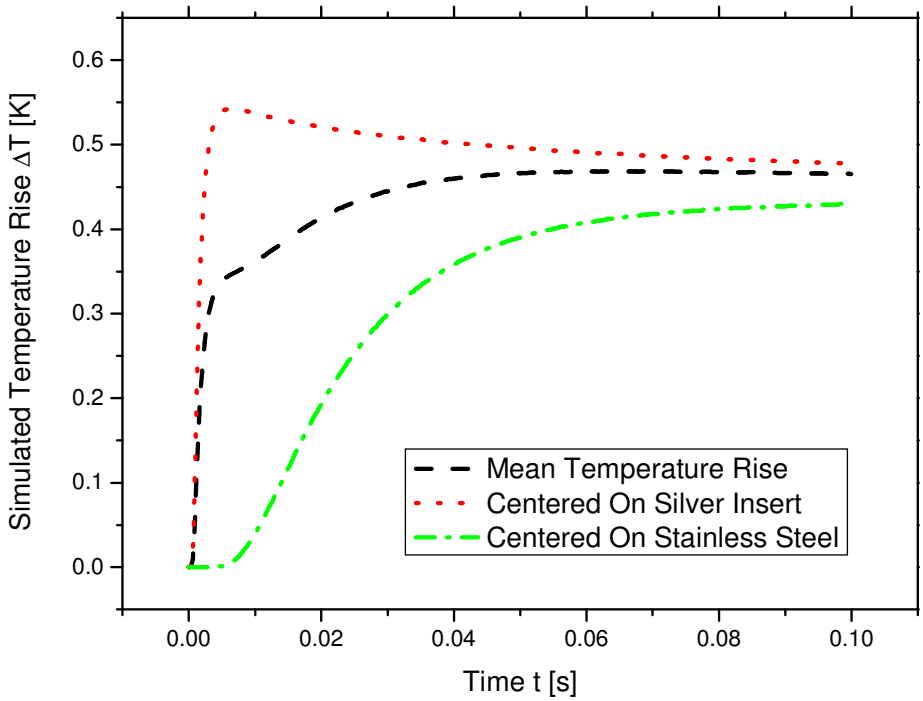

Figure 6: *Simulated temperature rise on the backside of the silver / stainless steel demonstrator sample for two positions.*

Figure 7 shows a typical laser flash curve with the detection unit focusing on to the region of the silver insert. The measurement curve can be split in two time ranges as it is expected from the simulation results. Firstly, the fast signal rise caused by the highly conducting silver, secondly the slower, but stronger, signal rise caused from the "slower" stainless steel. The thermal diffusivity in each of the two time ranges, extracted following the model in [4], is (163 ± 16) mm^2 s^{-1} for the fast rise and (16.4 ± 1.6) mm^2s^{-1} for the slower part. The continuous increase of the signal in the case of the fast rise is treated as heat gain comparable to the heat losses in [4].

By moving the sample off the centred position, the silver-signal vanishes.

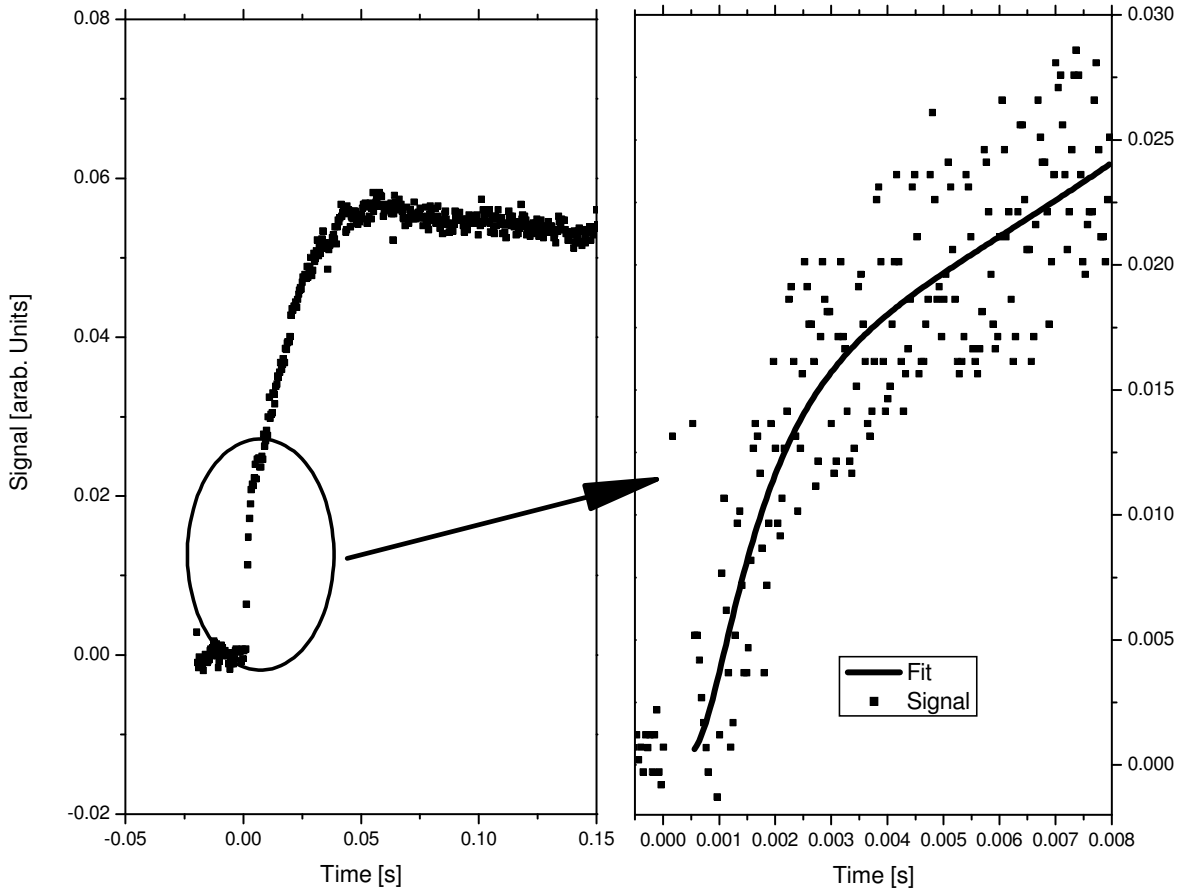

Figure 7: *Resulting detector signal for a laser flash measurement on the silver / stainless steel demonstrator sample. The detector was staring on to the silver spot. Two values for the thermal diffusivity can be derived by analyzing different time sections.*

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

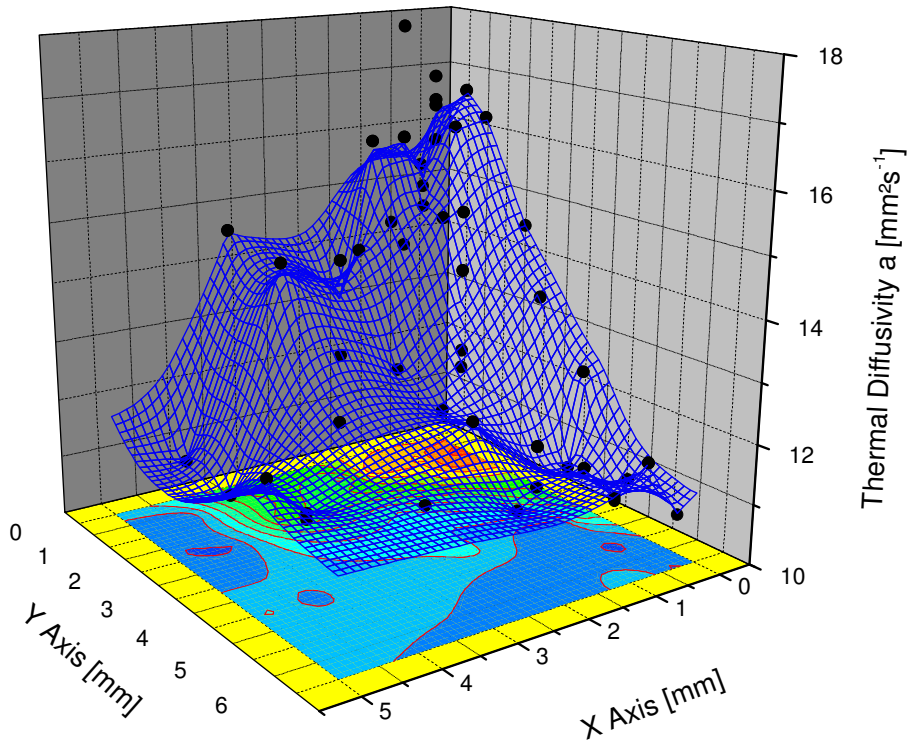

Figure 8: *Measured local thermal diffusivity of the silver/stainless steel demonstration sample.*

4.3. Measurement on Polycrystalline Aluminum Nitride

In Figure 9 a typical detector signal as function of time and the corresponding fit curve is shown for polycrystalline aluminium nitride (AlN). In contrast to the measurements on the demonstrator sample 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 sample on the measurement curve is not expected.

Figure 10 shows the thermal diffusivity along the y-axis from the centre of the disc-shaped sample to the rim.

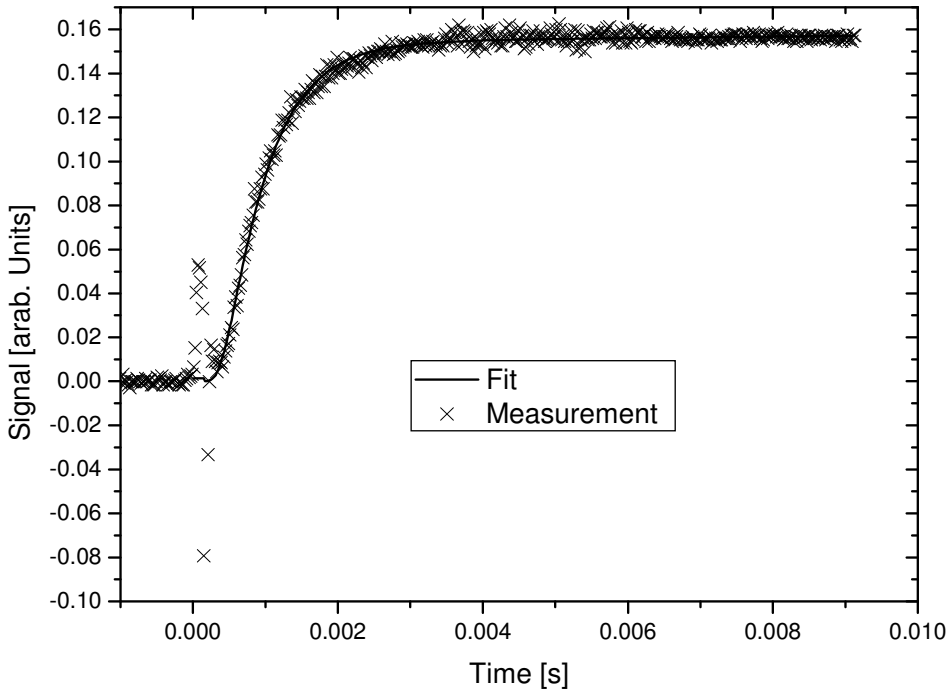

Figure 9: *Typical detector signal for a laserflash measurement on the polycrystalline AlN sample. The spikes in the measurement signal at about 0 s is caused by the recharging of the capacitor of the laser pump lamp and is omitted in the data evaluation. Also depicted is the fit curve (solid line) to the signal according to the analytical model in [2].*

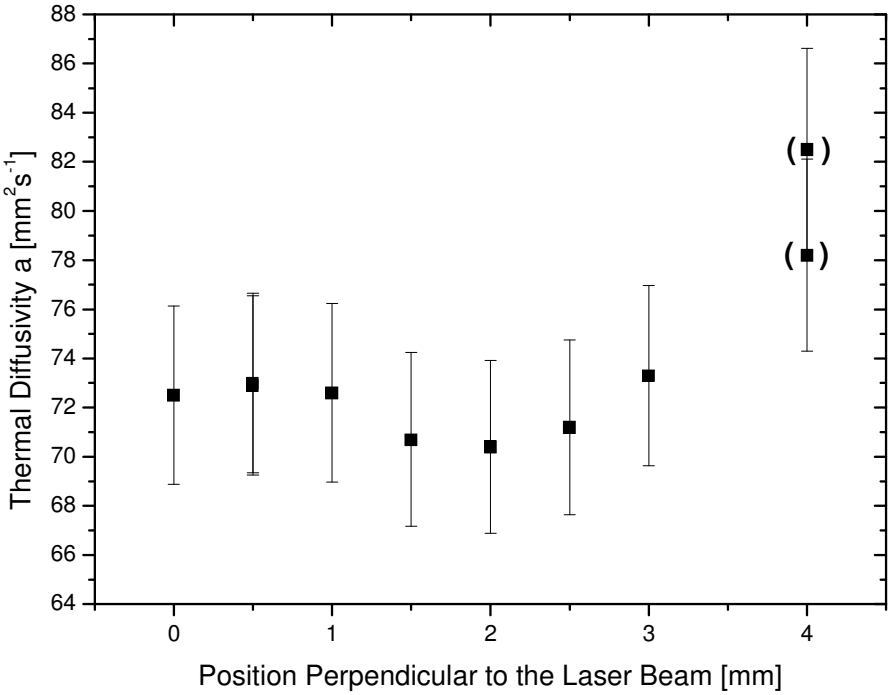

Figure 10 *Measured thermal diffusivity of the polycrystalline AlN-sample (12.5 mm diameter) as function of the sample position perpendicular to the laser-beam. The increase at 4 mm is caused by direct radiation either transmitted through small holes at the grain boundaries or transmitted between sample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

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

5. CONCLUSIONS

In this work first measurements of local thermal diffusivities within inhomogeneous samples were performed. It can be shown that the thermal diffusivity of a small sample area with a diameter of 2 mm can be deterimend sufficiently accurate. Measuments on a thermally well characterized stainless steel sample yields a thermal diffusivity value of (3.6 ± 0.1) mm^2s^{-1}, measured on different positions on the sample surface, which are in good agreement with the literature value of (3.50 ± 0.18) mm^2s^{-1}.

Measurements on a stainless steel sample with an insert of silver show that in 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 measuring 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 sample a mean value for the thermal diffusivity of (72.1 ± 3.6) m^2s^{-1} at room temperature was determined. A local variation of the thermal diffusivity could not yet be observed within the present optical resolution.

Future work has to mainly focus on (i) the 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 sample and (iii) to develop an automatical data evaluation routine.

ACKNOWLEDGMENTS

The authors would like to thank Matthias Bickermann at the Department of Materials Science 6, University of Erlangen-Nürnberg for providing the AlN samples and Matthias Reinhardt for his valuable help in the laboratory.

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